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Mechanical property and antibacterial activity of silver-loaded polycation functionalized nanodiamonds for use in resin-based dental material formulations

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ABSTRACT

Research on the cutting-edge nano-fillers for designing dental restorative materials with excellent mechanical property and antibacterial performance is demanding and provoking work. In this study, we fabricated silver-loaded polycation functionalized nanodiamonds (Ag/QNDs) that harbored dual biocidal modes for use as reinforced filler in dental resin. As a result, by reason of homogeneous dispersion and strong bonding with resin matrix, Ag/QNDs-reinforced resin exhibited great improvement of Vickers hardness, flexural strength and modulus as compared with pure resins. Due to the combined bactericidal effect of Ag^+ and cationic polymers, Ag/QNDs-containing composites had an excellent antibacterial activity even at a concentration of 1.0 wt% but without significant cytotoxicity. Therefore, development of the Ag/QNDs hybrid is expected to aid in significant improvement of lifetime and quality of dental restorative resin.

1. Introduction

Resin-based dental materials has been widely used in restorative dentistry, mainly due to their satisfactory aesthetics and easy operation process [1]. However, some disadvantages still remain, affecting the long-term performance in clinical dentistry. One disadvantage of resin composites is the insufficient mechanical property, such as low hardness and flexural strength [2]. Many inorganic nanofillers have been developed to enhance mechanical behavior [3]. Recently, nanodiamonds (NDs) have been considered to be a new nano-filler, because of its excellent properties such as exceptional mechanical property, chemical stability and biocompatibility [4]. Nevertheless, the major obstacle of using NDs is the ability to deliver nanodiamonds in the form of well-dispersed particles into polymer matrix. Some researches demonstrated that surface treatment of NDs was beneficial for improving the wettability and dispersion in the matrix, thereby improving the mechanical behavior of polymer [5].

In addition, resin composites are susceptible to accumulate bacterial plaques, causing secondary caries [6]. To settle this problem, lots of antibacterial agents have been incorporated into dental resins to improve the antibacterial activity. Among these prepared dental resins, materials possessing both contact-killing and release-killing capabilities have much better potential to meet requirements than those possessing one mode capability by incorporation of quaternary ammonium salt and silver nanoparticles (Ag NPs) [7]. Nevertheless, Ag NPs are limited to be used due to easy aggregation in resins, causing the decrease of mechanical properties [8]. Silver-supported fillers have been proved to effectively improve the dispersity of Ag NPs, resulting in enhancement of mechanical and antibacterial properties of resin composites [9].

Inspired by above findings, in this study, NDs were functionalized by a cationic copolymer, quaternized poly (4-vinylpyridinium-co-2-hydroxyethyl methacrylate) (QP(VP-co-H_{10})) to improve their dispersion in resin matrix, obtaining the contact-killing nanocomposite QNDs. Whereafter, Ag/QND hybrids with dual biocidal modes were prepared through an *in-situ* reduction method using QNDs as substrate and $AgNO_3$ as Ag^+ source. Finally, dental resin containing Ag/QND nanoparticles was fabricated and their mechanical properties, antibacterial performance and cytotoxicity were systemically evaluated.

2. Material and methods

As shown in Supplementary information.

3. Results and discussion

3.1. Characterizations of Ag/QND nano-hybrids

Fig. 1a shows FTIR spectra of particles. As compared to pristine NDs, the new absorption features at 1601, 1467 and 1417 cm^{-1}

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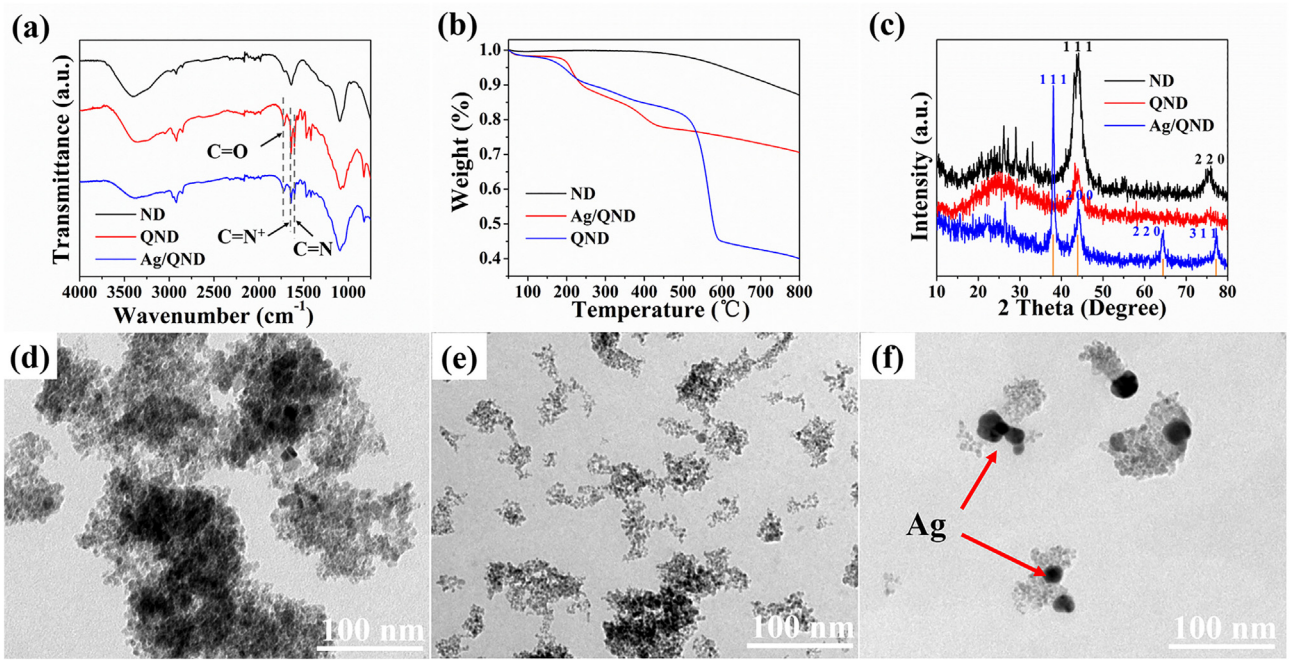

Fig. 1. (a) FTIR spectra, (b) TGA curves and (c) XRD patterns of NDs, QNDs and Ag/QNDs. TEM images of (d) pristine NDs, (e) QNDs and (f) Ag/QNDs.

observed in the spectra of QNDs and Ag/QNDs represented C=N and C=C stretching vibration in pyridine group. The surface bonding of 2-bromoethyl methacrylate (BEMA) and cationization were verified by increased C=O vibration at 1715 cm^{-1} and the stretching band intensity at 1639 cm^{-1}, respectively. Due to chemical bonding between Ag NPs and QNDs, such as Ag-N and Ag-O coordination bond, absorption peaks of Ag/QND were weaker than that of QND. As TGA curves shown in Fig. 1b, pristine ND were thermally stable and only decomposed over 500 °C. QNDs displayed a 48% weight loss between 250 and 500 °C as compared with NDs, which corresponded to the decomposition of organic compound. Ag/QNDs remained approximately 30 wt% more than QNDs at

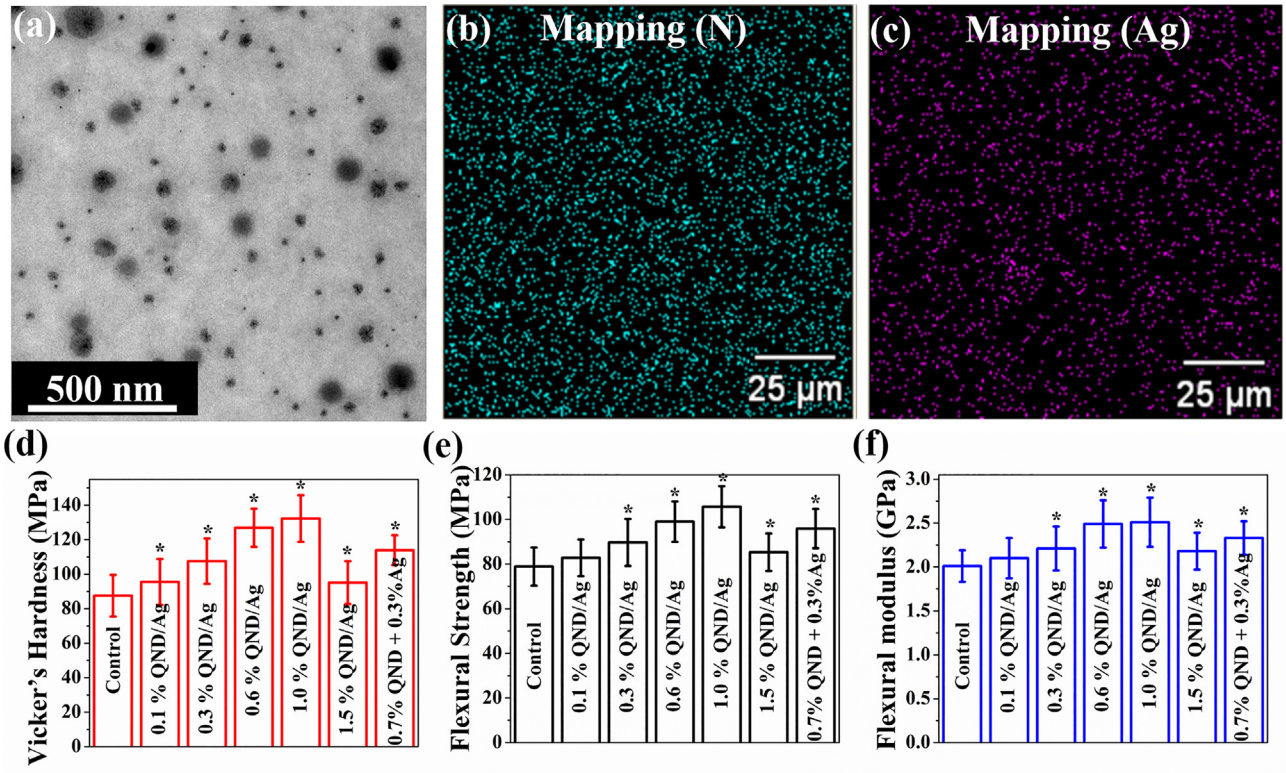

Fig. 2. (a) Representative TEM images of Ag/QND nanoparticles dispersed in the resin matrix at concentration of 1.0 wt%. (b and c) Mapping images of N and Ag on Ag/QND-incorporated resin surface. (d) Vicker's hardness, (e) flexural strength and (f) flexural modulus of all specimen. $^{*}P < 0.05$ as compared to pure resins.