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Research Paper

Evaluation of damage progression and mechanical behavior under compression of bone cements containing core–shell nanoparticles by using acoustic emission technique

O.F. Pacheco-Salazar[a], Shuichi Wakayama[b], Takenobu Sakai[b], J.V. Cauich-Rodríguez[a], C.R. Ríos-Soberanis[a], J.M. Cervantes-Uc[a,*,1]

[a]Unidad de Materiales, Centro de Investigación Científica de Yucatán, A.C., Calle 43 No. 130., Col. Chuburná de Hidalgo, C.P. 97200 Mérida, Yucatán, Mexico
[b]Tokyo Metropolitan University 1-1 Minami-Osawa, Hachioji-shi, Tokyo 192-0397, Japan

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ABSTRACT

In this work, the effect of the incorporation of core–shell particles on the fracture mechanisms of the acrylic bone cements by using acoustic emission (AE) technique during the quasi-static compression mechanical test was investigated. Core–shell particles were composed of a poly(butyl acrylate) (PBA) rubbery core and a methyl methacrylate/styrene copolymer (P(MMA-co-St)) outer glassy shell. Nanoparticles were prepared with different core–shell ratio (20/80, 30/70, 40/60 and 50/50) and were incorporated into the solid phase of bone cement at several percentages (5, 10 and 15 wt%). It was observed that the particles exhibited a spherical morphology averaging ca. 125 nm in diameter, and the dynamic mechanical analysis (DMA) thermograms revealed the desired structuring pattern of phases associated with core–shell structures. A fracture mechanism was proposed taking into account the detected AE signals and the scanning electron microscopy (SEM) micrographs. In this regard, core–shell nanoparticles can act as both additional nucleation sites for microcracks (and crazes) and to hinder the microcrack propagation acting as a barrier to its growth; this behavior was presented by all formulations. Cement samples containing 15 wt% of core–shell nanoparticles, either 40/60 or 50/50, were fractured at 40% deformation. This fact seems related to the coalescence of microcracks after they surround the agglomerates of core–shell nanoparticles to continue growing up. This work also demonstrated the potential of the AE technique to be used as an accurate and reliable detection tool for quasi-static compression test in acrylic bone cements.

*Corresponding author. Tel.: +52 999 9428330.
E-mail address: manceruc@cicy.mx (J.M. Cervantes-Uc).
[1]Postal address: Calle 43 No. 130 Col. Chuburná de Hidalgo, C.P. 97200 Mérida, Yucatán, México.

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1. Introduction

Acoustic emission (AE) technique is a nondestructive test (NDT) method commonly used to detect and locate flaws in mechanically loaded structures and components (Hellier, 2003; Qi and Wayne, 2014; Shen et al., 2015). In the biomedical field, this technique has been used to analyze biomaterials, tissues and tissue/biomaterial interfaces mainly those concerning to orthopedic discipline (Kohn, 1995). In this regard, the evaluation of cemented implants by using AE has helped to provide an understanding of failure mechanisms in bone cements during fatigue loading, during polymerization shrinkage cracking, and in entire cemented constructs (Browne et al., 2010). It has also demonstrated that is possible to distinguish between different failure processes and to locate damage sites. Moreover, this technique has the potential to monitor microcracks formation in the cement mantle in vitro for cemented femoral stems during fatigue loading. Finally, it has been used in clinical studies to predict the probability of bone fracture in patients (Sinnett-Jones et al., 2009.

Several authors have studied the fracture mechanisms of commercial bone cements by using AE technique. Roques et al. (2004) monitored the damage in CMW-1 bone cement during four point bending fatigue tests whereas Ng and Qi (2001) employed the wavelet-based acoustic emission (WBAE) technique to characterize the fatigue behavior of Palacos R bone cement using compact tension specimens. Furthermore, Jeffers et al. (2005) and Sinnett-Jones et al. (2009) reported the use of AE technique during uniaxial tensile fatigue tests performed on CMW-1 bone cement. It can be seen that all previous reports involved fatigue tests, i.e. dynamic conditions. Recently, Rios-Soberanis et al. (2014) reported the mechanical characterization of bone cements during a quasi-static tensile test by using AE technique and to our knowledge there are no more reports on this type of tests; also, this paper was not only focused on the study of the fracture mechanism of cements by means of AE technique using tensile coupons under quasi-static conditions, but also on the effect of adding a second monomer to bone cement formulations in order to yield materials with lower modulus; this feature will allow cement to withstand large strains without fracture.

An alternative method to reduce modulus and enhance the fracture toughness of bone cements consists of using core–shell nanostructured particles (Paul and Bucknall, 1999; Gutiérrez-Mejía et al., 2014); this approach improves ultimate properties but does not implies a significant reduction in the elastic modulus considering that they must fulfill the minimum bending modulus of 1800 MPa (International Organization for Standardization (ISO) 5833, 2002). It is well known that the core–shell nanoparticles result in an enhanced toughness which has been attributed to a mechanism of plastic shear and cavitation rather than crazing (Murakami et al., 1998). Despite this, the effect of the incorporation of core–shell nanoparticles on the fracture mechanisms of bone cements is unknown.

Therefore, the aim of this work was to identify the fracture mechanisms of acrylic bone cements containing core–shell nanoparticles during a quasi-static compression test by using the AE technique. To our knowledge, there are no reports neither on the mechanical evaluation of bone cements under compressive forces by means of AE nor on the effect of adding core–shell nanostructured particles on the fracture mechanisms of bone cements using this technique. Nanoparticles were composed of a poly(butyl acrylate) (PBA) rubbery core and a methyl methacrylate/styrene copolymer (P(MMA-*co*-St)) outer glassy shell, considering that the solid component of commercial bone cement Surgical Simplex® P contains this copolymer. This brand contains in its solid phase 73.5 wt% of P(MMA-*co*-St), 15 wt%. of poly(methyl methacrylate), PMMA, 10 wt% of barium sulfate, $BaSO_4$ and 1.5 wt% of benzoyl peroxide, BPO, whereas the liquid component consists of methyl methacrylate, MMA, at 97.4 vol.% and *N*,*N*-dimethyl-*p*-toluidine, DMPT, at 2.6 vol.% (Kuehn et al., 2005). Nanoparticles were prepared with different core–shell compositions (20/80, 30/70, 40/60 and 50/50) and were incorporated into the solid phase of bone cement at various percentages (5, 10 and 15 wt%).

2. Experimental

2.1. Materials

Core–shell particles were synthesized using butyl acrylate, methyl methacrylate and styrene. Inhibitors were removed by passing the monomer through a pre-packed column. Ethylene glycol dimethacrylate (EGDMA) was used as crosslinker, while potassium persulfate ($K_2S_2O_8$) and sodium dodecyl sulfate (SDS) were used as initiator and as surfactant, respectively. All chemicals were purchased from Aldrich.

2.2. Core–shell nanoparticles synthesis

The synthesis of core–shell particles was carried out in 2 L four-necked glass reactor equipped with a mechanical stirrer, a reflux condenser and a thermometer. The reactor was immersed in a thermostatic water-bath for controlling the reaction temperature.

Core–shell particles were prepared by two-stage seeded emulsion polymerization by growth of PBA seed latex previously polymerized. The first stage involved the synthesis of the PBA rubbery core at 80 °C during 2 h by adding the monomer after the initiator. The second stage corresponded to the 2 h polymerization of P(MMA-*co*-St) outer glassy shell. When the reaction finished, the obtained core–shell latex was diluted with distilled water and then they were precipitated after a defrosted process. The powdered core–shell particles were washed with abundant water to remove the excess surfactant, dried in an oven at 60 °C for 24 h, grounded and sieved. By changing the composition of feeding monomers, a series of core–shell latex particles were synthesized (20/80, 30/70, 40/60 and 50/50).

2.3. Characterization of core–shell particles

2.3.1. Scanning electron microscopy (SEM)

SEM micrographs of the powdered core–shell particles, obtained after precipitating the latex, were obtained with a JEOL 6360-LV scanning electron microscope at 20–25 kV. For