GOLD NANOPARTICLES AND SILICON AS EFFECTIVE MODIFIERS OF HYBRID-TYPE, CHEMICALLY BONDED BIOMATERIALS

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Introduction

Calcium phosphate (CaP) bioceramics is widely used in the field of bone regeneration, mainly due to its excellent biocompatibility. Usually, CaPs-based biomaterials are produced in the sintered form as porous and dense blocks. The attractive alternative for pre-shaped bioceramics are chemically bonded materials in the form of mouldable pastes. Calcium phosphate-based bone cements (CPCs) are the most widely investigated group of these bone substitutes [1]. Recently CPCs enriched with granules and microspheres i.e., biomicroconcretes, have gained huge interest [2,3]. Granules when loaded with biologically active agents, can serve as delivery vehicles in targeted therapies. To obtain antimicrobial activity nanoparticles such as silver (AgNPs), copper (CuNPs), and gold (AuNPs) may be introduced into biomaterials [4]. Whereas, to improve biological performance, CaPs modified with elements such as silicon, magnesium or zinc can be developed [5]. A combination of these strategies can lead to production of biomaterials with some superior properties.
The aim of our study was to obtain and examine hybrid-type, chemically bonded biomaterials composed of surgically handy silicon-modified CPCs and hybrid AuNPs-loaded hydroxyapatite/chitosan granules.

Materials and Methods

The silicon-modified α-tricalcium phosphate (Si-αTCP) powder was used as a setting phase of chemically bonded biomaterials. Si-αTCP (0.3 wt.%Si) was synthesized by the wet chemical method. Hybrid hydroxyapatite/chitosan granules (HA/CTS) containing 17 wt.% chitosan (Sigma-Aldrich) modified with 0.1wt.% gold nanoparticles (US Research Nanomaterials), were synthesized via a wet chemical method. The solid phases of the biomicroconcretes were obtained by mixing the hybrid granules with α-tricalcium phosphate powder in a ratio of 2:3, respectively. The 0.75 wt.% methylcellulose solution in 2.0 wt.% Na_2HPO_4 was applied as the liquid phase of the biomicroconcretes.
The phase composition (XRD), microstructure (SEM, TEM), setting times (Gilmore needles), mechanical strength (Instron), and *in vitro* bioactive potential of the composites in simulated body fluid were examined. Furthermore, based on the AATCC 100, the antibacterial activity of the materials against *Staphylococcus epidermidis, Staphylococcus aureus* and *Escherichia coli* was evaluated.

Results and Discussion

The XRD analysis revealed that the biomicroconcretes composed of two major crystalline phases: α tricalcium phosphate and hydroxyapatite. The amorphous halo in XRD patterns refers to the presence of chitosan. SEM and TEM studies showed good adhesion at the granule/matrix interface (FIG. 1).

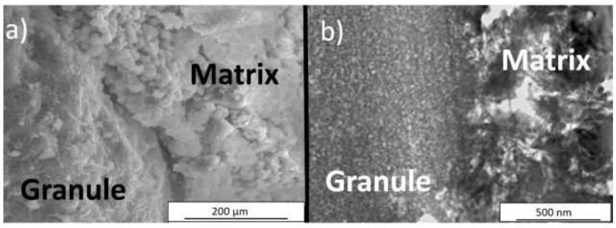

FIG. 1. SEM (a) and TEM (b) images of granule/matrix interface of AuNPs and silicon modified biomicroconcrete.

Furthermore, hybrid-type biomicroconcretes possessed acceptable setting times and mechanical properties appropriate for low-load bearing applications. The introduction of silicon and AuNPs led to a favourable shortening of the setting process (t_F = 10 min). Moreover, biomicroconcretes modified with gold nanoparticles and silicon possessed enhanced bioactivity, proven during *in vitro* studies in simulated body fluid. The *in vitro* tests revealed the antimicrobial activity of all developed biomicroconcretes against the tested bacterial strains i.e., *Staphylococcus epidermidis, Staphylococcus aureus* and *Escherichia coli*. This effect related to the presence of chitosan and gold nanoparticles.

Conclusions

The new hybrid-type, chemically bonded biomaterials based on hydroxyapatite and chitosan were successfully developed and investigated. Gold nanoparticles and silicon were proven to be effective modifiers of biomicroconcretes. Developed materials possessed physicochemical properties sufficient for non-load bearing applications. Furthermore, the bioactive potential as well as the antibacterial activity of all developed biomicroconcretes were confirmed in *in vitro* studies. The AuNPs and Si-modified composites were found to be promising candidates for further biological studies.

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