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Use of nano materials in concrete construction

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Abstract. Portland cement is the major chemical product processed in the world, more than 3.6 Bton at 2011, causing important environmental impacts. Several nano-particles have been tested in cement based materials in order to improve their performance and durability leading to an eco-efficiency use for this binder. Published researches for nano-particles in cement based materi-als have been demonstrated optimum volumetric contents considering the relative strength gain, directly related with packing and nucleation effect observed for nano-particles, and a poz-zolanic reaction is observed for nano-SiO_2.

1. INTRODUCTION

Nanotechnology is an emerging field of science related to the understanding and control of matter at the nanoscale, i.e., at dimensions between approximately 1 and 100 nm. Nanoscale particles are not new in either nature or science. Recent developments in testing materials at the nanoscale have led to an explosion in nanotechnology-based materials in areas such as polymers, plastics, electronics, car manufacturing, and medicine. Concrete, the most ubiquitous material in the world, is a nanostructured, multiphase, composite material that ages over time (Sanchez and Sobolev, 2010). It is composed of an amorphous phase, nanometer- to micrometer-size crystals, and bound water. The properties ofconcrete exist in, and the degradation mechanisms occur across, multiple length scales (nano to micro to macro) where the properties of each scale derive from those of the next smaller scale

The mechanical behavior of concrete materials depends to a great extent on structural elements and phenomena that are effective on a micro- and nanoscale. The size of the calcium silicate hydrate (C-S-H) phase, the primary component responsible for strength and other properties in cementitious systems, lies in the few nanometers range (Taylor, 1997). The structure of C-S-H is much like clay, with thin layers of solids separated by gel pores filled with interlayer and adsorbed water (Mehta, 1986). This has significant impact on the performance of concrete because the structure is sensitive to moisture movement, at times resulting in shrinkage
and consequent cracking if accommodations in element sizes are not made (Jennings et al., 2007). Hence, nanotechnology may have the potential to engineer concrete with superior properties through the optimization of material behavior and performance needed to significantly improve mechanical performance, durability, and sustainability. The development of nanotechnology-based concrete materials requires a multidisciplinary approach, consisting of teams of concrete materials experts: civil engineers, chemists, physicists, and materials scientists. Porro et al. (2010) presented an overview of how nanotechnology could be applied to concrete technology, emphasizing the multidisciplinary approach needed for successful breakthroughs leading to ultra high-performance materials and new multiscale models that enable the prediction of bulk material properties from composition and processing parameters. Grove et al. (2010) identified opportunities for nanotechnology leading to new concrete products and materials, and also for improving the sustainability and reducing the environmental footprint of concrete-based materials in the future. Finally, Birgisson et al. (2010) identified the following key breakthroughs in concrete technology that are most likely to result from the use of nanotechnology. The particles volume contend, illustrated by Fig. 1b con-sidering compressive strength of cement based materials containing nano-SiO2 and nano-TiO2 . So in order to study the nano-particles in cement based materials, this paper objective is a review for differ-ent nano-particles effects for cement based materi-als, discussing their mechanical and durability prop-erties, microstructural and rheological aspects pub-lished in the last years.

2. OXIDE NANO-PARTICLES

Nano-SiO2 is the main studied oxide nano-particles for cement based materials that could be delivered in powdered or liquid suspensions. The nano-SiO2 real density, about 2.25-2.54 g/cm3 [3,9], leads to the cheapest cost x volume relationship for the ox-ide nano-particles and the pozzolanic reaction di-rect related with its high surface area, between 50 to 750 m2/g [3,9] depending on the nano-particles size distribution, are two important issues for ce-ment based materials design, those could explain this bigger effort for nano-SiO2 researches.

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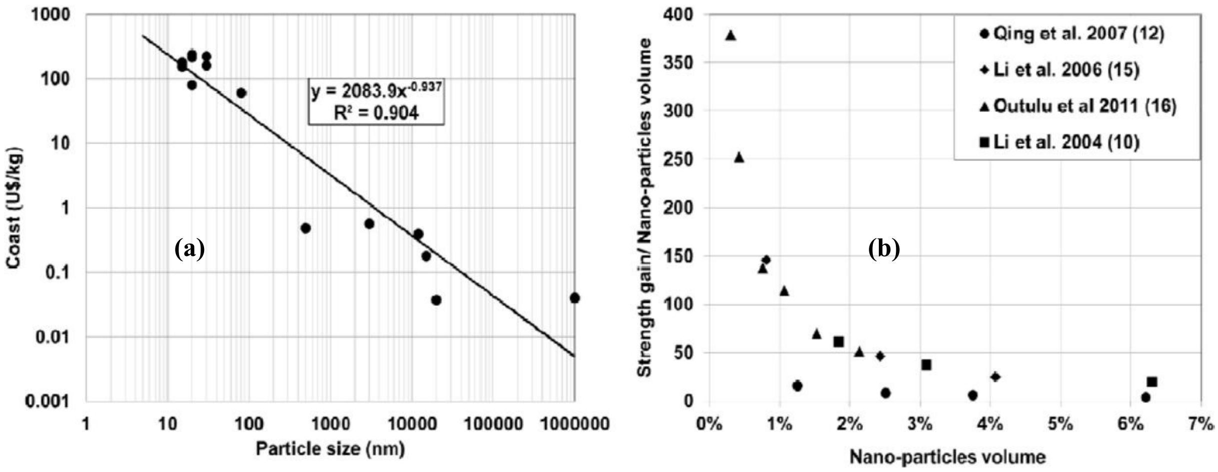

Fig. 1. (a) Raw-material coast of cement based materials [2-8]; (b) Nano-particles Efficiency [10,12,15,16].

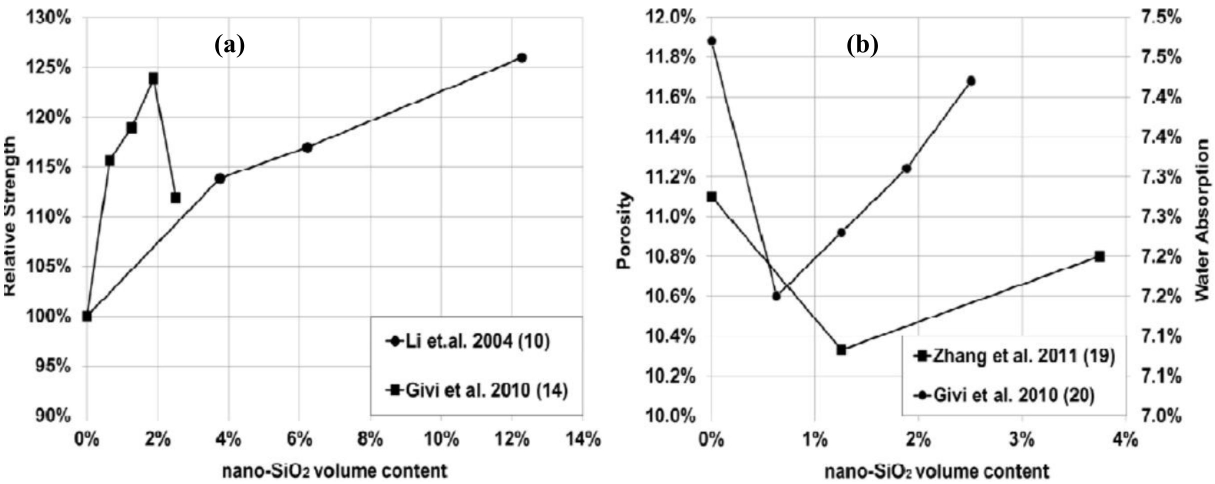

Fig. 2. (a) Nano-SiO_2 volume content x relative compressive strength [10,14]; (b) Nano-SiO_2 packing effect [19,20].

Nano-SiO_2 particles present a direct effect in cement based materials properties and durability, for large nano-SiO_2 volume content Li et al. (2004) [10]has demonstrated that the compressive strength increases, according with published results of Jo et al. (2007) [11], Qing et al. (2007) [12] and Litfi et al.

(2011) [13], Fig. 2a. Although, for low nano-SiO_2 volume content, Givi et al. (2011) [14], Li et al. (2006)

These properties and durability enhancement caused by the nano-SiO2 could be related with two important microstructural aspects of cement based materials: the packing effect and pozzolanic reac-tion of nano-SiO2 particles. Fig. 2b shows Zhang et al. (2011) [19] results of porosity determined by Mercury Intrusion Porosimetry (MIP) for concretes with 1 and 3 wt.% of nano-SiO2, similar results pub-lished by Givi et al. (2010) [20] considering the wa-ter absorption for cement based materials

with 0.5, 1, 1.5, and 2 wt.% of nano-SiO2 with two different nano-particles diameters 15 and 80 nm. These re-sults indicate a characteristic curve for packing ef-fect of nano-SiO2 in cement based materials con-sidering nano-SiO2 volume content and particle size.

Pozzolanic reaction has been observed by Senff et al. (2010) [21] using thermogravimetric analysis (TGA) Fig. 3a considering the higher mass loss peak for C-S-H hydrated products and the peak reduc-tion of Ca(OH)2, Fig. 3b, due the consumption in

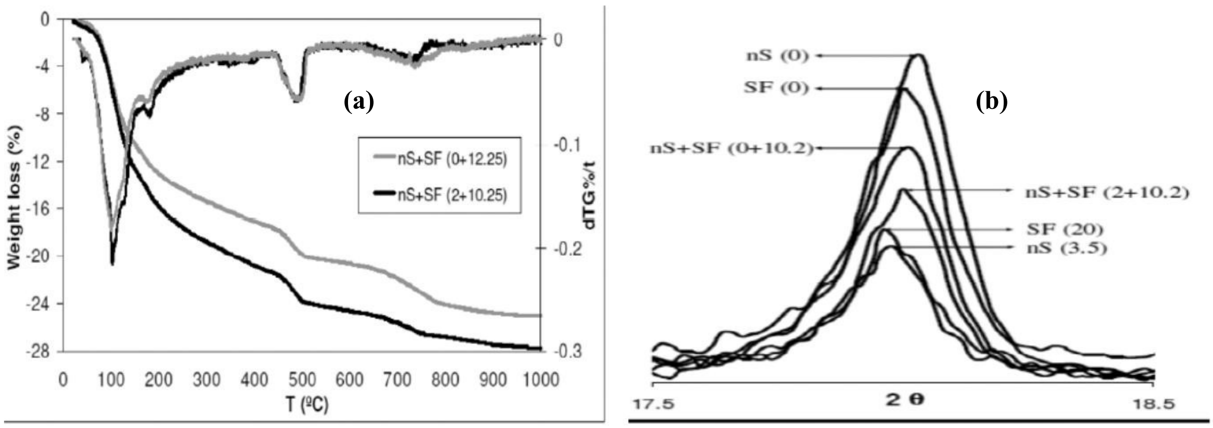

Fig. 3. (a) Thermogravimetric analysis results for cement pastes containing nano-SiO_2, (b) DRX of $Ca(OH)_2$ peaks, reprinted with permission from L. Senff,D. Hotza, W.L. Repette, V. Ferreira and J.A. Labrincha // *Advances in Applied Ceramics* **109** (2010) 104, (c) 2010 Maney.

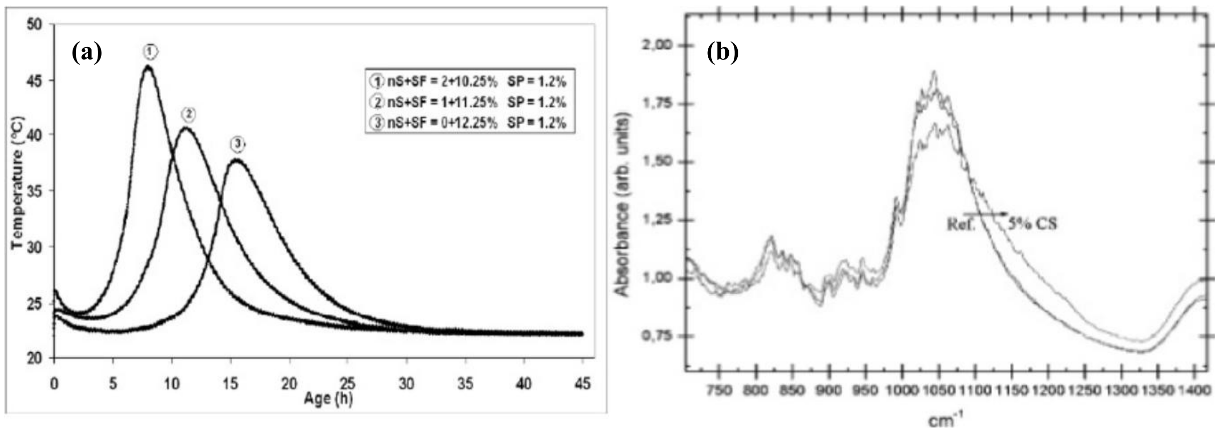

Fig. 4. Adiabatic Calorimetry of cement pastes with 0, 1, and 2% of nano-SiO_2, reprinted with permission from L. Senff, D. Hotza, W.L. Repette, V. Ferreira and J.A. Labrincha // Advances in Applied Ceramics **109** (2010) 104, (c) 2010 Maney.

pozzolanic reaction with nano-SiO_2. The nano-SiO_2 plays an important role in reaction kinetics acceler-ating the cement hydrated products formation; Fig. 4 shows this effect for cement pastes with 0, 1, and 2 wt.% of nano-SiO_2 measured by Senff et al. (2009)
[22] using adiabatic calorimetry. Similar results has been published by Bjornstrom et al. (2004) [23] con-sidering the tobermorite (C-S-H) characteristic ab-
sorption band (1200 cm^{-1}), measured by Infrared Spectroscopy (FTIR) for cement pastes after 12
hours containing 0, 1 and 5% of nano-SiO_2.

The cement based materials processing is di-rectly affected by nano-SiO_2, reducing the concrete bleeding for self-compact concretes as demon-strated by Collepardi et al. (2002) [18], but increas-ing suspensions yield stress and viscosity leading to a setting time (Senff, 2009) [24] and slump re-duction Li et al. (2006) [15], two important rheologi-cal properties for casting. Drying shrinkage results published by Collepardi et al. (2002) [18] for self-compact concrete containing 0, 1, and 2 wt.% indi-cates that self-compact concretes containing nano-SiO_2 has the same order of magnitude of drying shrinkage of self-concretes without nano-SiO_2.

Nano-TiO_2 particles is second nano-oxide par-ticles most used for cement based materials, rutile photo-catalysis effect is an important auto-clean feature needed for nano-engineered building materi-als. Nano-TiO_2 high density about 3.9 g/cm^3 and consequently its higher hardness has a direct im-pact on cement based properties and durability, Fig. 5a shows the negative effect of nano-TiO_2 on com-pressive strength for large volume content published by Meng et al. (2012) [25] for cement based mate-rials containing 0, 5, and 10 wt.% of nano-TiO_2. Chen et al. 2012 [26] published results shows a com-pressive strength gains about 10 and 20% results for cement based materials containing 0, 5, and 10 wt.% of rutile and anatase nano-particles. For small nano-TiO_2 volume content an optimum value, about 1% in volume, is observed by Zhang et al. (2011) [19] considering the compressive strength (Fig. 5a);

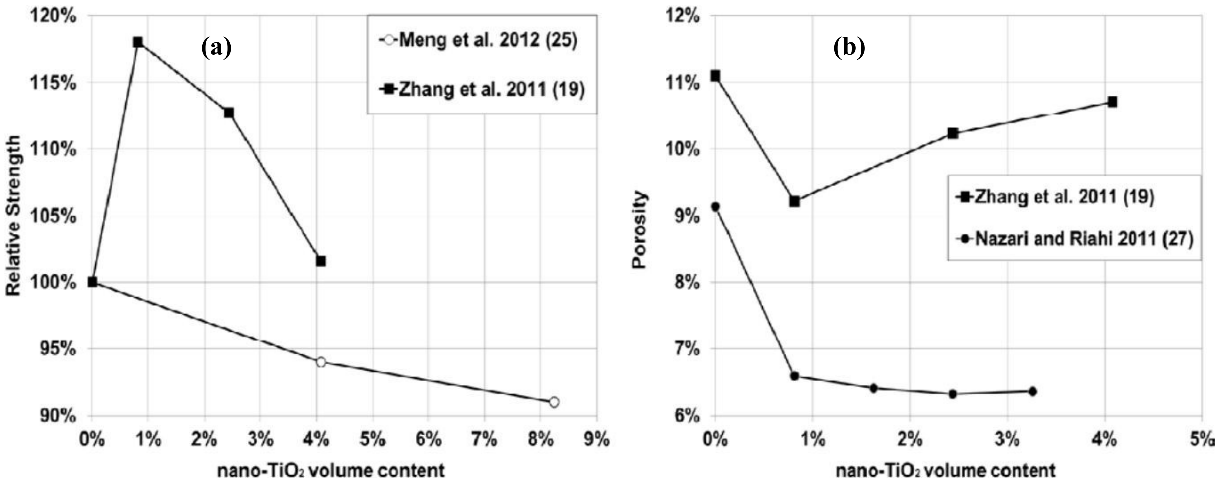

Fig. 5. (a) Nano-TiO_2 volume content x relative compressive strength [19,25]; (b) Nano-TiO_2 packing effect [19,27].

and concrete porosity (Fig. 5b) measured by Mer-cury Intrusion Porosimetry for cement based mate-rials with 0, 1, 2, 3, 4, and 5 wt.% of nano-TiO_2, as published by Zhang et al. (2011) [19], Nazari and Riahi (2011) [27]. These results indicates a similar packing effect of nano-TiO_2 particles in cement based materials reducing materials permeability and consequently the chloride diffusion coefficient as demonstrated by Zhang et al. (2011) [19].

Thermogravimetric analysis (TGA) between 110-650 iC for cement hydrates products C-S-H and $Ca(OH)_2$, shows that nano-TiO_2 contend increases total mass lost for this temperature range, as pub-lished by Nazari and Riahi (2011) [27] according with published results of Meng et al. (2012) [25]
considering the C_3S peaks reduction and $Ca(OH)_2$ peaks increases determined by X-Ray Diffraction for cement based materials containing 0, 5, and 10
wt.% of nano-TiO_2 after 28 days of curing. Cement reaction kinetics is directly affected by nano-TiO_2 content, calorimetry results published by Chen et al. 2012 [26] and Senff et al. (2012) [28], show that nano-TiO_2 accelerates the cement hydration and increases the reaction total heat indicating a nucle-ation effect of nano-TiO_2, according with Nazari and Riahi (2011) [27] results considering the X-Ray dif-
fraction peaks related to formation of the hydrated products $Ca(OH)_2$ N[Q4lDl9NaRNYfNTR N[
TiO_2 has a direct effect on materials based materi-als rheological properties, Senff et al. (2012) [28] results show that the nano-TiO_2 content increases the suspensions yield stress and viscosity, reduc-ing the mortars fluidity and concretes slump as pub-lished by T. Meng et al., 2012) [25] and Li et al. (2006) [15], respectively.

Many other nano-oxides particles have been tested in cement based materials nano-Fe_2O_3, nano-Al_2O_3, nano-ZnO_2, nano-CuO_2, and nano-$CaCO_3$, which present a direct effect on materials proper-ties and durability. Fig.6a shows a volume content

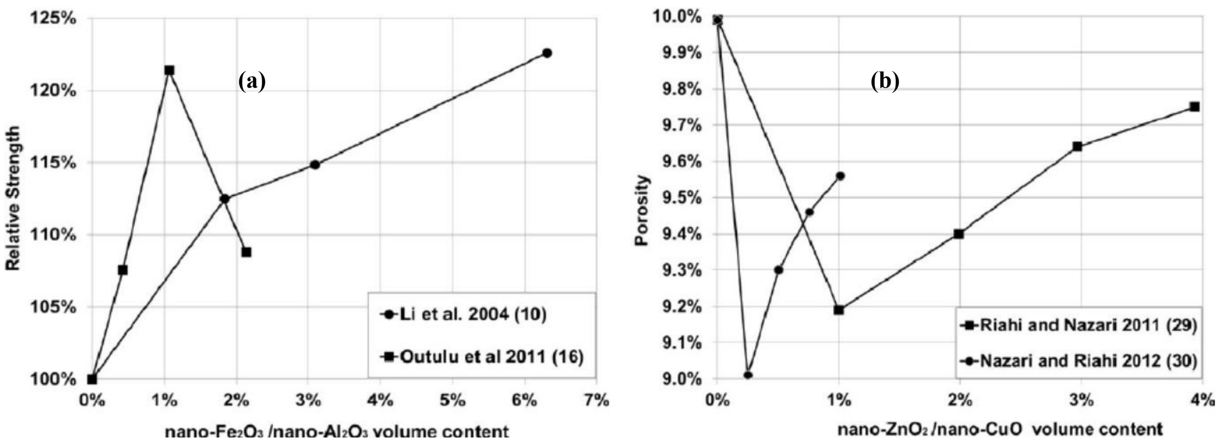

Fig. 6. (a) Nano-Fe_2O_3/Al_2O_3 volume content x relative compressive strength [10,16]; (b) Nano-ZnO_2/CuO Packing effect [29,30].

effect of nano-oxides particles on compressive strength, for small volume content of nano-Al_2O_3 and Fe_2O_3 an optimum volume could be observed based on compressive strength results published by Oltulu et al. (2011) [16], for cement based materials con-taining 0.5, 1.25, and 2.5% of these nano-particles oxides. Similar results have been published by Riahi and Nazari (2011) [29], Nazari and Riahi (2012) [30] and Xianoyan et al. (2012) [31] for cement based materials containing small quantities of nano-ZnO_2 and nano-CuO and nano-$CaCO_3$. For large volume content, Li et al. (2004) [10] present a positive ef-fect on the compressive strength for cement based materials containing 3, 5, and 10 wt.% of nano-Al_2O_3, according with published results of Sato and Beaudoin (2011) [32] considering the micro-hard-ness and elastic modulus of cement based materi-als containing 10 and 20 wt.% of nano-$CaCO_3$.

Packing effect could be identified for small vol-ume of nano-oxides particles; Fig. 6b shows mer-cury intrusion porosimetry results published by Riahi and Nazari (2011) [29] and Nazari and Riahi (2012)

[30] for cement based materials containing 0.5, 1,

1.5, and 2% wt.% of nano-ZnO_2 andnano-CuO. Oltulu et al. (2011) [16] has demonstrated an important effect of nano-oxide particles for cement based materials durability, considering the capillary per-meability reduction for concretes containing nano-

Al_2O_3 and nano-Fe_2O_3.

A nucleation effect has been identified by Riahi and Nazari (2011) [29] and Nazari and Riahi (2012)

[30] for cement based materials containing 0.5, 1,

1.5, and 2% nano-ZiO_2 and nano-CuO, considering the higher mass lost determined by

thermogravimetric analysis between 110 and 650 iC; the accelerate effect of nano-oxides in hydrated products kinetics and reaction total heat measured using an adiabatic colorimeter; and XDR peaks in-

tensity for hydrated products $Ca(OH)_2$ and CSH at early ages. Similar results about nucleation effect were published by Sato and Beaudoin (2011) [32] for cement based materials containing 10 and 20%

of nano-$CaCO_3$, considering adiabatic calorimetry and X-ray diffraction (XRD), carboaluminate peaks were not identified. Rheological properties are di-rectly affected by nano-oxides particles, slump, ini-tial and final setting time decreases for concretes containing nano-ZiO_2 [29] and nano-$CaCO_3$ [31].

3. CEMENT NANO-PARTICLES

Binder nano-particles of C_2S, C_3S, C_3A, and C_4AF compounds obtained by sol-gel processing and heating treated by flame spray reactor and oven heating have been tested by Halim et al. (2007) [33] and Huang (2006) [34], respectively; nano-C_2S (alita) and nano-C_3S (belite) particles obtained by sol-gel processing and submitted to oven heating treatment at 900 and 1400 iC has been analyzed by Perera et al.(2007) [35]; and sonochemical fabricated nano-pozzolan particles have been studied by A. Askarinejad et al. (2012) [36].

The high surface area of synthetized nano-binder particles leads to a considerable water demanding and consequently a higher water/binder ratio and porosity, reducing the compressive strength and micro-hardness of pure nano-cement based materi-als as demonstrated by Halim et al. (2007) [33] and Huang et al. (2006) [34]. Ultrasonic treatment for nano-pozzolan particles present a positive impact on compressive strength as compared nano-poz-zolan particles without a sonochemical treatment like demonstrated by Askarinejad et al. (2012) [36]. The surface area of nano-cement has an important impact on reaction kinetics considering accelerat-ing effect and the bigger total heat of nano-cement chemical reaction as published by Halim et al. (2007) [33] and Huang et al. (2006) [34].

4. CARBON NANO TUBES

Cement based nano-composites of carbon nanotubes (CNT) have been widely studied, mainly due the positive impact of carbon nanotubes on important composites properties like compressive and flexural strength. Fig. 7a shows the effect of carbon nanotubes on composites compressive strength, for very low volume contend, published results by Morsy et al. 2011 [37] show an optimum value for cement based materials containing 0.005, 0.02, 0.05, and 0.1 wt.% of CNT. Considering an-other volume content of carbon nanotubes studied by Melo (2009) [38], an optimum value about 0.3 wt.% has been achieved for cement based nano-composites containing 0.3, 0.5, and 0.75 wt.% of carbon nanotubes, similar results has been pub-lished by Manzur (2011) [39].

Fig. 7b shows the effect of carbon nanotubes on flexural strength of cement based nano-composites, for very low volume contend, results published by Cwirzen et al. (2008) [40] show a positive impact for volume contend lower than 0.01% of carbon nanotubes, similar results has been published by Konsta-Gdoutos et al. (2010) [41]. For carbon nanotube volume content higher than 0.5%, results published by Li et al. (2005) [42] and Musso et al.

(2009) [43] show a positive impact on cement based [N[

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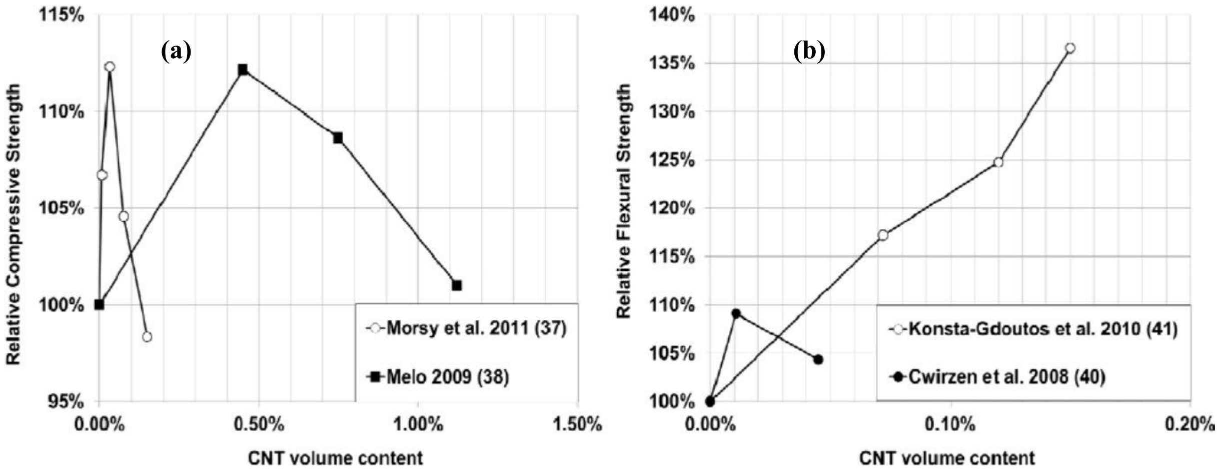

Fig. 7. (a) Carbon nanotube volume content x relative compressive strength [37,38]; (b) Carbon nanotube volume content x relative flexural strength [40,41].

nanotubes content, splitting tensile strength results published by Ince (2008) [44] for cement based nano-composites containing 2 wt.% of carbon nano-fibers (CNF) present approximately the same value for cement paste without carbon nanotubes.

These cement based nano-composites proper-ties improvement could be related with a total po-rosity reduction as demonstrated by Nochaiya and Chaipanich (2011) [45] applying Mercury Intrusion Porosimetry (MIP) technique for mixtures contain-ing 0.5 and 1% of carbon nanotubes, similar results has been published by Li et al. (2005) [42] for batches containing by 0.5% of CNT, Fig. 8a; and a carbon nanotube specific surface effect on materials strength as published by Manzur (2011) [39] for cement based nanocomposites containing 0.1, 0.2, 0.3 wt.% of Multi-Walled Carbon Nanotubes, 10-30 mm length and diameter varying between 8 and 30 nm resulting on specific surface areas of 40, 233, and 500 m^2/g, Fig. 8b shows this positive effect of carbon nanotubes specific surface on materials compressive strength. Surface chemical treatment of carbon nanotubes allows a possible interaction between COOH or C-OH groups of treated CNT and CSH cement phase as demonstrated by Li et al. (2005) [42] applying Infrared Spectroscopy (FTIR) technique.

A nucleation effect of Single Walled CNT has been verified by Makar and Chan (2009) [46] for cement nano-composites containing nanotubes, allowing a C-S-H hydration products growth on car-bon nanotubes surface as published by Makar and Chan (2009) [46] considering micrographs obtained by Scanning Electron Microscopy. $Ca(OH)_2$ mass loss measured using thermogravimetric analysis and adiabatic calorimetry published results by Makar and Chan (2009) [46] for cement pastes containing SWCNT, confirm the SWCNT nucleation effect. Ara-

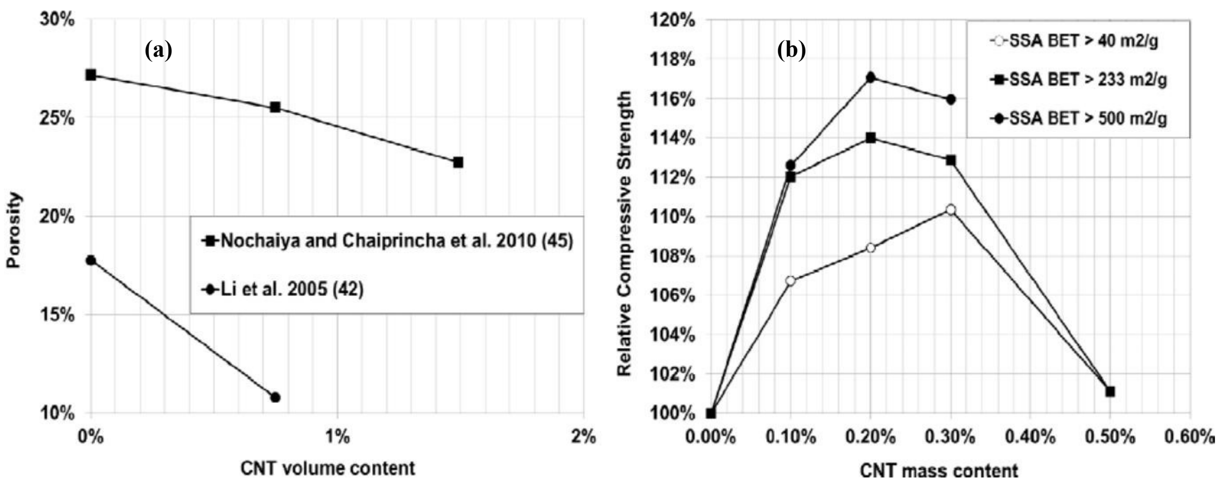

Fig. 8. (a) Carbon nanotube packing effect [42,45]; (b) Multi-walled Carbon Nanotubes Specific Surface Area x relative compressive strength [39].

gonite XRD peaks has been identified by Musso et al. (2009) [43] for cement pastes containing chemi-cal treated carbon nanotubes. Cement based nano-composites rheological behavior is directly affect by carbon nanotubes presence, as demonstrated by Konsta-Gdoutos et al. (2010) [47] considering the suspension viscosity increases for cement pastes containing MWCNT. A new technique for carbon nanotubes and nanofibers production called Carbon HedgeHog (CHH) allows growing the CNTs/CNFs from Fe catalyst particles naturally occurring in Portland cement, applying a chemical vapor depo-sition. Published results of Cwirzen et al. (2009)

[48] for cement based carbon hedgehog nano-com-posites has demonstrated a positive impact of these CNT/CNF on materials strength and electrical re-sistivity.

5. CONCLUSIONS

Mechanical performance, mainly compressive and flexural strengths, of cement based materials is improved by nano-particles use, reaching an opti-mum volume contend for all nano-oxides studied. Carbon nanotubes, present a very small optimum volume contend; and carbon nano tubes surface area present an important bonding effect on compres-sive strength.

These mechanical improvements are directly related with two important microstructural aspects of cement pastes containing nano-particles: the packing and nucleation effects observed for all nano-oxides and carbon nanotubes studied. For nano-SiO_2, pozzolanic reaction has been observed and lead to a mechanical improvement for large volumes contends.

Cement nano-particles does not present a me-chanical improvement due high water demand and large porosity. Kinetic reaction is directly affected due the high surface area of nano-particles modify-ing the suspensions rheological behavior: setting time, yield stress and viscosity, and consequently the flowtability and blending of concretes and mor-tars.

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