# An engineered production formula for enhanced artificial stone utilising stone cutting slurry and a superplasticiser

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**Abstract:** This paper demonstrates the technical feasibility of utilising local industrial wastewater (stone cutting wastewater and marble cutting wastewater) with a superplasticiser in manufacturing artificial stone of enhanced quality. 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. Based on that, an engineered production formula was developed and tested, based on stone product quality. The investigated experimental parameters included compressive strength, water absorption, and workability of fresh concrete. The experimental results indicate that a 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.

**Keywords:** artificial stone; stone cutting; marble dust; waste; recycling; compressive strength; water absorption.

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# 1    Introduction

Natural stone is one of the most important construction materials. 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 and industrial by-products are increasing dreadfully due to the population growth and the increase of economic activities. There is an increasing demand for coming up with innovative ideas to recycle the generated waste and to use it in developing useful materials such as artificial stone.

Artificial stone is a construction material produced from the 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 had been an initial building material for hundreds of years (Boyer, 2012; Al-Jabari and Sawalha, 2002). 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, since the latest 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 $SiO_2$) (Hanieh et al., 2014). The particle content in the wastewater has a range of 5–12 g/L. There are various wastewater treatment methods, which recover treated water and yield a concentrated slurry. These include, sedimentation, flocculation (through adding a polymeric flocculating agent) and filtration (Al-Jabari and Sawalha, 2002). Dumping these wastes into open areas has various environmental impacts on soil fertility and surface and ground water (Al-Joulani, 2014). Previous publications investigated the possibility of utilising such a waste in concrete (Islam et al., 2017; Fathi, 2014; Mahzuz et al. 2011; Barbhuiya et al., 2009; Cheah and Ramli, 2011; Spiesz et al., 2016), bricks (Binici et al., 2005; Rahman, 1987; Demir, 2006) and artificial stone (Al-Joulani, 2014). In a published review paper, these research direction were surveyed (Sanchez and Sobolev, 2010): investigated parameters included mechanical properties [e.g., compressive strength (CS), flexural strength (FS) and tensile strength (TS)], 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). The experimental procedure was based on casting concrete mixes, then curing. Then, the obtained concrete was tested for CS, WA, FS, etc. In some studies, other tests were performed such as splitting strength test and thermal conductivity test. CS was tested in all cases.

**Table 1** Recycling of industrial waste in production of various types of concrete

| Year | Ref. | Uses | Waste | Additives | Tests | Results |
|---|---|---|---|---|---|---|
| 1995 | Aspiras and Manalo | LWC | Textile cuttings | Portland cement | WA | CS = 4.781 MPa WA = 43.7% |
| 1999 | Shao et al. | Concrete | Finely ground glass | Mineral additives | Mortar bar | CS >= 4.1 MPa |
| 2001 | Bouzoubaa and Lachemi | SCC | Fly ash | None | Drying shrinkage | CS = 26–48 MPa |
| 2006 | Shayan and Xu | Concrete | Glass | Cement fly ash | - | CS = 32 MPa |
| 2004 | Sata et al. | HSC | Palm oil fuel ash | Silica fume | - | CS=79.5 MPa |
| 2005 | Zhu and Gibbs | SCC | Limestone chalk powders | Super plasticiser | - | Increased |
| 2007 | Turgut and Algin | LWC | Limestone powder wood sawdust | N/A* | - | CS = 7.2 MPa |
| 2007 | Felekoglu | SCC | Limestone powders | Portland cement | - | CS = 45–50 MPa |
| 2008 | Binici et al. | SCC | Marble dusts | Portland cement | - | Increased |
| 2009 | Barbhuiya et al. | Concrete | Fly ash silica fume | Water, super plasticiser | - | CS = 50 MPa |
| 2009 | Becchio et al. | LWC | Mineralised wood | N/A* | - | N/A* |
| 2011 | Mahzuz et al. | Concrete | Stone powder | N/A* | - | CS = 33.02 MPa |
| 2011 | Cheah and Ramli | LWC | Wood ash | Portland cement | FS and splitting strength | CS decreased |
| 2014 | Fathi | Concrete | Crushed Limestone | Portland cement | Slump test | CS increased |
| 2016 | Islam et al. | Concrete | Glass powder | CEM 1 super plasticiser | - | CS = 30 MPa |
| 2016 | Spiesz et al. | Concrete | Waste glass | Fly ash | FS | CS = 22 MPa |
| 2018a | Devi et al. | Concrete | Stone cutting slurry | N/A* | CS, split TS and electrical resistivity | Increasing electrical resistivity decreasing CS |
| 2018b | Devi et al. | Mortar | Stone cutting slurry | Calcium nitrate and triethanolamine | CS, consistency, setting time | Increasing consistency decreasing setting time calcium nitrate and stone slurry increased CS |

Notes: *N/A = no data available.

**Table 2**    Previous studies of artificial stone utilising mineral waste

| Year | Ref. | Uses | Waste | Additives | Tests | Results |
|------|------|------|-------|-----------|-------|---------|
| 2004 | Galetakis and Raka | Artificial stone | Limestone dust | Portland cement | Modulus of elasticity | CS > 7 MPa |
| 2008 | Lee et al. | Artificial stone slab | Waste glass powder and fine granite aggregates | Unsaturated polymer resins | FS | CS = 148.8 MPFS = 51.1 MPa |
| 2016 | Barani and Esmaili | LWS | Granite marble stone sludge | Unsaturated polymer | FS, TS and WA | CS > 90 MPa FS > 45 MPa WA < 0.64 TS > 35 MPa |
| 2018 | Silva et al. | Artificial stone | Marble calcite waste | Epoxy resin | FS | FS = 31.8 MPa CS = 85.2 MPa WA < 0.05% |
| 2018 | Abd Al-Majeed et al. | Cast stone | Glass wastes | Plasticiser admixture | CS, FS, WA and drying shrinkage | CS = 47.5–44.3 MPa WA = 5.3–4.7% |

Table 1 summarises potential recycling approaches of various industrial wastes 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). Various production formulas were used for producing artificial stone, as summarised in Table 2.

There is gab in the available literature regarding a practical production formula that can yield better stone characteristics than those obtained with 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, utilising stone cutting slurry and marble dust. It investigates the effects of adding stone cutting wastewater on stone CS, WA and workability.

**Figure 1** Images of the (a) stone particles and (b) marble particles obtained using scanning electron microscope
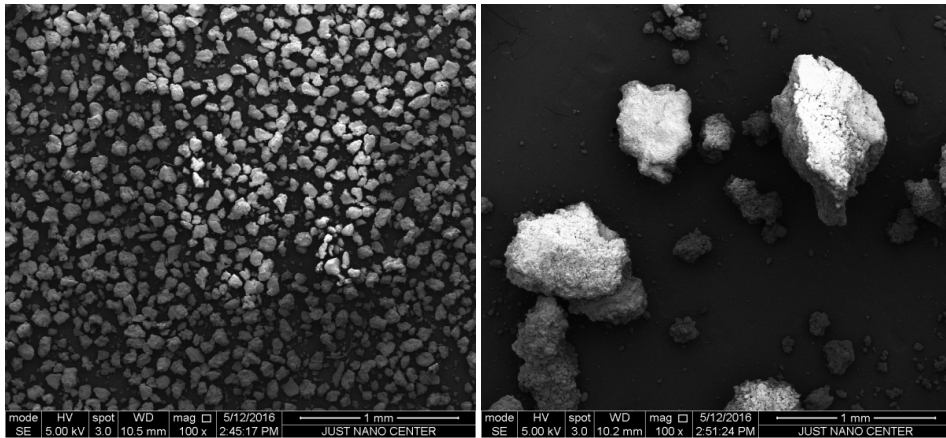


(a)                                        (b)

## 2    Materials and methods

### 2.1    Materials

Materials used included ordinary Portland white cement type CEMII/B-L32, 5R(br), local natural sand passing Tyler mesh #100, two sizes of angular crushed limestone aggregates, with equal proportions of medium aggregates passing mesh #1 and fine aggregates passing mesh #8, and tap water. Concrete materials were conformed to comply with ASTM specifications. The additives included superplasticiser (CF-12 from AFEC E.R.M.A Ltd, Matan), defined by the manufacturer as a liquid admixture for concrete mixes, which modifies the rheology of concrete and giving it thixotropic properties, sodium silicate ($SiO_2Na_2O$) (Imperplast, Italy), stone cutting slurry with flocculent, stone cutting slurry without flocculent, and marble cutting wastewater. The stone cutting slurry composed of water and negatively charged limestone particles containing about 85% of calcium carbonate ($CaCO_3$), determined using reverse titration. The remaining

composition included other mineral oxides such as $SiO_2$ and MgO. The solid content was about 23 g/L. The stone particles had an approximate particle size of about 50 μm. Figure 1 shows images of particles from stone cutting wastewater and from marble cutting wastewater obtained using scanning electron microscope.

## 2.2   Equipment

The used apparatus and equipment included oven to dry samples and materials, steel moulds to cast the mixes (10 × 10 × 10 $cm^3$), concrete compression machine (300 KN motorise, Matest, Italy), slump test apparatus and a water bath.

## 2.3   Experimental procedure

Table 3 lists all formulas of the investigated artificial stone specimens. Reference specimens of artificial stone were prepared using two suggested formulas (two control samples), obtained from local manufacturers (samples A and B in Table 3). White cement was mixed manually with sand, aggregate and tap water for 10 min, using a trowel. Other components were then added according to the selected formula. Then, the obtained mix was poured in steel moulds. Standard cubic specimens (10 × 10 × 10 $cm^3$) were made for control and modified samples, then tested for CS and WA.

The industrial formula A was improved by adding 14.5 ml superplasticiser (sample AS in Table 3). Such an addition of the superplasticiser was adopted after preliminary experiments of adding superplasticiser with a range of 12–15 ml then finding the best CS and workability, comparing it to the standard B300 concrete.

The effects of adding stone cutting slurry on the modified production formula (AS) was investigated by casting artificial stone specimens with various percentage additions as partial replaces of sand including 5, 10, 15 and 20% (labelled $ASP_5$, $ASP_{10}$, $ASP_{15}$ and $ASP_{20}$ in Table 3). These additions were determined from mass balance calculations, knowing that the measured particle content in the used slurry was 23 g/L. The water content in the used slurry was accounted for when adding the required mixing water. For the purpose of comparison, cast stone specimens were prepared with wastewater containing polymeric flocculent agent used in the industrial wastewater treatment at 10% addition, i.e., similar to $ASP_{10}$ sample. Other samples were prepared with addition of marble cutting wastewater (samples labelled $ASM_{20}$ and $ASM_{30}$ 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).

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

The WA test was carried out for fully cured specimens at 28 days curing, according to ASTM C1585: The dry cubic stone specimen was first weighted; the mass was recorded as $W_1$. The sample was then immersed in distilled water for 48 hours. Then, using a clean and dry towel, the surface of the specimen was tumble dry, the specimen was weighed

and the mass was recorded as $W_2$. The percentage WA was obtained from mass balance according to the following equation:

$$\text{Percentage water absorbtion (WA)} = (w_2 - w_1)/w_1 \times 100\%$$

For reducing WA, samples of the cast stone were treated by spraying the surfaces with sodium silicate solution (37.5 wt% sodium silicate ($SiO_2Na_2O$)). The treated ($SiO_2Na_2O$) were coded as above but by adding I letter before the above codes to distinguish between treated and untreated samples, e.g., I-ASP10 is specimen ASP10 with sodium silicate surface treatment.

**Table 3**     Formulas of the investigated artificial stone

| Code | Addition | Materials | | | Additives | | | Investigated experimental parameters |
|------|----------|-----------|------|-----------|-------|-------|------------------------|--------------------------|
| | | White cement | Sand | Aggregates | Water | Waste | Super plasticiser (ml) | |
| **A** | kg | 2.6 | 2.7 | 6.5 | 1.2 | None | none | Practicality of industrial formula 1 |
| | wt% | 20.0 | 20.8 | 50.0 | 9.2 | 0.0 | | |
| **B** | kg | 2 | 2.9 | 9.5 | 0.95 | None | none | Practicality of industrial formula 2 |
| | wt% | 13.0 | 18.9 | 61.9 | 6.2 | 0.0 | | |
| **AS** | kg | 2.6 | 2.7 | 6.5 | 0.95 | None | 14.5 | The addition of super plasticiser |
| | wt% | 20.4 | 21.2 | 51.0 | 7.5 | 0.0 | | |
| **ASP$_5$** | kg | 2.6 | 2.56 | 6.5 | 0.95 | 0.13 | 14.5 | Effect of stone waste addition percentage 5% |
| | wt% | 20.4 | 20.1 | 51.0 | 7.5 | 1.0 | | |
| **ASP$_{10}$** | kg | 2.6 | 2.43 | 6.5 | 0.95 | 0.26 | 14.5 | Effect of stone waste addition percentage 10% |
| | wt% | 20.4 | 19.1 | 51.0 | 7.5 | 2.0 | | |
| **ASP$_{15}$** | kg | 2.6 | 2.3 | 6.5 | 0.95 | 0.39 | 14.5 | Effect of stone waste addition percentage 15% |
| | wt% | 20.4 | 18.1 | 51.0 | 7.5 | 3.1 | | |
| **ASP$_{20}$** | kg | 2.6 | 2.16 | 6.5 | 0.95 | 0.52 | 14.5 | Effect of stone waste addition percentage 20% |
| | wt% | 20.4 | 17.0 | 51.1 | 7.5 | 4.1 | | |
| **ASF** | kg | 2.6 | 2.43 | 6.5 | 0.95 | 0.26 | 14.5 | Effect polymeric flocculating agent in stone waste – 10% |
| | wt% | 20.8 | 19.5 | 52.1 | 7.6 | 0.0 | | |
| **ASM$_{20}$** | kg | 3.12 | 3.24 | 7.8 | 0.912 | 0.624 | 17.5 | Effect of marble waste addition percentage 20% |
| | wt% | 19.9 | 20.6 | 49.7 | 5.8 | 4.0 | | |
| **ASM$_{30}$** | kg | 3.12 | 3.24 | 7.8 | 0.789 | 0.936 | 17.5 | Effect of marble waste addition percentage 30% |
| | wt% | 19.6 | 20.4 | 49.1 | 5.0 | 5.9 | | |

## 3    Results and discussion

Table 4 lists the slump test reults obtained from the experimental work. The slump is dependent on water content (or water to cement ratio) and on the proportions of aggregates. Slump is improved by the addition of fine materials. Obviously, production formula B has a very bad workability, while A has a good workability. This is attributed the low content of water and fine materials in formula B (lower cement particles and lower sand particles than formula A). In addition, specimen B contains a larger proportion of the aggregates in the mix design than specimen A (see Table 3). In all other formulas, the addition of wastewater from stone cutting increased the workability of the fresh mix; since the presence of fine calcium carbonate (limestone) particles in the mix design has a positive impact on workability. It is obvious from Figure 1 that these stone particles have a size close to that of cement particles, and thus their addition improves the workability, in a similar manner as when adding more cement particles. The impact of workability on cast stone is reflected on homogeneity and integrity of the cast stone: Figure 2 shows images of 4 types of examined artificial stone. Obviously, cast stone from formula B has a bad homogeneity and weak integrity, as reflected with large voids. No noticeable difference in appearance was observed with the addition of superplasticiser and stone cutting waste. The addition of marble particles has slightly higher workability than that for the case with stone cutting wastewater. However, the production formula is different.

**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 $ASP_5$ | 172 | High |
| Formula $ASP_{10}$ | 166 | High |
| Formula $ASP_{15}$ | 160 | High |
| Formula $ASP_{20}$ | 160 | High |
| Formula $ASM_{20}$ | 167 | High |
| Formula $ASM_{30}$ | 167 | High |

Results of the CS development with time are presented in Figure 3. Such a development in CS reflects the kinetics of cement hydration reactions and concrete maturity. Figure 3 compares the curves of CS development for artificial stone produced according to the two formulas obtained from local manufacturers (A and B) and for standard concrete B300. Obviously, for the two formulas, the CS increases with time in a similar manner as in typical concrete curve (B300). The CS for fully cured specimens at 28 days for the artificial stone is higher than that for typical concrete (B300 with a CS of 28 MPa). The CS 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 2** Images of the cast stone obtained from the two industrial production formulas (A and B), compared to the modified formulas AS and $ASP_{10}$ (see online version for colours)
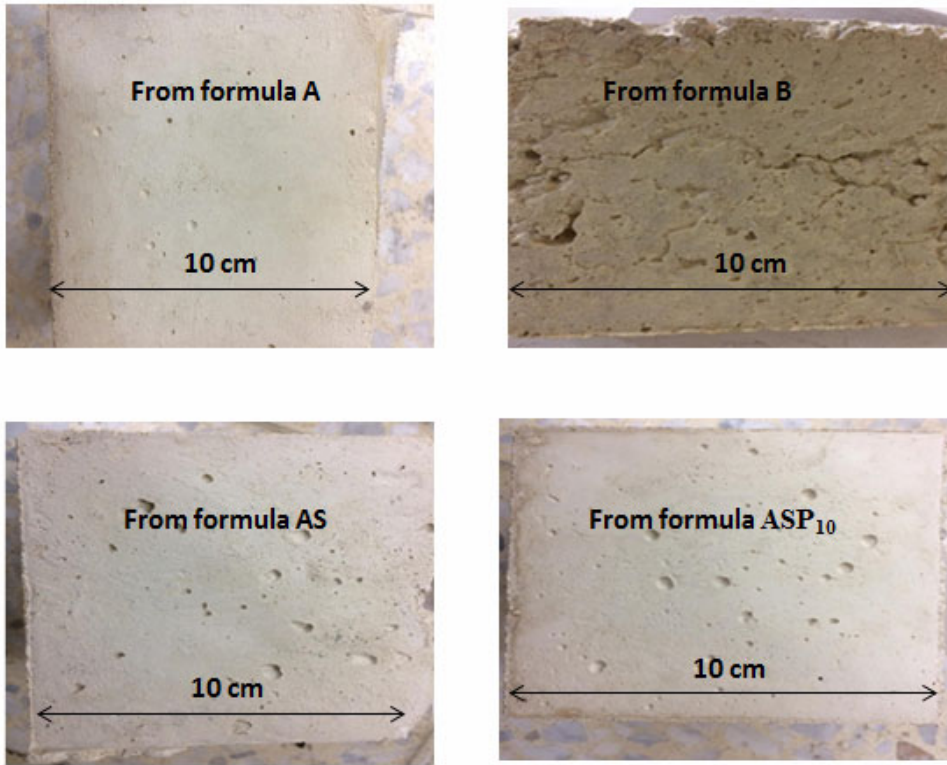


The higher CS s of artificial stone compared to concrete is attributed to the fact that water to cement ratio in artificial stone is lower than that in ordinary concrete. In addition, white cement has smaller particles than the ordinary Portland cement. This enhances hydration reactions and improves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

Figure 3 indicates that the CS for fully cured specimens at 28 days is above 40 MPa, for both cases of industrial artificial stone (A and B). These values are within the range of the CS for local natural stone. Such values of CS are higher than those of weak natural stones, as a lower limit. These results indicate that the two industrial formulas have competiveness in terms of mechanical characteristics compared to some natural stones. The obtained range of CS is much larger than that obtained in the previous research attempts that utilised waste and by-products in producing artificial stone (see surveyed publications in Table 2). For examples, when cast stone was prepared using limestone waste and Portland cement (Galetakis and Raka, 2004), it yielded only 7 MPa CS. On the other hand, other listed previous attempts were based on non-cementations ingredients (Lee et al., 2008), and thus they are not comparable.

The artificial stone produced with formula A has higher CS 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 3**   The measured CS (MPa) versus curing time (day) for artificial stone produced according to the two industrial formulas obtained from local manufacturers (A and B as listed in Table 3), compared to that for standard concrete B300 (see online version for colours)
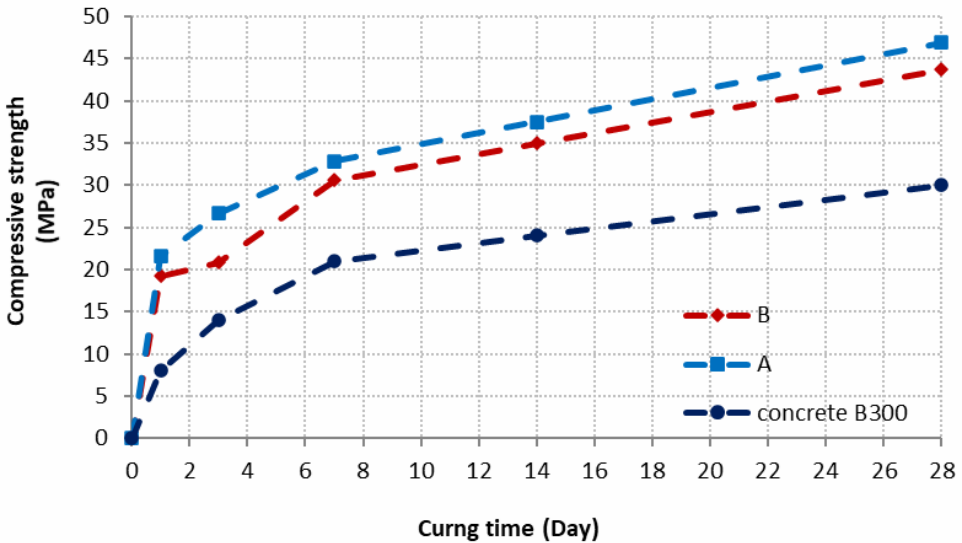


## 3.1   Enhancing CS of the artificial stone

Figure 4 shows the CS curve obtained for artificial stone produced according to industrial formula A modified with the addition of superplasticiser (formula labelled 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 3 indicates that adding a superplasticiser to the formula A increases the CS considerably. The 28 days CS jumps over 100 MPa. The used superplasticiser is a high range water reducer (HRWR).

The effect of adding stone cutting wastewater on the CS was investigated, by manipulating the formula (AS) with various ratios of stone cutting wastewater. Figure 5 presents experimental curves of the development of CS with age for various percentage additions of stone cutting wastewater (samples $ASP_5$, $ASP_{10}$, $ASP_{15}$ and $ASP_{20}$ 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 CS on stone particle content. However, at late stages, the CS is strongly dependent on the addition of stone cutting wastewater to the mix. With low percentage additions, the CS increased with increasing the percentage of wastewater from 5% to 10%. This might attributed to the positive effect of calcium carbonate on cement hydration. On the other hand, as the percentage was increased above 10% a trend of decreasing CS was observed. Among the investigated percentages, the 10% addition of stone cutting wastewater is the most appropriate production formula. The 28 days CS reached 84 MPa. 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 cutting slurry). In addition, this approach has a positive impact on

environment. In this case, the utilisation of the waste in production is associated with a large increase in CS, compared to the existing production formulas.

**Figure 4** The measured CS (MPa) versus curing time (day) for artificial stone produced according to industrial formula A modiefied with the addition of superplastisier (AS formula in Table 3), showing results of three replicates (see online version for colours)
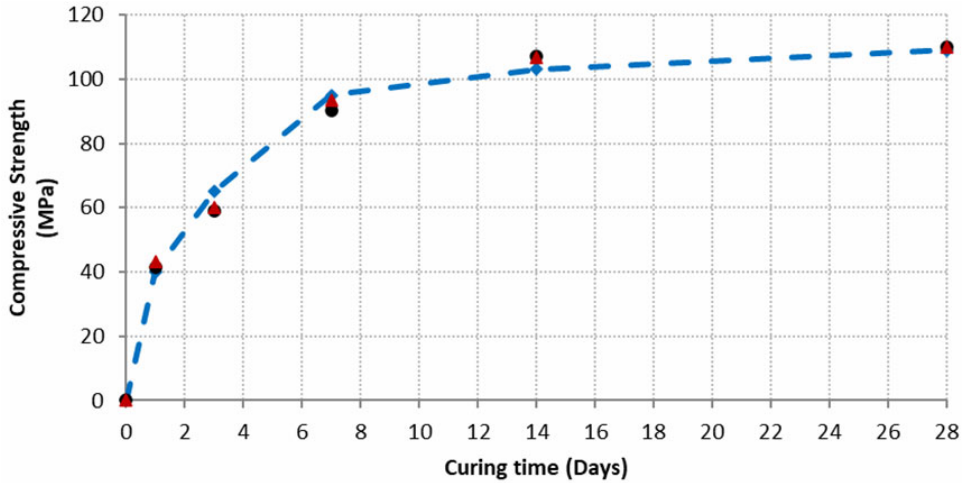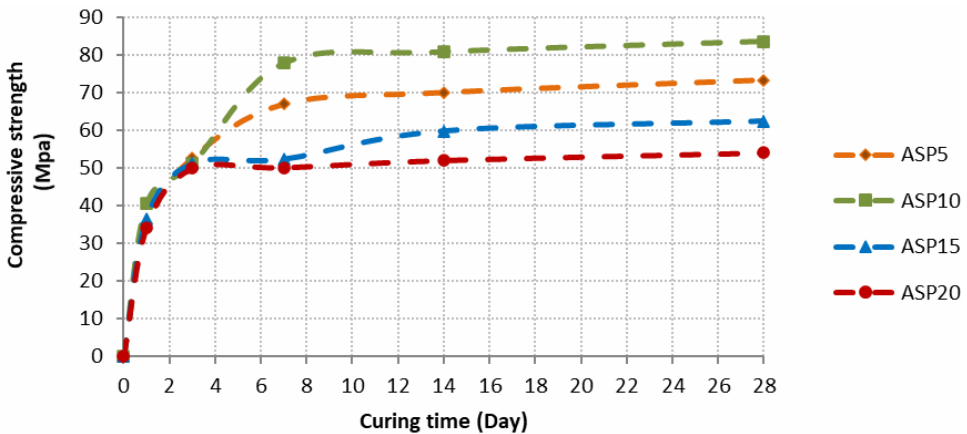


**Figure 5** The measured CS (MPa) versus curing time (day) for various addition percentages of stone cutting wastewater, without flocculating agent (see online version for colours)



Notes: Samples $ASP_5$, $ASP_{10}$, $ASP_{15}$ and $ASP_{20}$ in Table 3.

Figure 6 presents the results of CS for fully cured specimens at 28 days for the above cases, compared to the other cases listed in Table 3: when another source of stone cutting wastewater containing a flocculating agent was used (with 10% addition) i.e., sample ASF, a CS of 60 MPa was obtained. This is lower than that for the case of wastewater without a flocculating agent, at the equivalent ratio (i.e., $ASP_{10}$ sample). This is attributed to the fact that the addition of the polymer resulted in a clustered stone particles. These flocculated particles decrease the consistency of the mix, and then decrease

the bonding upon drying and solidification of polymer, since the polymer residues are non-cementations content.

**Figure 6**     The CS of fully cured specimens at 28 days for the investigated formulas as listed 3 and for standard concrete B300, compared to the range of CS for natural stone (see online version for colours)
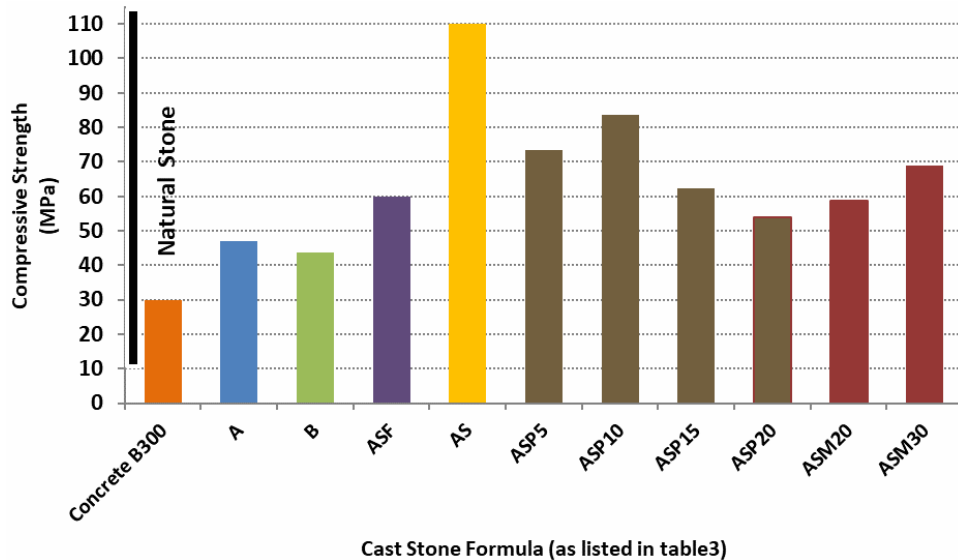


The effect of using marble cutting wastewater was explored first at 20% addition. The obtained CS for fully cured specimens at 28 days was 59 MPa. This is slightly larger than that for specimen $ASP_{20}$ sample (having 54 MPa CS). Marble particles are much stronger and harder than limestone particles. Increasing the ratio of marble cutting wastewater to 30% increased the CS to a value of 68 MPa. This behaviour of increasing CS with percentage additions at high values 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 and harder than lime stone particles.

## 3.2   *Reducing WA of the artificial stone*

Figure 7 compares percentage WA of the produced artificial stone for modified formulas ($ASP_5$, $ASP_{10}$, $ASP_{15}$ and $ASP_{20}$, as listed in Table 3) with that for other samples. The measurements were made after 28 days curing according to ASTM C1585 standard. Also shown is the reported range of percentage WA of natural stones (black bar). WA is dependent on the porosity, which is governed by the amount of water in the mix design. Increasing water content increases the size and the number of capillary pores and hence increases WA. The addition of controlled amount of the fine particles of calcium carbonate (e.g., 5 and 10%) reduced the porosity of the artificial stone by filling effect and possibly by better cement hydration. Thus, Figure 7 shows that specimens ASP5 and ASP10 have lower percentage WA than ASF (see Table 3). Clearly, the WA for all the investigated modified formulas is at the lower limit of that of natural stone. This indicates that these formulas have an additional competeveness factor over natural stone (in

addition to strength). Slightly larger WA was obtained for artificial stone produced with wastewater containing a polymeric flocculating agents.

**Figure 7** WA of cast stone samples for formulas (see online version for colours)
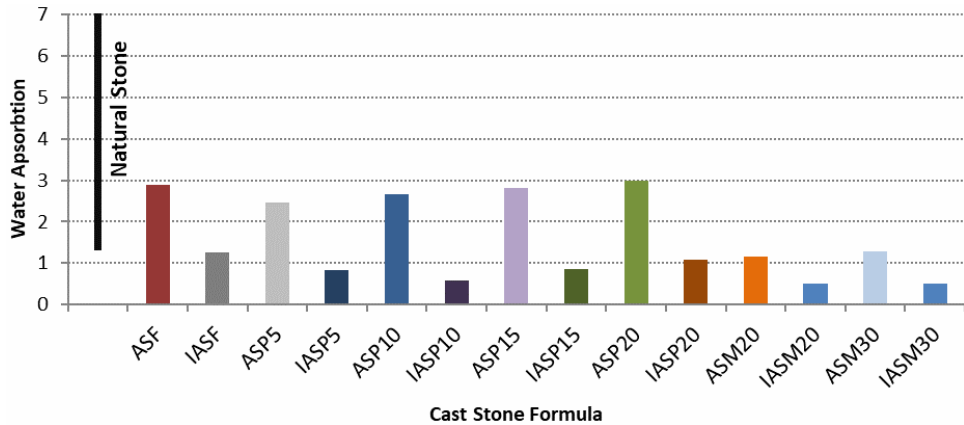


Notes: Samples $ASP_5$, $ASP_{10}$, $ASP_{15}$ and $ASP_{20}$, $ASM_{20}$ and $ASM_{30}$as listed in Table 3.

WA of the cast stone was reduced considerably when its surface was treated by spraying the produced artificial stone with sodium silicate ($SiO_2Na_2O$) solution. Such a surface treatment resulted in less than 1% WA 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 tight and thus less water is allowed to penetrate within the stone.

## 4    Conclusions

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

In comparison to artificial stone produced using limestone dust, 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, yields an artificial stone with 84 MPa, a value that is much higher than the industrially used production formula. WA values are at the lower levels of those for natural stone. The surface treatment of the cast stone with sodium silicate decreases WA.

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