A hydrophobic release agent containing SiO_2-CH_3 submicron-sized particles for waterproofing mortar structures.

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Abstract

Hydrophobic release agents were developed from SiO_2-CH_3 submicron-sized particles containing hydrophilic and hydrophobic functional groups dispersed into a vegetable oil as support. The SiO_2-CH_3 submicron-sized particles were synthesized by changing the molar ratio between the precursors Tetraethyl orthosilicate (TEOS) and Methyltriethoxysilane (MTES) from 0.66 to 5 (MTEOS-0.66 to MTEOS-5), being the relative amount of SiO_2-CH_3 quantified by FTIR technique. Mortar specimens having hydrophobic properties were manufactured using 3 wt.% of the above SiO_2-CH_3 submicron-sized particles. Additionally, the concentration effect was studied by using MTEOS-0.66 within 3-10wt.%. The hydrophobic properties of the mortar specimens were checked by measuring the contact angle within water droplets and surface. The particle size increased with the MTES/TEOS molar ratio and according to the hydrophobic properties, the proper release agent must be synthesized by using MTEOS-2.5 dispersed into the vegetable oil and having a concentration of 3wt.%. This hydrophobic release agent led mortar surfaces with contact angles higher than 145º. Waterproofing and mechanical studies on concrete specimens allowed to conclude that this demoulding agent do not have a high

penetration depth but improved the concrete waterproofing properties, without depletion in the compressive strength.

Keywords: Hydrophobic submicron-sized particles, Stöber process, release agent, mortar/concrete waterproofing

1. Introduction

The benefits and applications of nanomaterial technologies have been risen in recent years. They are exhibiting a large potential either in advanced applications such as biomedical, catalytic, separation, chemical sensing, microfabrication or in high-technology industries like electronics, sports equipment, aerospace, among others [1, 2]. Currently, the application of nanoparticles for improving the concrete waterproofing is gathering the interest of the construction industry in permanent seeking of concrete releasing formulations with water protection ability [3].

It is well known that water is one of the most aggressive external atmospheric agent that penetrates into mortar structure causing an important physical and chemical degradation that compromises its functionality [4-7]. In this way, materials such as superplasticizers [8], hydrophobic organic-inorganic materials and membranes based on EVA-copolymer [9] have been used to increase the mortar waterproofness and to enhance the infrastructure service life [10, 11]. These materials have been applied by different procedures: impregnation, immersion, additives and release agents. Among them, hydrophobic impregnation has demonstrated to be easily implemented because it is applied once the concrete structure is dried, favouring the depth penetration of the treatment [12]. The waterproofing ability of the surfaces is characterized by the water droplet contact angle technique, which is visually quantifiable by measuring the angle that forms a water drop with the studied surface [10]. The greatest contact angle has been reported for expensive monodispersed fluorinated silica nanoparticles into a

fluorine resin (165º) [13], being higher than those reached using SiO_2/polymethylhydrosiloxane synthesized by sol-gel (120º) [14] or Nano-SiO_2 within organic films (90º) [8].

Flores et al. [15] reported the achievement of a super hydrophobic concrete by immersion technique, having a contact angle of 156º when emulsions of hydrophobic polymethyl-hydrogen siloxane oil containing polyvinyl alcohol fibres combined with metakolin or silica fume (5%) were used. Isobutyltriethoxisilane and silica fume have been used as additive in the mortar mixture exhibiting large protection of the mortar materials [16]. This methodology allowed to reach mortars having lower water absorption due to the generation of more hydrated product and refined pore structure. This behaviour was attributed to the pozzolanic reaction that forms calcium silicate hydrate (C-S-H) and the covalent linkage or crosslinking between the hydroxyl groups of silicon and concrete, promoted by the $Ca(OH)_2$ presents in the cement [17, 18]. Hence, the interaction between the substrate surface and the hydrophobic agent determines the adhesion degree. It was also reported that nanosilicons added to silane or siloxanes compounds enhance the hydrophobic properties of bricks, tiles and concrete surfaces [19].

Finally, release agents as coatings having hydrophobic properties have been applied on the formwork surface used for producing the concrete. They are mainly petroleum-based form, like Sika Tite-BE, which consists of a water based bitumen emulsion modified with acrylic latex polymers [20], and it has been used to ensure a waterproofing protection and an easy demoulding of the concrete in only one step [21]. Nevertheless, this material presents the disadvantage of no remaining bonded to the mortar structure during long-time, being removed by rainwater.

Hence, taking into account that for obtaining the hydrophobic mortar, coatings require two steps, the immersion involves the use of a large amount of additive; mould release agents could attribute the waterproofing functionality in only one-step by using a thin-layer of product. In this way, a proper release agent seems to be the most feasible and cost-effective alternative for conferring hydrophobicity/waterproofing to concrete and mortar structures [22]. For this

purpose, the use and development of hydrophobic release agents for concrete and mortar specimens are being considered as a big challenge in the building industry. Consequently, these materials in combination with particles having hydrophobic and hydrophilic character could perfectly work in two ways: either as demoulding agent or as link with the mortar surface. It is important to point out that mould release agent's market size was at USD 1.1 billion in 2014 and is projected to reach USD 1.5 Billion by 2020, at a CAGR (Compound Annual Growth Rate) of 6.1% between 2015 and 2020 [23].

Taking in consideration the previous information, the addition of hydrophobic particles into a demoulding formulation to create a new releasing product with long time waterproofing effectivity is worthy to be studied. Thus, it has been confirmed that there are not reports in the technical literature about the influence of the hydrophobic/hydrophilic character of silica particles on the properties of hydrophobic release agents, being possible to move forward in the development of this kind of products. For this reason, in this paper, the surface modification of silica particles with Methyltriethoxysilane (MTES) and Tetraethylorthosilicate (TEOS) by the Stöber process was studied [24, 25]. The modified silica particles were incorporated into mortar demoulding formulations by using an eco-friendly vegetable oil, to attribute hydrophobicity and improve the ability to unmould the mortar specimens. In this way, different types of silicon particles were synthesized by changing the MTES/TEOS molar ratio from 0.66 to 5, and further, these particles were mixed with a vegetable oil using different concentrations from 3 to 10wt.%. The proper release agent formulation for leading hydrophobic mortar surfaces containing silicon particles from MTES and TEOS was found by chosen the best particles attending to their physical and chemical properties, and measuring the interfacial tension, the contact angle, the dispersability of the particles into the vegetable oil and the distribution of the particles on the mortar specimens. Finally, the performance of this release agent was checked in concrete specimens, analysing the penetration depth, the water absorption capacity, the resistance to alkali and the compressive strength.

2. Materials and method.

2.1. Materials

The materials used for the synthesis of hydrophobic silica particles were Tetraethyl orthosilicate (TEOS) reagent grade 98%, triethoxymethylsilane (MTES) 99% and Ammonium Hydroxide (NH_4OH) 33% which were obtained from Sigma Aldrich. Ethanol (EtOH) 96 % was purchased from Panreac (Spain) and water used was purified by distillation followed by deionization using ion-exchange resins.

The vegetable oil was commercially available and it presents the following properties shown in Table 1.

Table 1. Properties of the vegetable oil used.

Assay	Method	Results	Units
Density at 15 ºC	ISO 3675	905.45	kg/m^3
Viscosity at 40 ºC	ISO 3104	11.8273	mm^2/s
Viscosity a 100 ºC		4.3450	mm^2/s
Water content (Karl-Fischer)	ISO 12937	458	ppm
Oxidative stability	ISO 6886	4.98	h
Acid value	UNE EN 14104	2.87	mg KOH/g
Iodine number	UNE EN 14111	119	g I_2/100 g

Mortars were produced using Portland cement CEM II/B-L 32,5 N (Portland Valderrivas Cements, Spain), Standard sand CEN EN 196-1(Beckum, Germany) and tap water.

2.2. Preparation of Silicon particles.

Silicon particles were synthesized following the reported modified Stöber method [22, 26]. Firstly, TEOS and EtOH were added to a mixture of NH_4OH, H_2O and EtOH. This mixture is left under stirring at 30 ºC for 90 min to produce particles with only hydroxyl groups on its surface. The desired amount of MTES was then added to the above solution and maintained

under vigorous agitation during 19 h at 60 ºC. After this reaction period, the mother liquor is removed by centrifugation and finally, the product was dried at 110ºC. Synthesized silicon particles were called as MTEOS-x, where x is the MTES/TEOS molar ratio.

2.3. Preparation of hydrophobic release agent and mortar specimens.

Different masses of prepared silicon particles were added to the vegetable oil from 3 to 10 wt.% in order to produce the hydrophobic release agents. A homogeneous and stable dispersion of the particles in the oil was achieved by heating the mixture at under a vigorous agitation of 1400 rpm during 15 min, followed by sonication for 15 min at room temperature. Mortars were synthesized according to UNE-EN 12504-1:2009. First, the known masses of Portland cement, tap water and Standard sand CEN EN 196-1 (ISO Standard Sand) were weighted using a mass ratio of 1:0.5:3, respectively. Then, they were mixed during 5 min under vigorous agitation. Finally, the mixture was poured into a mould of 3x6x10 cm, that was previously impregnated with the corresponding release agent (Fig. 1).

Fig. 1. Mortar specimens production according to UNE-EN 12504-1:2009

2.4. Sample characterization

2.4.1 SEM Analysis.

The silicon particles morphology and its distribution into the base form release agent were obtained by means of Scanning Electron Microscopy (SEM) by using a FEI QUANTA 200.

2.4.2 Thermal Degradation of Silicon Particles.

Thermogravimetric analyses (TGAs) of hydrophobic particles were performed by using a TA Instrument equipment model SDT Q600. The used conditions for each analysis were a heating rate of 10ºC/min from room temperature to 700ºC under nitrogen atmosphere.

2.4.3 Infrared spectroscopy.

The chemical structure of each synthesis was confirmed by Fourier transform infrared spectrometer. Infrared spectra was obtained with a spectrophotometer Varian 640-IR type FT-IR in the range 4000 to 600cm^{-1}, 8 cm^{-1}.

2.4.4 Contact angle and interfacial tension.

The contact angle analyses were performed on mortars of 3x6x10 cm^3, each mortar was divided into three equal quadrants and three equidistant points in each area were taken. Therefore, the contact angle value as function of the water drop age is the average result from 9 different points. Interfacial tension between different synthesized release agents and water was measured twice. In both cases, an Attension Theta Optical Tensiometer from Biolin Scientific's, with a computer controlled by OneAttension software and provided with a high definition camera was used.

2.4.5 Particle size and Particle Size Distribution.

Particles size and Particle size distribution for MTEOS 0.66-2.5 where achieved by Z-Sizer Nano ZS, that uses the technique of microelectrophoresis of laser Doppler. MTEOS-5 where obtained employing Malvern Mastersizer 2000 equipped with a Scirocco 2000 and using a Low Angel Laser Light Scattering (LALLS) utilizing software based on the Mie theory to analyse the experimental data.

2.4.6 Waterproofing and Mechanical Testing of Concrete Specimens

The penetration depth, water absorption, resistance to alkali and mechanical strength for non-structurally reinforced concrete class C45 (water/cement ratio 0.45) prepared by using the best release agent have been studied.

2.4.6.1 Penetration depth. It was tested following requirements of EN 1504-2 and with test procedure described in EN 14630. The Specimens were fractured perpendicular to the treated surface and the then sprayed with water.

*2.4.6.2 Water absorption and resistance to alkali. T*hey were tested according to EN 13580. The absorption coefficient (AR) was measured after absorption in deionized water for 1 or 24 hour in case of untreated or treated specimens, respectively. The alkali absorption coefficient (AR_{alk}) was determined in three steps. Firstly, the specimens were exposure to a KOH solution of 5.6 g/L during 21 days, followed by conditioning the specimens into a controlled environment at 21 ºC and having a relative humidity (RH) of 60%. Finally, the specimens were placed in contact with deionized water for further 24 hours.

The above analyses were performed at *RISE CBI Swedish Cement and Concrete Research Institute* by using cubic specimens of 100x100x100 mm.

2.4.6.3 Compressive strength: Concrete specimens prepared with the selected release agent were tested according to the UNE-EN 12390-3 using cylinder samples having a size (diameter per height) of 150x300 mm at two curing times (14 and 28 days).

3. Results and discussion

3.1.Particles characterization.

Fig. 2 shows SEM photographs of the different synthesized particles. Fig. 2a) corresponds with silicon particles without superficial modification. It is observed that this product is formed by single spherical particles having a size ranged between 400 and 600 nm. It can be seen that the increase of methyl groups up to MTEOS-2.5 does not change the morphology and the size of the particles, finding particles in a wide scale within 90-600 nm. It is also important to point

out that silicon particles synthesized with the highest MTEOS molar ratio (MTEOS-5) presented an amorphous structure in which the single particles are not the major class.

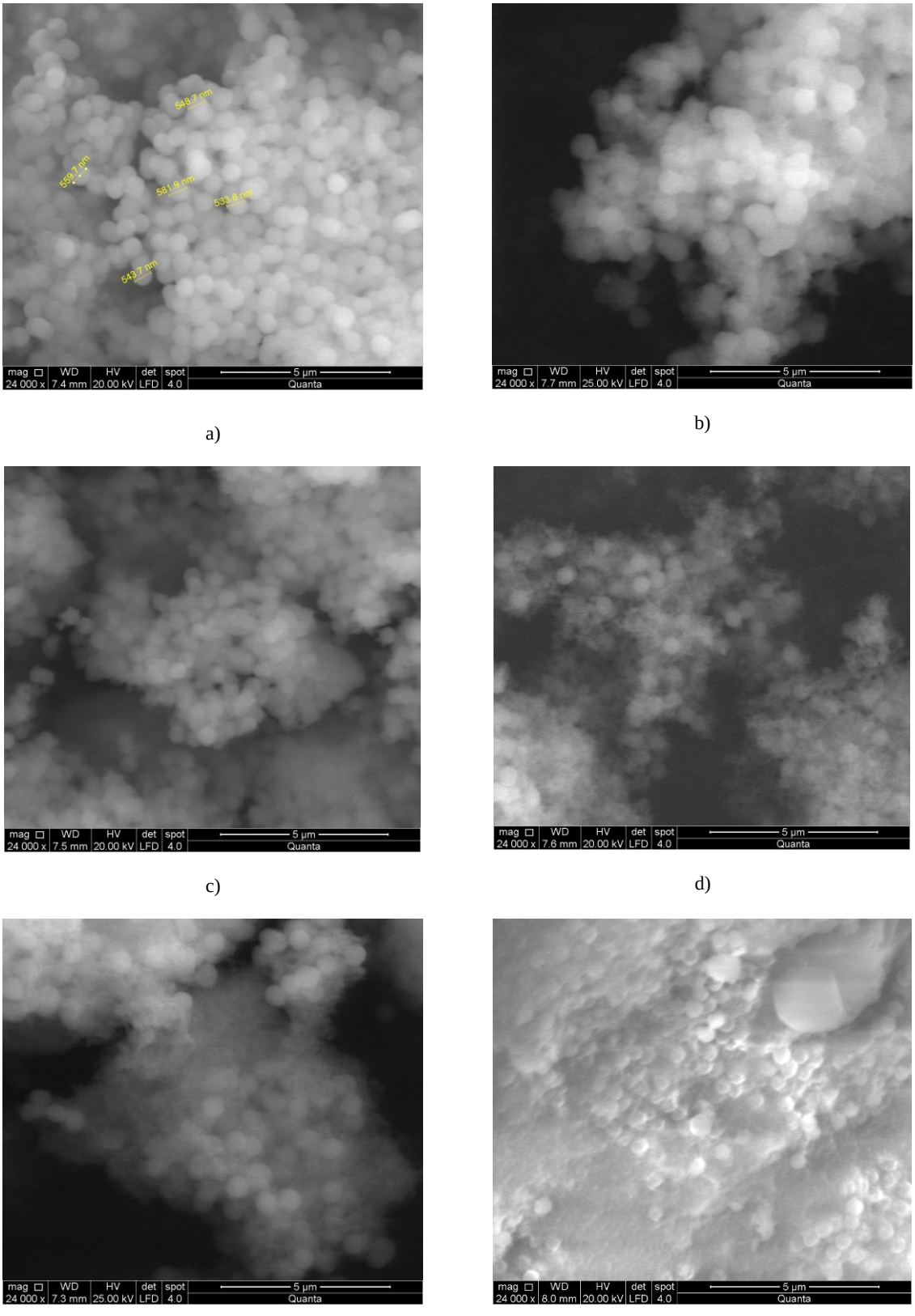
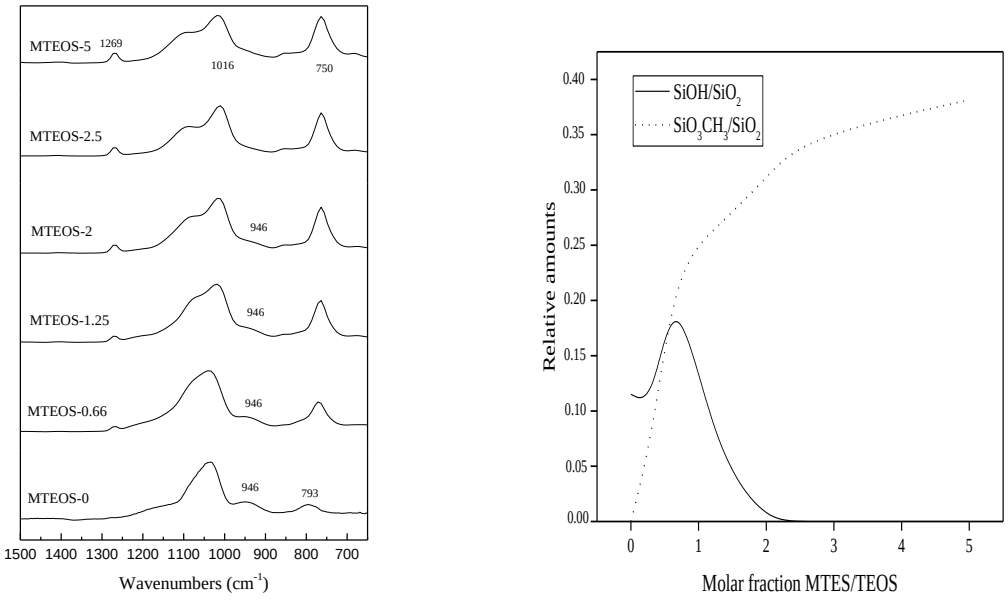


Fig. 2. SEM images of silicon particles varying MTES/TEOS molar ratio. a) 0, b) 0.66, c) 1.25, d) 2, e) 2.5, f) 5.

According to the particle size of these materials mainly within 100 nm-1.0 μm, they can be classified as submicron-sized particles instead of nanoparticles which is the name often used for particles having a size less than 100 nm [27].

The success in the surface attachment of alkyl groups can be confirmed by infrared spectroscopy. Characteristic peaks of Si-O_3CH_3 are the signals at 1250, 840 and 750 nm. At 950 nm it can be seen Si-OH groups, and at 1050 and 800 nm are the vibrations of the Si-O-Si can be identified [28]. The relative amounts of Si-OH and Si-O_3CH_3 have been obtained by deconvolution of each spectrum at least in four individual peaks as shown Fig.3b. As expected, the higher the MTES/TEOS molar ratio the higher the intensity of the methyl groups in the spectra. This increase in methyl groups leads to a depletion in the Si-OH group signal, confirming that methyl groups have perfectly replaced the Si-OH ones.



a) b)

Fig. 3. a) FTIR Spectra of submicron-sized particles b) Deconvolution and relative amounts between SiOH/SiO_2 and SiO_3CH_3/SiO_2 groups.

Thus, attending to the above results, the different properties of the synthesized submicron-sized particles will allow to check the influence of each synthesis on the mortar specimens surface,

trying to stablish a relationship between the amount of methyl groups on the surface and their influence on the surface contact angle.

TGA analyses allowed to know the modification in the surface of submicron-sized particles according to the weight loss promoted by the release of hydroxyl and alkyl groups as temperature function. It was also possible to quantify the water content and the thermal stability of the particles, mainly as the total residue.

Fig. 4 shows the TGA for each product synthesized by changing MTES/TEOS molar ratio from 0.66 to 5. Different regions of weight loss can be seen in all synthesized materials except in the particles without superficial modification, which only presents silanol groups (Si-OH) on the surface. In that case, the observed weight of loss at low temperature (100 ºC) is attributed to the evaporation of the adsorbed water (I). The weight loss within 200 and 400 ºC is due to the condensation of silanol groups (II), the degradation of methyl groups occurs at 500 °C (III) and the final one (IV), corresponding with the inorganic silica degradation occurs at temperatures higher than 600 ºC [29]. The presence of methyl groups is clearly observed in the products MTEOS-2.5 and MTEOS-5. Besides, attending to the thermal stability analyses it is possible to affirm that the most stable material is MTEOS-5 and this stability is enhanced by the large amount of silanol groups substituted by the alkyl ones.

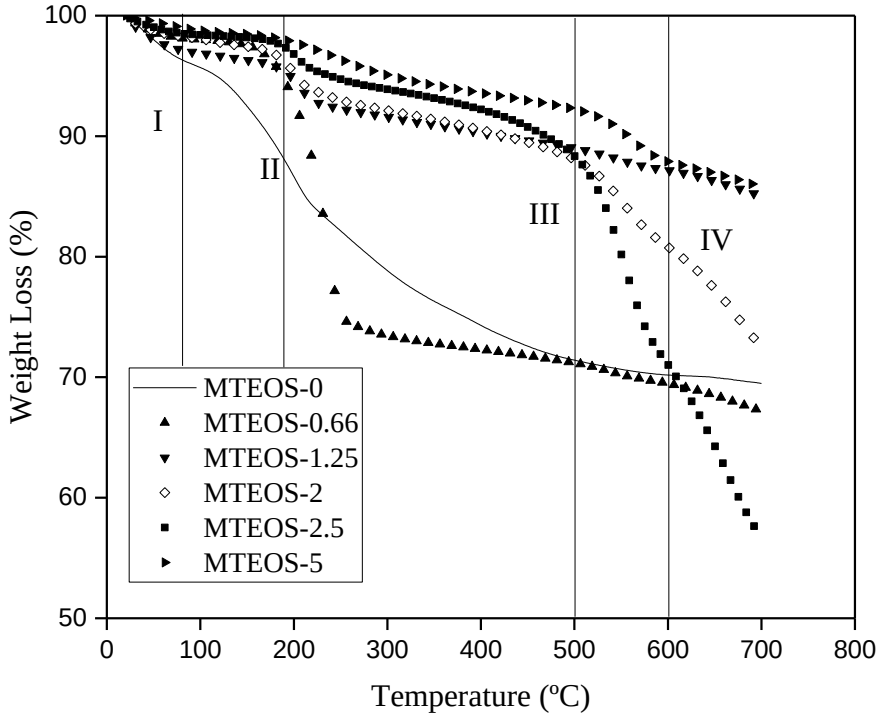


Fig. 4. TGA analyses for the synthesised submicron-sized particles.

3.2. Application on mortar specimen.

3.2.1 Particle Size distribution in the release agent.

The submicron-sized particles into vegetable oil after sonication were analysed by Z-Sizer Nano ZS for the release agents containing MTEOS within 0.66-2.5, whereas the particle size of the release agent formed with MTEOS-5 was analysed by using the Mastersizer technique, separating the solid particles from the oil after the sonication. Measurements in Z-Sizer must be lower than 10 microns for obtaining a stable measurement.

The particle size distributions of demoulding agents with different amounts of particles are shown in the Fig. 5.

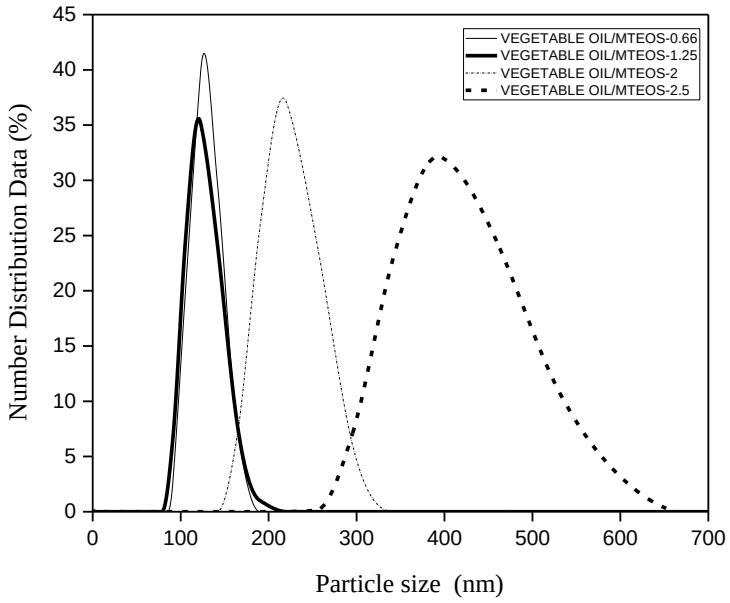


Fig. 5. Particle size distribution of silicon particles into the release agent containing 3 wt.% of MTEOS.

It is observed that all materials present a unimodal distribution although greatly differ in the particle size. According to this figure, the MTEOS-2.5 presents the higher particle size and a width distribution ranged within 250-650 nm, whereas the other materials have narrow particles size distributions within 150-350 nm for MTEOS-2, and ranged between 80 to 200 nm for MTEOS-0.66 and MTEOS-1.5. These results are quite similar to those measured using the SEM photographs. The size of these materials are in the range within 90 to 600 nm depending on the type, indicating that the sonication does not change the morphology of the particles and also that the particle size increases with the MTES/TEOS molar ratio. Regarding the MTEOS-5, they were poorly dispersed into the vegetable oil and the material was analysed as a dried powder once it was separated from the oil. The particle size was in the micrometre scale range, with $dn_{0.5}$ (50%) and $dn_{0.9}$ (90%) having values of 606.26 and 1003.58 µm, respectively, confirming the previous observation in which this product is not constituted by single particles. To know the dispersability of submicron-sized particles into the vegetable oil, SEM images of each release agent after sonication are shown in Fig. 6.

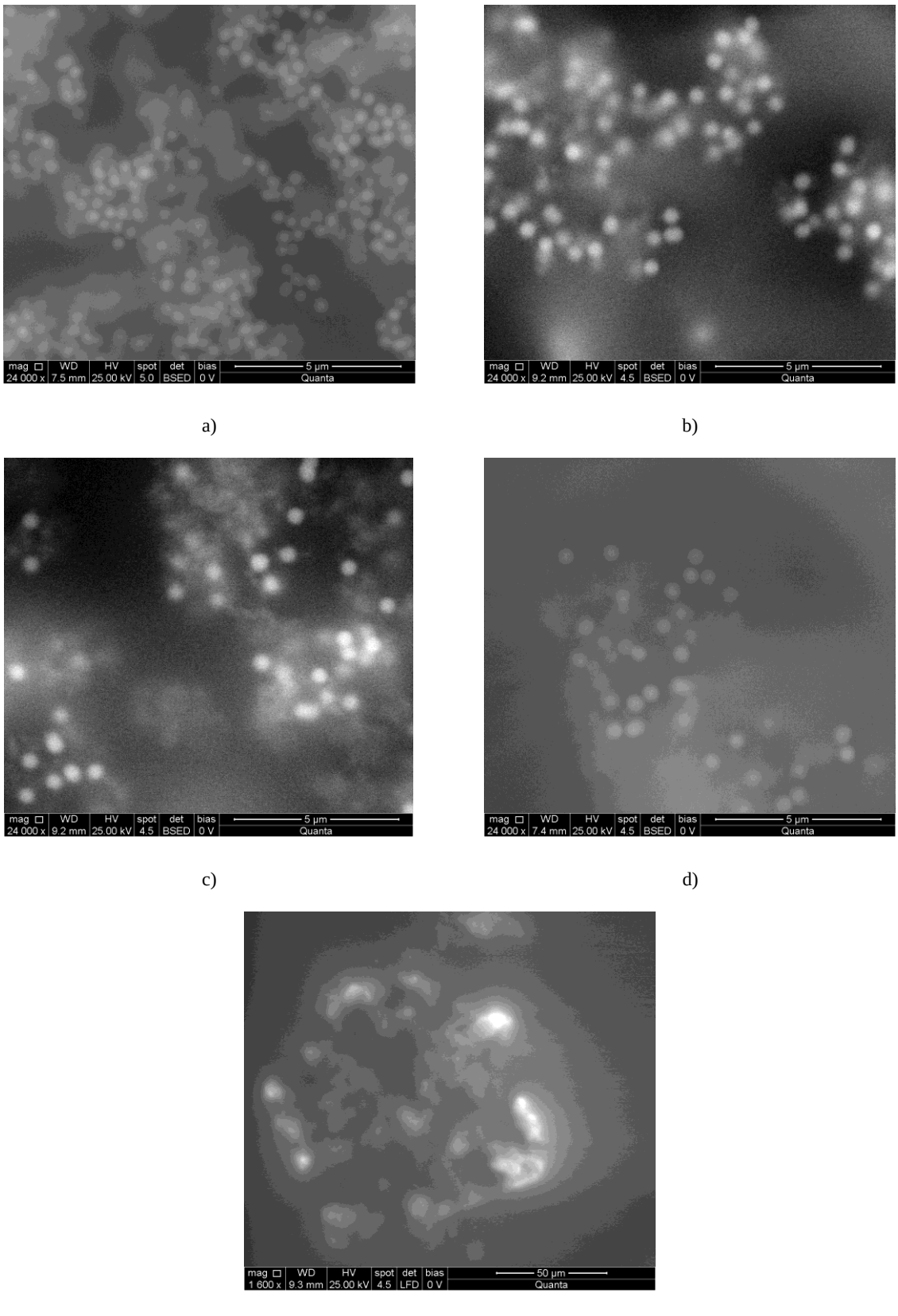


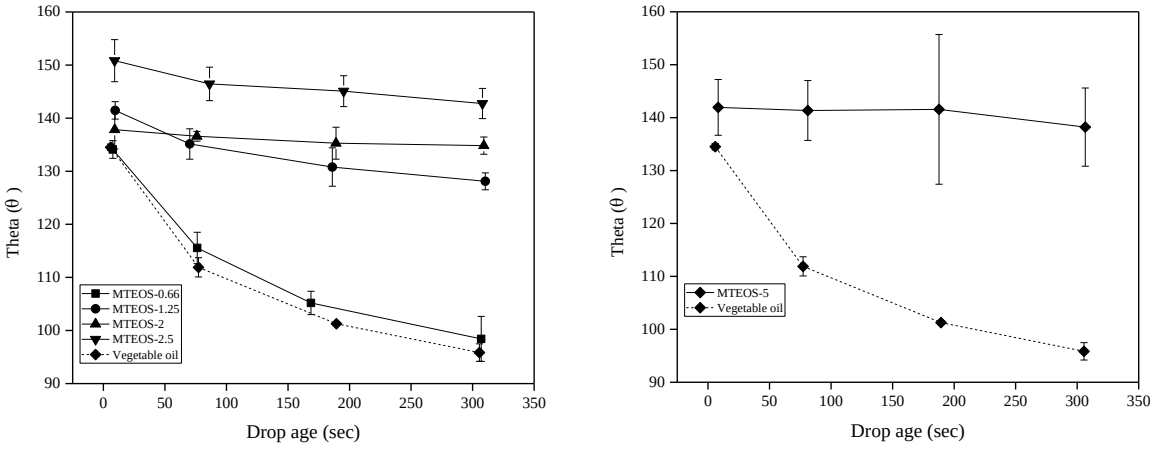
Fig. 6. SEM images of MTEOS into the vegetable oil. a) 0.66, b) 1.25, c) 2, d) 2.5, e) 5.

It can be observed that these particles have spherical shape and are uniform and perfectly dispersed into the base form release agent except when the MTEOS-5 is used. This result

indicates that this material is strongly agglomerated and that the sonication is not effective for their disaggregation. The particle size observed by SEM for the release agent containing MTEOS-0.66, MTEOS-1.25, MTEOS-2 and MTEOS-2.5 is quite similar to those previously showed.

3.2.2 Demoulded behaviour

Fig. 7 shows the evolution of the contact angle with time in unmould mortars using different synthesized release agents, being considered as reference, the specimen prepared using only the vegetable oil.



a) b)

Fig. 7. Evolution of the contact angle with time using release agents without and containing submicron-sized particles having different MTES/TEOS molar ratio. a) Up to 2.5 b) Equal to 5.0

It can be seen how the addition of submicron-sized particles clearly increases the contact angle of the mortar surface when they are included in the formulation of the release agent. In Fig. 7a) a small deviation of the contact angle measurements is observed, indicating a proper distribution of the particles onto the mortar surface. These results are the expected ones according to the Z-Sizer Nano ZS analyses, where SiO_2-CH_3 submicron-sized particles are perfectly dispersed into the vegetable oil. On contrary Fig. 7b) shows a highest deviation (±14) in the contact angle when the MTEOS-5 is used in the formulation of the demoulding agent, confirming the worse distribution of these particles on the mortar surface. Hence, when this material is used, its rate of precipitation in the release agent is faster than the possible formation of the calcium silicate

hydrate (C-S-H) or the covalent linkage or crosslinking with hydroxyl groups of cement due to its low content of silanol groups [17, 18]. The release agent containing MTEOS-2.5 exhibits the maximum hydrophobic power and as expected, the submicron-sized particles with the minimum content of methyl groups MTEOS-0.66 led the worse hydrophobic power. It is also observed that this hydrophobic power is maintained for longer time, being the decrease caused by water evaporation, when the release agent containing the MTEOS-2.5 was used.

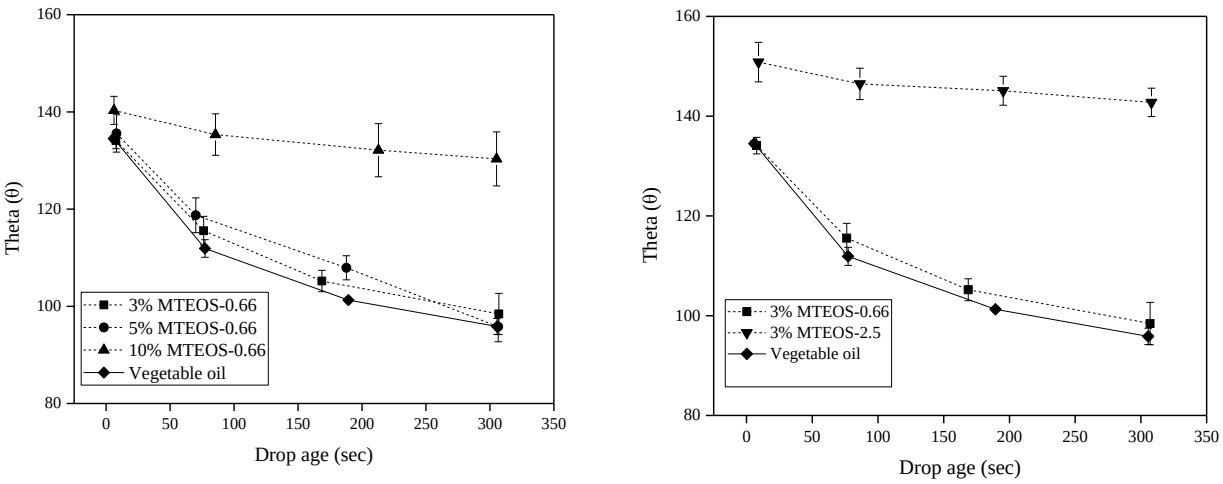
The hydrophobic behaviour of the specimens with and without submicron-sized particles (MTEOS-2.5) after 7 months of curing time is shown in Fig.8. It is observed that the waterproofing ability of the demoulding mixture continues active even after 7 months. This is, while in the specimen without hydrophobic particles the water penetrates immediately, the water drops on to the surface of the silicon treated specimens remain on the mortar surface up to its full evaporation. This result indicates that the MTEOS-2.5 is retained on the mortar surface whereas the vegetable oil disappears by diffusion into the solid phase or vaporization. Thus, it is possible that these submicron-sized particles must be stabilized on the mortar surface by either the pozzolanic reaction or covalent linkage between the hydroxyl groups of silanol and cement [17, 18]. Attending to these results, the improvement of the hydrophobic ability by using the proper release agent could be very important to avoid corrosion problems in mortar infrastructure since the particles are maintained linked to the mortar surface.

Fig. 8. Hydrophobic effect of mortar specimens after 7 months of curing time by using release agents with MTEOS 2.5 (left) and without silicon particles (right).

3.3.Effect of submicron-sized particles concentration in hydrophobic release agent.

Once demonstrated the effectivity of this form release mixture in creating a stable waterproofing layer on mortar specimens, the influence of the amount of submicron-sized particles on the waterproofing ability of the coating was studied. Fig. 9 shows the evolution of the contact angle of five mortar specimens using different types and percentages of submicron-sized particles. Three specimens were manufactured using the MTEOS-0.66 at concentrations of 3, 5 and 10 wt.%. The other two were produced without and containing 3wt.% of MTEOS-2.5 and considering the specimen free of silicon particles as the reference ones. Results showed that increasing the concentration of MTEOS-0.66 in the demoulding agent from 3 to 10 wt.%, the hydrophobicity of the mortar specimens was significantly increased. In addition, it was observed that this high concentration of MTEOS-0.66 was not enough to lead a similar hydrophobic capacity than that offered by MTEOS-2.5 with only 3 wt.%.

Thus, these results showed that the best release agent produced in this work must be constituted by MTEOS-2.5 dispersed into the vegetable oil at 3 wt.%.



a) b)

Fig. 9. Evolution of the contact angle with concentration and type of MTEOS, a) varying the MTEOS-0.66 concentration from 3 to 10wt.% and b) 3wt.% of MTEOS-0.66 and MTEOS-2.5.

The precision on the measurement of the contact angle also indicates that sonication is a good technique for achieving the dispersion of the SiO_2-CH_3 submicron-sized particles into the vegetable oil. Besides, this vegetable oil is a good media for leading the distribution of the particles around the whole surface of the mortar and it is not dependent on the particle concentration.

3.4.Influence of submicron-sized particles concentration on the interfacial tension.

In order to know the reason of the best behaviour as release agent of the submicron-sized particles from an MTEOS-2.5 and also, the maximum concentration in the vegetable oil, the interfacial tension between the different synthesized release agents and water was measured. Values of interfacial tension for each synthesized release agent and using submicron-sized particles concentration from 0 to 10 wt.% are shown in Fig.10. The addition of submicron-sized particles increased satisfactorily the interfacial tension of the release agent respect to that value of 21.23 mN/m exhibited by the original vegetable oil.

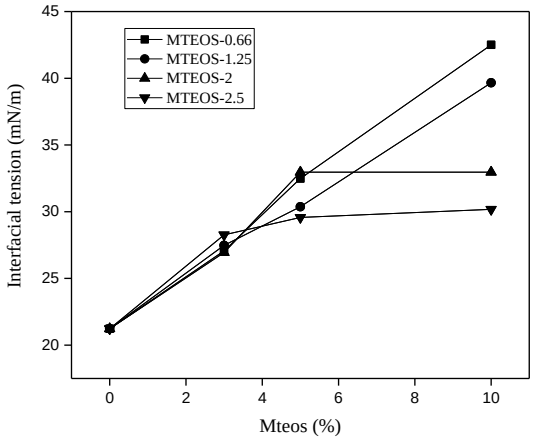


Fig. 10. Evolution of the interfacial tension of release agents versus submicron-sized particles concentration.

It was also observed that, for low MTES/TEOS molar ratios (0.66 and 1.25), the interfacial tension increases linearly, while for the MTEOS-2 and MTEOS-2.5, it tends to be practically constant for submicron-sized particles concentration higher than 5 wt.%. As it can be seen, for a concentration of 3 wt.% the synthesized MTEOS-2.5 reach an interfacial tension that explains why using this concentration the contact angle values are the highest when this material is used to form the release agent. The higher the interfacial tension, the larger the amount of water repelled by the release agent, reducing the linkage of the silanol groups with the hydroxyl groups of the mortar that takes place in water presence and alkaline media [17, 18].

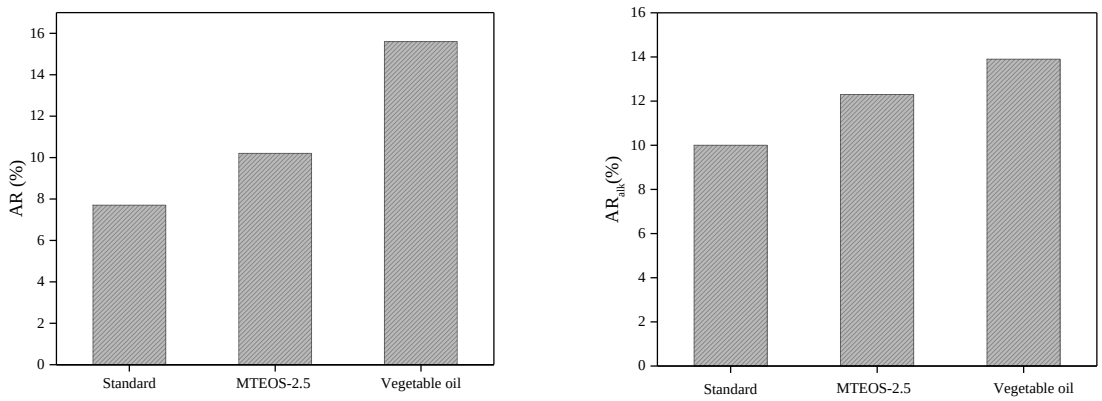
In that way, the demoulding agent manufactured from 3 wt.% of MTEOS-2.5 and the vegetable oil seems to be the best product for attributing hydrophobic characteristics to the mortar surface. These properties were strongly dependent on the relationship between hydrophobic/hydrophilic groups of the silicon particles, its dispersability into the support and dispersion on the mortar surface and on the interfacial tension of the whole release agent.

3.5.Waterproofing and Mechanical Testing of Concrete Specimens.

Once the best demoulding agent has been found, the penetration depth, the water absorption, the resistance to alkali and the compressive strength of concrete specimens manufactured by using this release agent has been studied.

No detectable penetration depths were obtained on the treated samples, being the concrete classified as a *Class I* according to EN1504, having a penetration depth lower than 10 mm. Hence, results suggest that sub-micron particles are mainly located on the concrete surface forming a new water protective film.

Fig. 11 shows water absorption and the resistance to alkali of concrete specimens prepared by using the demoulding agents (selected and vegetable oil) and compared with the standard values for waterproofing concrete materials.



a) b)

Fig. 11. Absorption studies on concrete specimens. a) Water absorption and b) Resistance to alkali.

As can be seen, by using this kind of demoulding agent containing 3 wt.% of MTEOS-2.5 in the vegetable oil, it is possible to reduce the water uptake from 16 to 10% which is closer to the standard value (8% reduction). An important improving in the resistance to alkali is also observed, reducing the AR_{alk} from 14% to 12%. According to these results, by using this demoulding agent is not possible to achieve standards for waterproofing concrete structures according to EN1504 threshold, but it could help to enlarge the concrete service life. Finally, Fig. 12 shows the compressive strength of the concrete specimens produced by using the above demoulding agent.

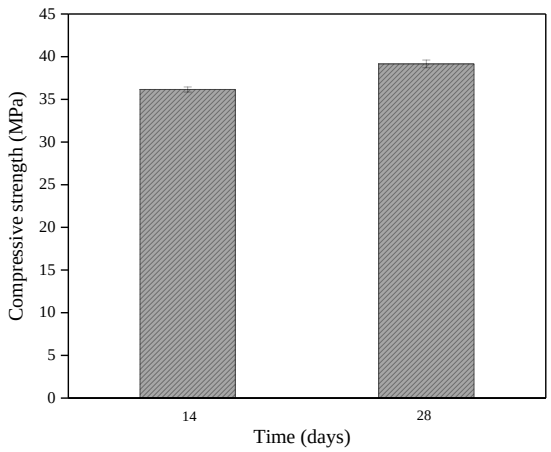


Fig. 12. Effect of the curing time on the compression strength of concrete specimens produced using an demoulding agent containing 3 wt.% of MTEOS-2.5 sub-micron particles in vegetable oil.

The values of the compressive strength indicates that this treatment do not affect the mechanical resistance of the concrete and these materials satisfy the requirement of 32 MPa stablished by a concrete class C45 even at curing time of 14 days. It indicates that this superficial treatment with a low penetration depth does not cause any depletion in the compressive strength.

4. Conclusions

Submicron-sized particles having hydrophilic and hydrophobic functional groups were obtained by modifying the molar ratio between the precursors methyltriethoxysilane and tetraethyl orthosilicate. Different hydrophobic release agents were developed dispersing by sonication these particles into the vegetable oil. The demoulding agent containing 3 wt.% of MTEOS-2.5 with a proper surface tension allowed to produce mortar specimens having contact angles higher than 145° with a good distribution on the mortar surface and creating surfaces more hydrophobic than those generated using 10 wt.% of MTEOS-0.66. The hydrophobic character of MTEOS-5 was not enough for leading a good result as demoulding product, either by the worse dispersion of this product in the vegetable oil or the low amount of silanol groups for promoting its linkage with the mortar. Attending to the contact angle of the mortar specimens, the hydrophobic/hydrophilic molar relationship of the particles and the particle size distribution, the demoulding agents containing submicron-sized particles MTEOS-2.5 can be

considered as a good chosen for being used as demoulding agent. Finally, concrete specimens prepared with this release agent exhibited an improved waterproofing respect to the demoulding oil and any depletion in the compressive strength.

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