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**SCHOOL OF ARCHITECTURE, URBAN PLANNING AND CONSTRUCTION ENGINEERING**

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**FINAL THESIS**

# Environmental Alternatives for Stone Slurry Circularity: from Waste to Resource

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ABSTRACT

The historical use of natural stone products in construction dates back to ancient times and continues to play a crucial role in modern urban infrastructure and the construction of tall buildings. In recent decades, with the rapid growth of the global economy, the demand for natural stone-based products has surged, leading to significant expansion in the natural stone quarrying industry. However, this growth is accompanied by substantial challenges, including inefficiency, high production costs, and substantial waste generation and resource depletion during the extraction and processing phases.

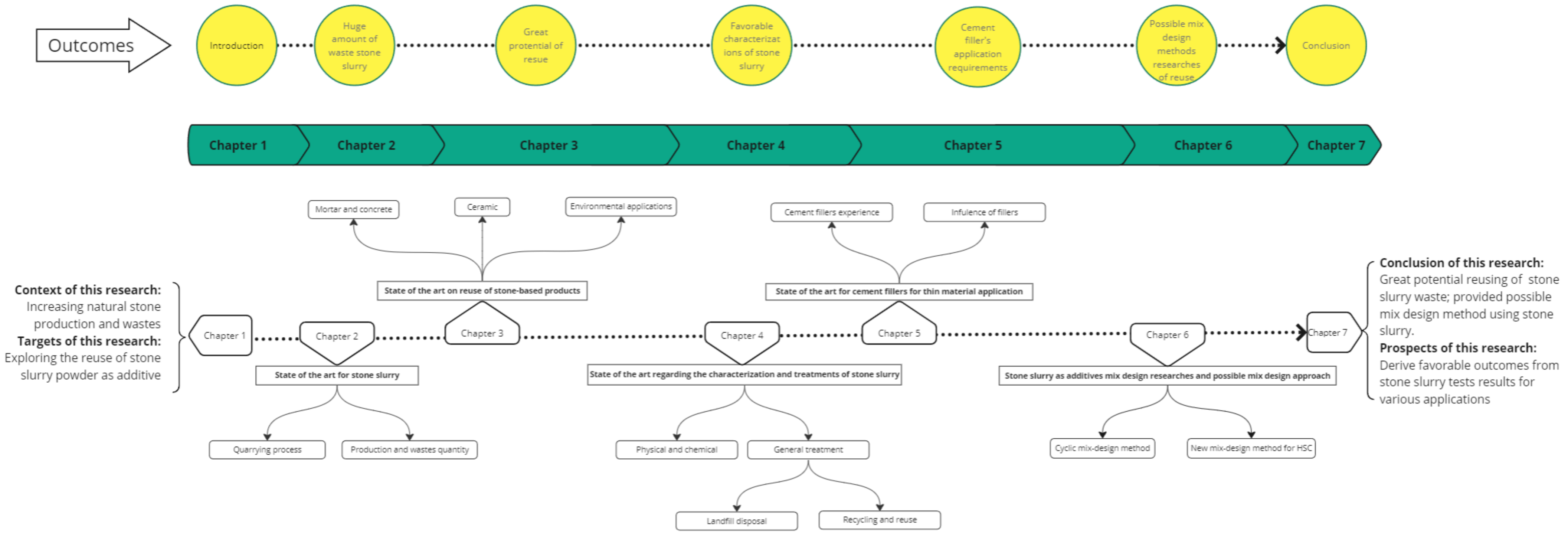
Globally, advancements in technology and the widespread adoption of innovative design concepts have transformed mining and quarrying methods for natural stones. Primitive techniques such as plug and feather have evolved into contemporary mechanized methods that employ diamond tools and specialized chemical agents, as well as hybrid methods that combine these techniques. Despite reaching peak production levels, the extraction and transformation process still generate substantial amounts of quarrying and processing wastes, encompassing solid/semi-solid debris, stone cracks, and stone slurry. This issue poses a serious challenge for many countries worldwide.

The extraction of natural stone and the production of stone floor tiles introduce a considerable amount of stone slurry, presenting a persistent challenge in waste disposal. Typically, stone debris mixed with slurry is landfilled, resulting in resource waste, land occupation, and potential harm to the natural environment. Consequently, the recycling of stone waste has garnered global attention.

This paper primarily focuses on the cutting-edge utilization of stone slurry as an additive in cement-based product mixtures. The objective is to comprehensively understand the basic formulation and process, exploring the optimal mix ratio of stone slurry powder to partially replace cement. This application specifically targets thin cement-tensile materials, enabling the investigation of the proportionate impact of stone slurry. The feasibility study highlights the potential contribution of utilizing stone slurry to enhance the sustainability of the natural stone quarrying industry.

Key words: Stone slurry; Cement-tensile products; Cement filler, Recycling, reduction of CO2 emission.

Workflow of this research



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1. Introduction

Derived from the Earth through quarrying, collectively termed Natural Stone, these products have functioned as construction materials and embellishments for countless centuries. Employed in construction projects and ornamental applications, this category includes marble, granite, limestone, travertine, quartzite, sandstone and various others. Several of these natural stone materials, including granite and marble, exhibit notable resistance to damage, making them a commendable investment for both exterior and interior home renovation endeavors spanning numerous years. (refer to: https://www.tilesdirect.net/natural-stone-faq/).

The connection between the production and utilization of natural stone and various global sectors, particularly construction, has consistently exhibited an upward trajectory since ancient times. Natural stone, prized for its distinctive colour, design, and physical and mechanical attributes, is favoured as both building and facing stone. According to the XXXIII World Marble and Stones Report 2021[1], the quarry production in the global stone industry reached 155 million tons in 2020, marking a 0.32% increase compared to 154.5 million tons in 2019. The production in 2019 rose by 0.98% from 153 million tons in 2018, which, in turn, increased by 0.66% from 152 million tons in 2017. The quarry production of 2017 experienced a significant growth of 4.83% compared to 145 million tons in 2016, and the production in 2016 saw a 3.57% increase from 140 million tons in 2015. Notably, when examining the data from 1996, the quarry production in the global stone industry was a mere 46.5 million tons, reflecting only 30.00% of the production in 2020. This clearly indicates a substantial rise in the demand for natural stone quarrying from 1996 to 2017, with demand remaining relatively stable in the years from 2017 to 2020.

Besides, the applications of products derived from natural stone quarrying are predominantly seen in floors and paving, external wall cladding, steps, internal wall cladding, special works, civil external uses, memorial art, and various other applications. Among these, floors and paving use account for almost one-third of the total quarry production in 2020.

Fig 1-1: World quarry production refer to XXXIII World Marble and Stones Report 2021[1]

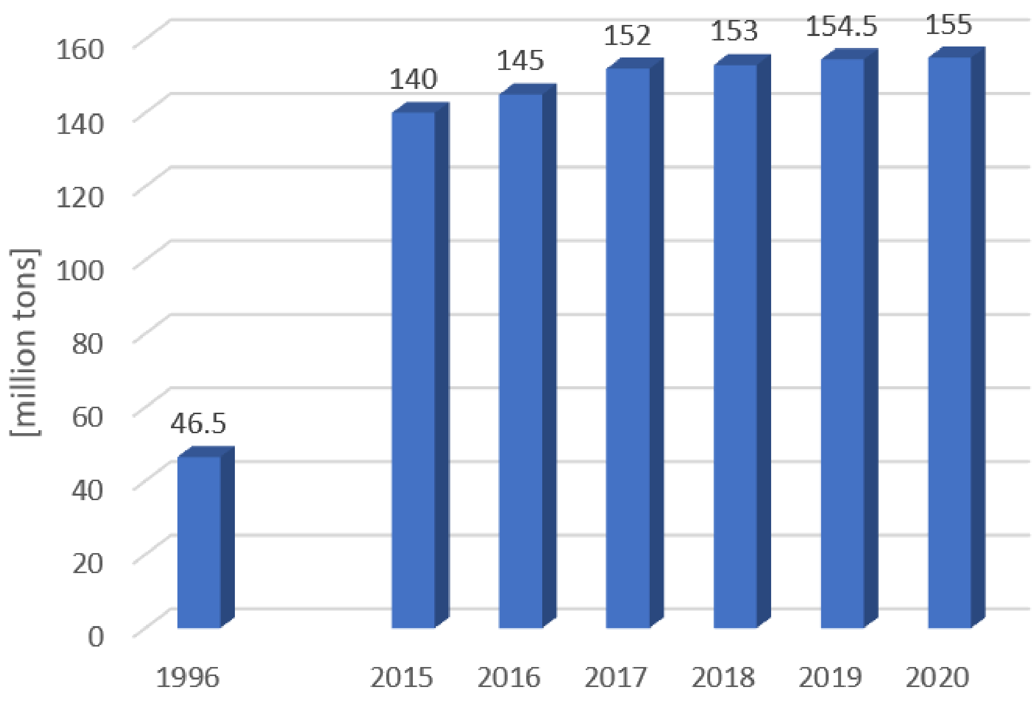
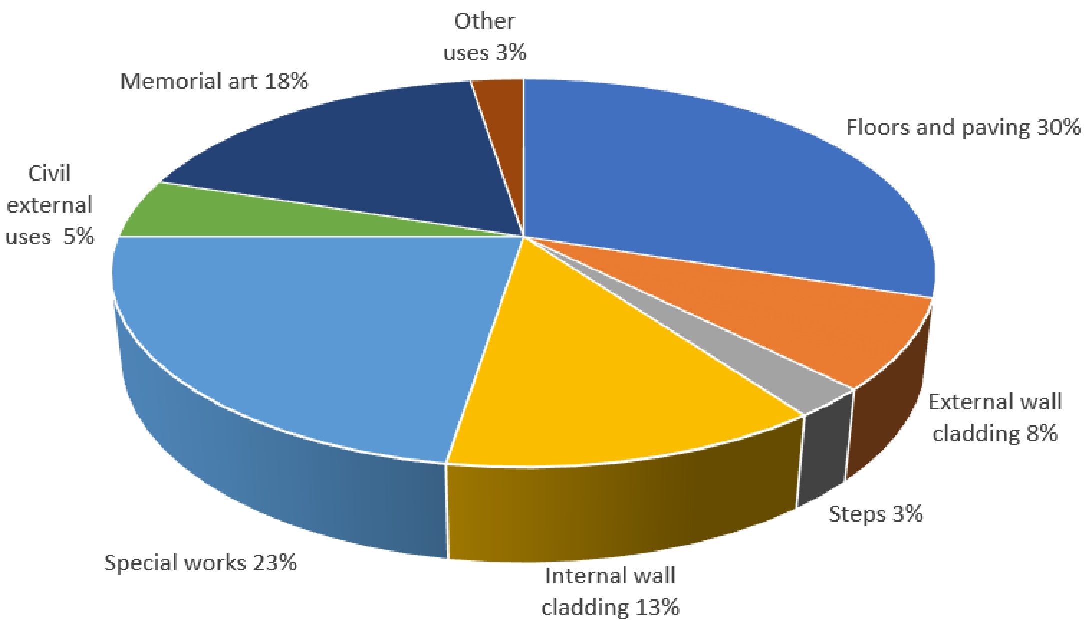


Fig 1-2: World main uses of natural stones products in 2020 refer to XXXIII World Marble and Stones Report 2021[1]




Annually, the extensive extraction of natural stone not only yields a range of practical products utilized industrially but also results in a significant volume of waste. Waste is produced at various points in the natural stone production process, stemming from factors such as operational efficiency, color disparities, inherent irregularities, and customer preferences in color and design at the quarry. Similarly, waste is generated during the sizing phase in marble processing facilities, arising from edge remnants, natural irregularities in the stone, manufacturing flaws, and customer-defined specifications[2].

Significantly noteworthy is the production of these waste materials during cutting and grinding procedures, encompassing water and diverse solid waste materials, including fly ash, slag, silica fume, crushed aged concrete, rock waste, discarded glass, waste rubber, and tailings. Given their relatively small particle size, industrial recycling experimentation proves challenging [3].

The issue of natural stone waste poses a pressing global environmental concern. Currently, enterprises predominantly employ the landfill method for waste disposal, a practice that not only contaminates the environment but also squanders valuable resources [4]. Hence, there is an immediate necessity to repurpose waste from natural stone in diverse industries and openly disclose the volume of waste produced during quarrying processes. Considering the significant material consumption in the construction sector, repurposing waste within this industry arises as the most advantageous solution. In 2004, Mitchell CJ et al. [5] described as residuals, releases, and other devoid materials originating from manufacturing and processing, mine wastes are delineated. Table 1-1 provides a depiction of waste classification, considering their economic importance, resource requirements, and potential ultimate applications. According to Mitchell et al. [5], wastes from natural stone fall within the initial and secondary categories of mineral wastes under this framework. However, this categorization oversimplifies the assessment of stone wastes, concentrating solely on solid waste accumulation in quarries and facilities. This oversimplification proves insufficient for establishing the environmental impact boundaries of wastes and implementing suitable waste management measures. A more intricate definition of stones and an exploration of the environmental consequences of their wastes become imperative.

Table 1-1: Categorization of mineral residues (from Mitchell et al. in 2004[5])

| Group | Description | Example | Potential end uses |
|---|---|---|---|
| Type 1 | Unprocessed wastes | Quarry scalpings, quarry blocks, colliery spoil | Fill, low grade road stone, armourstone, brick clay |
| Type 2 | Processed wastes—reclaimed mineral | Silica sand wastes, limestone wastes, building stone wastes | Silica sand, kaolin, brick clay, mineral filler, aglime, aggregate |
| Type 3 | Processed wastes—added-value products | Lead/zinc wastes, pegmatite wastes, silica sand wastes | Fluorite, barite, feldspar, rare earths, mica, heavy minerals |
| Type 4 | Beneficiated wastes | Certain mine wastes | Gemstones, high value metals |

In 2012, Zeki Karaca et al.[6]pioneered the classification of natural stone wastes, presenting the physical, chemical, and petrographically characteristics of these wastes in Table 1-2. This marked the first instance of systematically listing their environmental impacts and potential applications in other industries.

Table 1-2: Categorization of mineral residues (from Zeki Karaca et al. in 2012[6])

| Group | Description | | Properties | Composition | Contains metallic mineral | Environmental threat | Industry (usage area) |
|---|---|---|---|---|---|---|---|
| I | Solid | Quarry waste | Rejection, rice, diamond wire fragments, vidia bits, some chemicals, oil | Carbonated, silicified, others | Yes/no | Health, ecosystem, water resources | Stone (vase, trinket) |
| | | Plant waste | Valvestone, paladian, abrasive residues, saw and other cutter residues, oil | | | | Construction (aggregate, fill material, armourstone, gabion) |
| II | Dust | | Stone particle (dry and fly) | | | | Chemical (filling, whitening, abrasive) |
| | | | | | | | Agriculture and livestock (acid regulatory, fertilizer and animal breeding) |
| III | Semi-slurry, slurry, cake | | Stone particle, sawdust, resin, metal, some chemicals, oil | | | | Stone (pore filler) |
| | | | | | | | Construction (fine fill, mixture material) |
| | | | | | | | Chemical (fill material for ceramic and plastic) |

As outlined in Table 1-2, stone wastes have been systematically grouped into three distinct categories, diverging from the classifications commonly employed in the industry. The first category encompasses solid wastes, with separate considerations for quarry and plant solid wastes based on their quantities and characteristics[6]. Quarry solid wastes include stones devoid of economic value, overburden, weathered stone, and all stones other than the primary rock. The construction industry stands out as the most suitable sector for repurposing quarry solid wastes. If these wastes conform to specified standards, they can be utilized for road and sea filling. Following crushing and classification, aggregates can be extracted, and items such as ornaments or vases can be crafted from multivalent or rare wastes. However, the utilization of stone waste within the stone industry is generally restricted[6].

The second category involves dust wastes, representing the purest form of stone wastes. Dust can be collected using auxiliary equipment like exhausters[6]. Given their lack of other materials, carbonated dust wastes can find application in the chemical industry and its subindustries, provided they meet specific criteria. Dust wastes also hold potential for use in agriculture and animal breeding.

In general, the utilization of dust wastes is constrained by factors such as volume and the duration of accumulation. Nevertheless, the capture and retention of dust wastes hold significant importance for individuals with asthma and airborne insects like bees. Airborne dust presents a threat to those with asthma, especially in the spring and summer seasons.[7][8][9]. The third category involves semi-slurry, slurry, and cake wastes. Semi-slurry and slurry wastes are associated with fractured, ground, and abraded particles resulting from the introduction of cooling water. Semi-slurry waste is prevalent in quarries with minimal water usage, where soils absorb water, and evaporation is noticeable[6]. As with other groups of stone waste, the rock composition governs the potential applications of these wastes. Except for carbonated stones, siliceous and other types of stone can typically find use in the construction industry as aggregates, fill materials, and in the production of building materials[6].

Based on the preceding context, considering the favorable properties of natural stone slurry as a cement filler in cement-based products, this paper primarily focuses on exploring the reuse of stone slurry powder as additives to partially replace aggregates and sands in the application of a thin layer for cement-based products, drawing from previous extensive research experience.

## 2. State of the art for stone slurry

Throughout history, natural stone has maintained enduring popularity in the construction industry, thanks to its accessibility, performance, and ornamental features. In recent decades, the heightened interest in natural stone has led to increased production, prompting the use of various processing machinery and tools. The critical aspect lies in selecting and employing stone-processing machinery that is customized for the production of specific natural stone products, ensuring defined properties and parameters for the final elements.[10].

For diverse applications, natural stones undergo processing with various devices, shaping them with proper dimensions and surface textures. In the past, ancient societies depended on conventional techniques like thermal shock, hammer as well as plug and feather methods for extracting stone[10]. The processing technology for natural stone encompasses numerous procedures aimed at manufacturing stone products suitable for a variety of purposes[10]. The application of diamond-wire- cutting method, well-known for its efficiency in reducing waste and ensuring quality, has been complemented by explorations into alternative techniques such as circular diamond saws and frame sawing in the present era. Yet, the rise of contemporary technology has led to the dominance of automated approaches[11].

Nevertheless, in addition to the creation of functional items, the processing of stone gives rise to considerable amounts of stone waste. Effectively handling this waste poses a notable environmental dilemma in numerous nations. The quantity of produced stone waste is subject to variables such as the volume of processed material (and the efficiency of processing plants), the nature and dimensions of the waste, the geological characteristics of the stone, the machinery utilized in stone processing, the technology applied in dimensional natural stone processing, the degree to which the stone block is utilized in crafting the end product, and the requirements of the clientele[12]. To identify possible applications, it is imperative to comprehend the manufacturing procedures of natural stone products, unveiling the diverse forms of stone waste produced. Understanding the ratio of such waste in the overall volume of processed stone material elucidates the scale of the issue and emphasizes the need to formulate approaches for its repurposing[12].

### 2.1 World trend of natural stone production and wastes

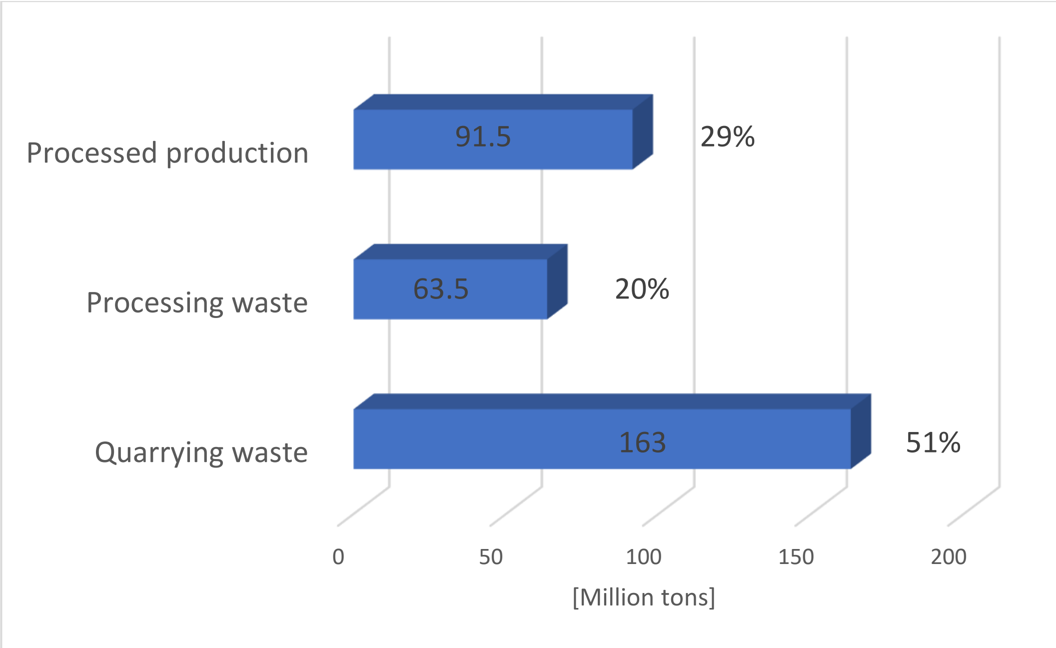
As per the XXXIII World Marble and Stones Report 2021[1], the worldwide stone network production statistics, covering total quarrying, quarrying by-products, initial production, waste during processing, and processed production, spanning the years 2003 to 2020, illustrates the remarkable surge in market demand for stones. The figure below demonstrates that the processed production is 91.5 million tons, constituting only 29% of the total gross quarrying quantity. In contrast, quarrying waste and processing waste account for 163 million tons and 63.5 million tons, representing 51% and

20%, respectively. The combined waste quantities surpass 70% of the gross quarrying, showcasing the substantial impact of modern quarrying methods, which is an astonishing figure.

Table 2.1-1: World stone industry: net production (million tons) in 2020 refer to XXXIII World Marble and Stones Report 2021[1]

| YEAR | GROSS QUARRYING | QUARRYING WASTE [1] | RAW PRODUCTION | PROCESSING WASTE [1] | PROCESSED PRODUCTION |
|---|---|---|---|---|---|
| 2003 | 153.750 | 78.750 | 75.000 | 30.750 | 44.250 |
| 2004 | 166.500 | 85.250 | 81.250 | 33.300 | 47.950 |
| 2005 | 174.750 | 89.500 | 85.250 | 34.950 | 50.300 |
| 2006 | 190.250 | 87.500 | 92.750 | 38.000 | 54.750 |
| 2007 | 212.000 | 108.500 | 103.500 | 42.500 | 61.000 |
| 2008 | 215.000 | 110.000 | 105.000 | 43.000 | 62.000 |
| 2009 | 213.750 | 100.250 | 104.500 | 42.850 | 61.650 |
| 2010 | 228.000 | 116.500 | 111.500 | 45.715 | 65.785 |
| 2011 | 237.200 | 121.200 | 116.000 | 47.560 | 68.440 |
| 2012 | 252.500 | 129.000 | 123.500 | 50.630 | 72.870 |
| 2013 | 265.800 | 135.800 | 130.000 | 53.300 | 76.700 |
| 2014 | 279.000 | 142.500 | 136.500 | 56.000 | 80.500 |
| 2015 | 286.200 | 146.200 | 140.000 | 57.400 | 82.600 |
| 2016 | 296.400 | 151.400 | 145.000 | 59.400 | 85.600 |
| 2017 | 310.700 | 158.700 | 152.000 | 62.300 | 89.700 |
| 2018 | 313.000 | 160.000 | 153.000 | 62.750 | 90.250 |
| 2019 | 316.000 | 161.500 | 154.500 | 63.350 | 91.150 |
| **2020** | **318.000** | **163.000** | **155.000** | **63.500** | **91.500** |

Fig 2.1-1: Wastes and production comparation in 2020 refer to XXXIII World Marble and Stones Report 2021[1]

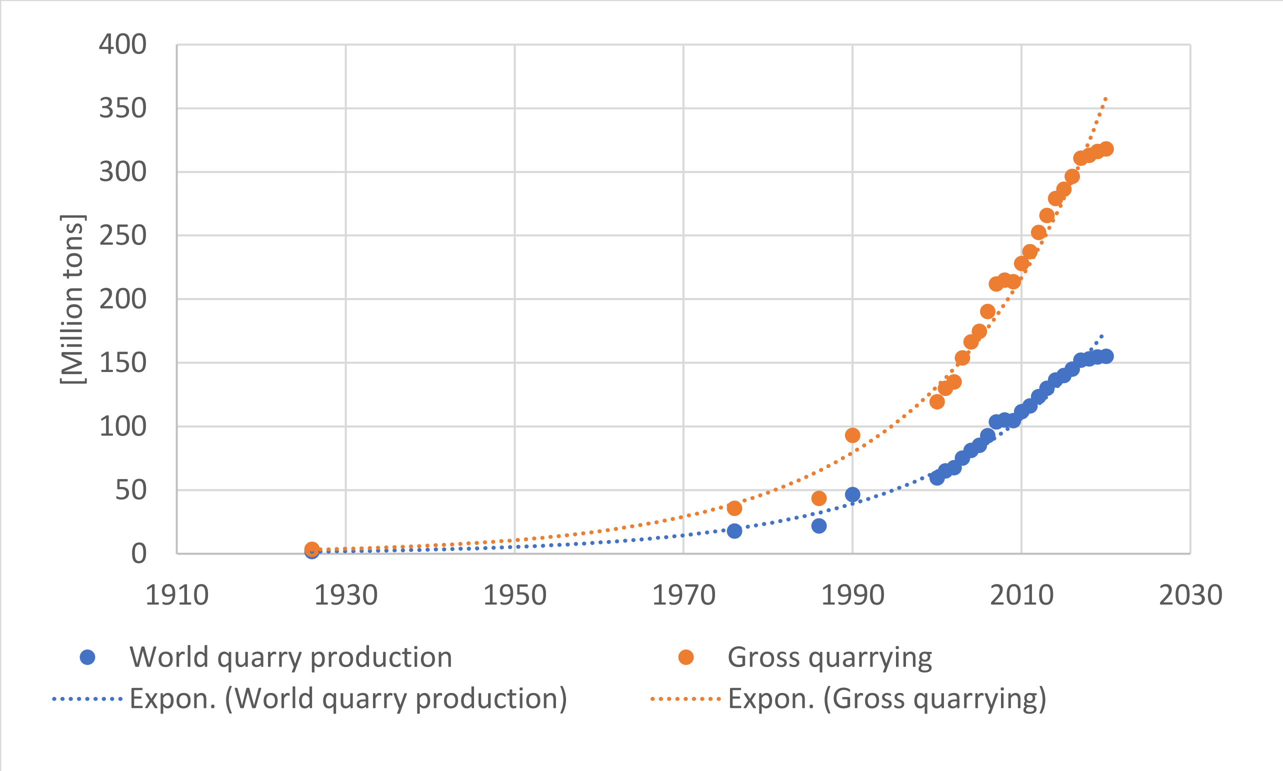



The XXXIII World Marble and Stones Report 2021[1] also furnishes a historical overview of the global stone industry's production. In 1926, the total raw production stood at a mere 1.79 million tons. Fast forward 50 years to 1976, and the world's total raw production soared to 17.8 million tons. A decade later, in 1986, the global total raw production reached 21.7 million tons. Moreover, by the year 2000, the number had nearly tripled compared to 1986, indicating substantial development during that period. From 2000 to 2017, natural stone raw production experienced annual rapid growth, peaking at around 152 million tons. Subsequently, from 2018 to 2020, the natural stone raw production maintained a high figure with slight increments. Ultimately, in 2020, the raw production of natural stone reached a pinnacle at 155 million tons, marking the highest point in the history of natural stone quarrying. The figure below illustrates the exponential growth over the past decades.

Table 2.1-2: World stone industry: historical production outline in 2020 refer to XXXIII World Marble and Stones Report 2021[1]

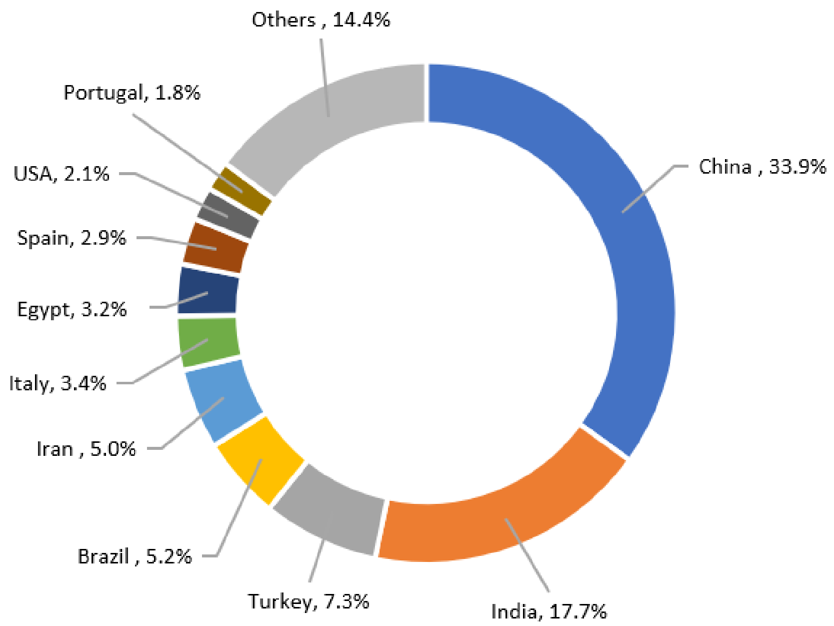
| YEARS | CALCAREOUS | | SILICEOUS | | OTHER | | TOTAL | |
|---|---|---|---|---|---|---|---|---|
| | 000 tons | % | 000 tons | % | 000 tons | % | 000 tons | % |
| 1926 | 1.175 | 65.6 | 175 | 3.3 | 440 | 24.6 | 1.790 | 10.1 |
| 1976 | 13.600 | 76.4 | 3.400 | 19.1 | 800 | 4.5 | 17.800 | 100.0 |
| 1986 | 13.130 | 60.5 | 7.380 | 34.0 | 1.190 | 5.5 | 21.700 | 122.0 |
| 1990 | 26.450 | 56.8 | 17.625 | 37.9 | 2.425 | 5.2 | 46.500 | 261.2 |
| 2000 | 34.500 | 57.3 | 21.700 | 36.3 | 3.450 | 5.9 | 59.650 | 335.1 |
| 2001 | 38.500 | 53.2 | 23.250 | 35.8 | 3.250 | 5.0 | 65.000 | 365.2 |
| 2002 | 39.000 | 57.8 | 25.000 | 37.0 | 3.500 | 5.2 | 67.500 | 379.2 |
| 2003 | 42.500 | 56.7 | 28.500 | 38.0 | 4.000 | 5.3 | 75.000 | 421.3 |
| 2004 | 43.750 | 53.9 | 33.000 | 40.6 | 4.500 | 5.5 | 81.250 | 456.5 |
| 2005 | 46.750 | 54.8 | 34.000 | 39.9 | 4.500 | 5.3 | 85.250 | 478.9 |
| 2006 | 53.350 | 57.5 | 34.300 | 37.5 | 4.600 | 5.0 | 92.750 | 521.1 |
| 2007 | 60.500 | 58.5 | 37.500 | 36.2 | 5.500 | 5.3 | 103.500 | 581.5 |
| 2008 | 62.000 | 58.0 | 38.000 | 36.5 | 5.700 | 5.5 | 105.000 | 589.9 |
| 2009 | 60.350 | 58.2 | 38.000 | 36.4 | 5.650 | 5.4 | 104.500 | 587.0 |
| 2010 | 63.230 | 58.5 | 40.500 | 36.3 | 5.750 | 5.2 | 111.500 | 626.4 |
| 2011 | 68.500 | 59.0 | 41.700 | 36.0 | 5.800 | 5.0 | 116.000 | 651.6 |
| 2012 | 72.250 | 58.5 | 45.750 | 37.0 | 5.500 | 4.5 | 123.500 | 693.8 |
| 2013 | 76.750 | 59.0 | 47.500 | 36.5 | 5.750 | 4.5 | 130.000 | 751.4 |
| 2014 | 79.200 | 58.0 | 51.900 | 38.0 | 5.400 | 4.0 | 136.500 | 766.8 |
| 2015 | 81.500 | 58.3 | 53.200 | 37.9 | 5.300 | 3.8 | 140.000 | 786.5 |
| 2016 | 83.750 | 57.8 | 56.000 | 38.6 | 5.250 | 3.6 | 145.000 | 814.6 |
| 2017 | 89.000 | 58.6 | 57.500 | 37.8 | 5.500 | 3.6 | 152.000 | 853.9 |
| 2018 | 89.250 | 58.3 | 58.250 | 38.1 | 5.500 | 3.6 | 153.000 | 859.6 |
| 2019 | 89.500 | 57.9 | 59.500 | 38.6 | 5.500 | 3.5 | 154.500 | 863.1 |
| **2020** | **88.000** | **56.8** | **62.000** | **39.9** | **5.000** | **3.3** | **155.000** | **867.0** |

Fig 2.1-2: The production of natural stone shown an exponential growth from 1926 to 2020 refer to XXXIII World Marble and Stones Report 2021[1]




Rapid expansion in the construction sector has spurred the growth of the dimension stone industry, presenting substantial economic opportunities fueled by ongoing urbanization. Consequently, the annual production output of this industry has exhibited a consistent upward trend. Such expansion is evident in numerous countries possessing extensive reserves of dimension stones or the capacity to generate them. The diagram beneath illustrates the top ten nations with the most substantial yields in dimension stone production.

Fig 2.1-3: The leading countries quarrying production of the world in 2020 refer to XXXIII World Marble and Stones Report 2021[1]



## 2.2 Natural stone quarrying execution

The extraction of dimension stones has experienced a significant evolution, progressing from rudimentary methods to contemporary mechanized approaches propelled by swift technological advancements in recent decades. The industry's transition to cutting-edge machinery and tools is clear, ensuring the effective and accurate extraction of dimension stones[37].

Historically, methods involving thermal stress, manual tools like hammers and chisels, and the use of plug and feather mechanisms have been employed for stone extraction[11], requiring arduous manual excavation with handheld tools, skilled craftsmen played an essential role in executing these tasks. Whereas, as methods for extracting stones advanced, more sophisticated tools were devised. The method involving a hammer and wedge and the approach using plug and feather [11] emerged as efficient methods for the regulated splitting of stones, incorporating systematic drilling[11] and customized procedures aligned with the typical characterization of the stone.

Contemporary quarrying operations heavily depend on sophisticated machinery and tools in the industry. Particularly, diamond-wire-cutting method has been used as a highly studied approach for extracting natural stones[37]. This method[11] entails a taut steel wire infused with diamond-engraved bits, consistently propelled by specialized machinery. The diamond wire method offers numerous advantages, such as improved machining precision, minimal production of waste, and a decreased requirement for unnecessary pre-processing in the final stage of production. Nevertheless, it is important to recognize the inherent limitations associated with diamond wire cutting, including increased operational expenses, complex procedures, wire breakage, restricted machinery lifespan, and the possibility of unevenly finished cutting surfaces[37].

In antiquity, ancient techniques[11], including thermal shock, the use of hammer and chisel, and the application of plug and feather, were utilized, requiring labor-intensive manual excavation using simple hand tools. Expert artisans played a crucial part in carrying out these activities. However, as stone extraction techniques advanced, more refined methods were devised. Through the emergence of both the method involving a hammer and wedge and the approach utilizing plug and feather, the effective splitting of stones was achieved, incorporating methodical drilling and personalized techniques adjusted to the specific attributes of the stone [37].

In contemporary quarrying activities, the industry heavily relies on state-of-the-art machinery and tools. Particularly diamond wire cutting has gained prominence as a popular approach for natural stones extracting. This method entails a tensioned steel cable infused with diamond-engraved tips, continuously propelled by specialized machines. The diamond wire technique offers various advantages, including enhanced quality of machining, minimized waste production, and a decreased necessity for redundant pre-processing in the concluding production phase. Nevertheless, it is imperative to know the drawbacks associated with cutting with a diamond wire, like increased operational costs, complex processes, wire breakage, restricted machinery lifespan, and the possibility of unevenly finished cutting surfaces [37].

Another noteworthy facet of extracting of natural stone revolves around the utilization of diamond saw cutters, characterized by steel blades embedded with diamond-encrusted segments[11]. Rotating diamond-edged saws, widely deployed across diverse industrial applications, deliver commendable production performance at a lower cost in contrast to other equipment utilized in the extraction of natural stone. Nevertheless, the choice of appropriate machines is contingent upon considerations including the stone's physical and mechanical properties, machine features, saw attributes, penetration rate, and tool usage[11].

Frame sawing, a deviation from the conventional diamond saw cutting, has garnered attention in the process of cutting natural stones[11]. A linear blade embedded with diamond bits was employed in this method, executing a back-and-forth movement at a precisely controlled sinusoidal speed[11]. Frame saw machines[11], with the capacity to handle multiple blades concurrently, facilitate the simultaneous production of numerous slabs This approach presents benefits such benefits encompass increased processing efficiency, enhanced cutting quality, and the potential to reduce operational expenditures[11].

In contemporary settings, novel techniques for concrete cutting have surfaced, encompassing methods such as abrasive water jets and plasma jet cutting[31][37]. These progressions offer significant prospects for inclusion in the post-extraction processing stage of dimension stone. Through the seamless integration of these sophisticated machinery and tools into dimension stone quarries, there is an increased potential for attaining extraction of dimension stones with superior quality [38].

Table 2.2-2: Comparation of the methods and equipment for rocks cutting[37]

| Method and equipment | Advantage | Disadvantage |
|---|---|---|
| Historical methods, including thermal shock, manual percussion using a hammer and chisel, and the application of plug and feather | Affordability and ease of access, regulated and accurate sculpting, capability to carve resilient stones | Necessitating labor-intensive manual excavation using hand tools, low efficacy, and reliant on a substantial workforce, constrained precision |
| Diamond wire cutting | Exceptional machining excellence, minimal waste generation, and diminished requirement for superfluous preliminary processing in the final stage of production. | Elevated operational expenditures, intricate procedures, wire fractures, restricted machinery longevity, and the likelihood of unevenly finalized cutting surfaces pose significant challenges. |
| Circular diamond saws | Provide outstanding production efficiency at minimal expenses when compared to alternative machinery employed in the production of natural stone. | Choosing appropriate machines relies on considerations such as the stone's physical and mechanical characteristics, machine features, saw properties, penetration rate, and tool consumption. |
| Frame sawing, a variant of diamond sawing cutting | Capable of supporting numerous blades, facilitating the concurrent generation of multiple slabs, thereby providing benefits such as heightened enhanced processing efficiency, superior quality in cutting, and the potential for reducing operational expenditures. | Increased material wastage is observed in contrast to precision cutting techniques such as diamond wire sawing or water jet cutting, owing to the thickness of the cutting blade. Frame sawing encounters limitations regarding the minimum achievable thickness. Thinner slabs may pose challenges with this approach. The cutting blades (teeth) of frame saws deteriorate over time, necessitating frequent |

| | | |
|---|---|---|
| | | maintenance and contributing to operational expenses. |
| Abrasive water jets | Considerate of the environment, cost-effective, devoid of thermal distortion, preserves material properties during cutting, water jet cutting possesses the capability to yield a polished surface finish, consequently minimizing the need for subsequent operations. This method is notably secure, consumes minimal energy, and is an economical process with a reasonably high rate of material removal. | Extended duration for cutting, economic feasibility limited to a select range of materials, potential for dimensional inaccuracies due to taper, and a slow-cutting process that may not meet the demands of situations requiring high production rates. |
| Plasma jet cutting | Remarkably compact and cost-effective, with no requirement for specialized cooling, it effectively executes thermal cutting or drilling. Its applicability extends beyond concrete structures to encompass substantial mineral materials like rocks and firebricks. | Establishing and operating plasma jet cutting can incur high costs, constraining the array of materials it is suitable for. Additionally, plasma jet cutting generates noise and fumes, giving rise to environmental concerns. |

## 2.3 Process of extracting and processing natural stones

The natural stone production process encompasses various stages, commencing from the quarry and concluding at the transformation warehouse where the stone undergoes processing for eventual distribution. Broadly speaking, the process of dimension stone production is comprised of 3 main stages[6]: exploration, quarrying, and processing. Following the initial stage of exploration, which involves quantification of volume and characterization of resources, and evaluation of environmental, economic, and socio-political factors linked to exploitation, the subsequent stage (quarry) involves site preparation, cutting, and the extraction of large stone blocks[58]. These blocks are then transformed into smaller units for convenient conveyance to the processing facility. The size of the blocks transported to the processing plant can vary from 2 to 15 $m^3$, dependent on the machinery and equipment utilized during the extraction and transportation processes.

Within the processing facility, the blocks undergo segregation derived from their distinctive traits and superior attributes, preparing them for diverse applications. Tailoring the incoming blocks to their intended purpose involves shaping them into sizable, sturdy, flat components, often in square or rectangular forms, commonly identified as slabs. Alternatively, they may be transformed into smaller counterparts termed tiles. The product surfaces are polished and smoothed in accordance with customer specifications or market demands [39].

### 2.3.1 Prospecting and exploration

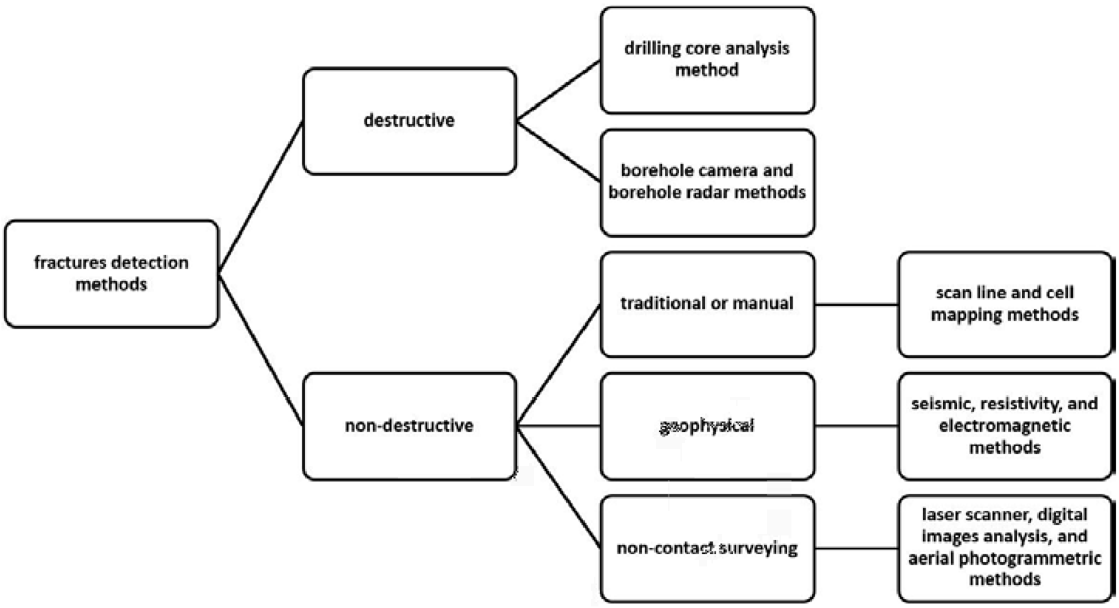
In spite of the significant technological progress achieved in prospecting and exploration, the current methods are underutilized when it comes to exploring stone dimensions in reserve[58]. Numerous challenges in this domain can be surmounted by collecting valuable geological data during the geological exploration of the target region.

In this domain, two pivotal decision support instruments that can wield a crucial influence are geological cartography and fracture analysis. The initial tool[58], mapping of geological features, can ascertain the extent and uniformity of the deposit. The subsequent tool, fracture analysis, is employed to examine discontinuities and fractures within the potential site, aiming to ascertain whether the resulting blocks will align with the preferred measurements.

The existence of cracks and fractures within a quarry for dimension stone[58] diminishes the value of the blocks to be fashioned and amplifies the volume of waste generated. To curtail waste and optimize block cutting, it is imperative to precisely identify and model discontinuities and fractures before the extraction phase.

Generally, methods for detecting discontinuities can be categorized categorized into two sets: those causing destruction and those not causing destruction[58]. Destructive techniques are both costly and time-intensive, yet they prove advantageous in gathering of information for expansive projects necessitating probability-based modeling of fractures. Methods that do not cause harm are also employed across a diverse array of projects, particularly in situations the region or the deposit itself is challenging to access. Figure 2.3-1 provides an overview of the various methods for detecting various forms of discontinuity and fractures [40].

Fig 2.3-1: Classification of discontinuity and fracture detection methods[40]

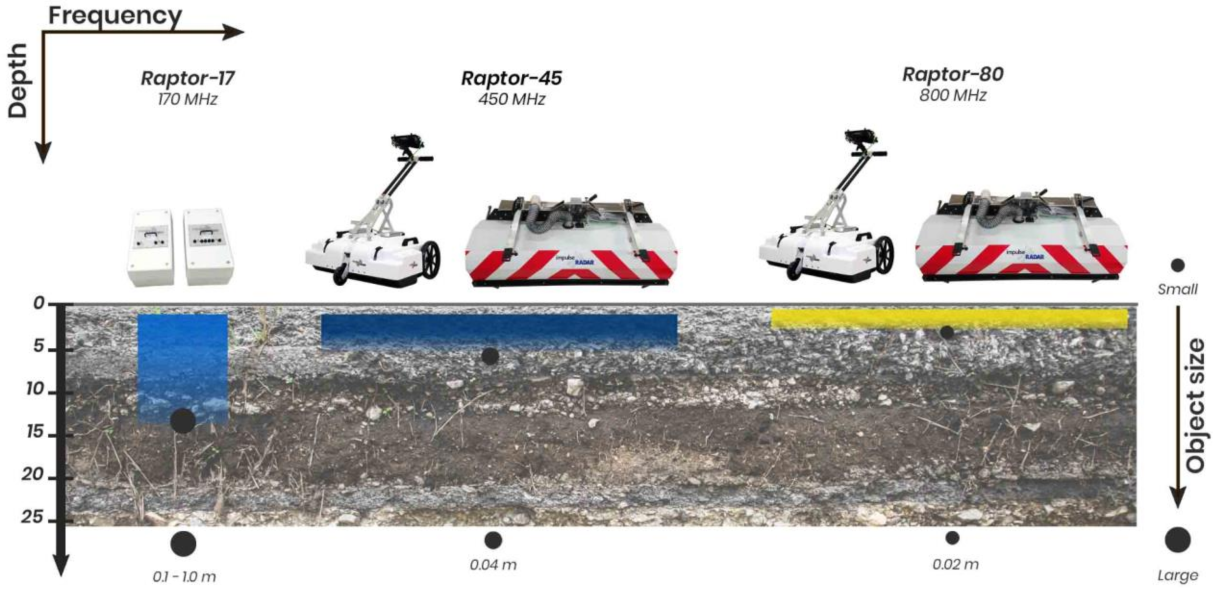



Among the non-destructive techniques for identifying discontinuities, Ground Penetrating Radar (GPR) stands out as one of the most valuable and widely embraced methods [41]. GPR has found extensive application in detecting fractures within rock masses in quarries.

The fundamental principle of GPR involves emitting radio waves into the soil and utilizing the waves that bounce back to generate a visual representation of fractures, joints, faults, and other underground structural attributes. This method[58] proves highly advantageous for dimension stone exploration, excelling in the detection of minute discontinuities such as cracks and fractures.

However, GPR is not without limitations, and its maximum penetration depth is contingent on factors like frequency of the antenna, the existence of water and the uniformity of the rock. The depth of penetration directly influences the resolution of subsurface feature images. Elevating the device frequency enhances image resolution but diminishes penetration depth, while reducing the frequency augments penetration depth at the expense of resolution [42].

Fig 2.3-2: The correlation among penetration depth, antenna frequency, and object size in Ground Penetrating Radar (GPR) (refer to Impulse Radar https://impulseradargpr.com/)




The utilization of Ground Penetrating Radar (GPR) for fracture detection within quarries extracting dimension stone traces its origins back to the early 1990s[58]. In the year of 1996, Grandjean and Gourry pioneered the identification of cracks observed in a marble quarry by extracting integrating information from Ground Penetrating Radar (GPR) profiles into a three-dimensional model. This study employed two radio wave frequencies: lower frequency (300 MHz) and higher frequency (900 MHz). The higher frequency proved instrumental in determining optimal marble block cutting strategies and quantities, facilitating temporary quarry administration. Meanwhile, the lower frequency data aided in distinguishing areas with fractures and areas without fractures, pinpointing regions with high extraction potential [42].

In 2006, Porsani and colleagues utilized Ground Penetrating Radar (GPR) profiles with frequencies set at 25, 50, and 100 MHz across different sections of a granite quarry. Their aim was to pinpoint regions exhibiting significant extraction potential and containing sizable blocks with uniform dimensions[58]. The outcomes demonstrated that identifying fractures led to cost savings by minimizing explosive consumption and streamlining the extraction of commercially viable blocks during operations[43].

In 2008, Kadioglu utilized the Ground Penetrating Radar (GPR) technique to examine fractures and voids in a carbonate rock layer located in a quarry in Turkey. This study involved the use of data derived from 2D profiles, which were then employed to develop a 3D model. The model was constructed by applying meticulously selected opacity functions and amplitude-color ranges.[58]. The model aided in identifying fractures, determining their orientation, and visualizing texture changes in the layers at varying depths [44].

In 2015, Rey et al. used Ground Penetrating Radar (GPR) to analyze textures and detect a heterogeneous layer in travertine quarries. This study employed modes with high frequency (800 MHz) and low frequency (250 MHz) [58]. Data from the 250 MHz frequency GPR were utilized to distinguish the travertine mass from the mica-schist layer, while probes with a frequency of 800 MHz were employed to identify anisotropies, such as minor fractures and porosities [45].

In 2017, Elkarmoty et al. utilized 400 MHz GPR data to detect and simulate disruptions in a sandstone quarry. The study employed a blend of Fractures Interpreted by the Tracing Technique (FITT) and Fractures Interpreted as Single Appearance (FISA) formulations, visualized in three dimensions using the Para View software[58]. The 3D model facilitated discontinuity frequency analysis and determination of the fracture index[46].

In the same year, Elkarmoty et al. employed utilizing a blend of Ground Penetrating Radar (GPR) and laboratory techniques for investigation bench fractures in a quarry. A survey grid employing Ground Penetrating Radar (GPR) was employed to encompass the overlap of 2D radargrams, leading to the creation of a 3D model that underwent validation through laboratory examinations. This model proved beneficial in pinpointing areas characterized by reduced fracture intensity.[40].

Furthermore, Elkarmoty et al. conducted GPR studies in two case studies in 2017, revealing that in one instance, benches were non-extractable due to severe fractures. Nevertheless, an overall planning strategy derived from the outcomes of fracture modeling was proposed. In the second case, interpolation of radargrams produced a three-dimensional model of the rock formation, enabling the detecting a three-dimensional zone with the lowest fracture intensity[47].

In 2018, Elkarmoty et al. employed the Ground Penetrating Radar (GPR) technique across an extensive region of an exploring and modeling fractures in an Italian quarry in bench formations. The results revealed clear trends in inclination, gradient, and spacing of joints within the three-dimensional model [48].

The consistent positive outcomes in detecting discontinuities and fractures across these studies underscore the significance of Ground Penetrating Radar (GPR) as a dependable and valuable method to enhance the effectiveness of the dimension stone production process and reducing waste generation in the course of exploration[58].

### 2.3.2 Extraction and transportation in the Quarry

Following the exploration of the dimension stone deposit, the subsequent phase involves extracting blocks of stone extracted from the quarry. This stage of the process can be optimized through measures aimed at enhancing efficiency, minimizing slicing, and reducing generation of waste, ultimately leading to lowered costs and heightened profitability[58].

When removing a block from a quarry ledge, employing non-destructive methods becomes imperative to mitigate damage to the blocks during the process. One frequently utilized technique for cutting substantial Diamond Wire Cutting (DWC) is the method used to extract dimension stone blocks from quarry benches. DWC offers several benefits in comparison to alternative approaches, including elevated cutting speed, enhanced recovery rates, and increased block yield [49]. This method involves drilling intersecting vertical and horizontal access holes in proximity to the block, threading the cutting wire through these openings, and using a machine to apply tensile force to separate the block from the bench. The figure below illustrates the block cutting process employing the Diamond Wire Cutting (DWC) method during the quarrying stage.

Fig 2.3-3: DWC method cutting of a block of dimension stone (Reference: https://optimaindia.com/)

In this stage, the main goal is to extract blocks with maximum uniformity, minimal presence of joints and cracks, and dimensions that are either standardized or economically favorable. The quest for this goal has driven the advancement of various methods for estimating the volume of rock mass between joints and cracks, determining the dimensions of extractable blocks, and evaluating the overall block yield potential of quarries[58].

Various studies have employed quarrying optimization methods, including algorithms for block yield potential. In the year 2009, Ülker and Turanboy employed data about fractures in a quarry ledge to recognize natural blocks bounded by these fractures and open surfaces. Using the 3D Linear Isometric Projection of Rock Mass (LIP-RM), they calculated the highest quantity of cubic blocks within the designated natural blocks and assessed the overall recovery rate of the bench accordingly. [50].

In 2011, Mosch et al. proposed a method to measure intact blocks in dimension stone quarries, showcasing its excellent performance in optimizing the extraction process [51].

In the year 2013, Fernández-de Arriba and colleagues utilized information concerning three primary sets of joints to create an automated computational algorithm. This algorithm predicts and enhances the cutting of commercially sized blocks from the primary block within a quarry. The technique estimates the maximum size of the block that can be extracted and recommends appropriate design alternatives to attain optimal extraction efficiency.[52].

Yarahmadi et al. (2014) created a computer program for assessing the geometry of rock mass blocks in two-dimensional space [53].

In 2015, Yarahmadi et al. explored various discontinuity modeling methods and rock block geometries, introducing a novel approach for evaluating building stone mines [54].

In a study conducted in 2018, Yarahmadi and colleagues examined commercially sized blocks in dimension stone quarries. They presented an algorithm designed to ascertain in-situ block geometry for the purpose of enhancing efficiency. This approach evaluated the comprehensive extraction potential of blocks within a quarry and identified the overall expansion direction on a broad scale. On a more localized scale, it determined the optimal spacing between quarry faces to maximize output and minimize waste generation, leading to a notable improvement in efficiency. [55].

In 2019, Yarahmadi et al. conducted

conducting cutting experiments using a diamond wire cutting machine on 12 granite samples, each in 12 different cutting directions at 15-degree intervals. The objective of the research was to identify the most effective cutting direction by examining how different cutting directions impact the achieved cutting rate. [56].

Elkarmoty et al. in 2020 developed a three-dimensional algorithm to discover the optimal cutting direction in dimension stone quarries, with the aim of maximizing block recovery ratios by addressing fracture challenges. The algorithm operates on adjusting the direction and cutting network to optimize the quantity of undamaged blocks. Through analyzing the intersections between identified fractures and designated blocks within the 3D cutting network, the algorithm offers the count of unbroken and undamaged blocks in various modes of the cutting network. Successfully tested in two case studies, the algorithm streamlines the determination of the most effective cutting direction to enhance the count of unbroken blocks, subsequently improving operational efficiency[57].

The table below provides a summary of key algorithms along with their main parameters.

Table2.3-1: A summary of crucial algorithms detailing the detection and optimization of yield potential in dimension stone blocks[58]

| Authors | Algorithm | Fracture detection method | Inputs | Steps of algorithm | Outputs |
|---|---|---|---|---|---|
| Ülker and Turanboy et al.2009[50] | Tree structure (TS), genetic algorithm (GA) | Scanline sampling technique | Dip, dip direction, cumulative spacing | (1) Modeling the rock mass in a three-dimensional fractured state<br>(2) Unique depictions of rock blocks in their natural setting<br>(3) Determination of the maximum volumetric capacity for the largest cuboids that can snugly fit into individual natural blocks | The retrieval efficiency of the extracted blocks |
| Mosch et al. 2011[51] | 3D-BlockExpert application | Window sampling, scanline sampling technique | Three points localized in a coordinate system | (1) Creating representations of 2D and 3D cross-sections of a distinctive block<br>(2) Identifying substantial volume natural blocks and enhancing the efficiency of raw block manufacturing | Volume of intact blocks and optimal cutting of the ultimate block |
| Fernández-de Arriba et al. 2013 [52] | CUT ROCK application | Manual | Inclination, directional dip, and separation of three principal dip families. | (1) Identification of the utmost block for extraction<br>(2) Enclosing parallelepiped of the utmost block for extraction<br>(3) Identification of the cuttable blocks<br>(4) Optimization of cutting: An examination of the progressive direction | Optimal progression direction for sizable blocks and diminutive blocks. |
| Yarahmadi et al. 2018[55] | 3D-Quarry Optimizer application, genetic algorithm | Manual | Ranges defined by the planes in the model, including Discontinuity planes (dip, dip direction, spacing, | (1) Modeling the geometry of all created blocks<br>(2) Categorization of the quality of all formed blocks<br>(3) Extensive optimization (categorizing the outcomes of each cutting pattern | Optimal quarrying orientation on a grand scale and the spacing between the cutting planes on a reduced scale. |

| | | | | | |
|---|---|---|---|---|---|
| | | | and...), optimization parameters ( dimensions of the cutting pattern cell and criteria for classifying block quality). | direction and identifying the optimal result)<br>(4) Limited optimization (determining the optimal result for the blocks formed during side cuttings) | |
| Elkarmoty et al. 2020b[70] | Block Cut Opt application | GPR, aerial photogrammetry | Ground Penetrating Radar (GPR) results and photogrammetric data. | (1) Fracture modeling in three dimensions<br>(2) Revolving the 3D cutting framework of blocks with a predetermined and constant increment in the cutting direction<br>(3) Shifting the 3D cutting framework horizontally (dx and dy) within a specified range for each simulated cutting direction<br>(4) Identifying the optimal cutting direction and shifting with maximum capacity the number of non-intersecting blocks with complete dimensions. | Optimal cutting orientation and repositioning to achieve the maximum recovery ratio. |

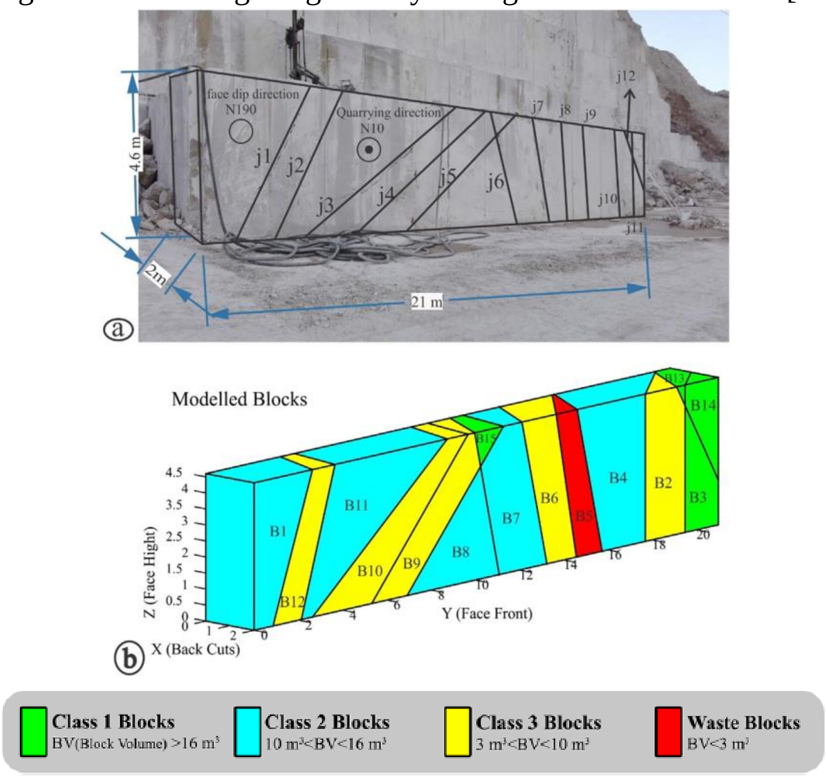
The initial step in examining the potential yield of blocks in dimension stone quarries involves computing assessing the in-situ block geometry using different algorithms[58].

Upon completion of surveying and detecting fractures in the rock formation, the characteristics of discontinuity planes, parameters like dip, dip direction, and average joint spacing are simulated. This modeling is accomplished by employing the 3D-QuarryOptimizer program within the MATLAB environment [55]. The program facilitates the intersection of discontinuity planes within three-dimensional space, leading to the creation of collision points and edges formed by these intersections.

In the subsequent stage, surfaces and edges that do not contribute to the formation of rock mass blocks are eliminated from the model[58]. The removal is based on the connectivity of these surfaces and edges with others. Following this, the enclosed surfaces along the planes of discontinuities are identified, consisting of boundaries created through the intersection of disparities.. Finally, the tracking of blocks in the rock mass is concluded through a combination of closed surfaces.

Figure 2.3-4 provides an illustrative illustration of representing the spatial structure of blocks in their original location, both on a significant and minor scale, within utilizing the previously mentioned approach in a quarry for extracting building stones [55].

Fig 2.3-4: Modeling the geometry of large-scale in situ blocks[55]




Where,

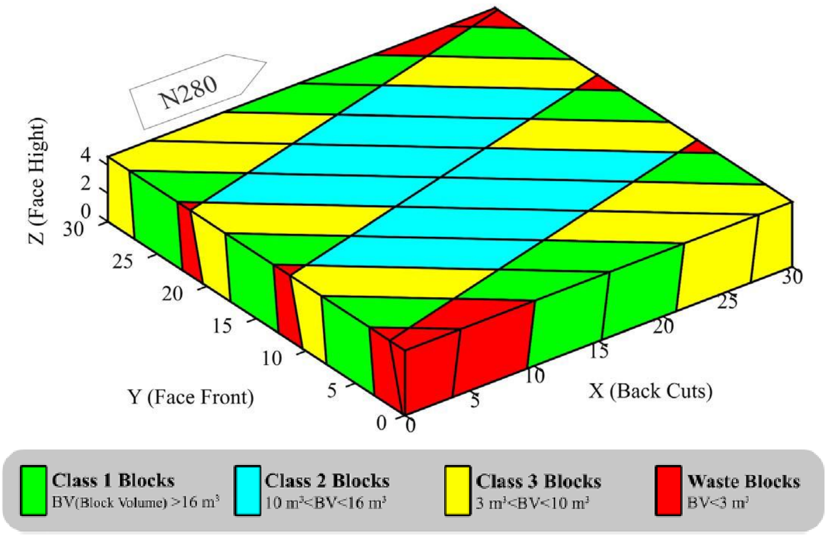
a. Portion of a stone facade with delineated joints.
b. Blocks represented in a modeled format

BV is block volume

The final phase of assessing the block yield potential of dimension stones involves two segments: small-scale and large-scale optimization[58]. Small-scale optimization, implemented within the active quarry bench, aims to enhance the excavation layout of the stone surface, maximizing the value of extracted blocks.

The categorization of recoverable blocks based on different cutting patterns within the rock mass depends on their usable volume[58]. This volume is determined by multiplying the block's volume by its shape factor. The block shape factor, ranging from 0 to 1, signifies the level of closeness and resemblance of blocks to a standard rectangular cube block with index dimensions. Larger block sizes and greater similarity in shape to the standard rectangular cube denote higher-quality blocks. Figure 2.3-5 depicts the outcomes of enhancing the quarry face cutting pattern is analyzed in comparison to the conventional pattern at an equivalent cutting distance[58]. In the depicted diagram, blocks classified as class 2 and 3 hold values of 0.25 and 0.1, respectively. Blocks categorized as class 1 and waste blocks are regarded as having no value.

Fig 2.3-5: Modeling the geometry of small-scale in situ blocks[55]




Where,

Class 1: Block Volume ≥ 16 $m^3$

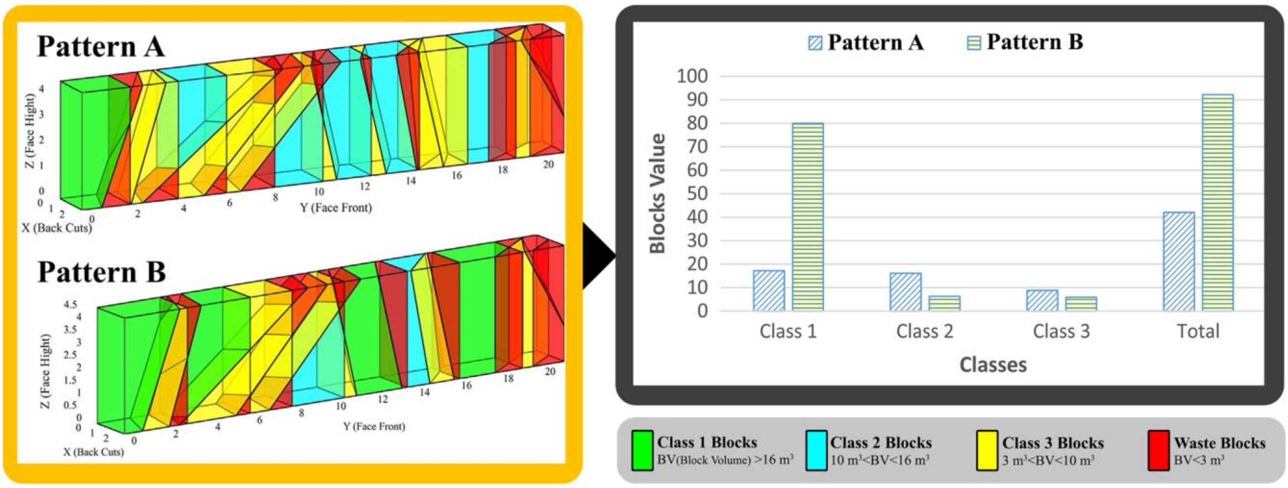
Class 2: 10 $m^3$≤ Block Volume < 16 $m^3$

Class 3: 3 $m^3$≤ Block Volume < 10 $m^3$

Waste: Block Volume < 3 $m^3$

Additionally, large-scale optimization is employed to ascertain the most effective direction of quarrying along with objective of optimizing the worth of recoverable blocks. In this regard, the overall worth of recoverable blocks in various direction of quarrying is juxtaposed, and the most effective orientation is identified. Figure 2.3-6 illustrates the outcomes of optimizing block cutting owing to the intersection of discontinuity planes in four separate directions within the cutting network[58]. The value assigned to blocks that can be extracted remains consistent with the valuation in small-scale optimization.

Fig 2.3-6: Contrasting the optimization outcomes of two cutting designs, A, maintaining a consistent cutting distance, and B, featuring an optimized variable cutting distance in a small-scale setting.[55]




Where,

Class 1: Block Volume ≥ 16 $m^3$

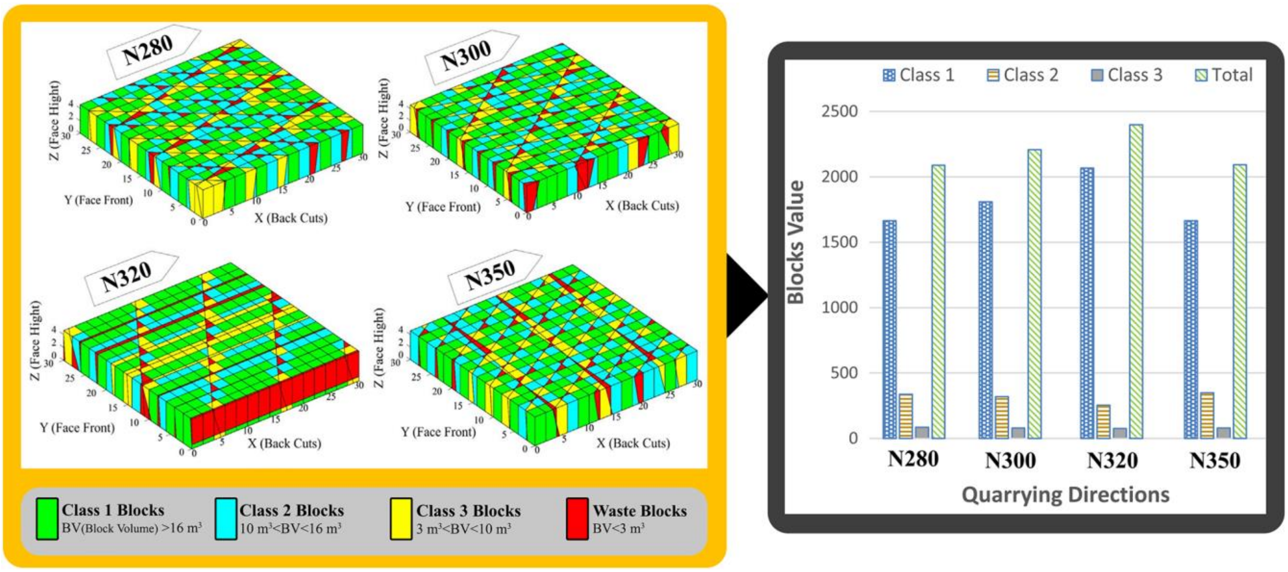
Class 2: 10 $m^3$≤ Block Volume < 16 $m^3$

Class 3: 3 $m^3$≤ Block Volume < 10 $m^3$

Waste: Block Volume < 3 $m^3$

Beyond the aforementioned methods, various other elements can influence the amount of generated waste during the extraction process. These elements encompass the utilization of refined extraction machinery, non-detrimental methods for transporting blocks, and strategic decisions and actions made by management. Overall, the stage of extracting dimension stone in the production process can profit from a holistic strategy involving the selection of optimal cutting techniques, assessment, and choice of the most suitable equipment, and the implementation of effective management measures to maximize efficiency.

Fig 2.3-7: Evaluating the optimization outcomes for four diverse quarrying directions on a large scale[55]




Where,

Class 1: Block Volume ≥ 16 $m^3$

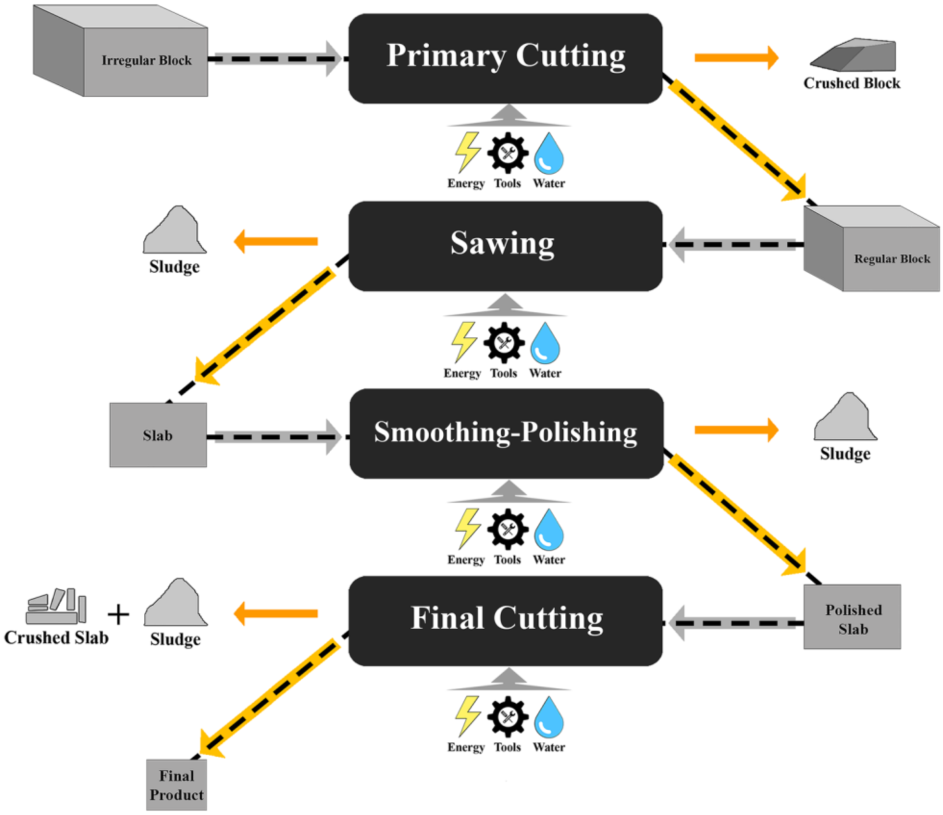
Class 2: 10 $m^3$≤ Block Volume < 16 $m^3$

Class 3: 3 $m^3 \leq$ Block Volume < 10 $m^3$
Waste: Block Volume < 3 $m^3$

### 2.3.3 Processing of the dimension stone with plants in Warehouse

Upon the extraction of dimension stone blocks and their transportation to the processing facility, the next step involves transforming them into slabs or tiles for final sale. This process entails initial cutting of the quarry-obtained blocks into regular forms, followed by placement within a cutting apparatus, for instance, a gang-saw machine. Subsequently, the blocks are sliced into slabs of varying thicknesses, aligned with market demand or intended applications. Following the sawing process, the surfaces of the slabs undergo polishing and smoothing, culminating in the final cutting to shape cutting the slabs into tiles of various dimensions. The figure below provides an overview of the operations conducted within a standard dimension stone processing facility, illustrating the generated waste at every phase of the procedure.

Fig 2.3-8: Schematic representation outlining the dimension stone processing sequence [58]




Illustrated in Fig 2.3-8, each stage of the facility for processing dimension stone can yield its own waste. The byproducts of this process encompass blocks that have been crushed, a result of joint and crack presence and suboptimal selections of cutting geometry (angle, direction, etc.), along with the byproducts such as sawdust and sludge resulting from the combination of water and dust during the processes of sawing and polishing[58]. Additionally, slabs that have been crushed emerge from the final cutting phase. The dimension stone processing also grapples with challenges such as heightened high water and energy consumption and generally inadequate efficiency.

Throughout the years, numerous studies have aimed to enhance the efficiency of the dimension stone processing by minimizing material loss, recycling generated waste, or decreasing water and energy consumption in the process[58]. In 2012, Tanthapan-ichakoon and Charinpanitkul scrutinized the efficacy of gathering particulate matter produced within the dimension stone processing facility at varying air velocities and dust concentrations, emphasizing the significant impact of increased water usage during cutting on dust collection[59].

Álvarez-Fernández et al. (2012) conducted a survey to pinpoint the optimal angle of cutting for achieving the best cutting arrangement and producing slabs maximizing their surface area, introducing a concept of a neutral region and developing a computational algorithm based on this concept [60].

In 2012, Gazi et al. proposed an approach to evaluate the energy intensity and environmental efficiency across different stages of dimension stone processing, highlighting the importance of optimization in each step[61].

Careddu et al. (2013) founded a facility dedicated to the storage and recycling of waste materials from different dimension stone production stages, repurposing them as raw materials for other industries [62]. Mendoza et al. (2014) compared the performance of various analyzing saw technologies with respect to their water usage, energy consumption, and overall efficiency., demonstrating the superiority of diamond multiwire saw (DMWS) technology over multi-blade gangsaw (MBGS) technology [63].

Sivrikaya et al. (2014) examined the impact of using marble and granite waste dust on soli Soil consolidation [64]. Careddu et al. (2014) investigated the potential use of lime sawdust from dimension stone processing plants in manufacturing industrial products [65]. Dino et al. (2015) explored the reuse of sludge from natural stone quarrying process in quarry rehabilitation [66].

Elkarmoty et al. (2018) used GPR to identify discontinuities in stone blocks at a processing plant, creating a 3D model to aid decision-making in the initial block cutting process[69].

Yurdakul (2020) scrutinized 19-dimension stone processing plants in Turkey, examining monthly waste production, types of production wastes, and their production stages[71].

Across these methodologies, there is a concerted effort to minimize material and resource loss during processing, thereby enhancing efficiency. This objective can be achieved through various means, including optimized methods and equipment usage, recycling of generated waste, and judicious use of available water resources.

Fig 2.3-9: Quarrying and processing of dimension stones[52]


## 2.4 Waste generation during the quarry site and warehouse

### 2.4.1 Total waste generation

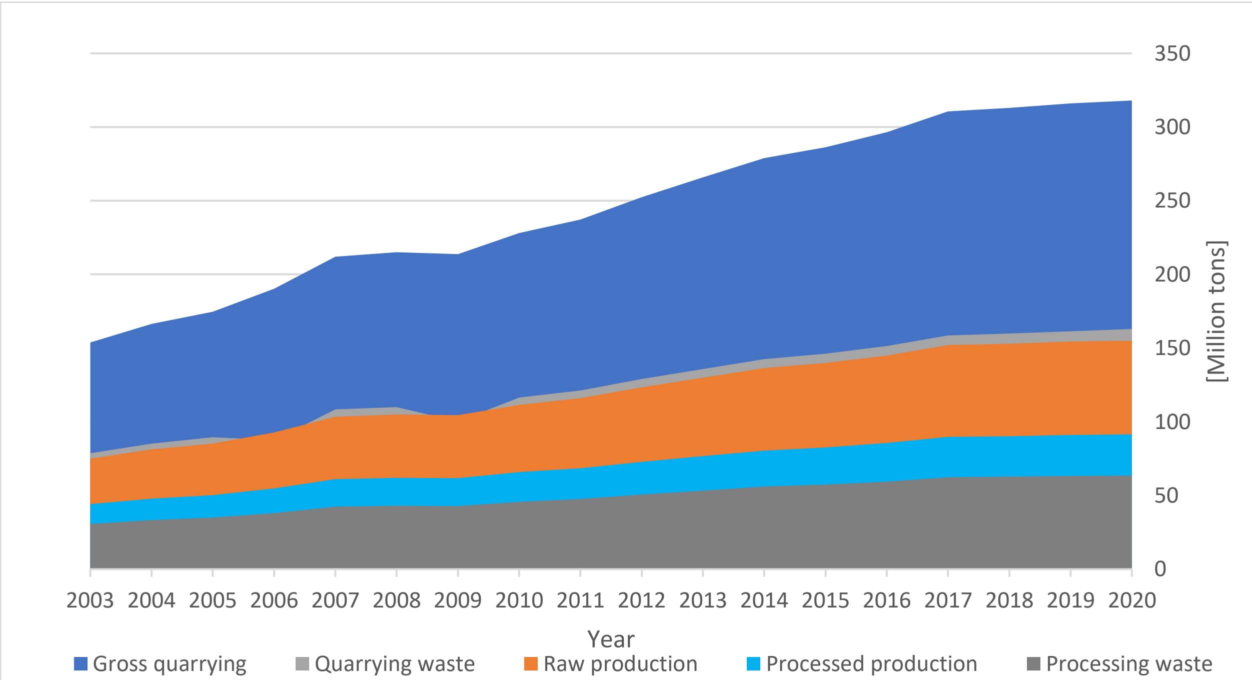
In spite of its substantial economic possibility and a notable world production quantity, the market of natural stone industry grapples with many challenges. Issues such as lack of quarrying practices, low operational efficiency, and elevated production costs pose potential threats to its future growth[58]. Adequate attention to production planning and resource preservation becomes crucial to overcome these challenges.

As per the 2021 statistics reported by countries actively engaged in the dimension stone industry, the total annual extraction from dimension stone quarries reached approximately 318 million tons in 2020. However, a significant portion of this, around 163 million tons, is designated as quarrying waste. The remaining 155 million tons transported to processing factories, about 63.5 million tons become slurry waste during the processing phase. Consequently, only 91.5 million tons culminate in the final product [1].

Tab 2.4-1: The quantities of both products and waste generated during various stages of dimension stone manufacturing vary from 2003 to 2020 (data from: (Montani 2021))[1]

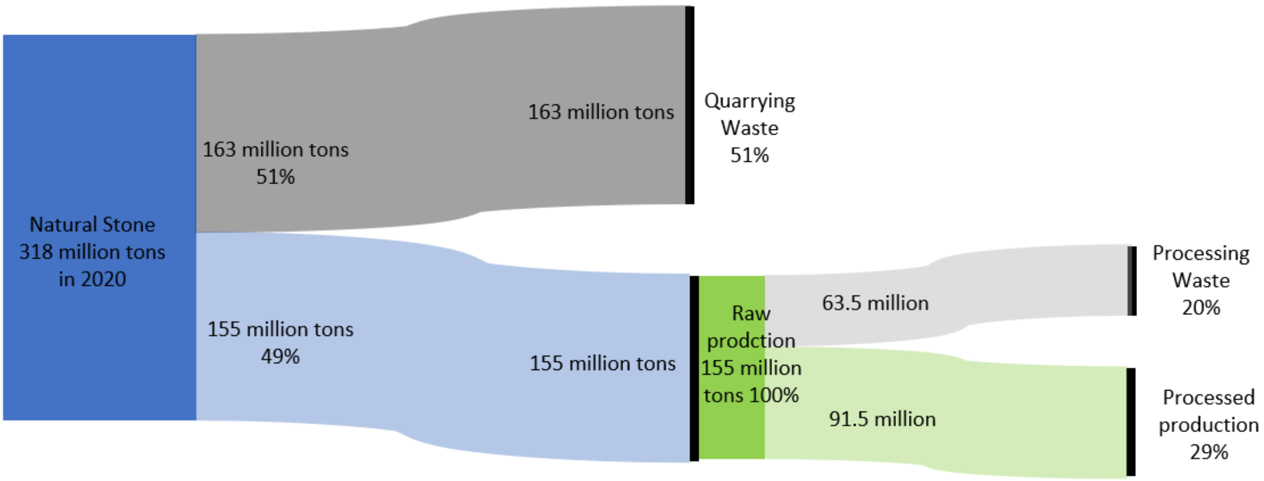
| Year | Gross quarrying [Million tons] | Quarrying waste [Million tons] | Raw production [Million tons] | Processed production [Million tons] | Processing waste [Million tons] |
|---|---|---|---|---|---|
| 2003 | 153.75 | 78.75 | 75 | 44.25 | 30.75 |
| 2004 | 166.5 | 85.25 | 81.25 | 47.95 | 33.3 |
| 2005 | 174.75 | 89.5 | 85.25 | 50.3 | 34.95 |
| 2006 | 190.25 | 87.5 | 92.75 | 54.75 | 38 |
| 2007 | 212 | 108.5 | 103.5 | 61 | 42.5 |
| 2008 | 215 | 110 | 105 | 62 | 43 |
| 2009 | 213.75 | 100.25 | 104.5 | 61.65 | 42.85 |
| 2010 | 228 | 116.5 | 111.5 | 65.785 | 45.715 |
| 2011 | 237.2 | 121.2 | 116 | 68.44 | 47.56 |
| 2012 | 252.5 | 129 | 123.5 | 72.87 | 50.63 |
| 2013 | 265.8 | 135.8 | 130 | 76.7 | 53.3 |
| 2014 | 279 | 142.5 | 136.5 | 80.5 | 56 |
| 2015 | 286.2 | 146.2 | 140 | 82.6 | 57.4 |
| 2016 | 296.4 | 151.4 | 145 | 85.6 | 59.4 |
| 2017 | 310.7 | 158.7 | 152 | 89.7 | 62.3 |
| 2018 | 313 | 160 | 153 | 90.25 | 62.75 |
| 2019 | 316 | 161.5 | 154.5 | 91.15 | 63.35 |
| 2020 | 318 | 163 | 155 | 91.5 | 63.5 |

Fig 2.4-1: Illustration depicting the production stages of dimension stone, highlighting both product and waste generation throughout the various phases from 2003 to 2020 (data from: (Montani 2021))[1]



In the dimension stone production process, a substantial volume of waste is generated. As detailed in earlier sections, citing the XXXIII World Marble and Stones Report 2021[1], approximately 51% of the material extracted from dimension stone quarries becomes waste during the extraction process. Furthermore, during the transportation of about 41% of the remaining material to dimension stone processing plants, it transforms into waste in the processing operation. Consequently, a mere 29% of the total material extracted from a stone quarry undergoes the transformation into the final product, while the remaining 71% is discarded as waste.

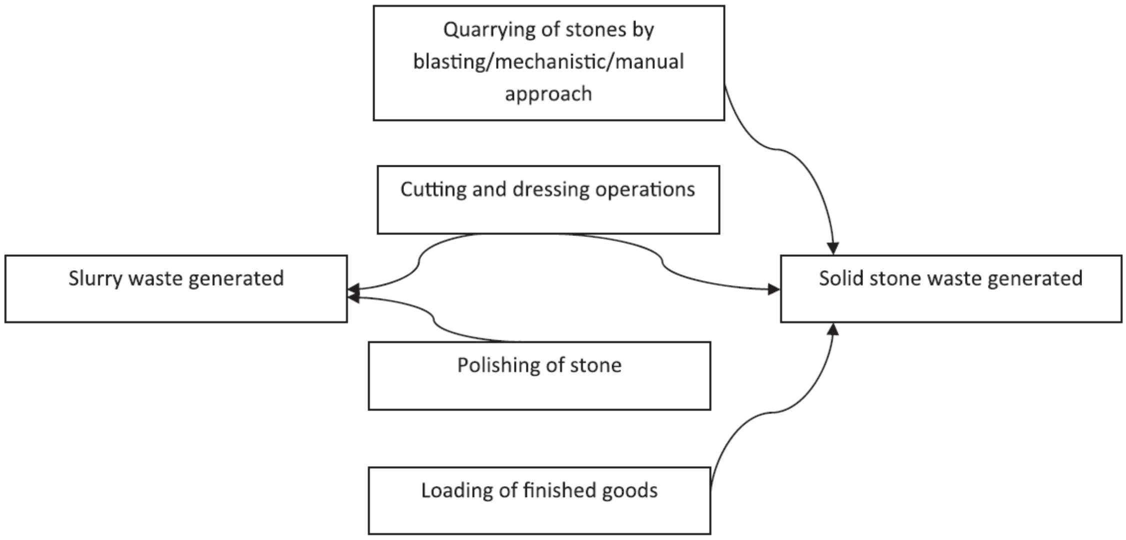
Fig 2.4-2: Sankey diagram of natural stone quarrying and processing production and wastes.




## 2.4.2 Classification of stone waste

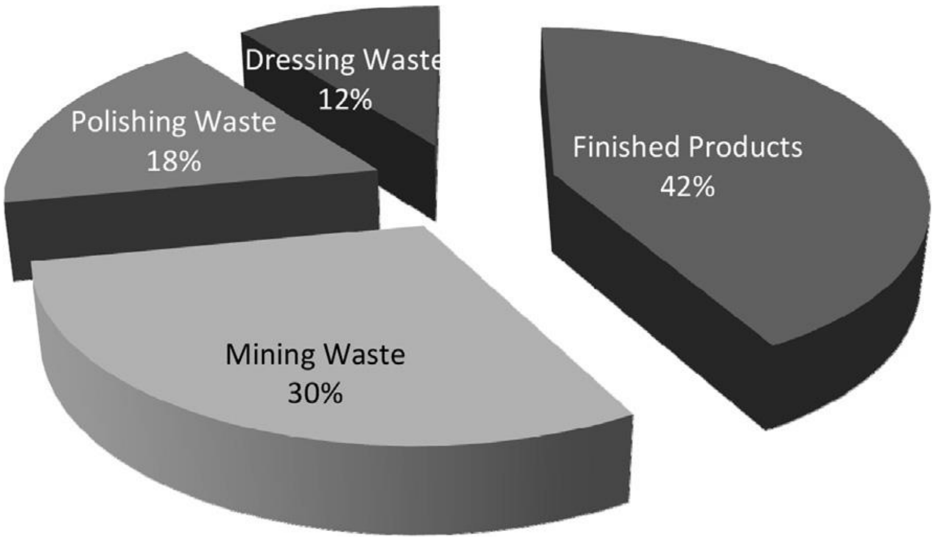
Careddu and Siotto (2011) [72] utilized the European Waste Catalogue (EPA, 2002)[73] to categorize stone waste. The European Waste Catalogue (EPA, 2002)[73] code 01 relates to "Wastes resulting from exploration, mining, quarrying, physical, and chemical treatment of minerals," consisting of five sub-codes, the generated waste, excluding topsoil, can be categorized under sub-code 01 04 "Wastes from physical and chemical processing of non-metalliferous minerals," including waste gravel, crushed rocks, particulate waste, and waste from the cutting and sawing of stone, among other unspecified waste materials. In every phase, spanning from quarrying to the loading of final products, is associated with the production of stone waste.

Fig 2.4-3: Illustrative chart illustrating the generation of stone waste throughout diverse mining and processing operations [72]




As per a report presented by the Department of Mines and Geology, Rajasthan, in 2011[74], approximately 58% of the extracted dimensional stone is designated as waste during mining, sawing, cutting, and polishing processes.

Fig 2.4-4: The distribution of diverse categories of stone waste generated and the finalized products acquired from a freshly mined-out stone (Rajasthan, in 2011)[74]




Stone waste from quarries and processing plants exhibits distinct characteristics. Karaca et al. (2012)[75]provided an exhaustive classification of this waste after conducting visits to over 50 quarries and 20 plants for classification purposes.

Table 2.4-2: Categorization of stone byproducts as per the study conducted by Karaca et al. (2012)[75]

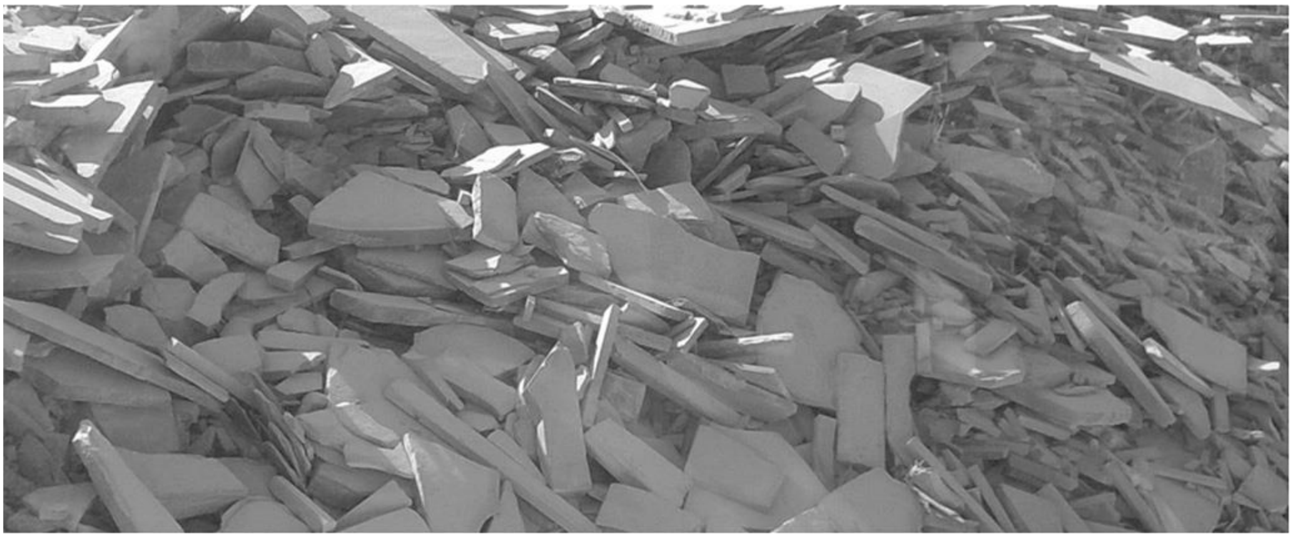
| Group | Sub group | Characteristics | Possible usage |
|---|---|---|---|
| Solid | Quarry | Rejected stones, rice, diamond wire fragments, vidiabits, chemicals, oil | Stone (vase, trinket) |
| Solid | Plant | Valvestone, paladian, abrasive residues, saw and Cutter Residues, Oil | Construction (aggregate, fill material, armourstone, gabion) |
| Dust | | Stone particles (dry and fly) | Chemical (filling, whitening, abrasive), agriculture and livestock (acid regulatory, fertilizer and animal breeding) |
| Semi-slurry, slurry, cake | | Stone particle, sawdust, resin, metal, chemicals, oil | Stone (pore filler), construction (filler), chemical (ceramic and plastic) |

Quarry wastes encompass stone fragments devoid of significance, further categorized into solid, rice, and dust forms. Solid quarry waste is easily separated from additional waste like diamond wire fragments, bits, chemicals, and grease. Rice waste, originating from cutters and drillers, is relatively minimal and incorporates bits, oil, and machine remnants, accumulating around the cutting edge. All block extraction techniques yield stone dust, with diamond equipment producing more than drillers. Stone dust particles are minuscule, measurable only in micrometres. Blasting can result in stone fragments up to 1 m in diameter, causing quarrying waste to vary widely in grain size and structure, from clayey soil particles to boulders (ranging from less than 0.1 mm to 1 m in diameter). Overburden waste, as well as the parent rock mass, undergo fragmentation through processes like drilling, ripping, and blasting. Overburden waste primarily consists of minerals containing dimensional stone and host material, generally inert but potentially containing toxic chemicals from extractive operations. The stripping ratio, representing the ratio of overburden waste to excavated mineral quantity, is a crucial metric, with a higher ratio indicating less profitable mining. The waste is then loaded into haulage trucks and disposed of in nearby areas, with sorting occurring during discharge. Fine particles accumulation occurs at the summit, whereas larger rock particles descend to the base of the dump, forming large heaps on open lands[76].

Plant wastes are categorized into solid and sludge waste. Solid plant waste results from cutting and sawing excavated stone blocks, where discarded solids not meeting required dimensions are considered waste. Approximately 20 to 22% of the stone block is lost during cutting operations, influenced by geological, textural, and petrographic aspects of the stone, plant capacity, technical specifications of machinery for cutting and processing, and correlations among block dimensions and quarry output products. Quarry blocks with larger dimensions inherently reduce waste.

The amount of waste is lower in comparison to the waste generated in quarries. Waste from the plant primarily includes paladians and valvestones [75]. Paladians [75]constitute materials discarded during cutting and other production operations, while valvestones are stones placed into block cutting machines for stability during cutting but are subsequently rejected. The solid waste produced in the plant during cutting and polishing operations is comparatively smooth and thinner than the waste from quarries. This characteristic aids in the separation[75] from saw bits, abrasives, and other residues.

Fig 2.4-5: Paladians(from https://ars.els-cdn.com/content/image/1-s2.0-S0959652616308034-gr4_lrg.jpg)



According to Careddu et al. (2013) [77], an estimated 20 to 30% of the ultimate product transforms into slurry. The composition of the sludge produced during sawing and finishing operations is contingent upon the attributes of the original rock mass and the equipment utilized for these procedures.

In 2021, Paweł Strzałkowski[10] conducted an examination of the quantity and composition of stone waste in 10 processing plants, considering the varieties of processed natural stone, stone products, and associated machinery in use. This investigation delved into data acquired from facilities for processing concerning the movement of partially processed items throughout various phases in the processing of stone, coupled with the author's firsthand observations.

Figure 2.4-6: Diagram illustrating the generation of waste during the natural stone processing

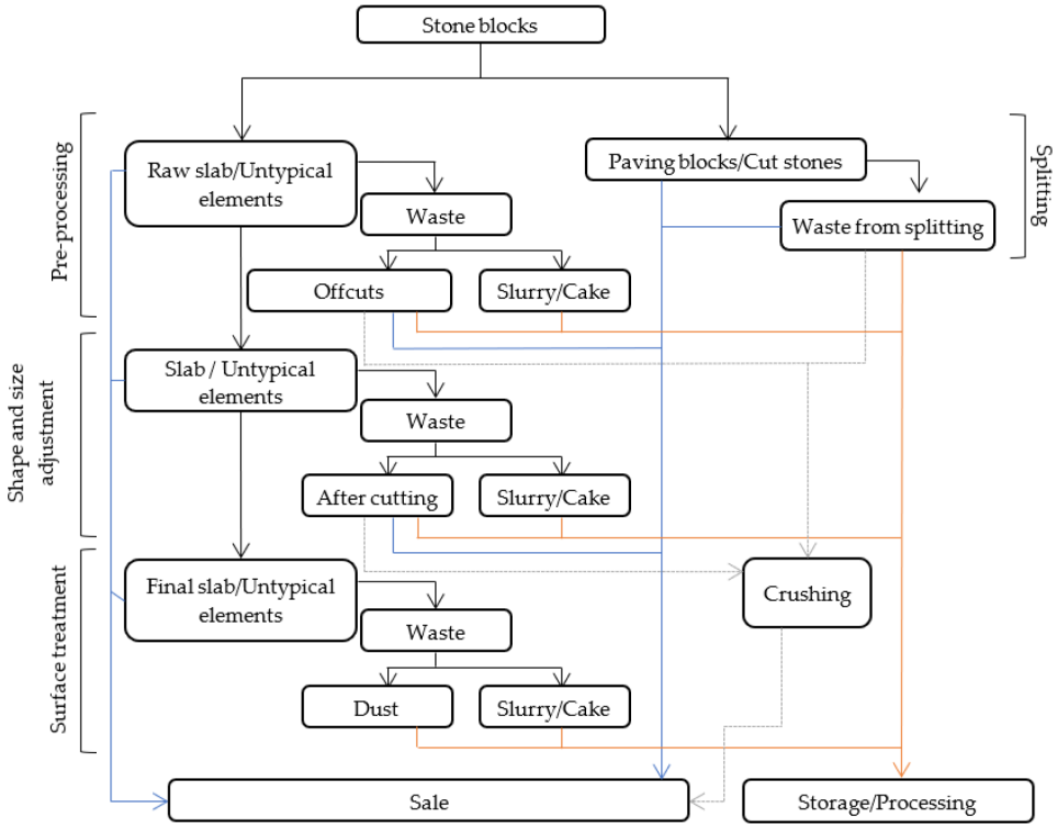



Table 2.4-3: Varieties of stone products and the machinery employed in plants were investigated by Paweł Strzałkowski, with a focus on analyzing the scale and type of stone waste [10]

| | | Plant 1 | Plant 2 | Plant 3 | Plant 4 | Plant 5 |
|---|---|---|---|---|---|---|
| Pre-processing | Stone slurry | 8.34% | 7.82% | 21.98% | 14.40% | 4.76% |
| | Solid stone scraps | 3.15% | 3.58% | 8.21% | 5.76% | 5.11% |
| Shape and szie adjustment | Stone slurry | 0.71% | 0.03% | 0.88% | 0% | 0.83% |
| | Solid stone scraps | 1.90% | 0.17% | 1.40% | 0 | 2.11% |
| Surface treatment | Stone slurry | 1.90% | 0.25% | 2.34% | 6.48% | 1.65% |
| | Solid stone scraps | 1.04% | 0.29% | 0.63% | 0.72% | 0 |
| | | Plant6 | Plant7 | Plant8 | Plant9 | Plant10 |
| Pre-processing | Stone slurry | 5.68% | 11.05% | 11.53% | 6.70% | 6.31% |
| | Solid stone scraps | 5.67% | 6.29% | 5.96% | 3.58% | 3.39% |
| Shape and szie adjustment | Stone slurry | 0 | 0.82% | 1.58% | 3.12% | 0 |
| | Solid stone scraps | 0 | 2.08% | 2.11% | 3.98% | 0 |
| Surface treatment | Stone slurry | 0 | 1.77% | 2.22% | 3.36% | 3.72% |
| | Solid stone scraps | 0 | 0 | 0.37% | 0 | 0.23% |

Table 2.4-4: Average total wastes during production process

| | Stone slurry | Solid stone scraps |
|---|---|---|
| Pre-processing | 11.46% | 5.16% |
| Shape and size adjustment | 0.49% | 1.12% |
| Surface treatment | 2.52% | 0.54% |

Through the conducted analyses, it was revealed that stone waste accounts for 10–35% concerning the processed stone material quantity. Notably, the volume of sludge is three times greater than that of solid scraps.

From the above analysis of researches, it is clear to see that the proportions of solid stone waste and stone slurry waste in quarrying vary based on several factors, including the type of stone being quarried, the quarrying methods used, and local regulations. Generally, solid stone waste includes blocks, fragments, and other large pieces of stone, while stone slurry waste consists of smaller particles and water generated during the cutting and processing of the stone.

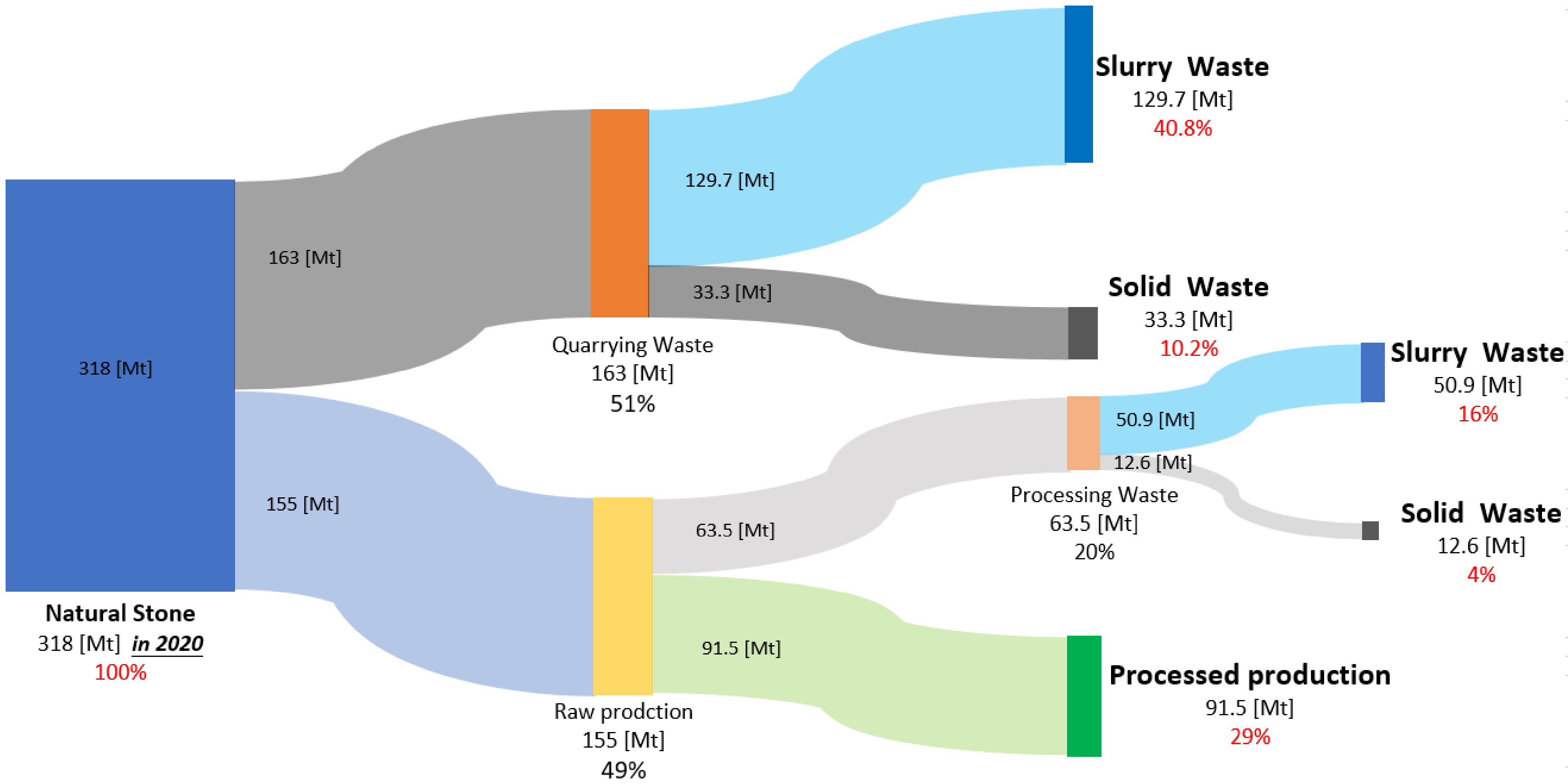
Exact proportions can vary significantly from one quarry to another. However, here's a general example to provide an estimate:

- Solid Stone Waste: It typically constitutes about 15% to 20% of the total quarrying waste. This includes larger stone fragments, unusable blocks, and other solid waste generated during quarrying operations.
- Stone Slurry Waste: This constitutes the majority of the quarrying waste, usually making up around 80% to 85% of the total. Stone slurry waste consists of water mixed with smaller stone particles, generated during cutting, shaping, and polishing operations.

These proportions can vary widely based on the type of stone being quarried (e.g., marble, granite, limestone), the quarrying techniques, and the efficiency of waste management practices employed at the quarry.

Therefore, the Sankey diagram of natural stone quarrying and processing production and wastes can be explained more in detail as follows:

Fig 2.4-7: Sankey diagram of natural stone quarrying and processing production and wastes with calculations of quantitively amounts in 2020 refer to XXXIII World Marble and Stones Report 2021[1]



## 2.5 Chapter summary

This chapter elaborates on the stat of the art for stone slurry in the aspects of stone quarrying market in the world till the year of 2020, main equipment for quarrying with their working performance, natural stone quarrying in mining site and processing in warehouse and final production and wastes estimation during the whole process.

Clearly to see from this chapter that the slurry waste takes up more than half (slurry waste from quarry site 40.8% plus slurry waste from warehouse 16%) of the total amount of natural stone quarried worldwide in 2020 which is 318 million tons, while the processed production of various products using for different fields is only 29% of the total amount of natural stone quarried worldwide in 2020.

In conclusion, the immense quantity (around 180 million tons worldwide in 2020) of waste in the form of stone slurry has significant environmental impacts if not managed properly, may cause water pollution, soil contamination, air pollution, habitat destruction, aesthetic and recreational impact, groundwater contamination, erosion and sedimentation, and so forth. To mitigate these environmental impacts, it's crucial to properly manage and treat stone slurry through appropriate disposal methods, recycling, or reusing the waste in an environmentally friendly manner. Implementing responsible waste management practices and adhering to environmental regulations can help minimize the negative effects of stone slurry on the environment.

# 3. State of the art on reuse of stone-based products

## 3.1 Mortar and concrete

Numerous inquiries have explored the integration of stone waste into mortars and concrete formulations, serving as either a complete or partial replacement for cement or fine aggregates [78]. Some studies even utilized marble fragments as coarse aggregates. With annual global concrete consumption reaching approximately 30 billion tons[79], and aggregates constituting 71% of the volume of traditional concretes, there exists theoretical potential for reusing all the generated stone waste (226.5 Mt in 2020, according to the XXXIII World Marble and Stones Report 2021[1] and calculations in the preceding chapters) in concrete formulations by substituting aggregate or cement.

Concrete aggregates are categorized into two distinct types, fine (predominantly smaller than 5 mm) and coarse (9.5–37.5 mm) [80]. Fine aggregates constitute a quarter of the total concrete matrix volume [81], with natural river sand considered the most suitable fine aggregate for concrete. However, its exploitation may lead to severe environmental impacts, prompting authorities to impose restrictions [82] and, consequently, encouraging the use of alternative materials. Similar concerns arise regarding finding alternatives to cement due to well-documented environmental problems associated with its production. Considering its high fineness, we concentrated on utilizing dimension stone waste in lieu of cement and fine aggregates.

While a consensus exists regarding the recycling viability of stone waste into cement-based matrices, numerous controversies surround optimal content, its impact on the workability of mortars and concretes, and potential chemical interactions between the cement paste and the waste. A literature survey indicates that the ideal stone waste content can vary based on waste characteristics, such as dispersion of particle dimensions, distinct surface area, and composition of chemicals, and the intended application for the concrete/mortar.

Various researchers have shown that the substitution of Portland cement with stone waste, up to 15%, does not cause substantial changes in the mechanical characteristics of mortar/concrete. In studies [83][84][85][86][87], enhancements of about 30% in the compressive strength of concrete were reported after a 3-day curing period, and approximately 20% improvement was observed after 7 and 28 days using 5 to 15 wt.% marble powder as an additive. Although the authors did not provide the particle size of marble powder, the measured specific surface area (11400 cm2/g) reveals its very high fineness, even surpassing typical OPC’s, which generally range from 3000 to 6000 cm2/g. In this context, the filler effect is likely associated.

Some researchers have observed positive outcomes with higher concentrations of stone waste. Khyaliya et al. (2017) [88] delved into the workability of mortars by partially substituting sand with stone waste. The authors provided data on the particle size and water absorption of marble powder compared to river sand, allowing for a more dependable comparison. River sand and marble powder exhibited water absorption and fineness modulus values of 9.9% and 8.23%, and 2.13 and 1.45, respectively. They discovered that incorporating marble waste from 25 to 50% resulted in improvements in mechanical performance due to reduced water requirements. At a 50% substitution rate[88], the water needed for the desired workability decreased by 6%, and compressive strength increased from 2.84 to 7.04 MPa. In this instance, the primary contribution of marble dust lies in enhancing workability rather than the filler effect, enabling a reduction in the w/c ratio. However, this improvement in workability contradicts the findings of most studies.

Sahan Arel et al. (2016)[89] noted a decrease in workability as the amount of marble powder, replacing fine aggregate, increased. Nevertheless, the loss of workability and, consequently, the higher water requirement due to the addition of marble waste appears to be overcome with the use of superplasticizers, as also observed by Ergün et al. (2011) [83]. Geso˘glu et al. (2012)[90] investigated the effect of up to 20% marble powder in place of the concrete binder, maintaining a water/cement ratio of 0.35. The authors observed that higher marble powder content necessitated increased amounts of superplasticizer to maintain the same slump flow. As per the findings of Rodrigues et al. (2015)[91] , the use of superplasticizers is strongly advised. In their observations, the absence of superplasticizers resulted in detrimental impacts on the mechanical properties of concrete when up to 10% of marble waste replaced cement. The authors further noted an augmentation in compressive strength in concretes incorporating up to 5 wt. % of waste as a partial substitute for cement.

The feasibility of utilizing this waste while still meeting the requirements set by specific legislations has already been explored. Alyamaç et al. (2015) [92] identified an ideal proportion of 40 vol% of waste while still complying with the TS EN 197-1 standard (TSE, 2002)[93]. This standard specifies compliance criteria, including physical and chemical requirements for commercial cement. Aruntas et al. (2010) [94] demonstrated that samples containing up to 10 wt. % of waste in cement mixtures resulted in compressive strengths exceeding 30 MPa at 7 days, also aligning with the TS EN 197-1 standard[93].

Li et al. (2018)[3]replaced an equal volume of cement with waste (as a filler) without altering the mixture proportions. In contrast to the typical approach (i.e., replacement of cement or aggregates with waste), the authors substituted the cementitious paste with stone waste. By employing this approach, a 33% reduction in the cement content within the mixture was achieved, leading to a decrease in shrinkage and enhancing resistance to water and carbonation. Rana et al. (2015) [95] substitute 10 wt.% of Portland cement with stone waste was accomplished without compromising the concrete's mechanical properties, as noted by the authors. They also observed that increased surface area of the stone waste resulted in decreased workability of the concrete. Mashaly et al. (2016)[96]replaced up to 20 wt% of cement with marble sludge and observed an enhancement in the mechanical properties of the resulting concrete without altering the hydration products. Vardhan et al. (2015) [97] revealed that replacing cement with marble powder up to 10% did not undermine the mixture's characteristics. In contrast to earlier research, it was observed that workability actually increased. The authors did not provide explanations for this enhanced workability, but the stone waste used in the study by Vardhan et al. (2015)[97] possessed a surface area that was approximately 50%

smaller than that used by Rana et al. (2015)[95]. The enhanced workability may be clarified by the larger particle size of the waste.

Singh et al. (2017b)[98] showed that marble sludge can effectively replace 10–15% of cement. The authors noted an elevation in chloride permeability resistance with a replacement of up to 15%. However, resistance to carbonation declined with a growing addition of waste. They attributed the improvement of chloride permeability resistance to pore refinement and the reduced carbonation resistance to the lower bicarbonate alkalinity of the waste (89 mg/L) compared to that of cement (113 mg/L). Mechanical properties were maintained for waste incorporation of less than 15%.

Some researchers have specifically delved into the endurance of hydration products, particularly in prolonged durations and challenging conditions such as high temperatures, marine environments, and adverse weather effects. Kelestemur et al. (2014)[99] explored the compressive strength of mortars with marble dust replacing up to 50 wt% of sand under elevated temperatures. Samples were exposed to the maximum temperature for 1 hour. Based on the authors' data, a noticeable increase in porosity is discernible, but the authors assert that marble dust enhanced compressive strength. However, this enhancement lacks robust evidence, as glass fibers were concurrently used in a random manner.

Khodabakhshian et al. (2018)[100] assessed the properties and durability of sixteen concrete mixtures incorporating powdered marble waste as a partial substitute for mortars, replacing cement and fine aggregates. Findings revealed that substitutions of up to 5 wt% improved the physical characteristics of the concrete, and with 10 wt% of marble powder, the properties were comparable to the control. Due to the high fineness of marble waste, exceeding that of fine aggregates, the filler effect is likely present. The authors also posit that the $CaCO_3$ from marble sludge can react with the cement paste to form calcium carboaluminates. They argue that calcium carboaluminates hydrate in the interfacial transition zone, altering the surface of the aggregate to a rougher texture and enhancing the bonding strength of paste-aggregate. Lastly, it is mentioned that the rate of hydration of alite is accelerated in the presence of $CaCO_3$, resulting in a shortened induction period. However, they did not scrutinize the microstructure of mortars to provide substantiating evidence for these claims.

Gencel et al. (2012)[101]partially substituted the aggregates of cement-paving blocks with marble waste. The authors noted that as the waste content increased, there was a decrease in mechanical strength. However, durability against freeze-thaw and resistance to abrasive wear exhibited an increase. Besides the technical characteristics, viability assessments demonstrated that the expense associated with concrete decreased from 34 to 30 US$/m3 (~10%) when utilizing 40% of marble waste as an aggregate, equating to 0.1 US$/m3 for each percentage of waste added.

Alternative investigations utilized marble waste to enhance the characteristics of fresh and cured states in self-compacting concretes (SCC). The widespread application of SCC in construction stems from its amalgamation of high performance and easy deployment without the necessity for vibration and additional procedures. Crafting this type of concrete demands the utilization of extremely fine aggregates and additives, which impact the fresh state performance of concrete and are typically costly. Hence, stone waste emerges as a potentially valuable resource due to its elevated fineness and particles smaller than 200 μm. These attributes prove effective in promoting the cohesion of mortar and concrete, devoid of energy loss during workability, a trait typical in other ultrafine mineral additions like silica fume [102].

Sadek et al. (2016) [103] employed three types of stone waste (single or mixed marble and granite powder) in SCC. The outcomes indicated that up to 50 wt% of the material could serve as a mineral additive. Corinaldesi et al. (2010) [102]demonstrated that a highest mechanical strength was achieved with a 10% substitution of commercial sand with marble powder (~52 MPa) with consistent workability in the fresh state. In the initial stages, the authors also identified a positive filling effect on the samples.

Literature has documented durability studies for SCC. Tennich et al. (2017)[104]assessed the integration of marbles, marble tiles, and gravel into distinct self-compacting concretes subjected to sulphate attack, with samples immersed in seawater and a sodium sulphate solution. The findings indicated that the inclusion of stone waste positively influenced the durability of these concretes, enhancing compaction and reducing sulphate ion permeation that might be present in the air, water, and aggressive soils.

Vardhan et al. (2019)[105]scrutinized the strength, permeation, and microstructure of concretes with marble waste partially substituting river sand as a fine aggregate. According to the authors, the maximum enhancement was achieved at a 40% replacement level. They attributed the strength improvement to the filler effect. The enhanced mechanical properties are likely attributed to the higher density of stone waste compared to sand. The increased fineness and angular-shaped particles, however, resulted in a loss of concrete workability.

Table 3.1-1: Compiled data from published works that used stone waste in mortar and concrete

| Reference | Stone waste as a replacement of | Content (wt.%) | Improvements on | Deterioration on |
|---|---|---|---|---|
| Ergün et al. (2011) [83] | Cement | up to 5.0 | Compressive strength | |
| Aruntas et al. (2010) [94] | Cement | up to 10.0 | Compressive strength | |
| Rana et al. (2015) [95] | Cement | 10.0 | Durability | |
| Mashaly et al. (2016) [96] | Cement | 20.0 | Mechanical properties | |
| Vardhan et al. (2015) [97] | Cement | up to 10.0 | Workability | |
| Singh et al. (2017b) [98] | Cement | 10.0–15.0 | Chloride permeability resistance | Carbonation resistance |
| Omar et al. (2012) [87] | Cement | up to 15.0 | Compressive strength | |
| Khodabakhshian et al. (2018)[100] | Cement | up to 10.0 | Compressive strength | |
| Rodrigues et al. (2015)[91] | Cement | Up to 5.0 | Compressive strength | Workability |
| Sahan Arel et al. (2016) [89] | Cement and Fine aggregates | 5.0–10.0 (in place of cement) ; 50.0–70.0 (in place of aggregates) | Compressive strength | Workability |

| | | | | |
|---|---|---|---|---|
| Gesoglu et al. (2012) [90] | Fine aggregates | 5.0–10.0 | Compressive strength | Workability |
| Gencel et al. (2012)[101] | Fine aggregates | 40.0 | Durability by freeze-thaw and the resistance to abrasive wear | |
| Corinaldesi et al. (2010)[102] | Fine aggregates | 10.0 | Mechanical strength | |
| Tennich et al. (2017) [104] | Limestone filler in selfcompacting concretes | | Durability | |
| Vardhan et al. (2019) [105] | Fine aggregates | 40.0 | Mechanical properties | |
| Khyaliya et al. (2017)[88] | Fine aggregates | 25.0–50.0 | Mechanical performance, workability, and durability | |

### 3.2 Ceramic formulations

Numerous investigations have scrutinized and characterized slurry originating from the cutting of granite, quartzite, and marble stones, contemplating their integration into conventional ceramic formulations. Positive outcomes were achieved by Yesilay et al. (2017) [106], who explored the feasibility of incorporating these materials (up to 27 wt%) into the fabrication of artistic stoneware clay bodies, sintered at 1160 ◦C. For these products, plasticity and formability are of paramount importance. The authors assert that the addition of up to 27% of stone waste did not adversely affect the dimensional stability of the product. However, a substantial increase in water absorption (ranging between 11 and 14%) was reported compared to the standard sample (~7%). Although porosity was not directly assessed by the authors, the elevated water absorption implies a probable occurrence linked to the release of CO2 from marble waste.

Beyond application in white ceramic formulations, certain researchers have explored the utilization of these materials in red ceramic formulations [107]. Menezes et al. (2005) [108]investigated the potential use of waste from granite cutting as an alternative raw material for manufacturing bricks and tiles. The authors observed a gradual reduction in plasticity with increasing additions of stone waste, yet not to the extent of surpassing the defined values required for extrusion. Results also indicated that the waste exhibits a chemical composition akin to commercial raw materials, encompassing clays, quartz, and feldspar. Samples with up to 50% waste content exhibited water absorption values below the recommended maximums of a percentage of 25% for bricks and 20% for tiles, along with minimum modulus of rupture values of 5.5 MPa for bricks and 6.5 MPa for tiles. Nonetheless, a discernible trend emerged wherein higher stone waste content correlated with increased porosity, particularly evident in the case of bricks. This phenomenon is ascribed to the increased resistance to heat, the higher refractoriness is acknowledged of stone waste compared to the clay used in the composition.

Segadaes et al. (2005)[109] harnessed waste resulting from the sawing of marble and granite in a concoction centered on red clay to manufacture floors and tiles. The outcomes indicate that the incorporation of approximately 30 wt% did not instigate adverse effects on the mechanical attributes of the end product and, additionally, facilitated the utilization of lower temperatures during the firing process. The porosity induced by the decarbonization of calcium carbonate was counteracted by the liquefied substance formed primarily due to the presence of alkaline and iron oxides, which functioned as fluxing agents.

Saboya et al. (2007)[110]introduced waste derived from marble into ceramic bricks fired within the temperature range of 750 to 950 ◦C. The authors documented a reduction in plasticity with the inclusion of waste, necessitating elevated water content for extrusion. Acchar et al. (2006)[111]employed waste compositions comprising marble and granite ranging from 10 to 50 wt.% in the firing of ceramic items under elevated temperatures spanning within the range of 950 to 1150 degrees Celsius. The authors asserted that residue from granite and marble processing could be incorporated into the material derived from clay without adversely affecting the properties of the sintered red-clay products. Nevertheless, there was an escalation in water absorption and a decline in both apparent density and flexural strength, except in the case of the composition featuring the utmost waste content (50 wt.%) at the maximum temperature (1150 ◦C) [110]. Interestingly, in this scenario, flexural strength exhibited improvement despite higher porosity levels. This phenomenon was attributed to the waste acting as a fluxing agent, fortifying the matrix.

Vieira et al. (2004)[112]scrutinized the integration of granite waste (up to 40 wt.%) in red ceramic formulations. The authors also detected a decrease in plasticity with the incremental addition of stone waste. However, as the collaborating factory, where the study took place, preferred excessively plastic formulations to alleviate equipment wear, the loss of plasticity posed no hindrance to extrusion. In contrast to prior works, the authors demonstrated an augmentation in thermal shrinkage and bulk density with a gradual reduction in water absorption with up to 40 wt.% of waste. Nonetheless, the flexural strength remained minimally altered, considering the standard deviation.

Torres et al. (2004)[113] delved into various formulations with granite-derived waste substituting feldspar in porcelain tiles, owing to the similarity between waste and feldspar chemical compositions. According to them, depending on the dosage of waste and sintering temperature, it is plausible to achieve superior properties compared to commercial porcelains, particularly regarding water absorption and flexural strength.

Torres et al. (2007) [114]employed granite and quartzite-based waste in stoneware tiles crafted from red clay with substantial levels (60–70 wt.%) of integration sintered between 1100 and 1200 ◦C. The authors noted flexural strengths surpassing triplicate and a reduction in absorption of water increased by a factor exceeding tenfold with the incorporation of granite sludge.

Bilgin et al. (2012)[115] incorporated marble waste (up to 80 wt.%) in the production of industrial bricks sintered within the temperature range of 900–1000 ◦C. According to the authors, the addition of up to 10 wt.% did not yield significant changes in the technical properties of the final product. Nevertheless, their results illustrated a conspicuous trend towards increased uptake of water and decreased the majority of the tested samples exhibited notable flexural strength.

Eliche-Quesada et al. (2012)[116] fashioned bricks containing marble waste (5–20 wt.%) that underwent sintering at temperatures of 950 and 1050 ◦C. They asserted the feasibility of incorporating a maximum of 15 wt.% of waste with no notable deterioration in mechanical characteristics observed when specimens are fired at 1050 ◦C. Lower temperatures resulted in an increase in open porosity, thereby diminishing mechanical properties.

Table 3.2-1 encapsulates the scrutinized literature that employed incorporating stone waste in the manufacturing of ceramic goods. The primary constituents utilized in crafting the ceramics, such as feldspars, kaolin, clays, and quartz, are predominantly characterized by silica and alumina. The amalgamation of these elements at elevated temperatures engenders mullite. Mullite, an aluminosilicate, remains stable under ambient conditions and spans a compositional range from 3:2 to 2:1 in the $Al_2O_3$ to $SiO_2$ ratio. While the generation of mullite from starting materials with micrometer-scale particle sizes necessitates temperatures surpassing ~1300 ◦C (or ~1100 ◦C with nano-scale starting materials), metastable mullite can manifest at lower temperatures within multi-phase systems, such as ceramic formulations, due to the presence of other components.

Wastes derived from granite and quartzite are similarly characterized by predominant compositions of silica and alumina. This characteristic permits their inclusion, even at high concentrations, without substantially altering the overall chemical and mineralogical composition of the intended product. Numerous authors have documented enhancements in the mechanical prowess of ceramic products utilizing this waste, demonstrating heightened flexural and compressive strength coupled with reduced water absorption. The rationale behind these accomplishments lies in the inclusion of fluxing agents in the waste's chemical composition, particularly the cumulative content of iron, sodium, and potassium oxides, predominantly falling within the range of 7–18%. Consequently, utilizing granite-derived waste in lieu of starting materials with fewer fluxing agents emerges as the pivotal factor in achieving ceramic products with augmented properties. Torres et al. (2007) [114], attaining optimal results marked by remarkable improvements in strength and water absorption, utilized waste with the highest fluxing agent content (~18% of iron, sodium, and potassium oxides combined).

To sum up, the presence of fluxing agents catalyzes the formation of phases with lower melting points. The resulting melted liquid infiltrates the pores, enhancing the bond strength of the ceramic matrix, thereby diminishing overall porosity and elevating mechanical performance.

Table 3.2-1: Compiled data from published works that used stone waste in ceramic products.

| Reference | Product | Content (wt.%) and type of dimension stone | Sintering temperature (◦C) | Main results |
|---|---|---|---|---|
| Yesilay et al. (2017) [106] | Artistic stoneware clay bodies | 10.0–27.0 (marble) | 1160.0 | Elevation in water uptake coupled with a reduction in thermal contraction. |
| Menezes et al. (2005) [108] | Bricks and floor tiles | 20.0–60.0 (granite) | 1150.0–1200.0 | Marginal elevation in water absorption alongside enhancements in mechanical performance using up to 35% of stone waste |
| Segad˜aes et al. (2005) [109] | Floor tiles | 10.0–30.0 (mix of marble and granite) | 1100.0–1150.0 | Negligible adverse alterations in physic mechanical attributes |

| | | | | |
|---|---|---|---|---|
| Saboya et al. (2007)[110] | Bricks | 5.0–20.0 (marble) | 750.0–950.0 | Augmentation in porosity and flexural strength |
| Vieira et al. (2004) [112] | Bricks | 10.0–40.0 (granite) | 970.0 | Diminution in water absorption and escalation in thermal contraction; no variations in flexural strength |
| Torres et al. (2004) [113] | Porcelain tile | 20.0–50.0 (granite) | 1140.0–1200.0 | Diminishment in water absorption and heightened flexural strength |
| Torres et al. (2007)[114] | Red-clay-based stoneware tiles | 60.0–70.0 (granite and quartzite) | 1100.0–1200.0 | A substantial upswing in flexural strength accompanied by diminished water absorption |
| Bilgin et al. (2012)[115] | Bricks | 10.0–80.0 (marble) | 900.0–1100.0 | Rise in water absorption concurrent with a decrease in flexural strength |

3.3 Environmental applications

The utilization of marble waste for environmental remediation purposes contexts, such as the elimination of hazardous extraction of elements from aqueous solutions [117][118] and gaseous environments [119], and the amelioration of acidic soils [120][121], has also been under consideration. Ercikdi et al. (2015) [122] delved into incorporating marble granules as a supplement to Portland cement employed in landfills stabilized with cement for the containment of sulphide tailings. Despite this material not demonstrating a pozzolanic effect (reactivity with calcium hydroxide), its application enhanced the mechanical performance of the landfill both in the short and long term, along with augmenting the acid buffering capacity of the tailings.

Kabas et al. (2012)[123] explored the feasibility of restoring native vegetation in a deactivated tailing pond in south-eastern Spain using marble waste and pig farming mud. The findings indicated the efficacy of these materials in fostering vegetation growth, increasing plant cover and biodiversity, and diminishing the mobility of heavy metals, such as Pb, Zn, and Cd.

The surge in industrialization has resulted in a rise in handling waste that includes environmentally toxic metals. Consequently, alternative treatments are needed to adjust the levels of these elements to comply with the standards established by environmental authorities [117].

Laboratory experiments were conducted in aqueous solutions to eliminate Al ions (Al3+) using marble powder as a sorbent material. The procedure proved successful, and stone waste did not leach under acidic conditions due to the solubility of the sorbent. Controlling this element is crucial as it is present in various compounds used globally for diverse applications, such as Al salts employed as coagulants in water treatment plants[117].

Similarly, other elements are regulated in drinking water worldwide. Fluoride is a chemical compound that can lead to adverse effects when present in high concentrations in the population's drinking water, causing dental fluorosis and, in severe cases, skeletal fluorosis. Accordingly, alternative techniques have been explored to address issues with fluoridated water. Mehta et al. (2016) [118] utilized marble powder after calcination at 650 ◦C as an adsorbent for fluoride ions in aqueous solutions. The obtained powder exhibited an adsorption capacity of 1.20 mg/g in a neutral medium after 3 h of contact and was deemed an effective adsorbent for fluoridated water.

Davini et al. (2000)[119] tested white marble stone waste from northern Tuscany "in nature" and calcined at 900 ◦C as an SO2 sorbent material for atmospheric emissions. Following calcination, the desulphurization properties of the waste surpassed ones derived from limestone available in the commercial market. This enhanced performance is attributed to a larger surface area combined with an adequate distribution of CaO pore size after heat treatment.

When contemplating these environmental applications, it is crucial to consider the other ions that the waste may solubilize during application. These considerations should be integral to the study to ensure that the removal of one toxic element does not result in the leaching of another. For these reasons, environmental applications are less explored compared to the numerous studies on traditional ceramics and cementitious materials.

Table 3.3-1: Compiled data from published works that used stone waste in environmental applications

| Reference | Product | Type of waste | Calcination temperature | Main results |
|---|---|---|---|---|
| Mehta et al., 2016[118] | novel adsorbent | marble waste powder (MWP) | 650°C, 850°C, 1000°C | MWP650 exhibited a higher removal capacity compared to MWP850 and MWP1000. Considering its economical cost and widespread availability, MWP650 stands out as an efficient adsorbent with promising prospects |
| (Davini et al., 2000) [119] | desulphurization properties of calcium hydroxide | white marble by-products | 900°C | The by-products generated during the production of white marble offer a favorable outlook for utilization as sorbents in the process of SO2 abatement for flue gases. Consequently, these by-products can be categorized as genuine secondary base materials |
| Ercikdi et al. (2015)[122] | Cemented paste backfill (CPB) | marble wastes (MW) (10–30 wt%) | 700°C-1100°C | Granulated MW and WB samples prove to be viable in mitigating the adverse effects of acid/sulphate, thereby enhancing the strength and durability of CPB samples derived from sulphide-rich tailings |
| Kabas et al. (2012)[123] | farming mud | marble waste | | Post-application, plots amended with pig slurry achieved a native vegetation cover of 25–30%, with a richness of 10 and the highest biodiversity (H=1.1–1.3) |
| (Ghazy et al., 2005)[117] | inorganic sorbent | powdered marble | 125°C | Investigations into solid marble wastes reveal their efficacy as cost-effective inorganic sorbents for the |

| | | | | |
|---|---|---|---|---|
| | | wastes (PMW) | | removal of $Al^{3+}$ ions from aqueous solutions |

3.4 Other applications

Examining the industrial applications of calcium carbonate opens avenues for specific uses of marble-derived stone waste. Notably, calcium carbonate serves as a filler or extender in paints, aiming to reduce costs while enhancing spreadability and strength. However, the presence of chromophoric agents in marble waste, such as chromium and lead, may alter the paint's color.

A recent study by Tressmann et al. (2020)[124]delved into utilizing untreated marble waste (up to 80 wt.%) as a mineral filler in pigment-infused paints for soil and functioning as an active pigment in waterborne paints. Analyses of opacity, resistance to abrasion, defense against microbiological degradation, and resilience to weathering revealed that paints incorporating marble waste met normative specifications for economical latex paint. Notably, the best-performing formula included 30% polyvinyl acetate resin and 70% marble waste.

Given the inert nature of marble-derived waste for paints, analogous tests can be conducted with other stone waste. Lopes et al. (2019) [125] explored the impact of granite waste as a mineral filler in polyvinyl acetate latex paints with soil pigments. Promising results were achieved with a paint composition of 29% PVA resin and 71% granite waste. However, studies on the content and influence of chromophoric elements from stone waste on paint color variations remain limited.

In another approach, efforts have been made to obtain pure calcium carbonate from marble waste, making it suitable for use as a filler in the paper industry, which demands high filler purity[126].

Beyond the realm of paints, calcium carbonate, alongside carbon black, silica, and clays, is employed as a filler in natural rubber compounds to enhance physical and mechanical properties. Ahmed et al. (2013)[127] assessed marble sludge as a filler in natural rubber, noting no significant improvements compared to silica but recommending its use for cost reduction.

Expanding into polymeric matrices, Awad and Abdellatif (2019)[128] utilized dimension stone waste as reinforcement (10–50 wt.%) in a low-density polyethylene matrix, achieving enhancements in mechanical and thermal properties. Abenojar et al. (2021) [129]explored a composite material based on marble waste in a polyester matrix, aiming for improved fire resistance in building applications. Marble's impact on mechanical properties and fire resistance was attributed to the endothermic decomposition reaction of $CaCO_3$ releasing $CO_2$ around 700–800 ◦C.

Finally, considering the common use of pre-calcined dolomite and calcite as solid base catalysts for steam reforming in $H_2$ production, researchers are exploring the use of stone waste as an alternative [131].

Table 3.4-1: Compiled data from published works that used stone waste in mortar and concrete

| Reference | Product | Type of waste | Content and ratio | Main results |
| --- | --- | --- | --- | --- |
| Tressmann et al. (2020) [124] | paints | marble waste | marble waste pigment 70%, 60%, 57.5%, 47.5% | Every paint incorporating marble waste pigment (MWP) as an active pigment exhibited hiding power (HP) surpassing normative specifications |
| Lopes et al. (2019[125] | paints | granite waste | 71% of granite waste and 29% of PVA resin | The introduction of waste into soil pigment-based paint positively impacted hiding power. |
| Ahmed et al. (2013) [127] | rubber | marble sludge (MS) | Loading the filler at a rate of 60 parts per 100 parts of rubber (phr) | Leveraging the superior percentage of silica to enhance the hybrid composites strength is essential. Considering the comprehensive properties of hybrid composites, propose the utilization of marble sludge (MS) as a cost-effective extender. |
| Awad and Abdellatif et al. (2019) [128] | low density polyethylene (LDPE) | marble dust | 10–50% | The incorporation of marble dust particles into the low-density polyethylene (LDPE) matrix elevated the mechanical and thermal properties, including flexural strength, compressive strength, and hardness, of LDPE composites. |
| Abenojar et al. (2021)[129] | a composite material as floor or wall in buildings | Marble waste powder | 50 wt.% of marble and 3 wt.% of glass fiber | Polyester's mechanical properties are enhanced by marble, and the influence of glass fiber, whether fiber or mesh. The fire resistance is notably high, extinguishing the flame upon cessation. |
| Kannapu et al., 2021[131] | catalyst | activated marble waste (AMW) | AMW/Si (6, 12, and 18 wt. %) | The modification of amorphous marble waste (AMW) with SBA-15 demonstrated exceptional catalytic performance in extending the C-C bond for the self-aldol condensation of n-propanal to C6 oxygenated hydrocarbon, coupled with a commendable hydrophobic nature and optimal basicity |

## 3.5 Chapter summary

Within this chapter, the primary applications suggested for products derived from stone are presented based on extensive literature studies. While numerous potential applications exist for stone waste, they can be condensed into three broad categories: serving as fillers for binders in concrete and mortar, contributing to ceramic formulations, and finding utility in environmental applications. Tables in each catalogue summarized these applications. There are also some specific applications related to the production of paper, paints, rubber, and other polymers which are shown at the end of this chapter.

In conclusion, various applications of the stone-based products using stone waste as additives were demonstrated above in the fields of concrete and mortar, ceramic formulations, environmental restoration and some other industries like paint, paper, rubber, composite material, catalyst and so forth.

# 4. State of the art regarding the characterization and treatments of stone slurry

Broadly speaking, there are two distinct sludge categories: waste slurry originating from carbonate natural stone (referred to as CS) and that deriving from silicate natural stones (referred to as SS). Both exhibit an exceedingly fine size distribution. CS primarily consists of identical compounds found in the processed stones, such as marble, limestone. In contrast, SS is marked by a high concentration of heavy metals, a consequence of both the tools utilized during processing activities and the inherent characteristics of the original rock. Moreover, residual sludge frequently contains recognizable levels of total petroleum hydrocarbons, as indicated by N. Careddu et al. in 2016[4].

Fig 4-1: Carbonate rocks quarrying site and marble stone(resources: left: https://stonegate.com.tr/en/corporate/our-vision right: https://www.pikist.com/free-photo-inluk)

Fig 4-2: Silicate rocks quarrying site and granite(resources: left: https://www.flickr.com/photos/jsjgeology/48906507488/in/photostream/ right: https://www.geocaching.com/geocache/GC7MB66_king-alfred-the-great )

In the detailed account provided in chapter 2 concerning the cutting and polishing stages, the production of turbid water occurs both at the quarry site and in the warehouse during the quarrying process. Separating the solid fraction from the water is crucial for two reasons[67]. Firstly, water should not be squandered; treated water is collected and reused throughout the cutting and polishing phases. Secondly, dewatered sludge is easier to manage, and the costs associated with transportation and landfill activities hinge on the material's weight.

The treatment of turbid water primarily involves three alternative activities[67]:

1) Thickening through the utilization of large bags
2) Thickening using settling basins/tanks
3) Implementing filter pressing for SS and CS

Fig 4-3: Sludge treatment: Big bag[67]

Fig 4-4: Sludge treatment: settling basins/tanks[67]

Fig 4-5: Filter-press SS; Filter-press CS[67]

Fig 4-6: Semi-fluid waste (sludge or slurry)(from : https://www.scirp.org/pdf/JEP_2019021814545054.pdf)

Essential for sustainable waste management and resource utilization is the investigation into the characterization and treatment of waste materials from natural stone quarries, encompassing carbonate varieties like limestone and dolomite, as well as silicate types such as granite and basalt.

## 4.1 Characterization of stone slurry from stone quarrying and manufacturing

### 4.1.1 Physical Characterization:

The physical attributes of remaining sludge are contingent upon the raw material and the undertaken operational activities. This study presents findings related to both CS and SS. Specifically, SS can be categorized into three distinct subgroups based on their production methods[4]: sludge originating from gangsaw via abrasive steel shot (GSS), slurry obtained from multi-diamond-saw block cutter (DBC), and a composite sludge (MS) originating from both gangsaw and block cutter..

According to the investigation by N. Careddu et al. in 2016[4]:

Concerning CS, N. Careddu conducted sludge sampling (Fig 4.1-1.c) at four diverse factories, each adhering to distinct production cycles. Samples of sludge were collected at the discharge port of the filter-press, the location where the dehydrated sludge was stored. Utilizing a Serigraph 5100 Analyzer, the analysis of grain size was conducted on the solid cake produced by the filter press across all processing facilities.

In the case of SS, six different materials were sampled[4], with two of them originating from DBC (Fig 4.1-1.a), two from GSS (Fig 4.1-1.b), and two from MS. These six samples originated from three different operational facilities, where granites and gneisses sourced from the quarry basin in Verbano Cusio Ossola (VCO—NW Piedmont) were examined underwent cutting and polishing. The distribution of grain sizes was assessed through analysis sieving in accordance with ASTM standards (ASTM D421-85(1998); D422-63(1998); D1140-00; D2217- 85(1998)).

Fig 4.1-1: Dry powder samples from GSS (a); DBC dry powder samples from DBC (b); CS dry powder sample from CS (c)( refer to N. Careddu et al. in 2016[4])


Both CS and SS exhibit a notably fine size distribution, specifically in silt–clay dimensions, as illustrated in Fig 4.1-2 and Table 4.1-1, where 40% of the solid fraction measures less than 25 μm. The sludge, characterized by its incoherent nature, possesses asphyxia properties[4].

Upon examination of Fig 4.1-2 and Table 4.1-1, several observations stand out[4]:

1) CS originating from operational activities utilizing diamond tools, as scrutinized in this study (derived from Orosei limestone), displays high uniformity and a smaller $d_{50}$.
2) CS from the honing and polishing line exhibits a size distribution slightly different from CS generated in activities employing diamond tools, presenting higher $d_{50}$ and U.
3) SS (including GSS, DBC, and MS from granites and gneisses in the VCO quarry basin) demonstrates a generally coarser size distribution compared to CS, with higher $d_{50}$ and U.

Table 4.1-1: Summarizing the size distribution of residual sludge based on characteristics such as the composition of rocks and the tools used in the processing ( data from N. Careddu et al. in 2016[4])

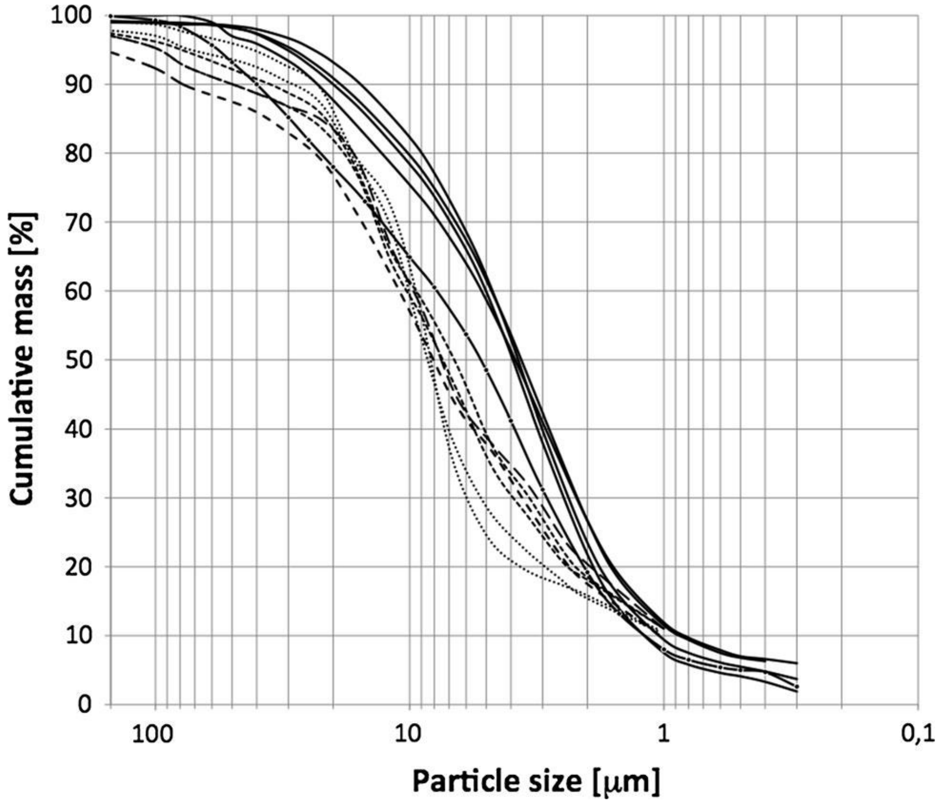
| | $d_{50}(\mu m)$ | U |
|---|---|---|
| Residual sludge from carbonate rocks (CS) | | |
| Coming from working activities employing diamond tools (average) | 3.8 | 4.8 |
| From honing and polishing line | 5.2 | 6.3 |
| Residual sludge from silicate rocks (SS) | | |
| From gangsaw using abrasive steel shot (GSS) (average) | 7.7 | 10.6 |
| From multi-diamond-saw block cutter (DBC) (average) | 8.3 | 10.0 |
| Mixed sludge (MS) (average) | 8.2 | 10.3 |

Where,

$d_{50}$ is the mean or average particle size of a mineral refers to the particle size corresponding to the cumulative frequency of 50%.

U is the uniformity of residual sludge

Fig 4.1-2: Residual sludge particle size distribution. (N. Careddu et al. in 2016[4])




Where,

Lines that are unbroken denote CS derived from the sawing or cutting process;

Uninterrupted lines featuring black dots represent CS originating from the honing and polishing lin;

Lines characterized by long dashes represent GSS;

Lines with short dashes indicate MS;

Dotted lines signify DBC.

Concerning SS, it is important to note that these materials exhibit extremely low permeability, as indicated by a very low value of hydraulic conductivity (k). The evaluation of hydraulic conductivity involved conducting falling head permeability tests (ASTM D 5084) using a modified triaxial cell, with two samples selected from the original six. The results of the falling head permeability tests revealed a hydraulic conductivity (k) of $2.3 * 10^{-8}$ m/s for DBC, $2.9 * 10^{-8}$m/s for GSS and $9.2 * 10^{-9}$10-9 m/s for MS. [132].

Except the above approach, various researches regarding the physical properties of stone slurry from marble stone, granite stone and limestone quarrying as shown in the following Table 4.1-2.

Table 4.1-2: Physical properties of stone waste used in different studies.

| Reference | Waste | Waste Process | Specific Gravity | Waster absorption (%) | Maximum Nominal Size of Particles | Fineness Modules | Specific Surface ($m^2$/Kg) |
|---|---|---|---|---|---|---|---|
| K.Chahour et al., 2020[133] | Marble | Crushing of white marble from quarry of north-east of Algeria | 2.73 | 0.39 | – | 2.2 | – |
| K. Yamanel et al., 2019[134] | Marble | Cevikler Marbles, Kayseri City of Turkey | 2.5 | 2.18 | 0.063 mm-4.00 mm | – | – |
| K. I. S. A. Kabeer, 2017[135] | Marble | Sludge | 2.65 | 0.18 | < 1.18 mm | 2.33 | – |
| R.P.S. Kushwah et al., 2016[136] | Marble | Slurry | 2.56 | 0.0 | 75 $\mu$m 4.75 mm | 0.91 | – |
| K.I.S.A. Kabeer et al., 2018[137] | Marble | Wet Marble Sludge | 2.7 | 9.89 | < 1.18 mm | 2.13 | 350 |
| V. Corinaldesi et al., 2010[138] | Marble | Powder | 2.55 | 3.0 | < 90 $\mu$m | – | 1500 |
| V. Shrivastava et al., 2014[139] | Marble | Slurry | – | – | – | – | – |
| A.A. Aliabdo et al., 2014[140] | Marble | Dust/Powder | 2.50 | – | 19.00 mm | 2.35 | 399.6 |
| A. Chawla et al., 2018[141] | Marble | Crushed Marble | 2.56 | 8.23 | < 4.75 mm | 1.45 | – |
| Y. Singh et al., 2017[142] | Marble | Dust & Slurry | | | < 4.75 mm | | |
| R. Lakhani et al., 2014[143] | Marble | Waste | 2.12–2.67 | 22–24 | 0.062 mm | – | 510–525 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| L.K. Gupta et al.，2018[144] | Granite | Granite crushed waste | 2.46 | 15.29 | – | 0.9 | – |
| J.Balasubramanian et al. ， 2016[145] | Granite | Powder | 2.19 | 0.032 | < 600 $\mu$m | – | 333.0 |
| M. R. Gowda et al.，2011[146] | Granite | Crushing | 2.56 | 9.0 | 150 $\mu$m – 4.75 mm | 2.32 | – |
| A.R.G. de Azevedo et al.， 2019[147] | Granite | Cutting Residue | 1.87 | – | 0.15 mm-9.5 mm | – | – |
| R. Lakhani et al.，2014[143] | Granite | Granite Stone Waste | 2–2.84 | 26–28 | 0.065 mm | 0.90–2.0 | – |
| H.S. Chouhan et al.，2019[148] | Limestone | Limestone Slurry | 2.7 | 8.8 | 0.01 mm 10 mm | 1.29 | – |
| | | Crushed limestone Sand | 2.7 | 3.5 | 0.01 mm 10 mm | 1.99 | – |
| E.B.C. Costa et al.，2017[149] | Limestone | Fines | | | | | |
| K. Turk et al.，2017[150] | Limestone | Limestone slurry Powder | 2.61 | 1.58 | 0.05 mm – o.3 mm | – | – |
| H.S.Chouhan et al.,2019[151] | Limestone | Slurry | 2.59 | 8.77 | 1.18 mm | 1.29 | – |
| M. Bederina et al.，2013[153] | Limestone | Limestone Crushed Sand | 2.7 | 4.3 | 0.08–5.0 mm | 2.30 | |
| B. Benabed et al.，2012[154] | Limestone | Crushed Sand | 2.68 | – | 0.04–4.75 mm | 2.21 | 350 |
| R. Lakhani et al.，2014[143] | Limestone | Waste | 2.58–2.65 | 2–4 | 0.2 mm | – | 390–412 |
| A.K.H. Kwan et al.，2014[155] | Limestone | Limestone Fines | 2.64 | 1.0 | 1.18 mm | – | 418 |
| P. Pliya et al.，2015[152] | Limestone | Eggshell Powder | 2.5 | – | < 2 mm | – | 367 |

### 4.1.2 Chemical Characterization:

Based on the study conducted by N. Careddu et al. in 2016[4], SS exhibits a notable concentration of heavy metals and total petroleum hydrocarbons (TPH), specifically:

Heavy metals are released from abrasive gangue, containing elements such as Fe, Ni, and Cr, which are associated with lime, serving as an antioxidant in steel shot gangsaws. Additionally, metal powders, including Co and Fe, employed in diamond alloys for diamond cutting tools, are also contributors to heavy metal emissions in diamond frame shots[4].

The three SS sub-categories (GSS, DBC, MS) present distinct issues related to heavy metal content. GSS is characterized by a high percentage of Ni, Cr, Cu, etc., while DBC exhibits high Co and Cu content (refer to the table below).

Unlike granite, the processing[4] of marble demonstrates a notably enhanced efficiency of diamond tools. This can be ascribed to the lack of silicate minerals, such as quartz and feldspars, which are highly abrasive, in carbonate rocks. As a result, carbonate sludge exhibits an almost negligible presence of heavy metals (refer to the table below) originating from the wear of diamond tools, in contrast to comparable sludge generated during the sawing, cutting, or polishing of granite.

The eluates were subjected to an assessment of all waste parameters, and the outcomes from leaching tests were juxtaposed with the threshold values outlined in Italian Legislative Decree no. 186 (2006). It was observed that the concentrations remained beneath the stipulated limits[4].

The TPH content is linked to mineral oils, lubricants (C12–C40), and similar materials stemming from losses in oil machinery. These substances do not mix with water and require a dedicated degradation method, such as a bioremediation process, to avert material pollution.[132].

The isolation and retrieval of heavy metals (Fe, Cr, Ni, etc.) can be achieved through magnetic or gravimetric separation. Experimental trials were carried out to investigate the potential for recovering the magnetic or metallic fraction from GSS[4]. This specific fraction, in conjunction with the 'clean sludge,' holds promise for utilization in SRM exploitation. It is noteworthy that Fe is primarily concentrated in the >100 mesh fractions, suggesting the potential for a 'size cut' for these fractions followed by wet magnetic separation.

In a broader context, CS is primarily composed of identical compounds present in treated stones such as marble, limestone, and travertine. It becomes economically compelling when it possesses a $CaCO_3$ grade exceeding 95%.

Table 4.1-3: Residual sludge main chemical characteristics (according to D.Lgs. 152/06, Italian Legislation)

| Samples | As( 4.1.3 mg/kg) | Co (mg/kg) | Cr tot (mg/kg) | Cr VI (mg/kg) | Hg (mg/kg) | Ni (mg/kg) | Cu (mg/kg) | Zn (mg/kg) | TPH (mg/kg) |
|---|---|---|---|---|---|---|---|---|---|
| GSS1 | **12.0** | **10.7** | **65.3** | ***26.3*** | <0.5 | 35.6 | **114.9** | **64.8** | **43.5** |
| GSS2 | **14.2** | **10.0** | **69.6** | ***28.1*** | <0.5 | **37.1** | 131.6 | **65.0** | 73.8 |
| DBC1 | 5.7 | 80.3 | 5.5 | **1.2** | **0.7** | 15 | **46.7** | **59.8** | **49.6** |
| DBC2 | 4.1 | 91.0 | <5.0 | **1.4** | **0.8** | 2.7 | 33.3 | **51.1** | **40.3** |
| MS1 | **10.1** | **12.2** | **59.1** | ***50.6*** | 0.6 | **40.4** | **86.6** | **61.6** | 77.9 |
| MS2 | **9.3** | **10.1** | **50.6** | ***36.9*** | <0.5 | **47.3** | **91.3** | **57.9** | **41.4** |
| CS1 | <5 | <1 | <20 | NA | NA | <20 | <10 | <30 | NA |
| CS2 | <5 | <1 | <20 | NA | NA | <20 | <10 | <30 | NA |

Where,

**Bold**: concentrations that are not negligible and fall below legal limits;

***Bold–italic***: concentrations exceeding the prescribed limits for industrial zones;

Except the above approach, various researches regarding the chemical properties of stone slurry from marble stone, granite stone and limestone quarrying as shown in the following Table 4.1-4.

Table 4.1-4: Chemical composition of Stone waste used in different studies

| Reference | Waste | $SiO_2$ | Cao | $Al_2O_3$ | $Fe_2O_3$ | MgO | $Na_2O$ | $K_2O$ | $SO_3$ | LOI |
|---|---|---|---|---|---|---|---|---|---|---|
| K.Chahour et al., 2020[133] | Marble | 0.15 | 54.86 | 0.08 | 0.04 | 1.03 | – | – | – | – |
| K. Yamanel et al., 2019[134] | Marble | 0.66 | 53.06 | 1.45 | 1.45 | 0.45 | 0.09 | 0.02 | 0.24 | – |
| K. I. S. A. Kabeer et al., 2017[135] | Marble | 1.57 | 32.19 | 0.18 | 1.18 | 19.85 | 0.03 | 0.03 | – | – |
| A.A. Aliabdo et al., 2014[140] | Marble | 1.12 | 83.22 | 0.73 | 0.05 | 0.52 | 1.12 | 0.09 | 0.56 | 2.5 |
| R.K. Khyaliya et al., 2017[88] | Marble | 3.75 | 33.12 | – | 0.13 | 17.91 | – | – | – | 45.07 |
| Lakhani et al., 2014[143] | Marble | 3–25 | 35–40 | 9.12–11.76 | 9.12–11.76 | 2–3 | 0.06–1.12 | 0.06–1.12 | – | 38–43 |
| L.K. Gupta et al., 2018[144] | Granite | 74.39 | 0.41 | 13.5 | 0.86 | 0.38 | 4.16 | 4.79 | – | – |
| J.Balasubramanian et al., 2016[145] | Granite | 2.464 | – | 1.05 | 0.57 | 1.01 | – | – | – | 2.96 |
| M. R. Gowda et al., 2011[146] | Granite | 54.65 | 11.31 | 2.17 | 0.89 | 0.23 | 0.59 | 1.15 | 1.15 | 27.84 |
| A.R.G. de Azevedo et al., 2019[147] | Granite | 63.23 | 3.34 | 15.34 | 3.53 | 3.19 | 3.04 | 5.34 | 1.52 | 1.34 |
| R. Lakhani et al., 2014[143] | Granite | 73–76 | 35–40 | 9.12–11.76 | 9.12–11.76 | 2–3 | 0.09–1.12 | 0.09–1.12 | – | 38–43 |
| H.S. Chouhan et al., 2019[148] | Limestone | 23.5 | 37.85 | 3.1 | 1.94 | – | – | – | – | 31.4 |
| K. Turk et al., 2017[150] | Limestone | 55.89 | 1.64 | 23.06 | 6.66 | 2.57 | 3.37 | 0.99 | 0.30 | 2.36 |

| | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| H.S.Chouhan et al.,2019[151] | Limestone | 23.5 | 37.85 | 3.1 | 1.94 | – | – | – | – | 31.4 |
| B. Benabed et al. , 2012[154] | Limestone | 1.0 | 52.6 | 0.2 | 0.2 | 2.1 | 0.06 | 0.04 | 0.07 | 43.63 |
| R. Lakhani et al. , 2014[143] | Limestone | 73–76 | 38–42 | 1.02–1.53 | 1.02–1.53 | 13.74–15.32 | 0.35–0.62 | 0.35–0.62 | – | 32–34 |
| P. Pliya et al. , 2015[152] | Limestone | – | 98.3 | – | – | – | – | – | 0.09 | 0.028 |

## 4.2 Treatments of stone slurry from stone quarrying and manufacturing

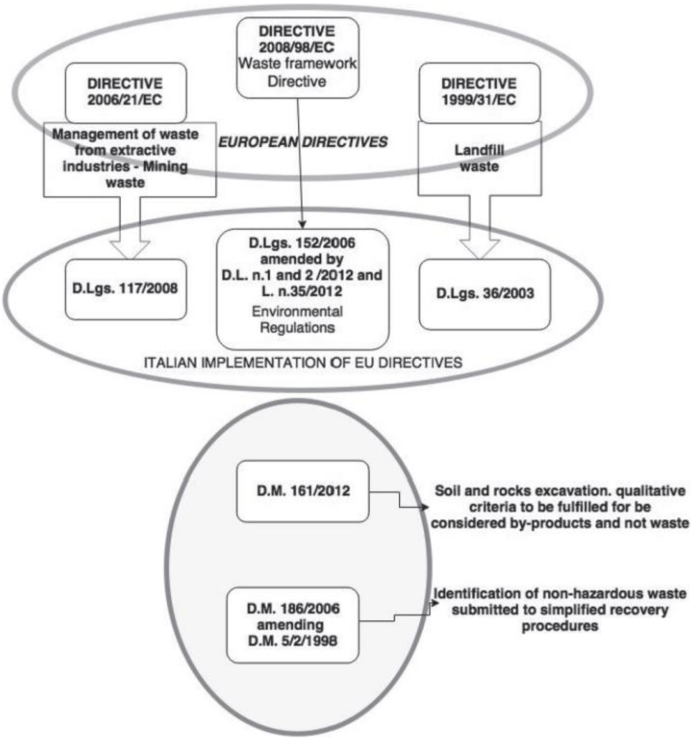
Effective handling and administration of stone slurry are imperative for reducing environmental repercussions and potentially reclaiming valuable resources.

### 4.2.1 Landfill disposal

In some cases, stone slurry can be disposed of in engineered landfills that comply with environmental regulations.

In certain instances, engineered landfills that adhere to environmental regulations may serve as a disposal option for stone slurry. The European Community, guided by Directive 2006/21/EC[156](pertaining to waste management in the extractive industries, i.e., Mining waste), Directive 2008/98/EC[157] (related to the Waste Framework), and Directive 1999/31/EC[158] (addressing Landfill Waste), furnishes directives to its member nations for the appropriate management of wastes, envisioning a sustainable future for the sector, particularly concerning critical raw materials (CRM) (EU Conference, 2016). The Italian incorporation of these European Directives is delineated in Fig 4.2-1.

Fig 4.2-1: Italian regulatory framework aligned with European Directives.




The Italian legal decree issued on April 3, 2006, under No. 152, which addresses environmental matters in Part IV and enforces the provisions of Directive 2008/98/EC [157], pertains to the “rules on waste management and remediation of contaminated sites.” It outlines measures for preventing (Article 180) and diminishing the production of hazardous waste, simultaneously supporting recycling, energy recovery (Article 181), disposal (Article 182), landfilling, and incineration. This decree allocates a six-digit CER code (matching the European Waste Catalogue, EWC) to each type of waste.

Sludge is associated with the waste code 010413[157], indicated as “wastes from stone cutting and sawing.” Current regulations expose deficiencies in the utilization of byproducts from mining and lithodid processing (such as gravel and rock dust in sludge form) for “by-products.” The European

Directive and its implementation in Italy provide guidance regarding the criteria for categorizing a substance as a by-product. The material should be utilized directly without undergoing additional treatment beyond regular industrial procedures, be an integral part of a production process, and its further use must adhere to health and environmental protection requirements (Annex 3 of D.M. 161 of 2012[159], encompassing activities like granulometric selection, volumetric reduction, and fostering natural biodegradation).

The management and disposal of sludge arising from silicate and carbonate rocks have been extensively researched[160][161]. However, an optimal solution for recovering this waste (CER 010413) or determining suitable disposal methods beyond authorized inert waste landfills, often located distant from sludge production sites, remains elusive.

The produced sludge comprises water, rock residues, and metallic fragments from cutting tools, exhibiting a moisture content ranging from approximately 35% to 45%. For landfill disposal, sludge must undergo dewatering until achieving a humidity percentage below 25% [162]. These generated wastes contribute to environmental, health, and economic challenges.

### 4.2.2 Recycling and reuse

Stone slurry frequently holds water that necessitates separation from the solid particles. Dewatering procedures encompass the utilization of mechanical or chemical methods to eliminate surplus water, rendering the slurry more manageable and transportable. Following dewatering, the solid fraction of the slurry can undergo additional treatment to isolate contaminants and reclaim valuable materials for potential reuse. Certain elements within the stone slurry may undergo recycling and find utility as construction or road base material, provided they undergo appropriate processing and cleansing.

Upon thorough treatment, stone mine waste possessing suitable stone application characteristics can be repurposed in the following manners:

1) Concrete Composite and Paving Blocks:

Hebhoub et al. [163]explored natural coarse aggregate with marble waste aggregate at levels of 25%, 50%, and 75%, while keeping a consistent water/cement ratio of 0.5, resulted in enhanced compressive and tensile strengths, according to the study. Nevertheless, as the replacement rates increased, workability declined due to the lower water absorption rates of marble waste aggregates in comparison to natural aggregates. The conclusion drawn was that marble waste has the potential to function as an alternative aggregate in concrete for purposes such as brick production, road construction, and landfill applications

Almeida et al. [164]investigated The mechanical characteristics of concrete mixtures were altered by replacing semi-liquid waste from natural stone (slurry) with sand aggregate, with volume percentages varying from 0% to 100%. Results revealed that a 5% substitution of the incorporation of stone slurry in place of sand content resulted in an improvement in compressive strength, splitting tensile strength, and modulus of elasticity.

2) Concrete Brick Making:

Alzboon et al. [165] delved into substituting both coarse and fine aggregates with marble waste of varying sizes, up to 40%, in the production of bricks made of concrete and tiles crafted from terrazzo. They also tested replacing dolomite for coarse aggregate and utilizing slurry powder in place of fine aggregates. Results indicated compliance with international standards for samples underwent testing for transverse strength, water absorption, and tile dimensions. However, an increase in marble waste sludge ratio led to a decrease in transverse strength and a rise in water absorption levels. The research findings indicated that the successful production of non-loaded bearing bricks in concrete mixtures can be achieved by substituting marble waste sludge with aggregates at levels of up to 50%..

Saboya et al.[166]investigated mixing clayey soil with marble powder at varying weight percentages. The findings demonstrated a relationship between water absorption, porosity, specific gravity, and strengths concerning both marble waste powder and firing temperature. The study found that adding 15-20% of marble waste powder to red ceramic raw material yielded optimal results within construction limits.

Under comparable circumstances, El-Mahllawy et al. [167] explored five distinct materials (clay, sand, marble cutting waste) alongside ordinary Portland cement and hydrated lime as stabilizers, the outcomes indicated a reduction in water absorption and an increase in bulk density with an extended curing period. Notably, the compressive strength exhibited a significant 50% improvement, achieving optimal results within a water content range of 10–13% for static compaction.

Further investigations by Arboleddas et al. [168] focused on producing lightweight structural clay ceramics involves incorporating marble cutting dust and utilizing waste from packaging paperboard/polyethylene.The results demonstrated feasibility and strength improvement, signaling potential for more extensive microstructural characterization and mechanical optimization.

3) Marble Sludge for Recycling in Relevant Applications:

In the preceding Chapter 3, the potential recycling and reuse of marble sludge were summarized. Key applications include:

a. **Cement Production:** Marble slurry, with its high calcium carbonate content, is considered a significant component for incorporation in cement manufacturing.
b. **Ceramics Industry:** Marble sludge powder, up to 45%, can be incorporated into ceramics production for forms, pottery, and artifacts.
c. **Brick Industry:** The addition of Marble sludge powder, with a maximum limit of 50%, is viable in the production of clay bricks.
d. **Paper Industry:** Incorporating finely ground dried Marble sludge powder, characterized by its elevated calcium carbonate content, up to 10% is feasible for fillers and pigments in the manufacturing and polishing stages.
e. **Fertilizers Manufacturing:** Finely ground marble sludge powder, distinguished by its high calcium carbonate content, is utilized in the production of calcium nitrate, a key ingredient in fertilizers.
f. **Marble Resin Products:** Mixing white calcium carbonate fine particles with polyester resin produces decorative plates in the manufacturing of marble resin products.

### 4.2.3 Chemical treatment

The application of chemicals in the treatment of waste from stone quarrying involves the utilization of diverse chemical agents or processes to address or alter the waste produced during quarrying activities. The objective is to minimize environmental impact, diminish the presence of hazardous elements, reclaim valuable materials, or repurpose the waste in an ecologically sustainable manner. Chemical treatments encompass a spectrum of processes, including stabilization, neutralization, separation, and extraction of valuable compounds. Here are several prospective chemical treatment methods for managing waste from stone quarrying:

1) Stabilization/Solidification (S/S):

Stabilization and solidification (S/S) represent methodologies in waste management that utilize chemicals to stabilize and solidify hazardous elements within waste, thereby curtailing or diminishing their mobility and potential environmental harm. The main goal of these techniques is to transfer the waste into a more steadfast and less leachable state.

In the realm of stone quarrying waste, stabilization and solidification find application in immobilizing hazardous substances, such as heavy metals or other pollutants inherent in the waste material. Several common chemicals employed in the stabilization and solidification process [169] include:

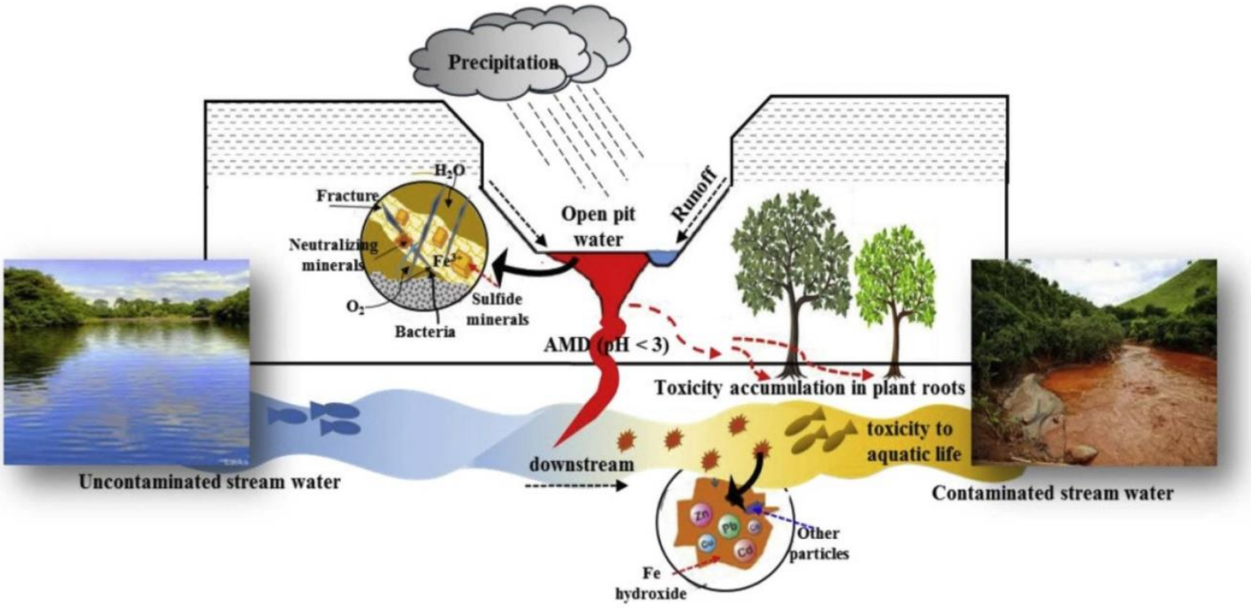
a. Lime (Calcium Hydroxide): Elevates the waste material's pH, facilitating the stabilization of heavy metals through precipitation or adsorption mechanisms.
b. Portland Cement: A frequently utilized binder that physically binds waste particles, enhancing stability and reducing susceptibility to leaching.
c. Fly Ash: A byproduct of coal combustion, employed as a binding agent to bolster the structural integrity of the waste material.
d. Bentonite: A clay mineral used to enhance the physical properties of the waste, aiding in stabilization and solidification.
e. Polymers: Employed to enhance the mechanical properties of stabilized waste, contributing to the solidification process.

These chemicals are blended with the waste material to achieve the desired stabilization and solidification. The resultant material is often utilized in construction or disposed of in a regulated manner, contingent on waste characteristics and local regulations.

2) Acid Neutralization:

Invariably, pollution arises during mineral resource extraction, with acid mine drainage (AMD) [170] standing out as a significant challenge for the worldwide mining sector. AMD primarily arises from the oxidative process of sulfide minerals upon exposure to the influence of air, water, and microbial activity, and it is prevalent in both active and disused mines extracting polymetallic sulfides and coal [170].

Fig 4.2-2: AMD formation and the corresponding contamination pathways[171]



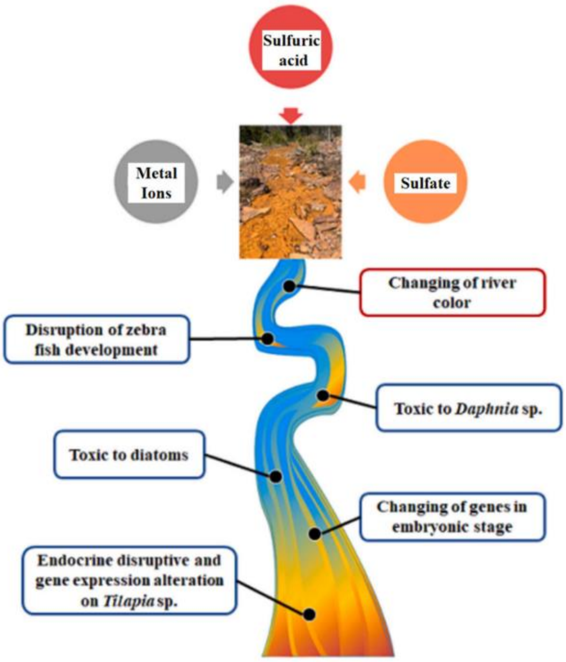

The characteristics of AMD exhibit considerable variation, influenced by diverse Environmental conditions, including weather patterns and geomorphological characteristics, at the site, and the volume of waste materials. The chemical, biological, and physical factors primarily impacting AMD generation encompass elements like air (oxygen), temperature including rainfall and water saturation levels, microbial activity, and the extent of metal sulfide exposure.

Over the past few decades, extensive reports have focused on preventing and treating AMD. The choice of a specific treatment method invariably hinges on the wastewater composition, metal ion concentrations, and the unique geographical environment. Consequently, achieving sustainable AMD treatment encounters formidable challenges.

Several sustainable treatment methods have been subject to scrutiny. Given the properties of acid mine drainage (AMD) [170], there is a significant focus on the retrieval of sulfuric acid, preservation of water resources, and the recovery of valuable components from AMD. Currently, there is a substantial amount of research[170] focused on recuperating and repurposing valuable resources from acid mine drainage (AMD), promising applications have been discovered in selective precipitation,

neutralization through chemical means, processes involving separation utilizing membranes, exchange of ions, methods involving chemical oxidation, chemical processes involving electricity, and the domain of fuel cell advancements[170]. While various treatment methods have yielded positive results, there are persistent technical challenges that require significant efforts for the practical implementation of these technologies in industry[170].

Fig 4.2-3: Summary of environmental effects of AMD[172]




Fenton oxidation technology boasts advantages such as swift response, gentle operational conditions, and straightforward apparatus[172]. The on-site creation of developing catalysts for electro-Fenton processes utilizing acid mine drainage (AMD), characterized by low pH and abundant $Fe^{2+}$, not only diminishes administering reagents for the Fenton oxidation process but also facilitates the reuse of valuable resources present in AMD[172]. However, additional investigation is necessary in the examination of the reaction mechanism linked to the creation of Fenton agents using AMD[172] as materials in processes for wastewater treatment. Subsequent research [172] should focus on the impact of various metal ions in AMD on the catalytic efficiency of the Fenton system and the optimal extension of the pH range for the reaction.

Employing acid mine drainage (AMD) [172] in beneficiation processes, like activating pyrite and chalcopyrite flotation, presents opportunities for the novel use of AMD in copper sulfide ore treatment. This method[172] not only ensures the effective retrieval of desired minerals, leading to increased profits in beneficiation, but also mitigates the adverse environmental effects of AMD at its origin, promoting environmentally friendly mining practices. Despite limited information on the application of AMD in beneficiation fields, most efforts are still at the laboratory stage. It is crucial to acknowledge that each method has its intricacies, and it is advisable to explore more integrated techniques for realizing the resource utilization of acid mine drainage[172].

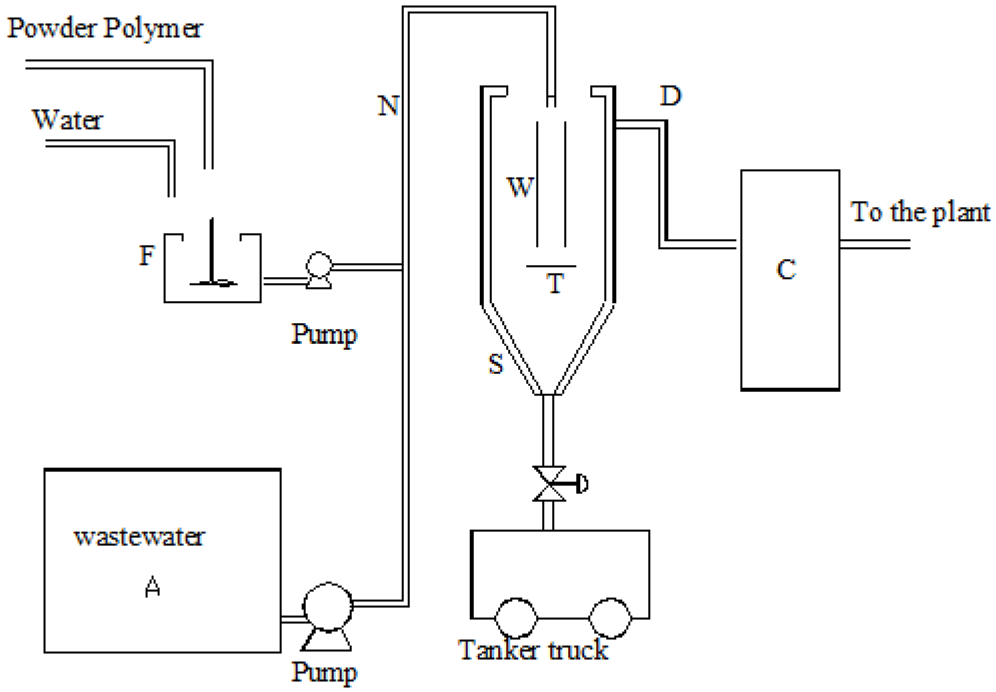
3) Flocculation and Sedimentation:

Flocculants or coagulants play a crucial role in inducing the aggregation of fine particles, ensuring efficient sedimentation and separation.

In the stone cutting industry, the water cycle initiates during the cutting process, where water serves as a cooling and lubricating agent, also acting as a medium for collecting stone dust[173]. The resultant wastewater comprises a suspension of stone powder, channeled and guided by gravity towards the wastewater treatment unit. Three distinct methods are employed for wastewater treatment [174]: plain sedimentation bonds, flocculation-sedimentation process (FSP), and filtration. The recent adoption of the latter two methods draws from the experiences of other nations, notably Italy. The flocculation sedimentation unit's flow sheet, applied in the local stone cutting industry, is depicted in Fig 4.2-4.

The effluent from the facility accumulates in tank (A) and is transferred via a feed conduit (N) into the settling basin (S) [174]. Prior to entering the sedimentation tank, the slurry undergoes treatment with a flocculation agent (polymer), dispersed in water within vessel (F) equipped with a stirrer. The resulting polymer emulsion is injected into the pipeline for feeding (N). The slurry that has undergone flocculation, fed into the sedimentation vessel, descends through an inner pipe (W), concentric with the sedimentation vessel. The flow changes direction upon impinging on a circular disk (T) at the bottom of the cylindrical section. Flocculated particles settle within the conical section, as clear water rises through the annular region into a specifically engineered upper channel. Subsequently, it gravity-flows into a clean water tank (C). Finally, water is gravitationally recycled back to the cutting process. Commercial polymers function as flocculating agents, enhancing the efficiency of particle separation in FSP. This enlargement of particle size allows calcium carbonate particles to flocculate, resulting in increased settling velocity. Recycling the resulting powder waste has not garnered practical interest thus far. The solid waste can be recycled either chemically into lime, gypsum, cement, or utilized directly as fillers in industries such as construction chemicals, paper, and paints[173].

Fig 4.2-4: The stone cutting plant in Palestine employs an industrial process involving flocculation and sedimentation [173]




This study proposes practical recommendations to enhance The planning and functioning of the process involving flocculation and sedimentation employed by the stone cutting industry in Palestine. Notably, high separation efficiency (95%) was achieved through plain sedimentation, without the use of flocculating agents, at flow rates generating a liquid upward velocity that is less than the settling velocity of the suspended particles[174]. This operational approach proves suitable for plants operating at partial capacities or for smaller facilities with total wastewater volumes below 185. Embracing this pragmatic approach can lead to cost savings on flocculating agents and a reduction in environmental impacts.

The separation efficiency experienced a decline with increasing flow rates due to the elutriation of fine particles. Interestingly, no significant variations in separation efficiency were noted when altering the depth of the impinging desk position. These findings underscore that separation efficiency hinges solely on the net upward velocity rather than the flow field. Consequently, it suggests that shorter sedimentation vessels can be employed for steady-state operation, as they are as effective as the presently utilized longer vessels. Such a proposed design modification not only enhances operational efficiency but also reduces the capital cost associated with sedimentation vessels[174].

### 4.2.4 Biological Treatment

The biological treatment of stone quarrying waste involves the utilization of microorganisms or biological processes to break down or alter both organic and inorganic components present in the waste. While this approach is gaining recognition for its environmental friendliness, it's crucial to acknowledge that the specific methods employed can fluctuate based on the attributes of the waste and the treatment objectives.

One strategy involves leveraging microorganisms to induce the precipitation of calcium carbonate or other minerals, thereby enhancing the structural stability of the waste. This process contributes to the stabilization of fine particles and a reduction in permeability.

When considering the broader context of biogeochemical processes, the challenges associated with their field implementation, and societal needs, the most promising applications are likely to be those that are straightforward to implement, offer unique solutions to specific problems, come with cost-effectiveness and the potential for swift adoption by both industries and society. In line with this perspective, a qualitative evaluation of 24 distinct biogeochemical applications was conducted, assessing them against criteria such as cost, The simplicity of implementation, likelihood of success, and social approval were considered during the 2012 Workshop activities. Table 4.2-1 delineates the applications and their rough ranking based on these criteria[175].

Table 4.2-1: Assessing alternative applications and their potential involves examining factors such as the feasibility of implementation, the likelihood of success, cost-effectiveness, and societal approval[175]

| Application | Implementation Easy: 5 Difficult: 1 | Probability of success High: 5 Low: 1 | Cost/viability Economic: 5 Expensive: 1 | Societal acceptance High: 5 Low: 1 | Total score out of 20 |
|---|---|---|---|---|---|
| Structural repair | 5 | 5 | 3 | 5 | 18 |
| Erosion control | 4 | 5 | 4 | 5 | 18 |
| Co-precipitation/immobilization of contaminants | 5 | 4 | 4 | 5 | 18 |
| Dust mitigation | 4 | 5 | 4 | 5 | 18 |
| Ground improvement for rural roads | 5 | 5 | 3 | 4 | 17 |
| Shallow carbon sequestration | 5 | 3 | 4 | 5 | 17 |
| Leak management | 4 | 3 | 4 | 5 | 16 |
| Rehabilitation of ancient monuments | 3 | 3 | 5 | 5 | 16 |
| Ground improvement for urban road sub-grading | 5 | 3 | 3 | 4 | 15 |
| Soil liquefaction mitigation (MICP) | 3 | 5 | 3 | 3 | 14 |
| Ground improvement for ash ponds | 1 | 4 | 4 | 5 | 14 |
| Recycling/reuse of dredging materials | 3 | 2 | 3 | 5 | 13 |
| Soil liquefaction mitigation (biogas) | 3 | 3 | 3 | 3 | 12 |
| Enhanced water/oil/gas recovery | 1 | 3 | 3 | 5 | 12 |

| | | | | | |
|---|---|---|---|---|---|
| De-desertification | 1 | 5 | 1 | 5 | 12 |
| Sediment weakening by fluidization | 3 | 2 | 3 | 3 | 11 |
| Underground creation (pipeline) | 3 | 4 | 1 | 3 | 11 |
| Stabilization of sinkholes | 1 | 3 | 2 | 5 | 11 |
| Landfills as new energy resource | 3 | 4 | 1 | 2 | 10 |
| Construction products (bricks) using soil-bio cementation | 2 | 4 | 1 | 3 | 10 |
| Water storage | 3 | 2 | 2 | 2 | 9 |
| De-swelling of clays | 1 | 1 | 1 | 4 | 7 |
| Deep carbon sequestration | 1 | 1 | 1 | 3 | 6 |
| Underground creation (tunnel) | 1 | 1 | 2 | 1 | 5 |

### 4.3 Chapter summary

This chapter summarized the physical and chemical characterizations according to researches approaches about the stone slurry quarrying waste and general treatments for these waste including landfills, recycling and reuse of stone slurry, chemical treatments and biological treatments methods for the recovery of these wastes in contribution to the sustainability construction.

## 5. State of the art for cement fillers for thin material application

Cementitious additives, known as fillers, play a crucial role in enhancing or modifying the attributes of cement-based mixtures. Their incorporation into cement products aims to elevate workability, durability, strength, or other targeted characteristics. Typically, these fillers consist of finely ground materials intimately blended with cement to attain specific performance objectives.

Presently, a diverse array of fillers is available in the market, including silica fume (micro-silica), fly ash, metakaolin, ground granulated blast furnace slag (GGBFS), limestone powder, natural pozzolans, rice husk ash (RHA), plasticizers, superplasticizers, cellulose fibers, nanoscale materials, and more.

The selection of a particular filler is contingent upon the unique application requirements and the envisioned properties of the final product. Achieving optimal proportions and combinations of fillers involves a meticulous process of testing and experimentation to ensure the best performance aligned with the intended use.

Table 5-1: comparations of common fillers application

| Reference | Fillers | Physical characteristics | Chemical characteristics | Functions | Pros | Cons |
|---|---|---|---|---|---|---|
| Mehta, P. K., & Monteiro, P. J. M. et al. (2014) [176] | silica fume | Very fine particles, usually with a high specific surface area. | Highly reactive amorphous silicon dioxide. | Strength and durability are augmented through the filling of voids and improvement of the cementitious matrix. | Enhances compressive and flexural strength while diminishing permeability and boosting corrosion resistance. | Elevated expenses and possible complications in handling owing to its finely granulated nature. |
| Malhotra, V. M., & Mehta, P. K. et al. (2002) [177] | fly ash | Finely divided powdery material. | Consisting of silicon, aluminum, iron, and calcium oxides | Workability is heightened, and the heat of hydration is diminished. | Elevates long-term strength and minimizes the reliance on cement, consequently mitigating the risk of alkali-silica reaction. | Composition inconsistencies contingent on the source and quality, leading to a variability in properties. |
| Alice T. Bakera et al. (2019) [178] | metakaolin | Finely divided powder. | Highly reactive amorphous aluminum silicate. | Early-age strength sees improvement, and permeability is reduced. | Augments strength, durability, and workability. | Greater expenditure in contrast to alternative supplementary cementitious materials. |
| Yanchen Oinam et al. (2022) [179] | ground granulated blast furnace slag (GGBFS) | Glassy granules. | Comprises calcium and other base silicates and aluminates. | Workability is enhanced, the heat of hydration is mitigated, and long-term strength experiences improvement. | Heightens resistance to sulfate and chloride attacks, offering superior protection in aggressive chemical environments. | Potential necessity for specialized storage and handling protocols due to its fine texture. |
| Barbara Lothenbach et al. (2008) [180] | limestone | Finely ground powder. | Primarily composed of calcium carbonate. | Functioning as a filler, it furnishes nucleation sites for hydration products. | Decreases clinker content, fostering sustainability and lessening environmental impact, all the while refining the workability | Possible impacts on setting time, demanding adjustments to mix proportions. |

| | | | | | | |
|---|---|---|---|---|---|---|
| | | | | | and durability of concrete. | |
| Blaise Ngwem Bayiha et al. (2023) [181] | natural pozzolans | Varied, depending on the specific natural pozzolan (e.g., volcanic ash, calcined clay). | Contains amorphous or crystalline silica and alumina. | Workability is refined, and supplementary cementitious properties are conferred. | Improves properties akin to other pozzolanic materials, leading to an overall reduction in the cost of concrete production. | Composition and properties may vary depending on the source. |
| Ayesha Siddika et al. (2020) [182] | rice husk ash (RHA) | Finely divided ash. | Contains high amounts of silica. | Strength is bolstered, and permeability undergoes reduction. | Utilizes agricultural waste to advocate for sustainability. | Quality fluctuations associated with the combustion process. |

5.1 Historical experience of cement fillers

The inaugural utilization of fillers as substitutes for binders can be traced back to the construction of Elephant Butte and Arrowrock Dams (Fig 5.1-1), overseen by the US Bureau of Reclamation from 1912 to 1916 [183]. In a bid to enhance economic efficiency, approximately 45–48% of the cement was substituted with ground granite or sandstone extracted during the construction phase. These materials underwent grinding to meet the 850 μm sieve requirements, followed by a blending process with coarse cement, and subsequent intergrading until 90% passed through the 75 μm sieve. The resultant concrete exhibited equivalent compressive strength at the one-year mark when compared to concrete employing solely the original coarse cement. This equivalence was achieved by compensating for dilution through an augmentation in clinker fineness during the second grinding [184]. In 1935, the downstream face and spillway channel of the Arrowrock Dam underwent repair due to frost-induced damage degradation [185]. Remarkably, these dams, now a century old, continue to serve their intended purposes.

Fig 5.1-1: 100-year-old Arrowrock arch Dam, Idaho, USA, portrayed in 2014. Water height is 78 m. Source: US Bureau of Reclamation https://c2.staticflickr.com/8/7308/13977827009_07278638a7_b.jpg.

In the 1930s, the Civil Engineering Department at the University of California, Berkeley, embarked on a significant and meticulous 10-year investigation led by R.E. Davis and team [186]. This groundbreaking study delved into cements enriched with diverse fillers and artificial pozzolans, including calcined clay and combinations thereof. A pivotal aspect of the experiment involved the inter-ground replacement in cement, with 20% limestone filler and 45% granite filler (LF and GF, respectively, as depicted in Fig 5.1-2a and Fig 5.1-2b).

The figures reveal that, despite the consistent fineness of the cement and nearly constant water content, a 20% filler replacement surpassed the original cement in compressive strength over the long term, under both wet and dry curing conditions. The authors noted favorable outcomes for blends involving limestone filler and various pozzolans like calcined Monterey Shale, siliceous clays, and pumicite [187]. It's worth noting that during the early 1900s in the USA, substituting clinker with inert materials or slag and limestone was labeled as 'cement adulteration' [188]. This ethical perspective likely introduced bias into technical discussions and may have adversely influenced the public perception of fillers in numerous cement markets.

Fig 5.1-2a: Influence relative tensile strength of filler mineralogy and content (Data from Davis et al. [187], University California, Berkeley.)

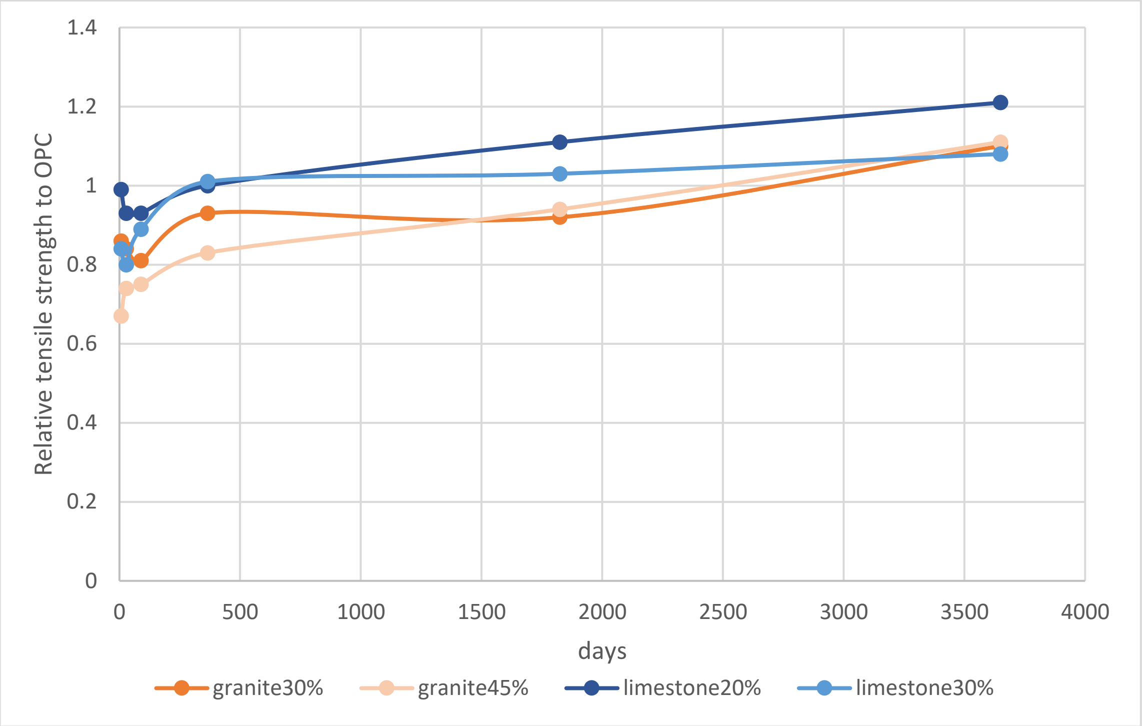
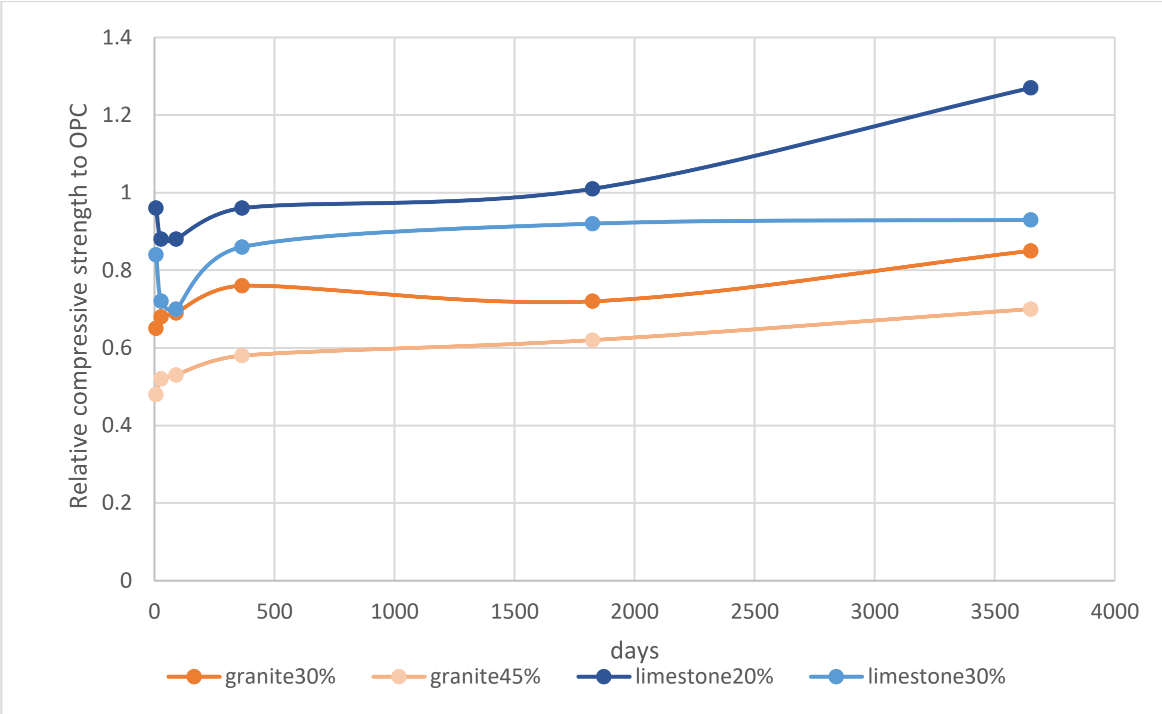



Fig 5.1-2b: Influence relative compressive strength of filler mineralogy and content (Data from Davis et al. [187], University California, Berkeley.)




Where,

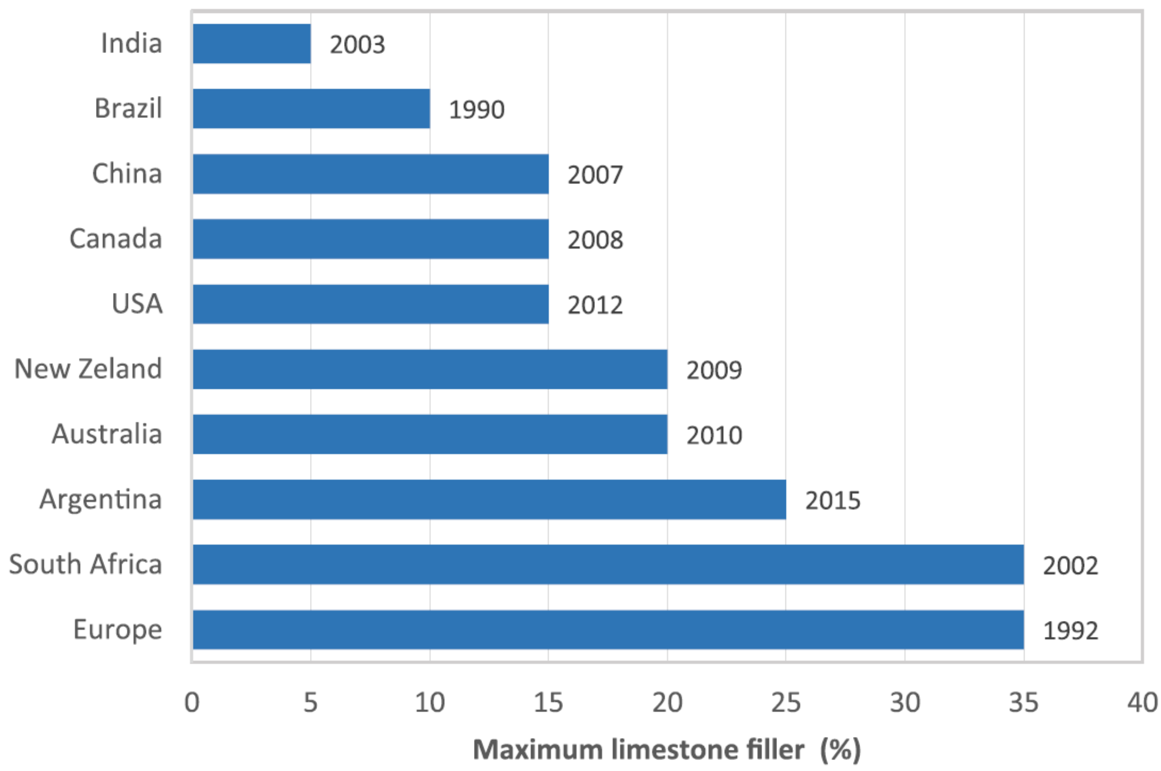
LF xx= limestone filler content, wt.%;

GR xx= granite filler content, wt.%;

Over a decade, the relative compressive strength of standard 1:3 Ottawa mortar remained consistent, with cements exhibiting nearly identical fineness. The integration of limestone as a clinker replacement likely emerged in response to the 1970s oil crisis, as suggested by P. Wedding, L. Mayfield, et al. in 1988 [188]. By 1983, Canada had incorporated a 5% limestone limit in normal Portland cement (CAN3-A5-M83). Concurrently, European discussions were underway in 1988, proposing to allow 20% filler in Portland composite or Portland-filler cements [188].

In the present scenario, numerous national and international standards include provisions for fillers. The EN 197-1 cement composition standard permits up to 35% limestone filler substitution in cements[189]. While this maximum is less than that allowed for fly ash and slag, limestone filler constitutes 7% of the global average cement, surpassing slag (5%) and fly ash (4%) rates [190]. Notably, Morocco leads in reported clinker substitution with filler, averaging above 20%. A recent trend indicates a growing interest in combining limestone with other supplementary cementitious materials, including calcined clay [191]. Figure 5.1-3 illustrates the maximum limestone filler content limits across different regions.

Fig 5.1-3: Maximum limestone filler (wt.%) in standard cements. Figures represent the year of first publication of the standard [189]




Limestone filler is subject to specific compositional restrictions in nearly all countries. However, the EN 197-1 standard permits the incorporation of non-limestone fillers. This standard sanctions the inclusion of "minor mineral constituents," up to 5%, encompassing natural materials that, following suitable preparation and owing to their particle size distribution, enhance the physical characteristics of cement. These constituents can either be inert or possess slightly hydraulic, latent hydraulic, or pozzolanic properties.

## 5.2 Up-to-date experience of cement fillers

Currently, various types of fillers are under investigation. The main objective is to economize on cement in composites, thereby diminishing the cost and energy requirements associated with concrete production. A significant portion of these fillers constitutes secondary or waste materials, contributing to the resolution of landfill-related issues through their processing.

### 5.2.1 Studies related to the influence of properties of fresh mortar/concrete

➢ **Workability**

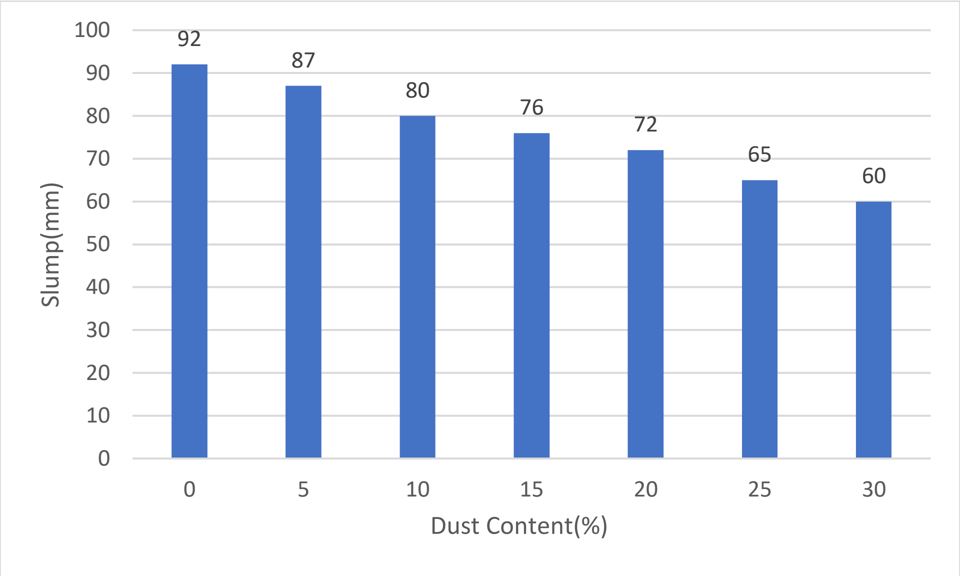
The utilization of stone dust as additive in concrete has a notable impact on workability.

In 1996, Tahir Celik and Khaled Marar et al. [192]conducted an investigation into the effects of crushed stone dust on concrete properties. Seven distinct mixes were formulated, incorporating varying percentages (0, 5, 10, 15, 20, 25, and 30) of dust used as a substitute for a portion of the fine aggregate. The results of the slump test (average of five tests) are presented in Table 5.2-1. It can be inferred that with an increase in the dust percentage in the concrete, the slump decreases (Fig 5.2-1). With an increase in the dust percentage, the fineness of the aggregate rises, leading to a higher specific surface area of aggregate particles. Consequently, more water is needed to wet the increased surfaces of particles, resulting in a decrease in workability.

Table 5.2-1: Slump and Air Content Test Results of Fresh Concretes (data from Tahir Celik and Khaled Marar et al., 1996 [192])

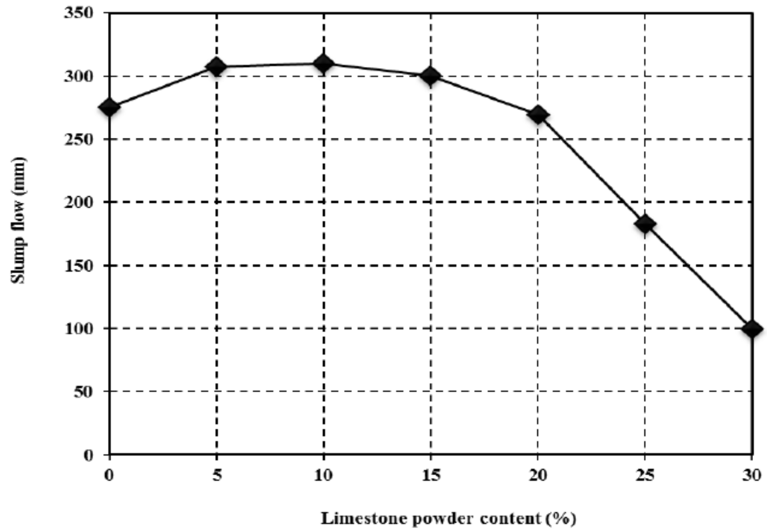
| Dust Content (%) | Slump (mm) | Air Content (%) |
|---|---|---|
| 0 | 92 | 2.77 |
| 5 | 87 | 2.40 |
| 10 | 80 | 2.15 |
| 15 | 76 | 1.9 |
| 20 | 72 | 1.73 |
| 25 | 65 | 1.48 |
| 30 | 60 | 1.28 |

Fig 5.2-1: slump test results with different dust contents



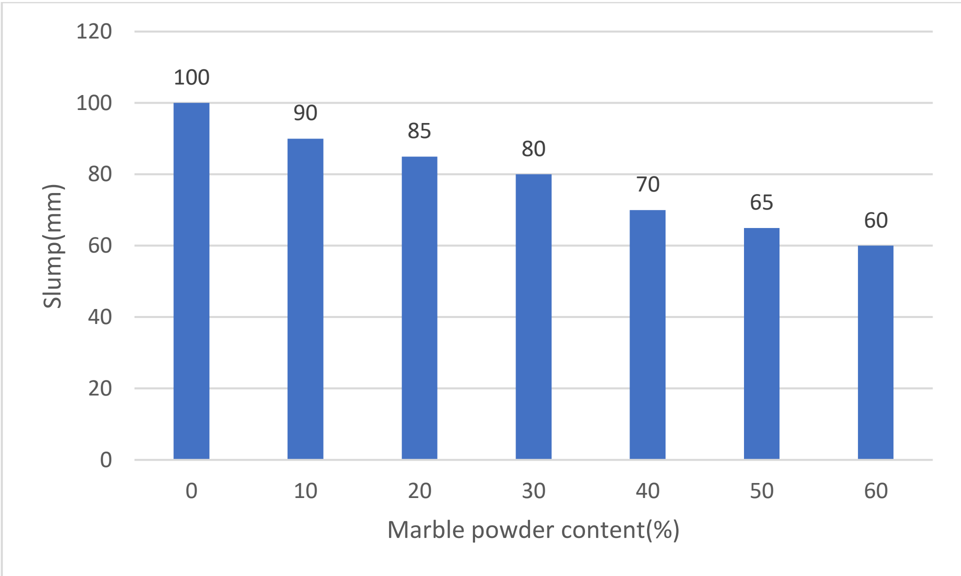

Numerous researchers conducted analogous tests, and in 2016, Benabed B et al.[193] observed that the slump flow in self-compacting repair mortar (SCRM) concrete rises with an increase in the percentage of limestone powder, up to 10%, and subsequently declines for elevated powder concentrations. This phenomenon may be elucidated by the crushed sand exhibits an elevated level of fineness and a greater specific surface area. resulting from increased fines content. Consequently, more water is needed to wet the particle surfaces, leading to a decrease in flowability. The corresponding test results are depicted below (Fig 5.2-2):

Fig 5.2-2: Conducting slump flow tests on SCRM with different proportions of limestone powder ( Data from Benabed B et al., 2016[193])



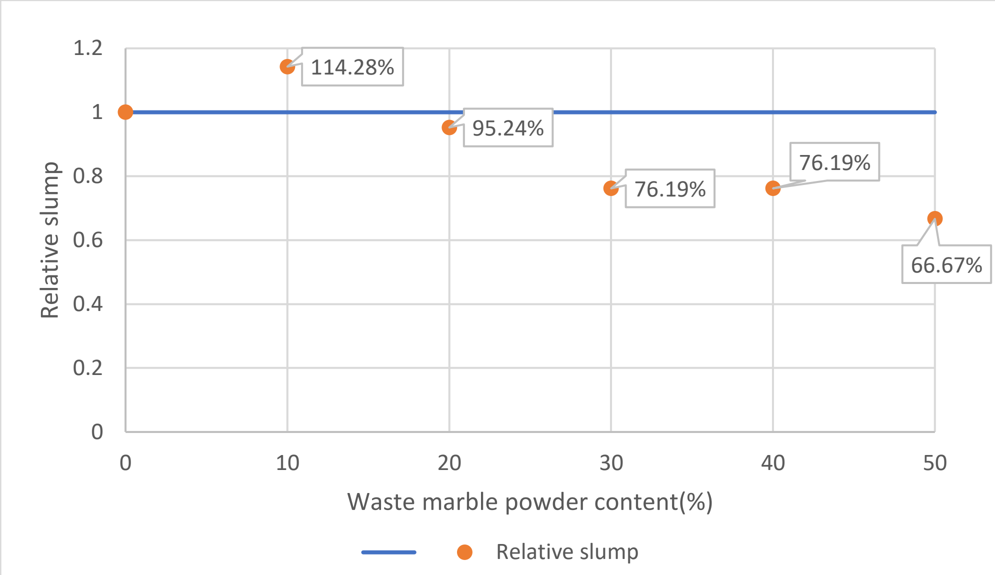

In the year of 2019, Kirti Vardhan et al.[192] argued that substituting waste marble for fine aggregates resulted in a decline in the mix's workability. This reduction occurred even though marble's lower water absorption was comparatively less than the water absorption of natural river sand.

Fig 5.2-3: slump test results with different dust contents( data from Kirti Vardhan et al.[192])




In the year 2021, Anitha Selvasofia S.D et al.[194] conducted An investigation into the mechanical characteristics of concrete through the substitution of cement with waste marble powder (WMP). The findings reveal that a 10% increase in WMP enhances workability by 14.28%. This reduction in WMP size influences the formation of concrete pores. Given its size, it ensures the thorough dispersion of materials and minimizes particle friction, consequently boosting concrete workability. However, a further increase in WMP results in decreased workability. The introduction of 20%, 30%, 40%, and 50% WMP leads to workability reductions of 4.76%, 23.81%, 23.81%, and 33.33%, respectively. The diminished workability is attributed to the irregular grain shape of WMP, contributing to a higher surface area due to its finer size, resulting in a more compact and less workable concrete, as illustrated in Fig 5.2-4.

Fig 5.2-4: the relative slump test results with different waste marble content (data from Anitha Selvasofia S.D et al., 2021[194])




➢ **Segregation and bleeding**

Segregation and bleeding, two manifestations of homogeneity loss in cement-based materials, pose challenges. Segregation becomes apparent through the settling of aggregates within mortars and concrete, whereas bleeding manifests as an excess of water rising in highly fluid concrete mixtures. Effective measures for control involve employing well-graded aggregates, using finer cement, maintaining an appropriate water-to-cement ratio, incorporating entraining agents, and adding mineral additives[195]. Ensuring homogeneous blending is crucial in minimizing the likelihood of bleeding and segregation. The incorporation of finely granulated substances plays a role in reducing bleeding and segregation by creating an extended path for water ascent, obstructing pores, and enhancing mix cohesion[196].

Various studies explore the impact of quarry dust, including marble dust, granite dust, crushed rock dust, and limestone powder, on the bleeding and segregation tendencies of cement-based composites.

In 2014, Hafez E. Elyamany et al. [197]conducted sieve stability and bleeding tests on various filler types in fresh self-compacting concrete, encompassing pozzolanic fillers (silica fume and metakaolin) and non-pozzolanic fillers (limestone powder, granite dust, and marble dust). The findings[197] suggest that incorporating granite dust and marble dust improves the resistance to segregation in Self-Compacting Concrete (SCC) compared to other fillers. For instance, in concrete with a cement content of 400 kg/m3 and a filler content of 10.0%, the segregated portion is 14.0%, 9.6%, 9.6%, 4.4%, and 3.8% for SCC with silica fume, metakaolin, lime powder, granite dust, and marble dust, respectively (refer to Fig 5.2-5). This pattern remains consistent across various filler content levels. The pattern remains consistent for concrete with 500 kg/m3 cement content. Silica fume SCC, despite its very fine nature, exhibits less segregation resistance than other fillers[197]. The outcomes of the bleeding test indicate that the properties of fresh concrete are markedly influenced by the type and amount of filler, with non-pozzolanic fillers notably enhancing resistance to segregation and bleeding. Refer to Fig 5.2-6 for a visual representation of how filler type and content impact the bleeding resistance of SCC with a cement content of 400 kg/m3. This figure underscores the significant influence of filler type on bleeding resistance, especially at a high filler content of 15.0%, while the filler type has no apparent effect on bleeding resistance at a lower filler content of 7.5%.

Fig 5.2-5: Sieve stability test results of Flow-able concrete. [197]

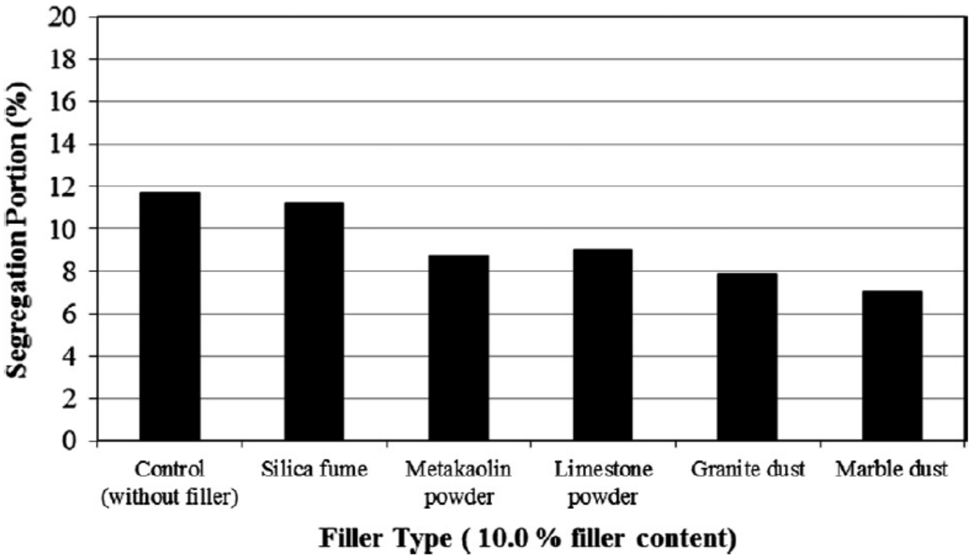
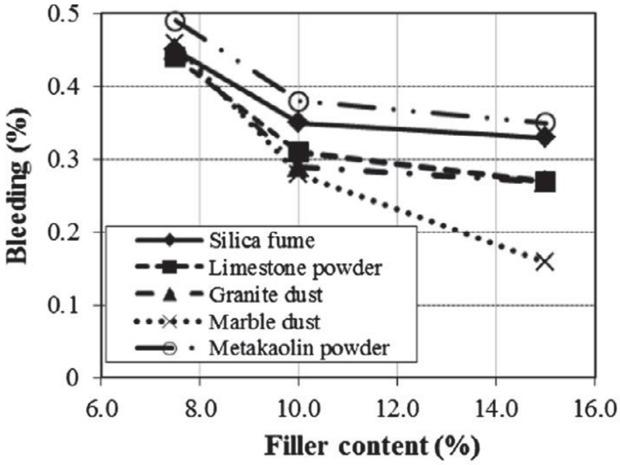
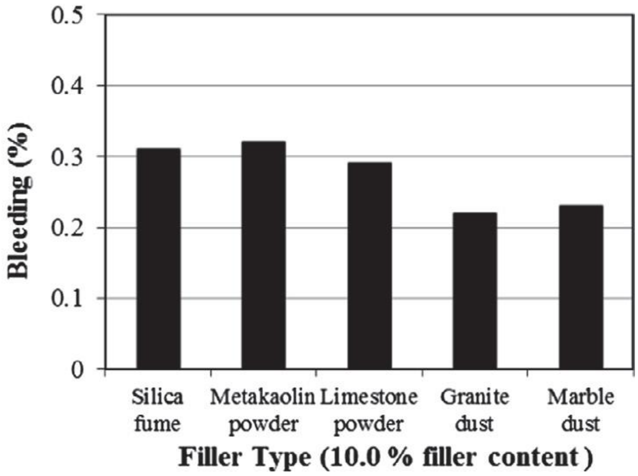



Fig 5.2-6: Influence of filler type and content on the percentage of bleeding in SCC. a.(left) 400 kg/$m^3$ cement content, b(right) 500 kg/$m^3$cement content. [197]




In 2016, Suma Paralada et al. [198] underscored the potential use of waste products from the granite industry, such as granite powder, to successfully attain Self-Compacting Concrete (SCC) properties in the fresh state. This finer material aids in preventing segregation, promoting the sustainability of natural resources. The study emphasized that incorporating The incorporation of non-pozzolanic fillers into mortar or concrete mixtures improves resistance to bleeding and segregation.

Rudiele Aparecida Schankoski et al. [199] noted in 2017 that quarry dust pastes exhibited no bleeding, featuring lower viscosity compared to those with limestone fillers. The prevention of bleeding was attributed to the increased surface area and elongated morphology of quarry dust particles.

In 2018, Hoang-Anh Nguyen et al. [200] highlighted that replacing 30% of dolomite powder with pozzolanic fillers could provide sufficient bleeding resistance for SCC.

Danish and Mohan Ganesh et al.[201] reported in 2021 a 65.2% reduction in bleeding resistance in achieving Self-Compacting Concrete (SCC) by incorporating marble powder. Both pozzolanic material, such as Metakaolin (MK), and a non-pozzolanic filler such as Waste Marble Powder (WMP), demonstrated a positive impact on segregation and bleeding resistance in SCC. The incorporation of fine materials was deemed essential to enhance the stability of fresh concrete (cohesiveness).

In summary, all these studies consistently highlight the significant influence of filler type on the resistance to bleeding in Self-Compacting Concrete (SCC). The use of non-pozzolanic fillers, especially granite dust and marble dust, exhibited notable bleeding resistance compared to other filler types.

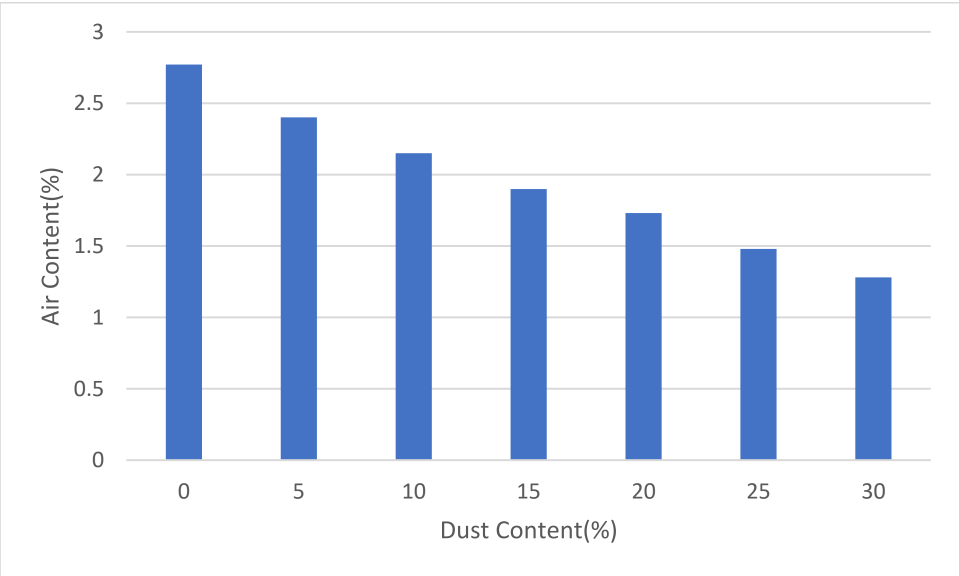
- **Air content**

Moreover, the air content plays a crucial role in cementitious materials as it directly impacts their mechanical and durability characteristics. Cementitious composites consist of two types: air-entrapped and entrained. Entrapped air naturally occurs during mixing operations, resulting in intricate and interconnected voids[195]. Conversely, entrained air is generated by introducing air-entraining admixtures into the mixture, resulting in spherical and independent voids.

In conventional concrete (non-air-entrained concrete) with an appropriate mixture and adequate compaction, the air content typically ranges from 1.5% to 2%. Through the utilization of air-entraining admixtures, the air content can be elevated to 4–8%, enhancing freeze-thaw resistance, especially in cold weather concreting[195][196]. Several factors contribute to the air content of concrete. These include water and cement proportions, the maximum size and grading of aggregate, the mixing and compaction of concrete, its temperature, the presence of admixtures (both mineral and chemical), and the introduction of fillers like stone powder and rock dust.

As mentioned earlier, in 1996, Tahir Celik and Khaled Marar et al. [192] investigated the effects of crushed stone dust on concrete properties. Seven different mixes were formulated, with fine aggregate partially replaced by dust at varying percentages (0, 5, 10, 15, 20, 25, and 30). The air contents were also examined for these mixtures. The test results (Fig 5.2-7) reveal that the total air content is influenced by the percentage replacement of dust. Acting as a filler material, dust fills the voids in fresh concrete, leading to a decrease in total air content as the percentage of dust content replacement increases.

Fig 5.2-7: Air content test results with different dust contents(data from Tahir Celik and Khaled Marar et al.,1996 [192])




Rock powders and quarry dusts emerge as highly desirable filler materials, functioning as ultrafine aggregates to occupy voids and manage or reduce the air content in cementitious materials when employed as a replacement for sand[192]. Introducing extremely fine materials with a larger specific surface area than cement, in suitable proportions, contributes to lowering the air content in concrete.

Contrary to these findings, studies by K. De Weerdt et al. (2011)[203], A.K.H. Kwan et al. (2014) [204], and K.I. Syed Ahmed Kabeer et al. (2018) [202] suggest that an excessive presence of rock dust particles concerning the voids between cement and sand particles can diminish pore filling, leading to increased air content due to reduced packing density. Consequently, determining the optimal substitution of fine rock powder in cementitious composites becomes a crucial consideration.

In summary of the effects of these fine powder additives, conflicting interpretations regarding workability are evident in the literature. Generally, the use of fillers tends to negatively impact workability by altering the water/cement ratio in the concrete. Therefore, when employing stone dust, preliminary tests are imperative, and adjustments to the water/cement ratio should consider the stone dust used instead of adhering strictly to a fixed ratio.

Furthermore, the literature generally agrees highlighting the significant impact of the impact of stone dust on resistance to segregation and bleeding. The use of stone dust is observed to reduce filling voids in the concrete to alter the air content. While this reduction appears positive for minimizing permeability, it warrants further investigation concerning durability concerns, such as freezing and thawing.

### 5.2.2 Studies related to the influence of fillers on the strength of hardening mortar/concrete

The incorporation of fillers in mortar and concrete mixtures is a widespread practice in the construction industry, prompting numerous studies aimed at comprehending their impact on the strength and characteristics of solidified mortar/concrete.

Numerous scholars advocate the use of rock dust as a substitute for a portion of sand, positing that it contributes to enhancing the mechanical properties of mortars and cement concretes. In 2012, Omar M. Omar et al. [87] delved into the mechanical strength implications of using limestone waste as a partial replacement in concrete. Their findings indicated that employing limestone waste for up to 50% replacement increased compressive strength by approximately 12% at 28 days.

In 2013, M. Vijayalakshmi et al.[205] investigated the mechanical properties of waste granite powder (GP) as an alternative material in the production of concrete. Notably, the compressive strength of blends with 5% CGP, 10% CGP, and 15% CGP exhibited superior strength gain at an early age (i.e., 7 days) compared to CM. This improvement could be attributed to the denser matrix of the GP by-product and the improved dispersion of cement grains.

Anzar Hamid Mir et al. [206] conducted a study in 2015 to examine the suitability of quarry dust as a sand-replacing material. Their experimental investigations revealed that quarry dust enhances the mechanical properties of concrete. The compressive strength exhibited an increase ranging from 55% to 75%, depending on the extent of sand replacement with quarry dust. Notably, a 100% replacement of sand showed variations in compressive strength contingent on the location of the quarry dust source.

In 2018, Hanifi Binici et al.[207] observed the mechanical properties of concrete composed of natural granular granite, silica sand, and powders derived from waste marble and basalt, utilized as fine aggregates. The compressive strength of the control sample was observed to be the lowest, while samples with 10% NG, SS, MP, and GB additive incorporation ratios exhibited higher compressive strengths at 7, 28, and 90 days.

In the year of 2021, Anitha  et al.[208] attempted to reduce the utilization of fine aggregates in concrete by substituting them partially with marble wastes. Their[208] analysis of the mechanical properties of the concrete revealed that compressive strength was enhanced an increase of 26.36% and 61.02% during curing periods of 14 and 28 days, respectively. The compressive strength exceeded the required concrete strength at both 14 days and 28 days. With the addition of waste marble powder (WMP), compressive strength showed improvements of 15.04%, 9.85%, and 2.81% at 7 days, 14 days, and 28 days, respectively.

Conclusively, various authors have drawn conclusions from studies on mortars and concretes employing additives as sand replacements, including granite dust [205][206][207], marble dust [87][207][208], lime dust [87], silicate sand [207], and basalt dust [207].

Fig 5.2-8: The compressive strength of cement mortars using natural stone dusts as additives after a curing period of 7 days [87][205][206][207][208]

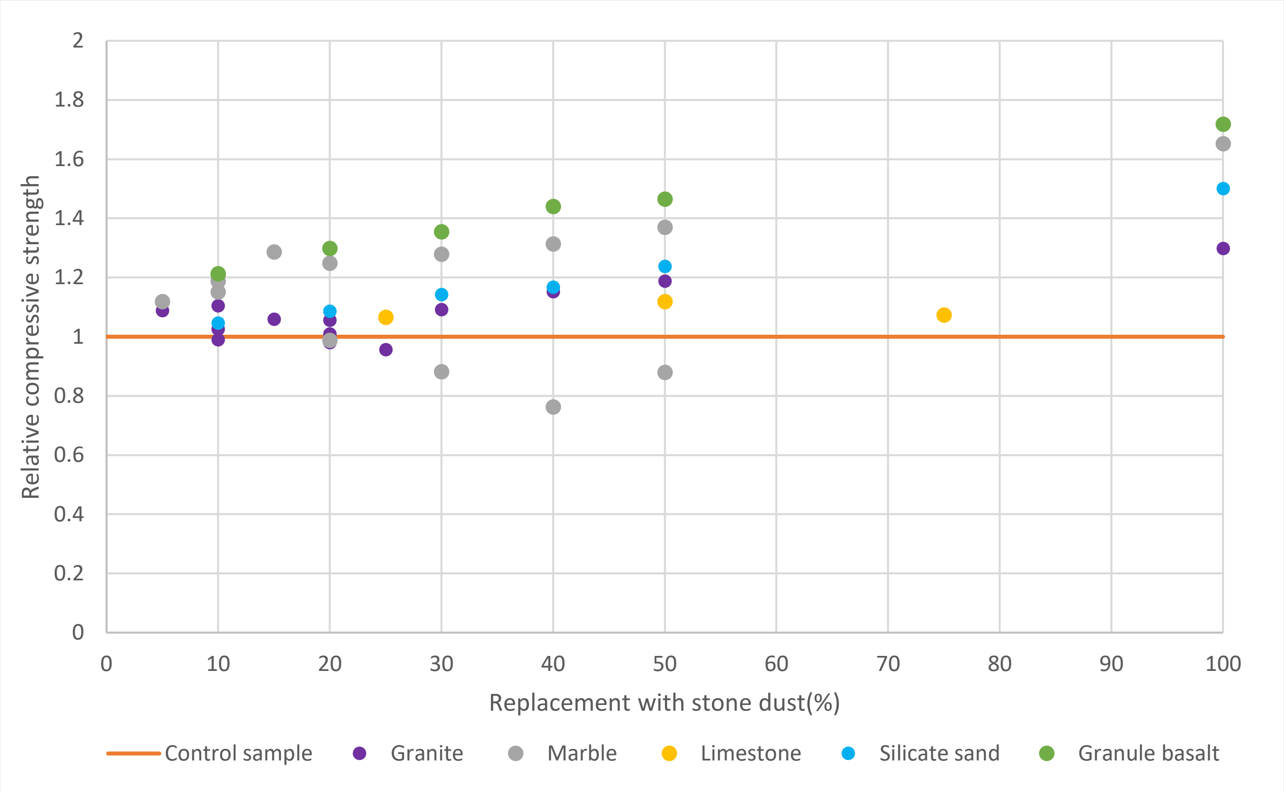



Fig 5.2-9 : The compressive strength of cement mortars using natural stone dusts as additives after a curing period of 28 days [87][205][206][207][208]

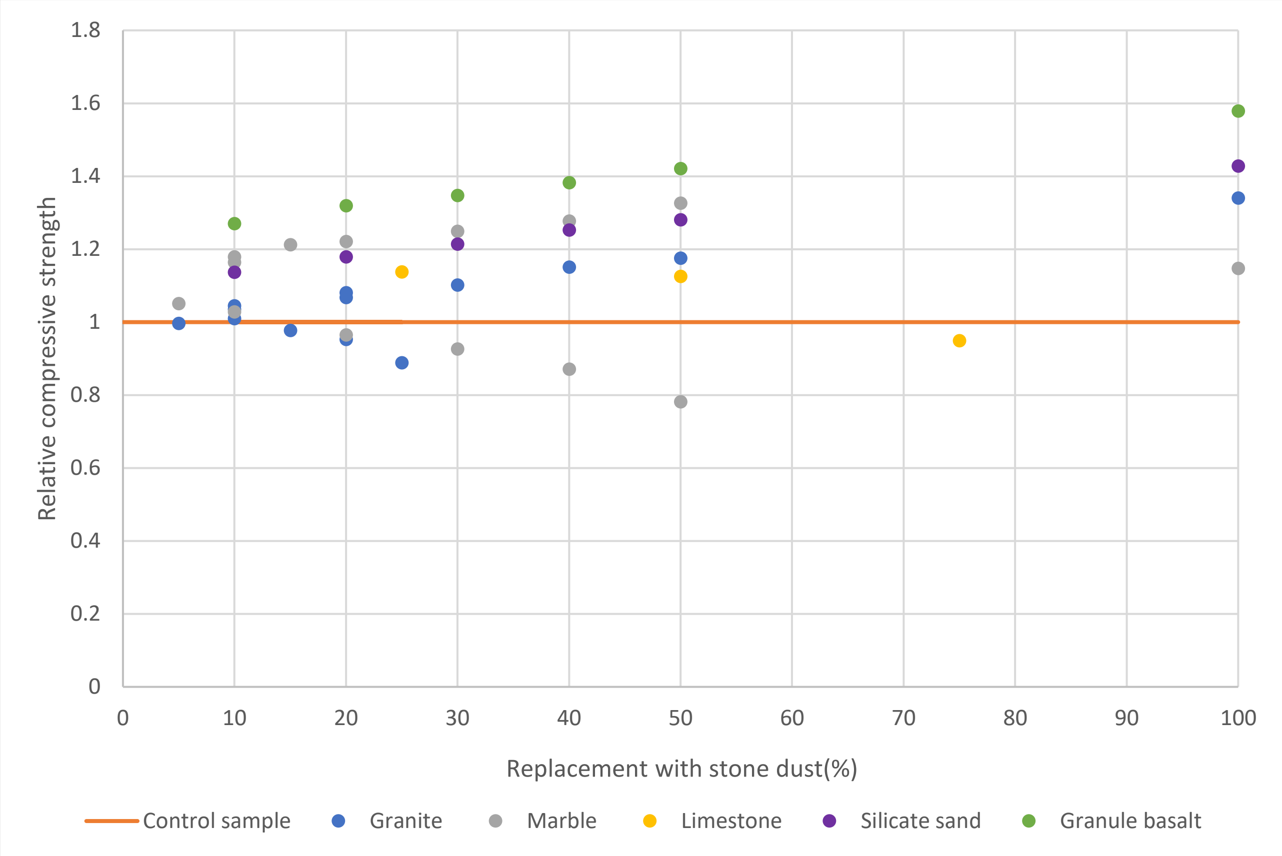
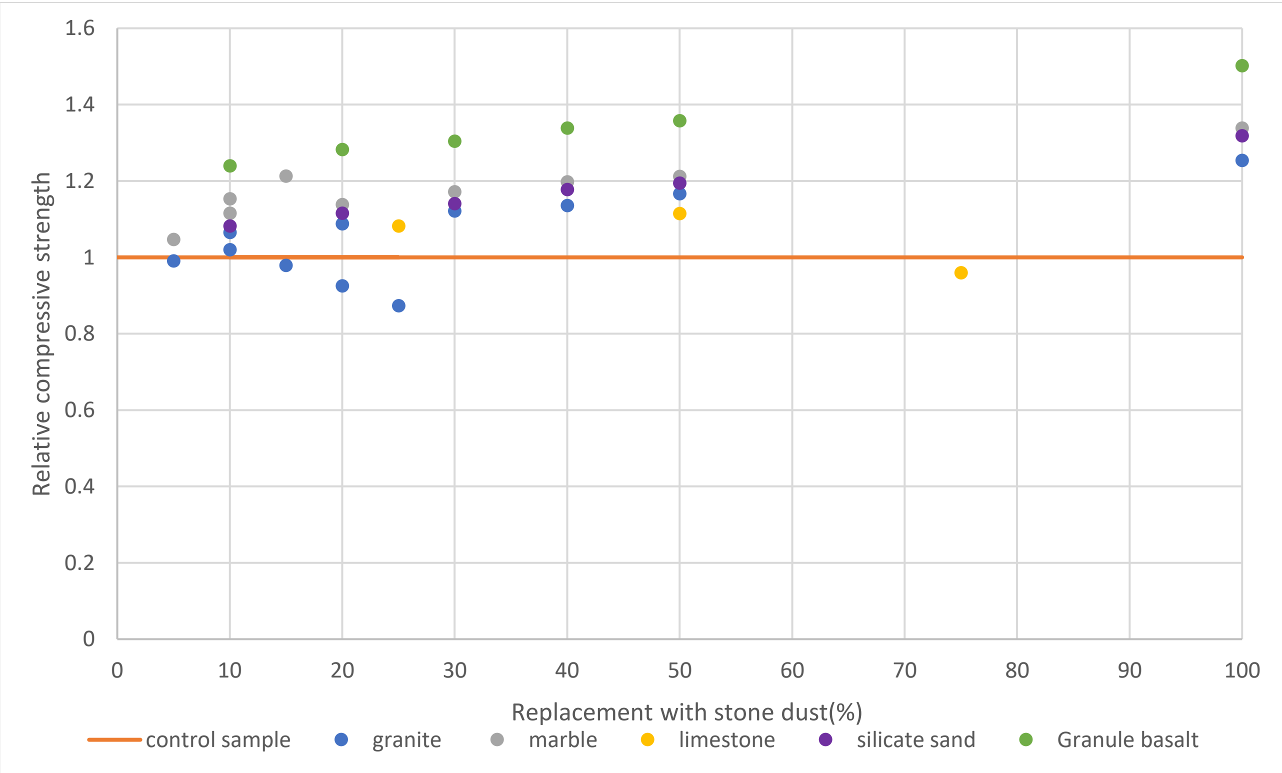



Fig 5.2-10: The compressive strength of cement mortars using natural stone dusts as additives after a curing period of 90 days [87][205] [207]




Broadly speaking, the incorporation of rock dust tends to enhance strength by occupying voids in the concrete and diminishing porosity. However, if the water/cement ratio remains unadjusted to accommodate the introduced stone powder, the cement hydration process may be affected. This is because rock dust utilizes the water necessary for cement hydration, potentially leading to a notable decrease in both compressive strength and concrete workability.

## 5.3 Chapter summary

This chapter summarized the common fillers applications from various researches by many authors, from the view of historical experience of cement filler in Arrowrock arch Dam to the up-to-date studies, according to these studies, the effects of various fillers like limestone dust, marble dust, granite dust, silicate sand, granule basalt on the workability, segregation and bleeding, air content of the fresh concrete/mortar, as well as the mechanical compressive strength were analyzed, after comparing the latest researches, these experimental indicates that the utilization of such dust not only enhances certain physical and mechanical properties of concretes/mortars but also reduces the depletion of natural raw materials. Consequently, the incorporation of these dusts contributes to the consumption of industrial waste, resulting in a dual advantage.

# 6. Stone slurry as additives mix design researches and possible mix design approach

The challenge of global warming necessitates collaborative global efforts to mitigate emissions and adapt to ongoing changes. The cement industry has actively pursued strategies to reduce CO2 emissions since long before global warming gained prominence. The Cement Sustainability Initiative (CSI), launched in 1999 at the World Business Council for Sustainable Development (WBCSD), has played a crucial role in evidence collection and strategy enhancement.

In 2009, the IEA/WBCSD Roadmap proposed various CO2 emissions reduction scenarios, indicating that achieving the target of a 50% global reduction to keep global warming below 2°C would require an 18% overall reduction in the cement sector's CO2 emissions by 2050 (compared to a 2006 baseline). In 2013, the CSI launched its inaugural regional roadmap in India, aiming for a reduction of 210 Mt of CO2 compared to the business-as-usual scenario, with a projected clinker factor of 58% by 2050. Subsequent roadmaps were launched in Brazil in 2014.

The landmark COP21 agreement in Paris in 2015 had the objective of restricting the global average temperature rise to significantly less than 2°C. In response, the WBCSD released a Global Statement of Ambition in the same year, calling for cooperative endeavors from all cement companies to attain a 20–25% reduction in CO2 emissions by 2030, surpassing business-as-usual projections.

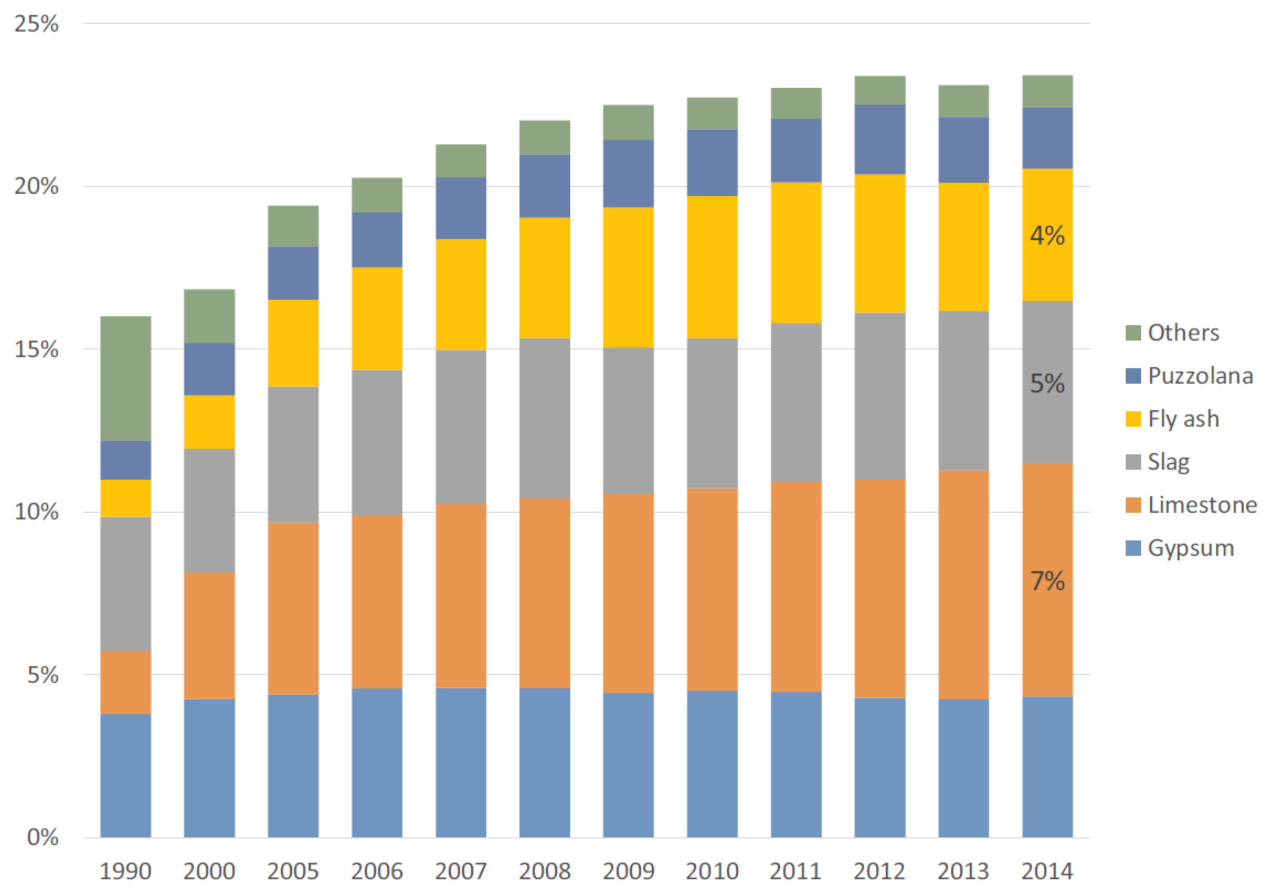
A UN Environment study in 2018 by Karen L. Scrivenera et al. identified two key areas for substantial CO2 emission reductions in cement and concrete without relying on costly CCS investments over the next 20–30 years:

1. Enhanced incorporation of low-CO2 supplements, known as supplementary cementitious materials (SCMs), to partially substitute Portland cement clinker.
2. Improved efficiency in the utilization of Portland cement clinker in mortars and concretes.

Regarding the first topic, the substitution of clinker by mineral additions/SCMs, commonly used substitutes include granulated blast furnace slag (GBFS) and fly ash (FA). However, the most prevalent supplementary cementitious material is the nearly inert limestone filler.

Over the last 25 years, the graph in Figure 6-1 illustrates the stabilization in the adoption of clinker substitutes[213]. This trend aligns with the low estimate of clinker substitutes' contribution to further CO2 reduction, indicating a challenge due to the modest supply of desirable substitutes compared to total cement production. In 2006, a significant the ratio of these alternatives were already integrated into cement or concrete, serving as the baseline for the IEA study.

Fig 6-1: The trend of substituting clinker observed among companies linked to the CSI WBCSD. [213].

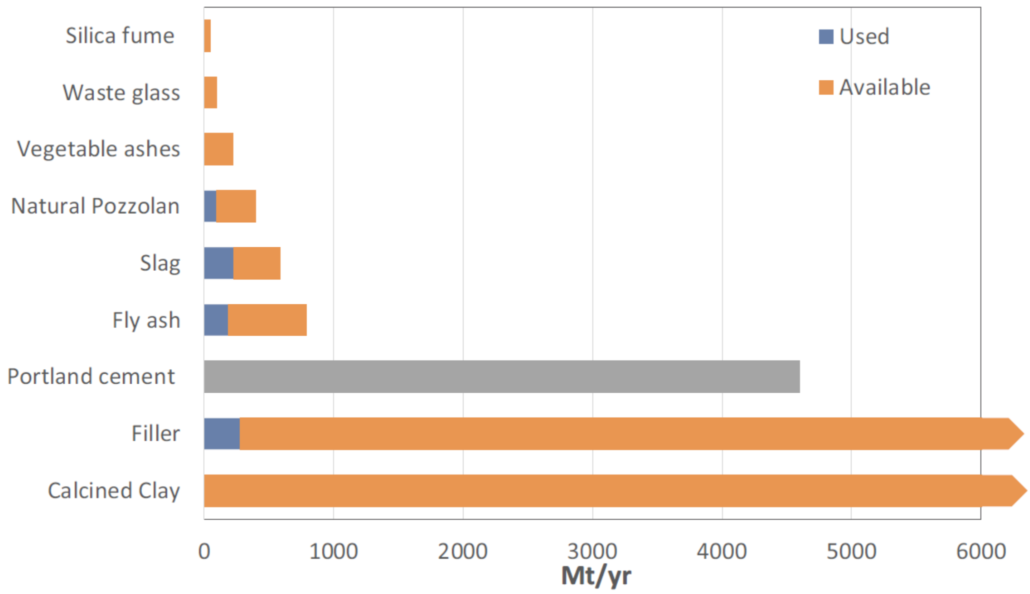



Thus, the central focus revolves around broadening the substitution of clinker with mineral additions or supplementary cementitious materials (SCMs) [213].The modification in the clinker fraction becomes evident by integrating 4–5% added as calcium sulfate, such as gypsum into the total ratio of supplementary cementitious materials (SCMs) [213]. The depicted figure [213] above emphasizes that only three materials: limestone, granulated blast furnace slag (GBFS), and fly ash (FA), currently are predominant in the domain of mineral supplements. However, it's noteworthy that the global availability of blast furnace slag hovers around 330 Mt/year, a figure [213] that has dwindled from constituting 17% of cement production in 1980 to a mere 8% in 2014. Projections suggest a decrease in the production of iron and slag from blast furnaces, owing to the increased availability of scrap steel for recycling and the implementation of more advanced steel-making technologies. In the long-term outlook, it is expected that the availability of blast furnace slag will stay below 8% of cement production [211].

At present, over ninety percent of blast furnace slag is currently utilized as a supplementary cementitious material (SCM), either blended into cement at manufacturing plants or added to various cement-based mixes. [212][214]. Consequently, there's limited potential for additional decrease in CO2 emissions resulting from the utilization of blast furnace slag. Unlike granulated blast furnace slag (GBFS), fly ash (FA) presents larger quantities, approximately 900 Mt/year; however, its variable quality restricts current usage to approximately one-third of this quantity in cement and concrete. It's imperative to consider that coal combustion for electricity, a major source of anthropogenic CO2, is gradually being phased out in some countries. Although coal will persist globally in the energy mix for the medium term, the recent shale gas availability in North America has created a deficiency in the availability of fly ash [211]].

Crucially, the availability of new sources offering high-quality SCMs could significantly alter the current landscape. Fig 6-2 delineates the estimated availability of potential SCMs and fillers in relation to the volume of cement produced.

Fig 6-2 : Utilization and anticipated availability of potential supplementary cementitious materials (SCMs) and fillers. The practical application will be contingent on logistical considerations, precise chemical and mineralogical compositions, contamination levels, and the local accessibility of other raw materials[211]



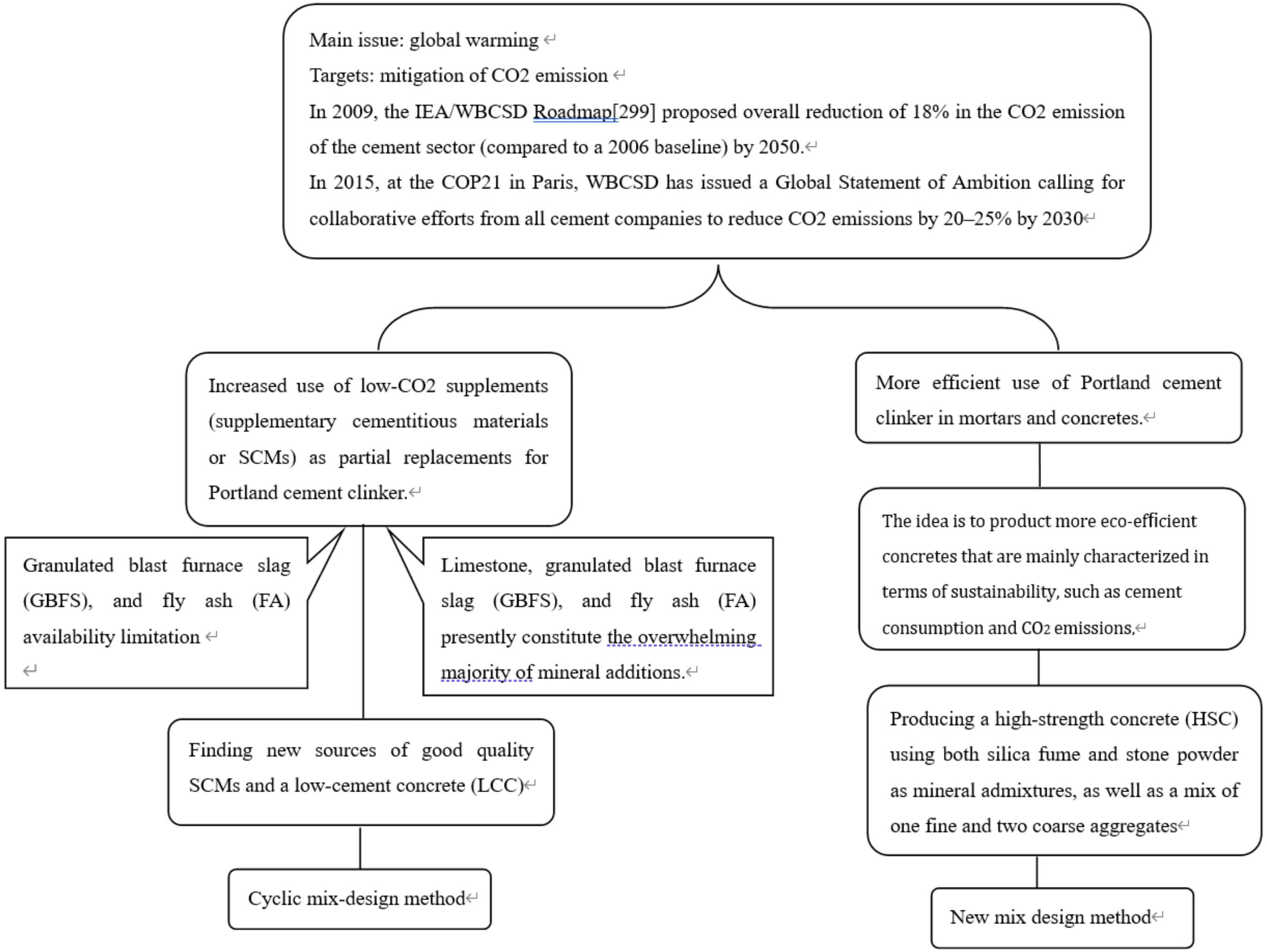

In addition to the aforementioned avenue of expanding the use of supplementary cementitious materials (SCMs), another identified approach involves reducing the clinker content in concrete by enhancing mix designs that facilitate increased filler incorporation. This can occur through the addition of filler either through the cement or directly during the mixing of concrete [211].

This chapter delves into the optimization of mix design procedures, drawing on experiences with the utilization of stone slurry wastes as additives, and explores potential mix design approaches. As highlighted earlier, numerous recent research endeavors have aimed to develop mix design methods for cement-based products, targeting low-cement consumption in the next phase of the cement industry's development[215].

Over the years, various mix-design methodologies have emerged to reduce cement consumption through particle packing optimization. Dhir et al. in 2005 [216]and Fennis et al. in 2011 [217] delved into the reduction of cement usage in concrete, incorporating mineral admixtures via particle packing techniques. They [216] [217] aimed to utilize fillers to decrease voids and chemical admixtures to lower water content in concrete. The objective was to achieve mixtures with properties in the hardened state comparable to those of reference concretes. Fennis et al. in 2011 [217] created a method for designing concrete mixes employing methods based on particle packing, permitting the systematic and organized substitution of cement with mineral additives. By introducing quartz powder and fly ash at levels of 34% and 24% [217], respectively, the author facilitated mixtures with a 57% reduction in cement, resulting in consumption below 200 kg/m3 and compressive strength exceeding 30 MPa at 28 days.

Embracing high-strength concrete (HSC) offers an environmentally efficient alternative, requiring reduced binder contents per unit of compressive strength.. H.F. Campos et al. in 2020 [218] Proposing an innovative approach to mix design for sustainable high-strength concrete (HSC), aiming to decrease cement consumption and $CO_2$ emissions in comparison to HSC formulated using current methods. In this newly introduced approach, the relationship between fine materials and aggregates is determined through particle packing techniques. The validation process included the production of high-strength concrete (HSC) using silica fume and stone powder as mineral admixtures, combined with a mixture of one fine and two coarse aggregates. This method achieved a cement consumption of only 3.2 kg to achieve 1 MPa of compressive strength, a significantly reduced value compared to those documented in existing literature for HSC. This highlights its effectiveness in generating more sustainable HSC.

Fig 6-3: Roadmap for the reason to explore new mix design method



## 6.1 Cyclic mix-design approach utilizing particle packing techniques

The cyclic mix-design approach, introduced by Fennis et al. in 2011 [217] and further detailed in their work in 2012[219], employs particle packing techniques for the formulation and assessment of properties in low-cement concrete (LCC). This method offers a systematic and rational optimization of the granular composition. The investigation commenced with an initial concrete ($C_0$) dosed using the IPT/EPUSP method [220], where particle packing techniques were not initially applied, and no supplementary cementitious materials were incorporated. The properties of the resulting LCC were evaluated in both the fresh and hardened states.

### 6.1.1 Materials

To manufacture the concrete, the components employed included industrial sand derived from granite, limestone gravel with a maximum dimension of 9.5 mm (designated as gravel 9.5), and basaltic gravel with a maximum dimension of 19 mm (designated as gravel 19). The particle size distribution of these three aggregates is illustrated in Fig. 6.1-1, and Table 6.1-1 provides a detailed characterization of these materials.

Fig.6.1-1: Particle size distribution of the aggregates and the limits recommended by ASTM C33

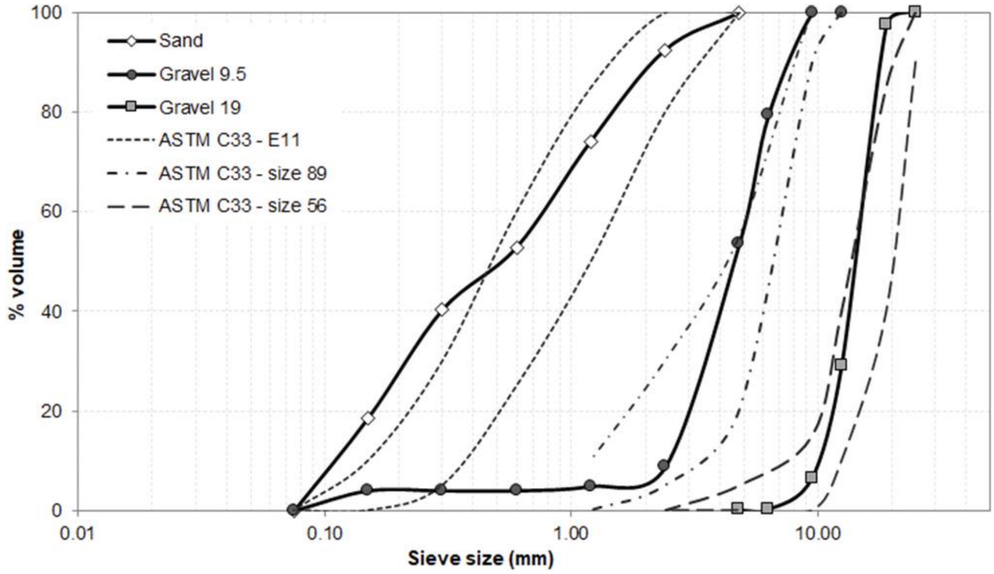



Table 6.1-1: Aggregates characterization

| Properties | Standard regulations | Sand | Gravel 9.5 | Gravel 19 |
|---|---|---|---|---|
| Maximum size (mm) | ASTM C136/ C136 M | 4.8 | 9.5 | 19 |
| Fineness module | ASTM C136/ C136 M | 2.22 | 5.21 | 6.95 |
| Specific gravity (g/ $cm^3$) | ASTM C128-15 and ASTM C127-15 | 2.62 | 2.8 | 2.67 |
| Compacted Bulk density (g/ $cm^3$) | ASTM C29/ C29 M | 1.723 | 1.711 | 1.517 |
| Voids index (%) | ASTM C29/ C29 M | 0.342 | 0.388 | 0.43 |
| Packing density | | 0.658 | 0.612 | 0.57 |

| | | | | |
|---|---|---|---|---|
| Water absorption (%) | ASTM C128-15 | 0.4 | 0.57 | 0.28 |
| 0.64Material finer than 75 µm (%) | ASTM C117-17 | 8.17 | 1.37 | 0.64 |
| Shape index | ASTM C33 | - | - | 2.16 |

For the creation of the concrete, Portland cement type CP II-F 32 was employed, adhering to the specifications outlined in ASTM C150/C150 M. The mineral additives incorporated were rice husk ash (RHA), obtained through the controlled incineration of rice husks, and quartz powder (QP). RHA, an agricultural by-product found in the southern region of Brazil, results from the controlled combustion of rice husks. Utilizing this material as a mineral additive aligns with sustainability objectives, as its incorporation reduces the necessity for environmental disposal. The fine materials' particle size distribution was evaluated using laser particle testing, and the results are depicted in Fig. 6.1-2. Table 6.1-2 provides key physical characteristics of the fine materials utilized [221].

To enhance the workability of the concrete, a superplasticizer admixture with a chemical base composed of polycarboxylic ether was employed, exhibiting a density of 1.087 kg/$m^3$ as per the manufacturer's technical specifications.

Fig.6.1-2: Particle size distribution of fine materials[221]

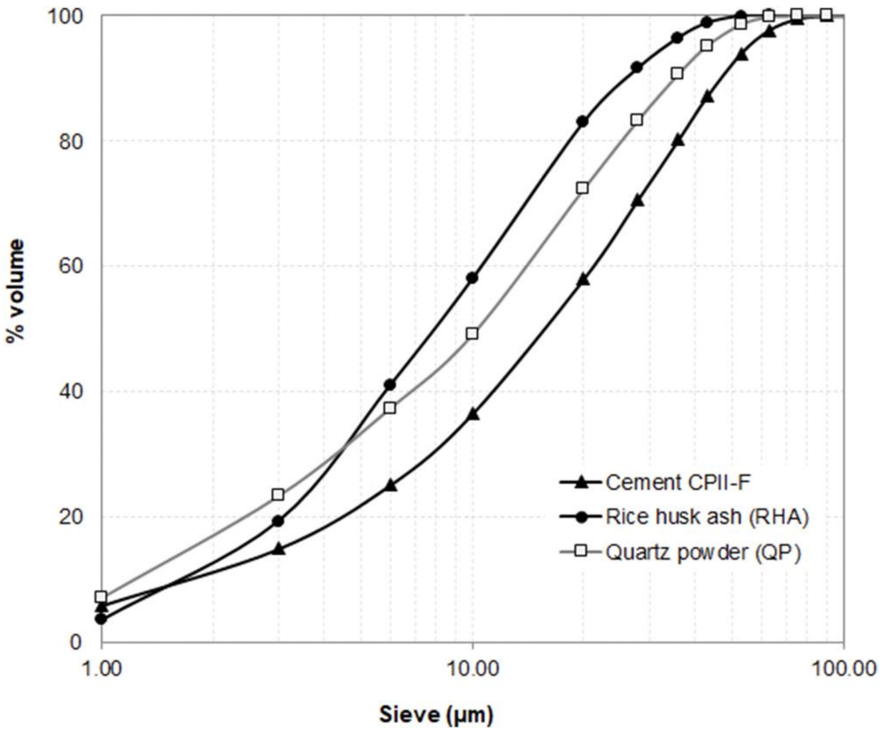



Table 6.1-2: Physical characteristics of the cement and mineral admixtures [222]

| Material | Physical characteristics | | |
|---|---|---|---|
| | Specific gravity (g/ $cm^3$) | $D_{50}$ (µm) | Blaine specific surface($m^2$/kg) |
| Cement CP II-F32 | 3.11 | 15.86 | 330 |
| RHA | 2.16 | 7.89 | 696 |
| QP | 2.60 | 10.34 | 795 |

### 6.1.2 Mix-design of the initial concrete ($C_0$)

To adhere to a systematic and structured approach aimed at reducing cement consumption through particle packing techniques, the investigation commenced with an initial concrete ($C_0$), formulated using the IPT/EPUSP method [220]. The slump test was set at (100 ± 10) mm, employing gravel 19 and industrial sand due to their prevalent usage in the region. The mix-design of $C_0$ did not involve any mineral or chemical admixture, and particle packing techniques were not applied. Consequently, when optimizing the concrete granular skeleton (a process detailed in subsequent sections), the study followed the procedure outlined by the cyclic mix-design method proposed by Fennis et al. [217] and Fennis et al. [219]. This method allowed for an expansion in the range of grain sizes within the mixture, leading to a reduction in cement consumption.

The optimal dry mortar content, determined in line with the mix-design method recommendations [220], was established at 53%. This value represents the volume of mortar required to fill the voids between the gravel grains. It's worth noting that, given the characteristics of the aggregate used, this value could be decreased by incorporating coarse aggregates of varying sizes. The combined use of different coarse aggregates, known as granular optimization, is endorsed by the cyclic mix-design method proposed by Fennis et al.[217] and Fennis et al.[219].

The selected characteristic compressive strength ($f_{ck}$) was 25 MPa, corresponding to a dosage compressive strength ($f_{cd}$) of 31.6 MPa, calculated considering a standard deviation of 4.0 MPa under the adopted production conditions. The proportioning of $C_0$ concrete, as derived from the mix-design study, is presented in Table 6.1-3.

Table 6.1-3: $C_0$ concrete proportioning(data from Fennis et al. [217] and Fennis et al. [219])

| Materials | Cement | Sand | Gravel 19 | Water |
|---|---|---|---|---|
| Specific gravity (kg/ $m^3$) | 3110 | 2620 | 2670 | 1000 |
| Consumptions (kg/ $m^3$) | 410 | 754 | 1038 | 217 |
| Unitary proportion | 1 | 1.84 | 2.52 | 0.53 |

Examining the data presented in the table above reveals a notable cement consumption stemming from the implementation of the IPT/EPUSP mix-design method[220]. It's crucial to recognize that this outcome is a direct consequence of relying solely on gravel 19 as the coarse aggregate. The IPT/EPUSP [220]mix-design method itself could be reapplied, employing combinations of coarse aggregates to achieve reduced voids and, consequently, diminished cement consumption.

$C_0$ samples, with dimensions of (100 × 200) mm, were manufactured, and the concrete was manually compacted. All specimens underwent a wet curing regimen for 28 days, aligning with ASTM C192/C192 M standards. Compressive strength tests were conducted at 28 days, adhering to the recommendations outlined in ASTM C39/C39 M.

### 6.1.3 Granular optimization of the aggregates and the fine materials

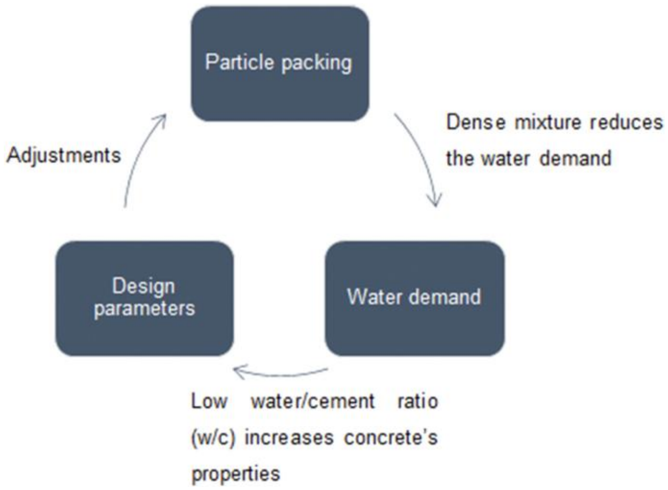
Commencing with the initial composition of $C_0$, particle packing techniques were applied to curtail the cement consumption of concrete. The objective was to ascertain the aggregate ratio that exhibited the least voids through a series of experiments. Experiments were undertaken to evaluate the compacted bulk density and voids index of various combinations of small and large aggregates, with proportions varying at 10% intervals. The investigation focused on optimizing the ratio between gravel 9.5 and gravel 19. Subsequently, having identified the proportion yielding the least voids among the coarse aggregates, the industrial sand was introduced, and the test cycle was repeated until the minimum voids were achieved with the amalgamation of all three aggregates[215].

With the aggregate ratios defined, the optimization of fine materials ensued. Utilizing the experimental method devised by Wong and Kwan et al. [223], the packing density of each fine material (cement, rice husk ash, and quartz powder) was determined. Packing density, denoting the volume of solids within a unitary volume, was calculated for each fine material. Subsequently, employing the CIPM model [217], the proportion among these fine materials was computed to maximize the packing density. Each fine material was treated as an independent class of grains, characterized by its diameter $D_{50}$.

### 6.1.4 Design of low-cement concrete (LCC)

Commencing with the determined proportions of aggregates and fine materials from the preceding section, it became imperative to ascertain the mixture packing density and water demand. Subsequently, the parameters deemed desirable for concrete were adjusted; in this article, the considered parameter was the required compressive strength. To achieve this, the cyclic mix-design method proposed by Fennis et al. [217]and Fennis et al. [219] was employed, as illustrated in Fig 6.1-3. The method comprises three steps: calculating the packing density of the particles constituting the mixture to minimize voids, establishing the amount of water needed to occupy the gaps between the particles and facilitate the flow of concrete, and, finally, comparing the results predicted by the method with the desired concrete parameters in this study, compressive strength. The subsequent subsections provide more detailed explanations of each step.

Fig 6.1-3: Cyclic mix-design process for low-cement concretes, adapted from Fennis et al. [217] and Fennis et al. [219]




### 6.1.5 Step 1: calculating the packing density of the granular set

The CPM model (Compressible Packing Model), introduced by De Larrard et al. [222], offers a predictive capability for determining the packing density of polydisperse granular sets. To apply this model effectively, there is a prerequisite to systematically arrange, identify, and characterize all size classes of grains constituting the mixture. Each size class (i) is treated as dominant in every calculation step. The particles within each size class must be meticulously ordered based on their grain size, necessitating knowledge of the packing density value for each size class ($\beta_i$), as determined by Equation (1).

$$\beta_i = 1 - VI \qquad (1)$$

Where,

$\beta_i$ is the value of the packing density of each size class

VI is the void index (VI)

The void index (VI) for aggregates is determined following ASTM C29/C29 M guidelines, which factor in the aggregates' specific gravity and compacted bulk density. However, when employing delicate materials like cements and admixtures, one must account for factors such as particle agglomeration induced by cohesive forces. In the realm of minute particles, cohesive forces may outweigh those linked to gravity. Agglomeration's occurrence hinges on particle size, distribution, shape, and roughness. This phenomenon complicates mixing and compaction, leading to the possible emergence of porous spaces between agglomerates, thereby diminishing packing density [217]. Consequently, for fine materials, it is advisable to determine the voids index (VI) in the presence of water and the chemical admixtures planned for use in concrete production.

In this study, we adopted the experimental method proposed by Wong and Kwan et al. [223].

To capture the intricacies of fine materials, Fennis et al.[217] and Fennis et al. [219] introduced modifications to De Larrard et al. [222]packing model. They formulated the Compaction-Interaction Packing Model (CIPM), which facilitates the calculation of granular set packing density through equations (2)-(5). The CIPM model accounts for interactions among fine particles and is endorsed by the cyclic mix-design method applied in this study.

$$\gamma_i = \frac{\beta_i}{1 - \sum_{j=1}^{i=1}\left[1 - \beta_i + b_{ij} * \beta_i\left(1 - \frac{1}{\beta_i}\right)\right] * y_i - \sum_{j=i+1}^{n}\left[1 - a_{ij} * \frac{\beta_i}{\beta_j}\right] * y_j} \quad (2)$$

$$a_{ij.c} = \begin{cases} 1 - \frac{\log(d_i/d_j)}{\omega_{0,a}} & \log(d_i/d_j) < \omega_{0,a} = \begin{cases} \omega_a * C_a & d_j < 25\mu m \\ \omega_a & d_j \geq 25\mu m \end{cases} \\ 0 & \log(d_i/d_j) \geq \omega_{0,a} \end{cases} \quad (3)$$

$$b_{ij.c} = \begin{cases} 1 - \frac{\log(d_i/d_j)}{\omega_{0,b}} & \log(d_i/d_j) < \omega_{0,b} = \begin{cases} \omega_b * C_b & d_j < 25\mu m \\ \omega_b & d_j \geq 25\mu m \end{cases} \\ 0 & \log(d_i/d_j) \geq \omega_{0,b} \end{cases} \quad (4)$$

$$k = \sum_{i=1}^{n} \frac{y_j/\beta_i}{1/\varphi - 1/\gamma_i} \quad (5)$$

In equations (2)–(5), where,

$a_{ij.c}$ and $b_{ij.c}$ are coefficients related to the loosening and wall effects, respectively. The wall effect describes the influence of the bigger particles on the smaller ones causing a decrease in the packing density of the mixture in the vicinity of the larger grains. Plus, the loosening effect happens when the thin particles are too big to occupy the voids in between the larger particles, collaborating for these larger grains to move away from each other, also causing a higher porosity and lower density.

$\gamma_i$ is the virtual packing density of the mixture when class $i$ is dominant (-);

$C_a$, $C_b$, $\omega_a$ and $\omega_b$ are constants related to loosening and wall effect according to the CIPM model and superplasticizer, $\omega_a$ = $\omega_b$= 1,0, $C_a$ = 1,5 $C_b$ = 0,2 (-);

$y_i$ and $y_j$ are the relative volumes of the classes $i$ and $j$ (-);

φ is the actual packing density in a stable composition De Larrard et al. [222].

The compaction index ($k$) utilized in applying the CIPM method was set at 12, denoting the compaction of pastes created with water and admixtures, according to Fennis et al.[217]. Every material utilized in forming the concrete was treated as a grain size category (n), resulting in a total of 6 categories (3 for fine materials and 3 for aggregates). The individual packing density of each category ($\beta_i$) was experimentally determined, as outlined in equation (1) earlier. The diameter chosen to represent each size category ($d_i$) was the $D_{50}$ diameter, extracted from the particle size distribution curves for each material.

### 6.1.6 Step 2: calculating the water demand

To determine the water demand of the mixture, it's essential to distinguish the value of the actual packing density (φ) in a stable composition (devoid of water and obtained from equation (5)) from the relative volume of solids ($\phi_{mix}$)) in a mixture. In this mixture, a portion of the water is utilized to fill the voids between the particles, while any excess water contributes to the fluidity of the mix. According to Fennis [217], the fluidity of concrete can be expressed through the ratio $\phi_{mix}/\varphi$, representing the relationship between the relative volume of solids in an actual fluid mixture and the actual packing density of the granular set when grains are tightly packed. This ratio signifies the proportion of relative water present in a specific concrete, allowing for the calculation of the water volume ($V_w$), in $m^3$, as per Equation (6).

$$V_w = 1 - \phi_{mix} \quad (6)$$

Where,

$\phi_{mix}$ is the relative volume of solids of mixture

$V_w$ is the volume of water, in $m^3$

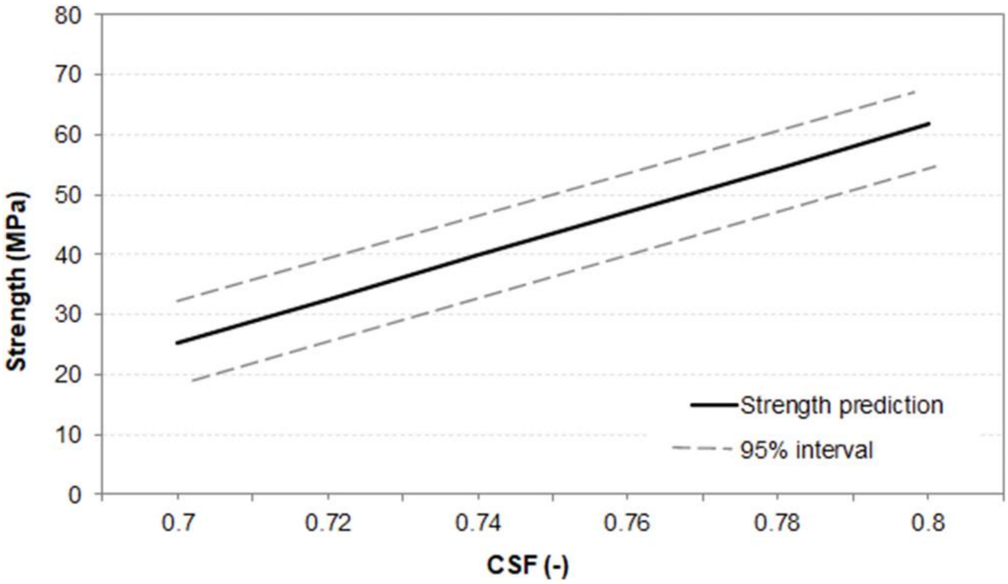
In this study, the ratio $\phi_{mix}/\varphi$ was chosen to be equal to 0.95, which corresponds to a plastic concrete with a slump test varying from 100 to 150 mm, according to the recommendations of the cyclic mix-design method.

### 6.1.7 Step 3: Prediction of concrete's compressive strength

In forecasting the compressive strength of concrete, the approach takes into account the gaps between cement particles and/or binders, as well as the volume of water in the mix, ensuring the formation of cement hydration products. According to Fennis et al.[217] and Fennis et al. [219], cement serves as a linking bridge among aggregates, and the concrete's strength hinges on the available space for this connection. In mixtures characterized by high packing density, particles are closely situated, minimizing the space requiring filling with cement hydration products. Consequently, such mixtures exhibit heightened mechanical strength [215].

The procedural calculation for predicting concrete's compressive strength is tied to the cement spacing factor (CSF), correlating directly with concrete strength, as illustrated in Fig 6.1-4.

Fig 6.1-4: Experimental relation between CSF and 28 days concrete's compressive strength (adapted from Refs. Fennis et al.[217] and Fennis et al. [219])




Fennis et al. [217] establishes a lower threshold for the CSF factor at 0.70, corresponding to a minimum mechanical strength of 25 MPa at 28 days, and an upper limit of 0.80, approaching 65 MPa at the same age. Fig 6.1-4, crafted by Fennis et al. [217], derives from experimental outcomes involving various concrete mixtures, incorporating Portland cement with diverse strength classifications.

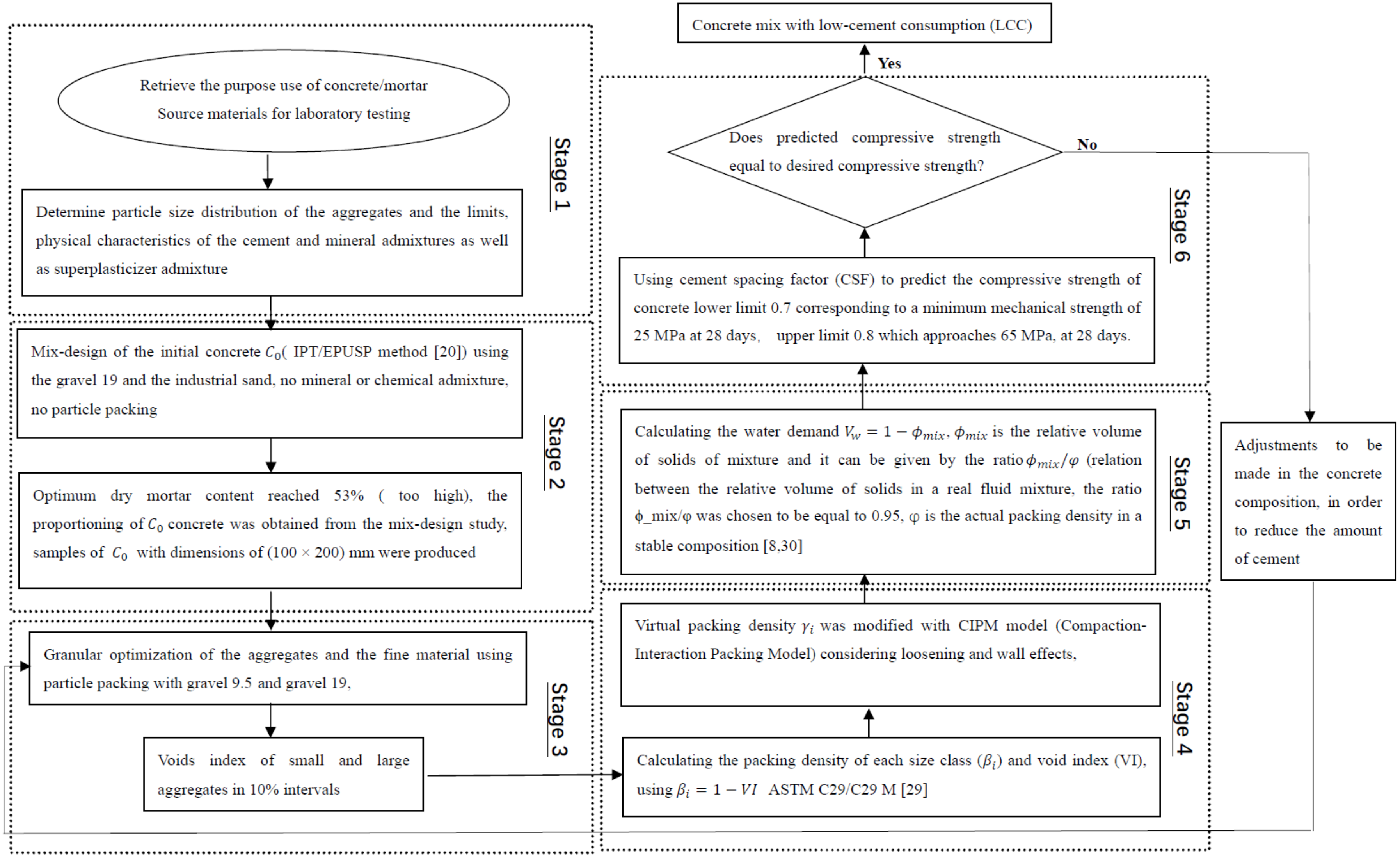
The computation of the CSF involves multiplying the $\phi_{mix}/\varphi$ ratio, aligned with the consistency specified for the concrete (a value determined in the preceding design step), by the correlation between the partial volume occupied by cement in a structure of stable particles ($\phi_{cem}$) and the maximum volume that cement can occupy due to other particles $\phi^{*}_{cem}$, as demonstrated in Equation (7).

$$CSF = \phi_{cem}/\phi^{*}_{cem} * \phi_{mix}/ \varphi \quad (7)$$

Upon completing the calculation, the derived value undergoes scrutiny against the targeted compressive strength. If the anticipated strength surpasses the desired value, the cyclic mix-design method permits adjustments to the concrete composition, aiming to curtail the cement quantity. As elucidated by Fennis et al.[217], alterations in the mixture's material volume bring about shifts in particle distribution and, consequently, the packing density of the set. Thus, the cyclic process is reiterated until the mixture's composition remains unaltered following the final step, aligning the anticipated compressive strength with the desired benchmark.

Concluding all the stages of the cyclic mix-design method yields a concrete mix characterized by low cement consumption (LCC). In this study, the quest for LCC commenced from the initial concrete ($C_0$), with the goal of maintaining a consistent compressive strength of 31.6 MPa but with diminished cement usage. The reduction in cement content stemmed from alterations in the packing of both coarse aggregates, incorporating Gravel 9.5, and fines, employing mineral admixtures. The proportions of LCC constitute integral outcomes of this investigation. It is noteworthy to reiterate that optimizing the granular set of the $C_0$ concrete could have been pursued through alternative design methods, including the IPT/EPUSP method itself (utilized in $C_0$ concrete design). However, the decision was made to adopt the cyclic mix-design method by Fennis et al.[217] and Fennis et al. [219], given its rational and systematic approach to optimizing particle packing. Additionally, the packing density of the granular set is computed in accordance with the CIPM model recommendations, enabling the anticipation of packing densities multiple times through computational assistance, eliminating the necessity for an extensive array of experiments, thereby saving time and resources.

Fig 6.1-5: Flow chart of the proposed cyclic mix design procedure.



## 6.2 A new mix design method for sustainable high-strength concrete (HSC)

In the year 2020, H.F. Campos et al.[218] introduced a mix design methodology for sustainable High-Strength Concrete (HSC), employing particle packing techniques to optimize the granular components of concrete. This approach is tailored to the creation of eco-efficient concretes, primarily characterized by sustainability aspects such as cement consumption and CO2 emissions. It aims to meet stipulated criteria in both the fresh and hardened states.

The suggested mix design method advocates a concurrent exploration to ascertain the material proportions for both paste and aggregate compositions. Concrete is conceptualized as suspensions comprised of fine materials (reactive or inert) with diameters <125 mm, possessing larger surface areas and predominantly surface forces, alongside aggregates with diameters >125 mm, exhibiting lower surface area and a dominance of gravitational forces [224]. Consequently, our mix design method posits that addressing the packing density of both fine materials and aggregates individually is imperative. Water's role in fine material packing density analysis is emphasized due to the presence of surface forces[223]. In this context, the initial step is delineating the compositions for paste and aggregates, followed by determining the material consumption for the concrete mix design. Fig 6.1-1 illustrates the schematic representation of the proposed method, with each subsequent step outlined in detail.

As depicted in Fig 6.2-1 (a), when determining the paste composition, it is imperative to establish the packing density for each material using the wet packing method [223]. Campos et al. (2019) [218] recently showcased that the most effective composition of fine materials, concerning sustainability, aligns with the one exhibiting the highest packing density according to the Compressible Packing Model (CPM) [222]. Consequently, it is recommended to define the composition of fine materials based on this model, as illustrated in Fig 6.2-1 (a).

For designing High-Strength Concrete (HSC), the inclusion of silica fume (SF) or a similar mineral admixture is advised. SF stands out among commonly used mineral admixtures for HSC due to its fineness, enabling particles to infiltrate the pores of coarse aggregates and enhance strength in the interfacial transition zone [225]. The incorporation of inert fillers proves beneficial in reducing voids between cement particles and contributes to the reduction of cement consumption, a primary objective of the proposed method.

In the application of the Compressible Packing Model (CPM)[222], the volume of each material should be determined through random compositions. Recommended variations for SF content range from 6% to 16% in 2% intervals, aligning with typical recommendations in the literature [226][227][228][229]. Volumes of other mineral admixture types should be adjusted based on specific type literature, also in 2% intervals. Approaching the ideal content, adjustments can be made in 1% intervals for higher precision. The compaction index K, for packing densities obtained using the wet packing method [223], should be set at 12 [217]. The optimal proportion between fine materials will be the one leading to the highest packing density, as emphasized in Fig 6.2-1 (a).

The paste volume comprises fine materials, water, air, and solids from the chemical admixture. It is recommended to consider the water content from the chemical admixture alongside the total water volume in concrete. The determination of the superplasticizer saturation dosage should follow Kantro's Miniature Slump Test[231], as illustrated in Fig 6.2-1 (a). Test results serve as a starting point for the chemical admixture percentage when producing concrete. However, adjustments to the admixture quantity can be made during concrete production to achieve the desired consistency in the fresh state. It is crucial to adhere to the supplier's maximum recommended dosage to prevent setting issues. The air content for High-Strength Concrete (HSC) is considered to range between 1.5% and 2% of the paste volume[232][225].

Concerning aggregates, the packing density for each size class is typically determined through the bulk density test, allowing for the calculation of the void index following ASTM C 29 (ASTM, 2017), as depicted in Fig 6.2-1 (b). The aggregate composition should be defined based on the Compressible Packing Model (CPM) [222] to achieve the lowest void index. The relative volume of grains for each class is determined through random compositions, starting with an equal volume of material for all aggregate classes. Further combinations involve increasing 5% in the class with the highest individual packing density and reducing 5% in the class with lower packing density. As the ideal content is approached, adjustments can be made at 1% intervals for greater precision. The selection of the compaction index value K depends on the compaction method to be employed, following the model recommendations[222].

Finally, after outlining the compositions for paste and aggregates, the concrete composition can be established, as indicated in Fig 6.2-1 (c). This involves determining the ideal paste volume ($V_p$). The minimum paste volume required to fill the voids between aggregate particles, plus some excess paste for workability, should be analyzed. The minimum paste volume aligns with an aggregate's void index, calculated according to ASTM C 29 (ASTM, 2017). Excess paste volumes up to 8% should be experimentally verified at 2% intervals. Previous research has indicated a significant decrease in the properties of concretes in the hardened state beyond an 8% paste excess [233]. The ideal paste content is the one that maximizes efficiency in terms of CO2 emissions per unit of compressive strength at 28 days, highlighted in Fig 6.2-1 (c).

The water/fines (w/f) ratio hinges on the desired compressive strength and the composition of fine materials. The minimum w/f ratio is determined to produce homogeneous concrete in the plastic state. Lower compressive strength may be achieved by increasing the w/f ratio. The consumption of each material required for concrete production can be calculated using equations (1)-(4).

$$V_f = \frac{R_f * (V_p - V_{Air} - V_{S.Ad})}{1 + w/f_{(v)}} \quad (1)$$

$$\sum_{i=1}^{n} R_f = 1 \quad (2)$$

$$V_{Agg} = R_{Agg} * (1 - V_p) \quad (3)$$

$$\sum_{i=1}^{n} R_{Agg} = 1 \quad (4)$$

From equations (3) and (4), the volume of each aggregate composing the concrete can be calculated.
Where,

$V_f$ is volume of the fine material ($m^3$);

$w/f_{(v)}$ is water/fines ratio by volume ($m^3$);

$R_f$ is volumetric ratio of the fine material (-);

$V_p$ is paste volume ($m^3$);

$V_{Air}$ is air content ($m^3$);

$V_{S.Ad}$ is solids content of the chemical admixture ($m^3$);
$V_{Agg}$ is volume of the aggregate ($m^3$);
$R_{Agg}$ is volumetric ratio of the aggregate (-).

Fig 6.2-1: The scheme for the proposed mix design method [218]

(a) PASTE COMPOSITION
Packing density of each individual material: the wet packing method[223]
Proportion between the fine materials: CPM [222], K-12[217] → The highest packing density
Superplasticizer saturation dosage: Kantro's Miniature Slump Test [231]

(b) AGGREGATES COMPOSITIONP
Packing density of each size class according to the bulk density test and voids index calculation;
Proportion between the aggregates: CPM, K=4.5 according to the compaction method used (DE [222] → The highest packing density

(c) CONCRETE COMPOSITION
Volume of paste ($V_p$): experimentally defined→The lowest value-CO2/MPa
Initial volume of paste: minimum volume of paste (VPm) + 2%
VPm according to voids index calculation
Add paste 2% by 2% up to 8% paste excess
Minimum water/fines (w/f) ratio: minimum water required for the production of a homogeneous concrete in the plastic state
Lower compressive strength may be obtained by increasing the w/fines ratio
Consumption of each material:

$$V_f = \frac{R_f * (V_p - V_{Air} - V_{S.Ad})}{1 + w/f_{(v)}} \qquad \sum_{i=1}^{n} R_f = 1$$

$$V_{Agg} = R_{Agg} * (1 - V_p) \qquad \sum_{i=1}^{n} R_{Agg} = 1$$

Where,
$V_f$ is volume of the fine material (m3); $w/f_{(v)}$ is water/fines ratio by volume (m3); $R_f$ is volumetric ratio of the fine material(-);$V_p$ is paste volume (m3); $V_{Air}$ is air content (m3); $V_{S.Ad}$ is solids content of the chemical admixture (m3); $V_{Agg}$ is volume of the aggregate (m3); $R_{Agg}$ is volumetric ratio of the aggregate (-); $V_{Air} + V_{S.Ad} = 2\%$ of the paste volume.

## 6.3 Chapter summary

This chapter commences by highlighting the $CO_2$ emission reduction targets delineated in the IEA/WBCSD Roadmap. A comprehensive statement of ambition has been issued, urging collaborative efforts from all cement enterprises to achieve a 20–25% reduction in $CO_2$ emissions by 2030. The exploration of sustainable cement-based products is proposed through two key concepts: the incorporation of supplementary alternatives with low $CO_2$ content as partial substitutes for clinker in Portland cement and the enhanced utilization of Portland cement clinker in mortars and concretes. Building upon these concepts, the cyclic mix design method, based on particle packing techniques, has been introduced and examined for the production and evaluation of the properties of low-cement concrete (LCC).

Furthermore, an innovative method for designing sustainable High-Strength Concrete (HSC) [218] was introduced. This method utilizes particle packing techniques to enhance the optimization of concrete's granular components.

In recent years, the underlying rationale for the evolution of mix design methods utilizing mineral dusts as additives aims to diminish the voids between cement particles by incorporating fine materials, whether reactive or inert. Hence, the substantial volume of stone slurry generated by the natural stone industry, as discussed in previous chapters, holds significant potential for reuse, contributing to the development of low-$CO_2$ emission cement-based products.

# 7. Conclusion and prospect

Following the aforementioned studies, the subsequent conclusions can be drawn:

- ✓ In the global market of natural stone quarrying, stone slurry waste accounts for more than half (40.8% from quarry sites and 16% from warehouses in 2020) of the total natural stone extracted. This presents a significant environmental challenge but also holds substantial potential for recycling and reuse with the exploration of appropriate methods in the future.
- ✓ Currently, the disposal and utilization of this stone waste can be broadly categorized into three main applications: as fillers for binders in concrete and mortar, in ceramic formulations, and in various environmental applications spanning concrete, mortar, ceramic formulations, environmental restoration, as well as industries such as paint, paper, rubber, composite materials, catalysts, and more.
- ✓ Among these stone-based wastes, stone slurry, encompassing silicate and carbonate, possesses a finely distributed size, low permeability, and inert chemical properties. These qualities make them suitable as additives in cement-based products and as fine aggregates in mix design.
- ✓ The historical use of stone slurry as cement fillers dates back more than 100 years, with ongoing studies indicating not only improvements in the physical and mechanical performances of concretes/mortars but also a reduction in the utilization of natural raw materials. The incorporation of these dusts results in a dual benefit by addressing industrial waste consumption.
- ✓ The exploration of a novel mix design method for cement-based products utilizing natural stone slurry as fillers, aiming to enhance or sustain the acceptable mechanical and chemical properties of mortar/concrete, holds significant potential in contributing to the targeted reduction of $CO_2$ emissions in the cement industry sector.

Drawing upon the preceding conclusion, our future endeavors encompass:

- ✧ Embracing the substantial volume of existing wasted natural stone slurry as fillers in cement-based products. Considering the mechanical and chemical properties of these stone slurry powders sourced from diverse quarries, we aim to categorize these slurry wastes for distinct applications. The objective is to derive favorable outcomes such as enhanced compressive/tensile strength and improved workability.

- Pioneering innovative mix design methods that incorporate various types of stone slurry. These methods may find inspiration from the cyclic mix design and High-Strength Concrete (HSC) mix design approaches, leading to the development of low-cement concrete (LCC). This initiative aligns with our commitment to contributing to the reduction of CO2 emissions within the cement sector.

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