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An Engineered Production Formula for Enhanced Artificial Stone Utilizing Stone Cutting Slurry and a Superplasticizer

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ABSTRACT

The purpose of this paper is to demonstrate the technical feasibility of utilizing some of the local industrial waste (stone cutting wastewater and marble) in manufacturing artificial stone and enhancing its quality, with the addition of a superplasticizer (liquefier). The paper reviews the previous work and presents new experimental work.

Artificial stone samples were casted according to two starting formulas obtained from local production facilities. Later on, an engineered production formula was developed and tested, based on product quality. The investigated experimental parameters included: compressive strength, water absorption, and workability of fresh concrete. The experimental results indicated that high compressive strength and a low water absorption for artificial stone is technically feasible. In term of the compressive strength of artificial stone the marble dust contributed better than limestone wastewater.

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Introduction

Natural stone is one of the most important construction materials in the Middle East and many other regions around the world. However, the excessive use of such a natural resource has led to its enormous depletion, causing devastating impact on environment and biodiversity. On the contrary, the amounts of solid waste, especially industrial by-products are increasing dreadfully due to the population growth and the increase of economic activities. There is an increasing need to come up with innovative ideas to recycle the generated waste and use it in developing useful materials such as artificial stone.

Artificial stone is a construction material mix of white cement, aggregates, mortar and potentially a specific admixture. It is manufactured to simulate natural cut stone. It is used as an architectural feature, trim, ornament or facing for buildings or other structures. Artificial stone was first produced in the 18th century; it has been an initial building material for hundreds of years [1, 2]. The process of manufacturing artificial stones can be tightly controlled in terms of the ingredients, physical conditions, and curing time, to produce steady stone, which has an advantage over the natural stones, which suffers from visual and physical variations.

Marble and stone cutting industry generates a huge amount of wastewater with stone particulate material, consisting mainly of mineral oxides (e.g. $CaCO_3$,MgO, and SiO_2) [3]. The particle content in the wastewater has a range of 5-12 g/L. There are various wastewater treatment methods, resulting in concentrating particles and yielding a concentrated slurry. These include, sedimentation, flocculation (The process by which small particles of fine soils and sediments aggregate into larger lumps by adding a polymeric flocculating agent) and filtration[2]. Therefore, Dumping these waste into open areas has various environmental impacts on soil fertility, surface and ground water. Previous research investigated the possibility of utilizing such a waste in concrete, bricks and artificial stone [4]. In a published review paper, these research direction were surveyed: Investigated parameters included mechanical properties(e.g. compressive strength (CS), flexural strength and tensile strength), physical properties (e.g. porosity, water absorption (WA) and water penetration), workability of fresh mix (e.g. slump test), and thermal characteristics (e.g. thermal conductivity) [5-14].

Table 1 summarizes the potential recycling of various industrial waste in the production of several types of concrete. These include self-compacting concrete (SCC), lightweight concrete (LWC), high strength concrete (HSC) and ordinary concrete products (CP).

The experimental procedure is well defined in the literature: It was based on casting concrete mixes, then curing. Then, the obtained concrete was tested for compressive strength, water absorption, flexural strength...etc. In some studies, other tests were performed such as splitting strength test and thermal conductivity test. Compressive strength (CS) was tested in all cases.

Various production formulas were used for producing artificial stone, as summarized in Table 2.

Table 1: Recycling of Industrial Waste in Production of Various types of Concrete

Year	Ref.	Uses	Waste	Additives	Tests	Results
1995	[15]	LWC	Textile cuttings	Portland cement	Water absorption	CS=4.781MPa WA=43.7%
1999	[16]	Concrete	Finely ground glass	Mineral additives	Mortar bar	CS>= 4.1MPa
2001	[17]	SCC	Fly ash	None	Drying shrinkage	CS=26-48MPa
2003	[18]	Concrete	Glass	Cement Fly ash	-	CS= 32MPa
2004	[19]	HSC	Palm oil fuel ash	Silica fume	-	CS=79.5MPa
2005	[20]	SCC	Limestone chalk powders	Super plasticizer	-	Increased
2007	[21]	LWC	Limestone powder wood sawdust	N/A*	-	CS=7.2MPa
2007	[22]	SCC	Limestone powders	Portland cement	-	CS=45_50MPa
2008	[23]	SCC	Marble dusts	Portland cement	-	Increased
2009	[8]	Concrete	Fly ash Silica fume	Water, super plasticizer	-	CS=50MPa
2009	[24]	LWC	Mineralized wood	N/A*	-	N/A*
2011	[7]	Concrete	Stone powder	N/A*	-	CS=33.02MPa
2011	[9]	LWC	Wood ash	Portland cement	Flexural strength Splitting strength	CS Decreased
2014	[6]	Concrete	Crushed Limestone	Portland cement	Slump test	CS Increased
2016	[5]	Concrete	Glass powder	CEM 1 super plasticizer	-	CS= 30MPa
2016	[10]	Concrete	Waste Glass	Fly ash	Flexural strength	CS= 22MPa

*N/A= No Data Available.

Table 2: Previous Studies of Artificial Stone Utilizing Mineral Waste

Year	Ref.	Uses	Waste	Additives	Tests	Results
2003	[25]	Artificial stone	Limestone dust	Portland cement	modulus of elasticity	CS> 7MPa
2008	[26]	Artificial stone slab	Waste glass powder and fine granite aggregates	Unsaturated polymer resins	Flexural strength	CS=148.8MP F S= 51.1MPa
2015	[27]	LWS	Granite Marble stone sludge	Unsaturated polymer	Flexural strength WA Tensile strength	CS> 90MPa FS> 45MPa WA< 0.64 TS> 35MPa
2003	[18]	Concrete	Glass	Cement Fly ash	-	CS= 32MPa
2004	[19]	HSC	Palm oil fuel ash	Silica fume	-	CS=79.5MPa
2005	[20]	SCC	Limestone chalk powders	Super plasticizer	-	Increased

There is gab in the available literature for a practical production formula that can yield better stone characteristics than those obtained from existing formulas. The manufacturers of artificial stone require such an engineered industrial formulas. This paper investigates the technical feasibility of producing artificial stone with a new production formula that improves product quality, utilizing stone cutting slurry and marble dust. It investigates the effects of adding stone cutting wastewater on stone compressive strength, water absorption and workability.

Materials and Methods

Materials

Materials used included: ordinary Portland white cement type CEMII/B-L32, 5R(br), local sand passing Tyler mesh #100, two sizes of angular aggregates (equal ratio of medium and fine gravel),tap water, CF-12superplasticizer(AFEC, "Netanya" city), sodium silicate (SiO2Na2O)(Imperplast, Italy), stone cutting slurry with flocculent, stone cutting slurry without flocculent, and marble cutting wastewater. Concrete materials were conformed to comply with ASTM specifications.

Equipment

The following apparatus and equipment's were used: oven to dry samples and materials, steel molds to cast the mixes (10X10X10cm), concrete compression machine (300 KN motorize, Matest, Italy), slump test equipment's.

Experimental Procedure

Artificial stone mix was prepared according to two suggested formulas (two control samples), obtained from local manufacturers (samples A and B in table 3). White cement was mixed with sand, aggregate and tap water for 10 min manually using a trowel. Other components were later on added according to a selected formula. Then the obtained mix was casted in steel molds, using control and modified samples.

The industrial formula A was improved by adding superplasticizer at 14.5% (sample AS in table 3). Such a ratio of the superplasticizer was adopted after preliminary experiments for the best compressive strength and workability, within the 12-15% ratios, according to standard B300 concrete.

The effects of stone cutting slurry addition on the modified production formula (AS) was investigated by casting stone at various addition ratios i.e. 5, 10, 15 and 20% (labeled ASP5, ASP10, ASP15 and ASP20 in table 3). The solid content of these additions replaced equivalent amounts of sand, according to mass balance calculations (the measured particle content in the used slurry was 23g/L). The water content of slurry was accounted for when adding the required water. For the purpose of comparison, cast stone samples were prepared with wastewater containing polymeric flocculent agent used in the industrial wastewater treatment at 10% ratio, i.e. similar to ASP10 sample. Other samples were prepared with addition of marble cutting wastewater (samples labeled ASM20 and ASM30 in table 3), for the purpose of comparison with limestone slurry. In these exploring experiments, wastewater was added as a partial replacement of tap water for artificial stone mixture (e.g. 20% and 30% replacement of water).

Table 3: The Investigated Artificial Stone Production Formulas

Formula Code	Materials				Alternatives		Fundamental experimental parameters
	White Cement (Kg)	Sand (Kg)	Aggregates (Kg)	Tap water (ml)	Waste	Super plasticizer (ml)	
A	2.6	2.7	6.5	1200	None	None	Practicality of industrial formula 1
B	2	2.9	9.5	1200	None	None	Practicality of industrial formula 2
AS	2.6	2.7	6.5	950	None	14.5	The addition of Super plasticizer
ASP5	2.6	2.56	6.5	950	5% stone slurry	14.5	Waste addition-ratio
ASP10	2.6	2.43	6.5	950	10% stone slurry	14.5	Waste addition-ratio
ASP15	2.6	2.3	6.5	950	15% stone slurry	14.5	Waste addition-ratio
ASP20	2.6	2.16	6.5	950	20% stone slurry	14.5	Waste addition-ratio
ASF	2.6	2.43	6.5	950	10% flocculated stone slurry	14.5	Utilizing wastewater with polymeric flocculating agent
ASM20	3.12	3.24	7.8	912	20% Marble	17.5	Utilizing wastewater with marble dust
ASM30	3.12	3.24	7.8	789	30% Marble	17.5	Waste addition-ratio

The workability of the fresh mixture was tested using slump test according to ASTM C143. The casted samples were left to dry in steel molds for 24 hours. Then, they were allowed to cure by immersing in water bath for 24 hours at room temperature. The compressive strength of the cured samples (for control and for cases with the waste additions) was measured at 1, 3, 7, 14, and 28 days according to ASTM C39M. The kinetic curves of compressive strength versus time were plotted. The obtained kinetic curves were confirmed to be reproducible.

The water absorption test was carried out after 28 days curing, according to ASTM C1585: The stone sample was first weighted; the mass was recorded as W1. The sample was then immersed in distilled water for 48 hours. Then, using a clean and dry towel, the surface of the sample was tumble dry, the sample was weighed and the mass was recorded as W2.The percentage water absorption (WA) was obtained from mass balance according to the following equation:

Percentage water absorption (WA) = $(w_2 - w_1)/w_1 \times 100\%$

For reducing water absorption, samples of the cast stone were treated by spraying the surfaces with sodium silicate (SiO2Na2O) solution (37.5% sodium silicate by weight in water). The treated samples are coded as above but by adding I letter before the above codes to distinguish between treated and untreated samples.

Results and Discussion

Table 4 lists the slump test results obtained from the experimental work. Obviously, production formula B has a bad workability, while A has a good workability. In all other formulas, the addition of wastewater from stone cutting increased the workability of the fresh mix; the presence of fine calcium carbonate (limestone) particles in the mix design has a positive impact on workability. The impact of workability on cast stone is reflected on homogeneity and integrity of the cast stone: Figure 1 shows images of 4 types of examined artificial stone. Obviously, cast stone from formula B have a bad homogeneity and weak integrity, as appears with large voids. No noticeable difference in appearance was observed with the addition of super plasticizer and stone cutting waste. The addition of marble particles has better impact on workability.

Table 4: Slump Test Results

Formula (mix design according to table 3)	Slump value (mm)	Classification of the Workability (ASTM)
Formula B	0	Very low
Formula A	75	Medium
Formula AS	89	Medium
Formula ASF	150	High
Formula ASP5	172	High
Formula ASP10	166	High
Formula ASP15	160	High
Formula ASP20	160	High
Formula ASM20	167	High
Formula ASM30	167	High

Results of the compressive strength development with time are presented in figure 2. Such a development in compressive strength reflects the kinetics of cement hydration reactions and concrete setting rate. Figure 2 compares the kinetic curves for artificial stone produced according to the two formulas obtained from local manufacturers (A and B). They are also compared to the kinetic curve for standard concrete B300. Obviously, for the two formulas, the compressive strength increases with time in a similar manner as in typical concrete curve. The 28-days compressive strength values for our artificial stone are higher than that for typical concrete (B300 with a compressive strength of 28MPa. The compressive strength of the artificial stone develops faster than that of concrete. This is an essential characteristic in manufacturing: productivity is strongly dependent on quick deforming after curing.

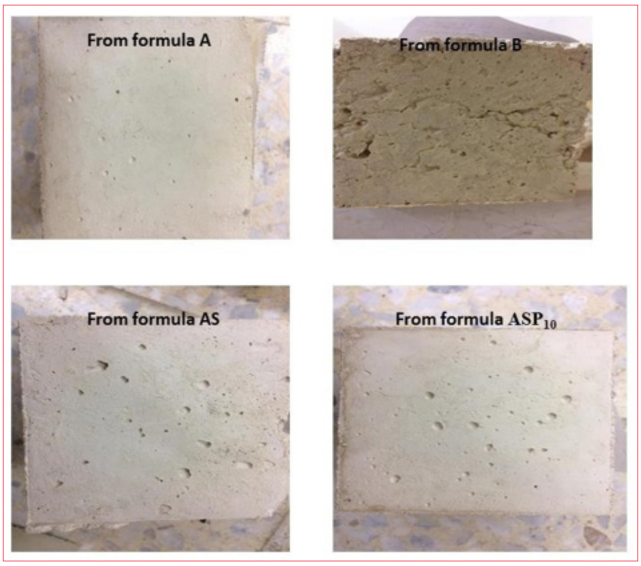

Figure 1: Images of the cast stone obtained from the two industrial production formulas (a and b), compared to the modified formulas AS and ASP$_{10}$

The higher compressive strengths of artificial stone compared to concrete is attributed to the fact that water to cement ratio in artificial stone is lower than that in ordinary concrete. In addition, white cement has smaller particles than the ordinary Portland cement. This enhances hydration reactions and improves the strength.

Figure 2 indicates that in both cases of industrial artificial stone (A and B), the 28-days compressive strength is above 40MPa. These values are within the range of the compressive strength for local natural stone. Their compressive strength values are higher than those for weak natural stones. These results indicate that the two industrial formulas have competitiveness in terms of mechanical characteristics with some natural stones. For examples, when cast stone was prepared using limestone waste and Portland cement, it yielded only 7MPa compressive strength. However, the other listed previous attempts were based on non-cementations ingredients, e.g , and thus they are not comparable [25, 26].

The artificial stone produced with formula A has higher compressive strength than that of B; since it is with higher water /cement ratio than that with formula B. Thus, production formula A was adopted as a base line in the subsequent work.

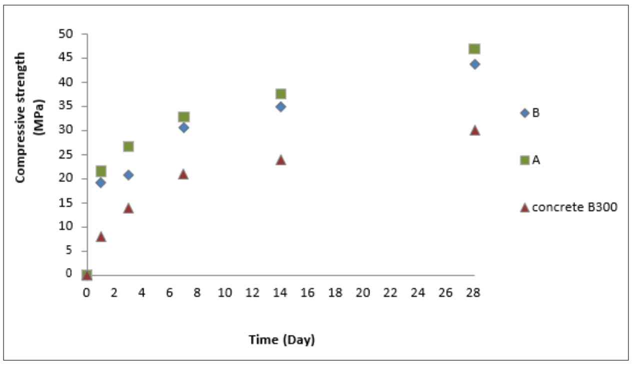

Figure 2: The measured compressive strength (MPa) versus time (day) for artificial stone produced according to the two industrial formulas obtained from local manufacturers (A and B in table 3), compared to kinetics for standard concrete B300.

Enhancing Compressive Strength of the Artificial Stone

Figure 3 shows the compressive strength curve obtained for artificial stone produced according to industrial formula A modified with the addition of superplasticizer (formula labeled AS in table 3), for three replicates. The points nearly superimpose on each other, confirming the reproducibility of the experimental results. Comparing these results with those in Figure 2 indicates that adding a superplasticizer to the formula A increases the compressive strength considerably. The 28-days compressive strength jumps over 100MPa. The used superplasticizer is a high range water reducer (HRWR).

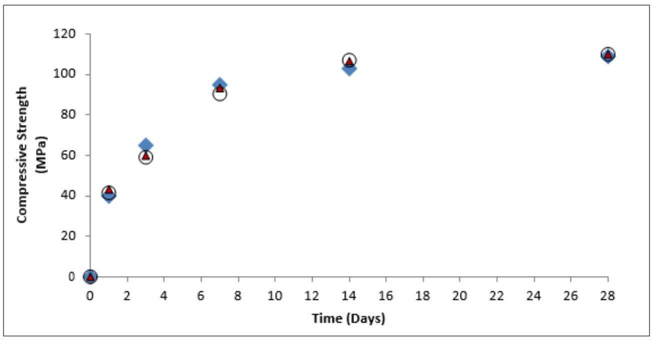

Figure 3: The measured kinetic curve of compressive strength (MPa) versus time (day) for artificial stone produced according to industrial formual A modiefied with the addition of superplastizier (AS formula in table 3), showing results of three replicates

The effect of adding stone cutting wastewater on the compressive strength was investigated, by manipulating the formula (AS) with various ratios of stone cutting wastewater. Figure 4 presents the experimental kinetic curves of compressive strength for various percentage additions of stone cutting wastewater (samples ASP5, ASP10, ASP15 and ASP20 in table 3). In these experiments, wastewater was from a local source without flocculating agent. At early stages, (up to 3 days), there was no strong dependence of compressive strength on stone particle content. However, at late stages, the compressive strength is strongly dependent on the addition of wastewater to the mix. With low percentage range, the compressive strength increased with increasing the percentage of wastewater from 5% to 10%. On the other hand, as the percentage was increased above 10% a trend of decrease in the compressive strength was observed. Among the investigated percentages, the 10% addition of stone cutting wastewater is the most appropriate production formula. The 28-days compressive strength reached 84MPa. This is nearly two times more than the existing industrial formula for artificial stone. This improvement in the strength is associated with a reduction in the cost of raw materials since part of the mixture is waste (stone slurry). In addition, this approach has a positive impact on environment. In this case, the utilization of the waste in production is associated with a large increase in compressive strength, compared to the existing production formulas.

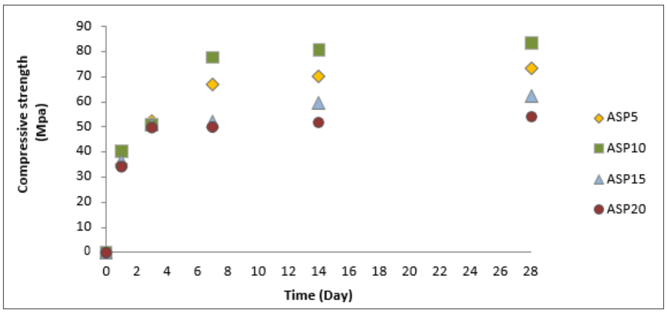

Figure 4: The experimental kinetic curves of compressive strength (MPa) for various percentages of stone cutting wastewater, without flocculating agent (samples ASP$_5$, ASP$_{10}$, ASP$_{15}$ and ASP$_{20}$ in table 3).

Figure 5 presents the results of 28-days compressive strength for the above cases, and compares them to the other cases investigated in this work: When another source of stone cutting wastewater containing a flocculating agent was used (with 10% addition) i.e. sample ASF, a compressive strength of 60MPa was obtained. This is lower than that for the case of wastewater without a flocculating agent, at the equivalent ratio (i.e. ASP10 sample). This is attributed to the fact that the addition of the polymer resulted in a clustered stone particles. Those interns decreased the consistency of the mix, and then decreased the bonding upon drying and solidification of polymer, since it residues as non-cementations content.

The effect of using marble cutting wastewater was explored first at 20% addition. The obtained 28-days compressive strength was 59MPa. This is slightly larger than that for equivalent ration of stone cutting wastewater (i.e. ASP20 sample with 54MPa). Marble particles are much stronger and harder than limestone particles. Thus, for the same addition ratio, the resulting strength is larger. Increasing the ratio of marble cutting wastewater to 30% increased the compressive strength to 68MPa. This behavior of increasing compressive strength with high percentage seems to be different than that observed with stone cutting wastewater. This can be interpreted to the fact that the marble particles are much stronger than lime stone particles.

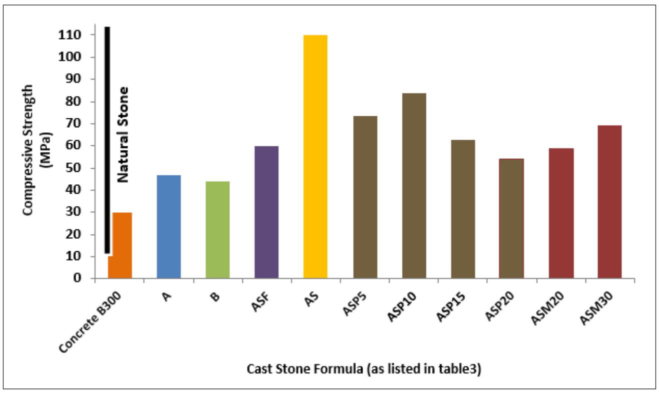

Figure 5: The 28-days compressive strength of various types of investigated formulas and for standard concrete B300, compared to that of natural stone

Reducing Water Absorption of the Artificial Stone

Figure 6: compares water absorption percentage of the produced artificial stone for modified formulas (ASP5, ASP10, ASP15 and ASP20, as listed in table 3) and compares it with other samples. The measurements were made after 28 days curing according to ASTM C1585 standard. In addition to strength, these formulas have an additional competiveness factor over natural stone the figure also shows the reported range of water absorption of natural stones (brown bar). Clearly, the water absorption for all the investigated modified formulas is at the lower limit of that of natural stone. Slightly larger water absorption was obtained for artificial stone produced with wastewater containing a polymeric flocculating agents.

Water absorption of the cast stone was reduced considerably by its surface treatment. This was accomplished, in this work, by spraying the produced artificial stone with sodium silicate (SiO2Na2O) solution. Such a surface treatment resulted in less than 1% water absorption for all samples for formulas. It is well known in waterproofing technology, that sodium silicate is a hydrophobic material. Upon its application, the surface becomes water repellent and thus less water is allowed to penetrate within the stone.

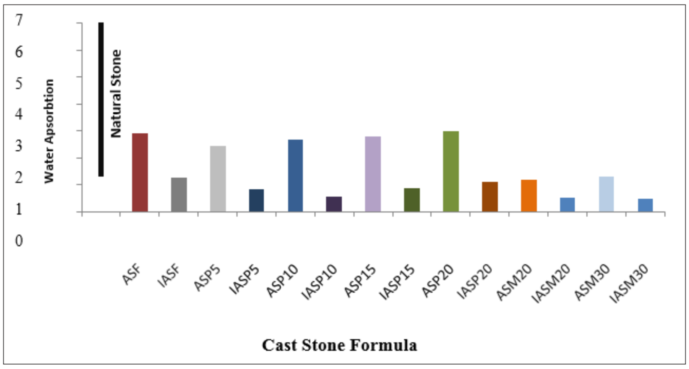

Figure 6: Water absorption of cast stone samples for formulas (samples ASP$_5$, ASP$_{10}$, ASP$_{15}$ and ASP$_{20}$, ASM$_{20}$ and ASM$_{30}$as listed in table 3).

Conclusions

Industrial waste from stone cutting industry is a potential additive for producing environmentally friendly construction stone. An engineered production formula for manufacturing artificial stone was developed. It is based on stone cutting wastewater, marble and superplasticizer.

In comparison to artificial stone produced using limestone dust, using marble dust produces artificial stone with better properties. Stone cutting wastewater without a flocculating agent yielded better products. The addition of 10% stone cutting wastewater, as a replacement of sand, yielded an artificial stone with 84MPa, a value that is much higher than the industrially used production formula. Water absorption values were at lower levels than those for natural stone. The surface treatment of the cast stone with sodium silicate decreased water absorption.

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