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#### **ACCEPTED MANUSCRIPT**

# Effect of graphene on setting and mechanical behaviour of tricalcium phosphate bioactive cements

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#### Abstract

The potential reinforcing effect of graphene on calcium phosphate cements (CPCs)-based for injectable bone substitutes and scaffolds is presented. The influence of graphene (0-3.84 vol.%) on the microstructural development during setting and the resultant mechanical properties of CPCs constituted by  $\alpha+\beta$ tricalcium phosphate is analysed. Optimum setting conditions were established using uniaxial compression strength of CPC and composites with pristine and functionalized graphene and liquid/solid ratios (L/S = 0.5-0.6 mL/g) that allowed the mixing and spatulation of the powders. Tensile strength of optimised materials has been determined using the Diametric Compression of Discs Test (DCDT). X-ray diffraction, Raman spectroscopy and FE-SEM-EDS on fracture surfaces were used to investigate phase composition and morphological changes in set specimens. Strengthening occurs for functionalized graphene additions up to 1.96 vol.% due to different toughening mechanisms. Crack deflection, bridging and branching by graphene and, finally, the pull-out of the unbroken graphene sheets have been identified. Interlayer sliding between the graphene before pulling-out is an additional toughening process. Main effect of graphene on strength is the increase of reliability.

Key Words: Calcium phosphate cements; Graphene; Composites; Setting; Microstructure; Mechanical properties

#### **1. Introduction**

Calcium phosphate (CaP) based ceramics are currently used in clinical practice for bone repair (e.g., dentistry, traumatology, and maxillofacial surgeries) due to their biological behaviour -biocompatibility, bioactivity and osteoconductivity- (see references [1-4] for review). In particular,  $\alpha$ - and  $\beta$ -

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