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Relationship between Oxygen Content of Graphene and Mechanical Properties of Cement-Based Composites

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Abstract
Cement composites were realized containing graphene-based materials having different oxygen content (from 0 to 45%). It was found that the polarity of the reinforcement influences both the dispersion in water and the chemical interaction with the hydrated cement matrix, and thus the mechanical performance of the composites. In particular, the reinforcements with the highest oxygen content result aggregated while the cement with the highest strength is obtained with functionalized graphene, containing 5% oxygen, where the surface has sufficient polar groups to guarantee proper dispersion and interaction with the hydrated cement.

Keywords: Cement and concrete; Mechanical properties; Carbon; Composites; Graphene

Introduction
The modification of cement and concrete-based materials with carbon-based nanomaterials is proposed as a simple option to improve their mechanical performance, with applications in civil and oil&gas applications [1−3]. In fact, compressive and flexural strength can increase significantly when a very low amount of nano-reinforcement is added. This effect derives from the combination of improved nucleation of cement hydrates and of crack propagation prevention [4−6]. In particular, it was demonstrated that graphene and graphene oxide may reduce the brittleness and enhance toughness, tensile and flexural strength of cement composites, providing the foundations for the production of cement-based materials with improved performance and durability [7−12].
Furthermore, some types of graphene-based materials have good electrical conductivity [13]. Thus, they could help decreasing the electrical resistivity of the cement thanks to the formation of conductive networks [14]. The coupling of high mechanical properties with acceptable electrical conductivity could help in the creation of self-monitoring structural materials, that would represent a turning point in the safety management of structural materials [15].
The incorporation of nanometric conductive materials can thus allow nano-scale control of matrix cracks [16,17] and improve the electrical properties. In order to have a significant effect on the mechanical properties with a limited impact on the cost of the final product, the content of graphene in the cement-based composites was chosen to be 0.1% by weight of cement (bwoc), in line with values used in the literature [11,18,19]. This quantity is much lower than percolation level, as required in self-monitoring applications.
The key concepts for the implementation of graphene-based nano-reinforcement in cement are the correct dispersion in the matrix and the interaction with the hydrated cement. It is well-known that an improvement of the properties of cement can be achieved only if the graphene is well dispersed in the matrix [20], i.e. if graphene remains dispersed during the whole process of preparation of cement paste and of setting of cement. The dispersion plays a key role in standardizing the properties of graphene-based composites for different applications, and has been extensively studied [6,21,22]. Two main methods can be used to guarantee a stable dispersion of graphene in water: the use of an organic surfactant, that creates an interface layer around the reinforcement [20,23] or the optimization of the polarity of the surface through functionalization [24]. However, studies suggest that surfactants reduce the interaction between matrix and reinforcement, so that to maximize the

interface adhesion it would be better if no surfactant were used [25,26]. In fact, the interaction between matrix and reinforcement in composites is known to be a very sensitive parameter, since it has a marked influence on the crack propagation and toughness [27–30].
In the literature several works are present on graphene-based materials, but in all cases are used either graphene nanoplatelets [8] (GNP, i.e. pure carbon) or graphene oxide [31] (GO, with very high oxygen content, around 50%). This paper is focused instead on the effect of oxygen content of graphene-based materials on the mechanical properties of cement composites. By using commercially available materials, it is proved that the best case is neither zero oxygen (GNP) nor high oxygen (GO), but instead a low/intermediate oxygen content. The lower oxygen content also ensures the maintenance of good electrical conductivity, as shown in the literature [13]. The dispersion of selected graphene-based reinforcements (oxygen content equal to 0%, 5%, 20%, 45.5%) in water was analyzed, followed by the assessment of the mechanical properties (flexural strength and toughness) of cement-based composites containing 0.1% bwoc of reinforcement.

Materials and methods

Graphene nanoplatelets (GNP) and COOH-functionalized graphene nanoplatelets (GNP-COOH) were purchased by Cheaptubes, while reduced graphene oxide (RGO) and graphene oxide (GO) were purchased by Graphenea. The oxygen content of these materials is, respectively, 0%, 5%, 20%, 45.5%, as declared by the producers. The NaOH and the $Ca(OH)_2$ were purchase from Sigma-Aldrich. Raman spectra of all these materials were obtained with a Horiba Jobin Yvon HR800 instrument equipped with an Olympus BX41 microscope. The samples were excited with a red HeNe laser (wavelength 633 nm, power 20 mW) and a Nd solid state green laser (wavelength 532 nm, power 250 mW) with a magnification ratio of 50× with 30 acquisition of 30 seconds. The hydrodynamic size and zeta potential of the carbon-based nanomaterials were measured by using a DLS Zetasizer Nanoseries ZS90, Malvern Instruments, UK, at a controlled temperature of 25.0 ± 0.1 °C in a thermostatic cell; analyses were done in triplicate and the average value was considered.
The graphene-based materials were dispersed in distilled water at the concentration of 2.22 g/L with an ultrasonic tip (Vibra-cell™) for 15 minutes at 100 W power in a cooled beaker. This concentration is required to produce cement-based composites containing 0.1% bwoc graphene, with a water-to-cement ratio equal to 0.45. The cement used in this study is an American Petroleum Institute (API) oil-well cement Class G (Lafarge North America). To prepare cement composites, the suspension of graphene-based material in water was mechanical stirred while cement powder was slowly added to the continuously stirred solution. The cement/graphene paste was then poured into prismatic molds of 75x20x20 mm size and cured for 24 h at 85 °C and 100% relative humidity.
Mechanical properties were tested with a single-column Zwick-Line z050 testing machine with a load cell having a maximum capacity of 1 kN. Flexural tests were performed on notched specimens, in 3-point bending, with 65 mm span, by controlling the crack mouth opening displacement (CMOD) (Figure 1), following the procedure prescribed by the Japanese standard JCI-S-001-2003 [32,33]. For every test at least three specimens were analyzed. Compression tests were performed on 20x20x20 mm cubic samples, obtained from the prisms used for bending tests after failure. At least four cubic specimens for each sample were tested.

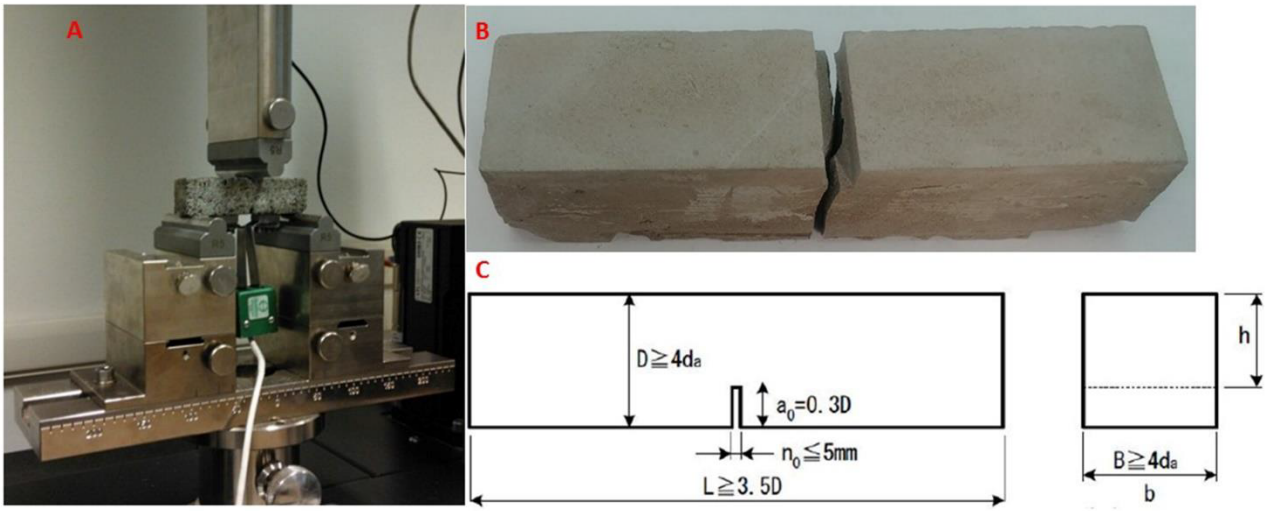

Figure 1. Prismatic sample of cement composite undergoing flexural testing in CMOD control mode (A); broken sample showing crack propagation up to fracture (B); geometry of the sample (C).

Results and discussion

In order to understand the structure of the graphene-based nanomaterials, Raman spectra of GNP, GNP-COOH, RGO and GO were collected, and are shown in Figure 2 together with their I_D/I_G ratio. Each spectrum is characterized by two characteristic bands, namely the D-band around 1300 cm^{-1}, arising from the disorder-induced phonon mode (A_{1g}-band), and the G-band around 1600 cm^{-1}, assigned to the Raman-allowed phonon mode (E_{2g}-band) [34]. The extent of the graphitic defects can be evaluated by the ratio of the D and G band intensities (I_D/I_G) [35]. For graphitic materials, the I_D/I_G ratio decreases with the degree of order of the graphitic structure [36], however for very small in-plane correlation length (<20 Å), corresponding to small hexagonally ordered carbon clusters, the I_D/I_G ratio decreases again reaching zero. Figure 2 confirms this behavior: GO has a I_D/I_G ratio of 1.01, due to its quasi-amorphous structure (very small in-plane correlation length), and this ratio further increase to 1.44 upon transforming into RGO, due a partial and local ordering of the structure [37,38]. For highly ordered graphitic structures, like GNPs and GNP-COOH, however, the I_D/I_G ratio decreases substantially, to a value close to 0.5. The fact that the ratio is similar for GNPs and GNP-COOH suggests that the carboxylic functionalization of GNP did not modify substantially the graphitic structure of the graphene, being probably confined to the sides of the graphene flakes [39].

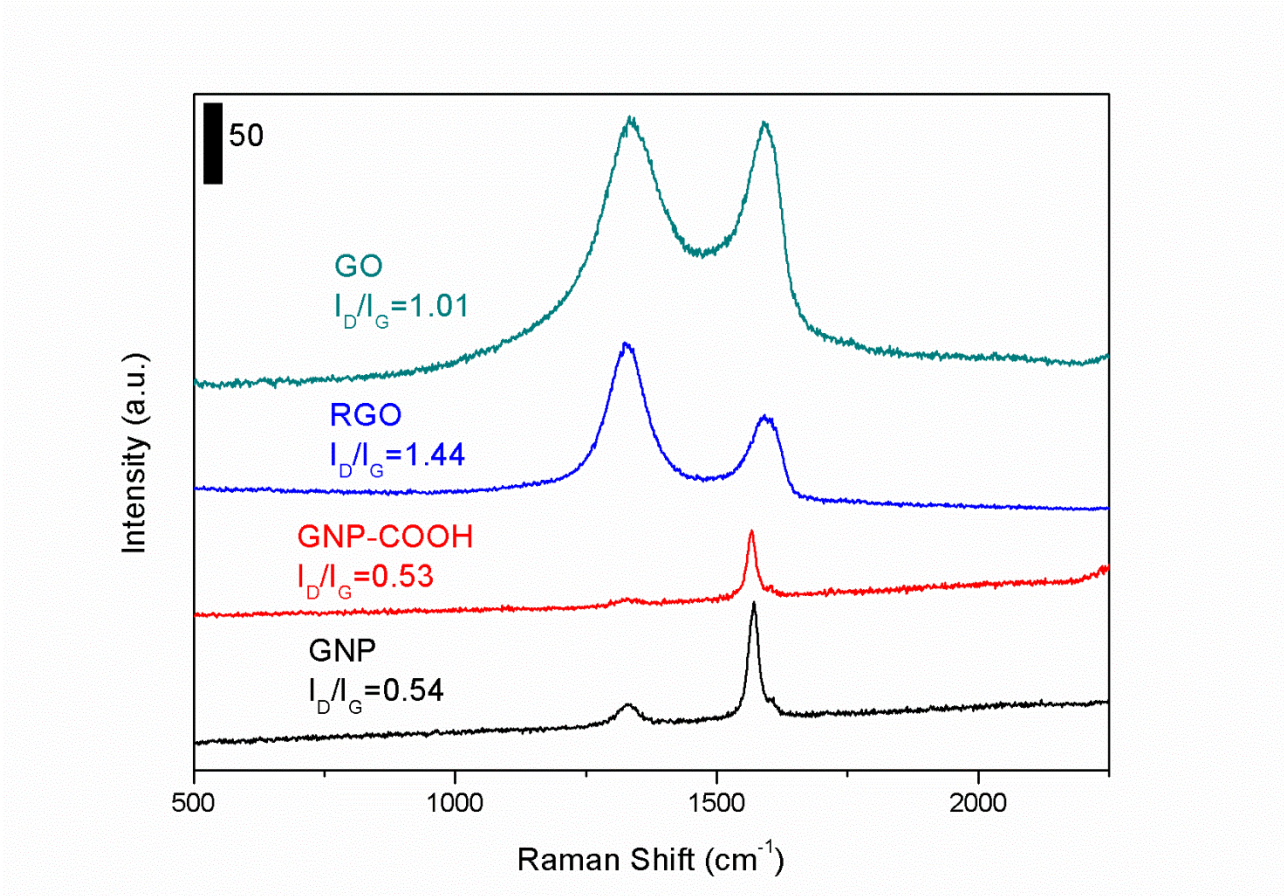

Figure 2. Raman spectra of the used graphene-based materials. For each sample, the average I_D/I_G ratio is calculated.

The dispersion in pure water of graphene-based materials yielded apparently stable solutions; however, zeta potential was also measured in order to quantify the suspension stability, that is considered acceptable when the zeta potential absolute value is higher than 30 mV [40]. The results are shown in Table 1. All the materials presented negative potential, demonstrating a negative charge forming on the surface. However, only GO presents a zeta potential absolute value close to the 30 mV limit, indicating a stable suspension. GNP suspension is the less stable, as expected due to the very low polarity of its surface. GNP-COOH, notwithstanding the rather low oxygen content, presents a rather high value of zeta potential, probably due to the presence of –COOH groups on the sides of the flakes. RGO, instead, notwithstanding the rather high oxygen content (20%), has lower stability than GNP-COOH; this is probably due to the fact the RGO presents an inhomogeneous microstructure, with alternate zones of high and low oxygen content [38].

Table 1: zeta potential of the suspensions of graphene-based materials.

Sample	Zeta potential (mV)
GO	-28.8
RGO	-16.3
GNP-COOH	-21.5
GNP	-12.9

The mechanical properties of cement-based composites containing GNP, GNP-COOH, RGO and GO are shown in Figure 3, with the error bars indicating the standard deviation. Figure 3D provides also typical stress-crack mouth opening displacement curves for the different samples. The results show that the mechanical properties are generally improved by the presence of low oxygen content graphene-based materials, while GO and RGO present little or no improvement in properties with respect to the base cement.

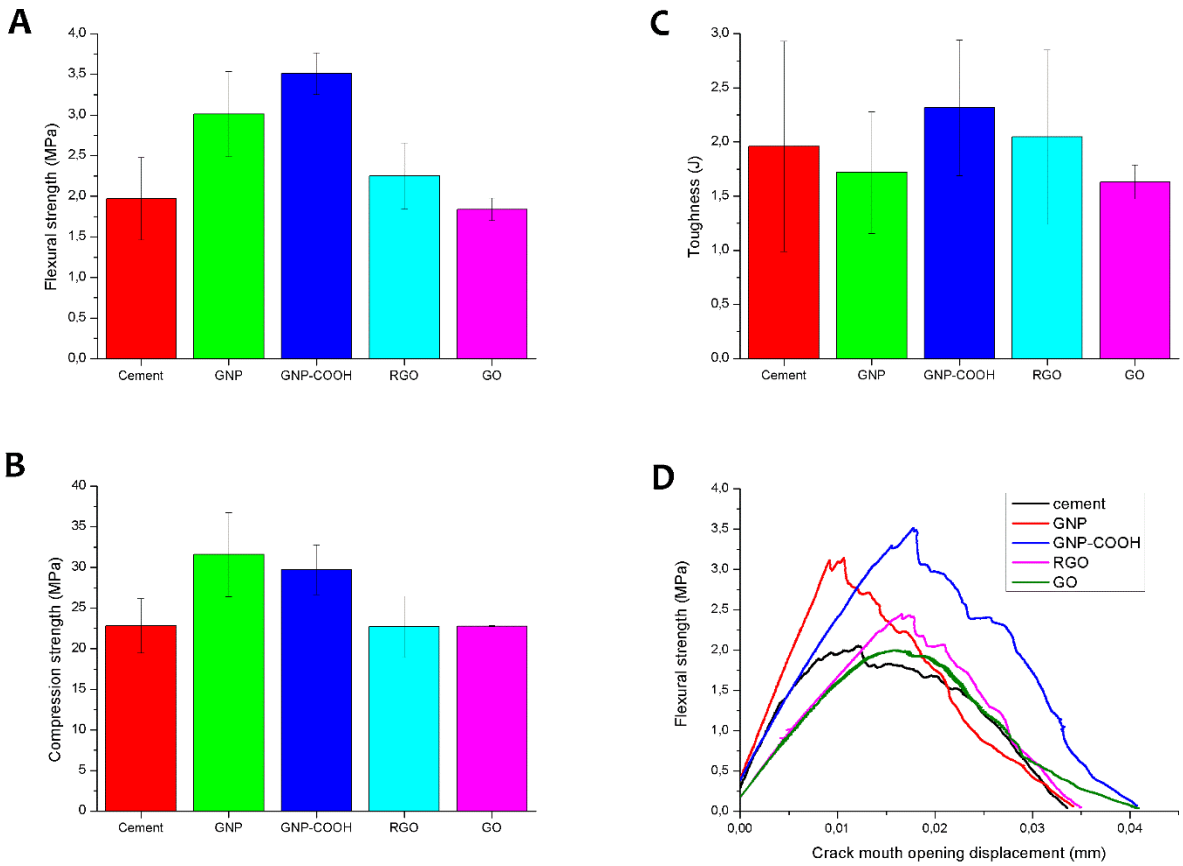

Figure 3. Flexural strength (A), compression strength (B), toughness (C) of pure cement and of cement composites containing GNP, GNP-COOH, RGO and GO; typical stress-crack mouth opening displacement curves (D) of pure cement and of cement composites containing GNP, GNP-COOH, RGO and GO.

Since all the chosen materials have similar size and shape factor, a positive effect would be expected in all cases, as suggested in the literature, where it is demonstrated that platelet-shaped reinforcements improve flexural strength [41] and that nano-sized powders have a positive effect on cement strength [42]. However, it is known that when GO comes in contact with a $Ca(OH)_2$ solution, flocculation occurs [43,44]. This effect is shown in Figures 4 and 5, where agglomeration of both GO and RGO flakes is observed upon addition of $Ca(OH)_2$ to a stable water-based suspension.

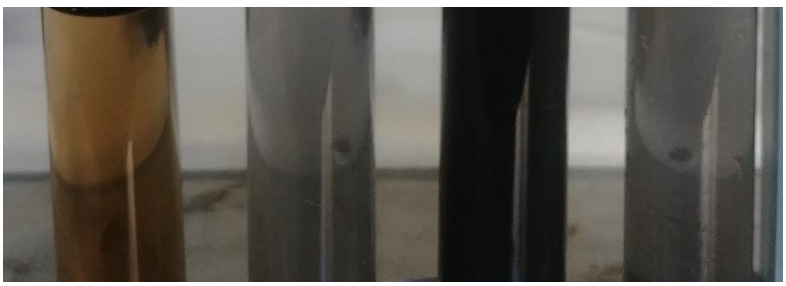

Figure 4: Dispersion of different graphene-based materials in distilled water, from left: GO, RGO, GNP-COOH, GNP

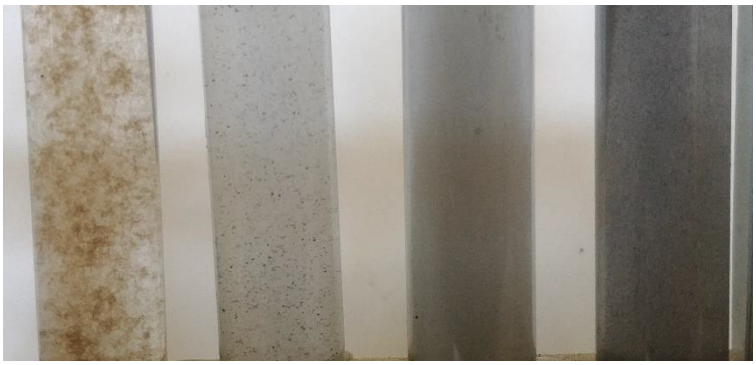

Figure 5: Dispersion of different graphene-based materials in water after adding 3 ml of calcium hydroxide solution, from left: GO, RGO, GNP-COOH, GNP.

Since the pH of the cement paste reaches in a very short time values close to 13 due to the dissolution of hydroxides [45, 46], GO and RGO can start to agglomerate while the cement is still in paste form. A test was also made on a GO suspension by adding 3 ml of a saturated NaOH solution instead of $Ca(OH)_2$. As illustrated in Figure 6, also in this case GO aggregation occurs.

Figure 6: Dispersion of GO in water after adding 3 ml of sodium hydroxide solution.

This behavior explains the low mechanical performance of cement samples containing GO (stronger agglomeration) and RGO (milder agglomeration): it is known that the presence of agglomerates is a source of crack initiation and propagation [37].
Regarding the improved performance of GNP-COOH with respect to the other materials considered, the reason can be found in the improved adhesion between matrix and reinforcement, that determines the properties of brittle composites [34]. The presence of functional groups such as carboxylic acids can react in cement with hydrated calcium silicates (C-S-H phase) or with portlandite, $Ca(OH)_2$, leading to a strong interfacial bond between reinforcement and matrix [6].
By keeping into account all these effects, the behavior of the different graphene-based nano-reinforcements can be explained.

- In the case of samples containing GNP, an improvement of both flexural (50%) and compression (40%) strength is observed due to the presence of nano-sized platelet-shaped particles, as also observed in literature [4,8]. Toughness is slightly lower than the base

cement, but due to the rather high standard deviation of the toughness data the difference is not very significant.

- In the case of GNP-COOH containing samples, the improved dispersion and interaction with cement allows to further increase flexural strength with respect to samples containing GNP and to have an appreciable improvement in toughness. The increase of flexural strength of samples containing GNP-COOH with respect to base cement is close to 80%.
- In samples containing RGO and GO, the flocculation due to the presence of portlandite leads to the formation of aggregates, that cause a reduction of strength with respect to GNP-COOH. In the case of RGO the strength remains slightly higher (15%) than pure cement, while in the case of GO the dispersion issues completely nullify the effect of the nanoreinforcement. Some works in the literature [25,27] suggest that another effect could play a role in the case of GO: its high hydrophilicity can trap water between the platelets, thus sequestering water that results not anymore available for the complete and uniform hydration of cement paste.

Conclusion

Overall, these results confirm the literature on cement composites containing GNP and GO [8,31,47,48], but also demonstrate that, in order to optimize the mechanical properties, it is important to take into account the oxygen content of graphene-based materials. In particular, a moderate oxygen content in graphene provides the best mechanical properties improvement for cement-based composites, thanks to a good dispersion and interaction with hydrated cement. In fact, when 0.1% bwoc of functionalized graphene nanoplatelets (GNP-COOH) containing 5% of oxygen were added to a class G cement, both strength (flexural and compression) and toughness were significantly improved with respect to pure cement (80% increase in flexural strength, 30% in compression strength, 20% in toughness). Non-functionalized GNPs are somewhat less effective in reinforcing cement, due to their limited dispersibility in water and their low chemical compatibility with the hydrated cement surface. On the contrary, RGO and GO suffer from severe dispersion issues, related to their strong interaction with calcium hydroxide, that brings to agglomeration. This behavior affects the properties, and in fact both the strength and the toughness of RGO and GO-containing cement composites are not significantly different from the one of pure cement.

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