



Quaternary ammonium silane-functionalized, methacrylate resin composition with antimicrobial activities and self-repair potential

Shi-qiang Gong^{a,1}, Li-na Niu^{b,1}, Lisa K. Kemp^c, Cynthia K.Y. Yiu^d, Heonjune Ryou^e, Yi-pin Qi^f, John D. Blizzard^g, Sergey Nikonov^h, Martha G. Brackett^h, Regina L.W. Messer^h, Christine D. Wuⁱ, Jing Mao^a, L. Bryan Brister^c, Frederick A. Rueggeberg^h, Dwayne D. Arola^e, David H. Pashley^h, Franklin R. Tay^{h,*}

^a Tongji Hospital, Huazhong University of Science and Technology, Wuhan, People's Republic of China

^b Fourth Military Medical University, Xi'an, People's Republic of China

^c Mississippi Polymer Institute, The University of Southern Mississippi, MS, USA

^d The University of Hong Kong, Hong Kong SAR, People's Republic of China

^e University of Maryland Baltimore County, Baltimore, MD, USA

^f Sun Yat-sen University, Guangzhou, People's Republic of China

^g Quadsil Inc., Midland, MI, USA

^h Georgia Health Sciences University, Augusta, GA, USA

ⁱ University of Illinois at Chicago, Chicago, IL, USA

ARTICLE INFO

Article history:

Received 1 March 2012

Received in revised form 23 April 2012

Accepted 23 May 2012

Available online 29 May 2012

Keywords:

Quaternary ammonium
Organic modified silicate
Antimicrobial
Sol-gel technique
Self-repair

ABSTRACT

The design of antimicrobial polymers to address healthcare issues and minimize environmental problems is an important endeavor with both fundamental and practical implications. Quaternary ammonium silane-functionalized methacrylate (QAMS) represents an example of antimicrobial macromonomers synthesized by a sol-gel chemical route; these compounds possess flexible Si–O–Si bonds. In present work, a partially hydrolyzed QAMS co-polymerized with 2,2-[4(2-hydroxy 3-methacryloxypropoxy)-phenyl]propane is introduced. This methacrylate resin was shown to possess desirable mechanical properties with both a high degree of conversion and minimal polymerization shrinkage. The kill-on-contact microbiocidal activities of this resin were demonstrated using single-species biofilms of *Streptococcus mutans* (ATCC 36558), *Actinomyces naeslundii* (ATCC 12104) and *Candida albicans* (ATCC 90028). Improved mechanical properties after hydration provided the proof-of-concept that QAMS-incorporated resin exhibits self-repair potential via water-induced condensation of organic modified silicate (ormosil) phases within the polymerized resin matrix.

© 2012 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Antimicrobial polymers address increasing concerns from industrial, healthcare and consumer end-use sectors regarding infections and contamination [1]. The global demand for healthcare antimicrobial polymers was 15,500 metric tons in 2007 [2] and is forecast to reach 221,758 metric tons by 2017, with emphasis on technological advancements that are more eco-friendly and nontoxic to humans [3].

Polymeric biocides can be made by grafting an antimicrobial agent to their surfaces or blending with a nonleaching biocide

* Corresponding author. Address: School of Graduate Studies, Georgia Health Sciences University, Augusta, GA 30912-1129, USA. Tel.: +1 706 721 2033; fax: +1 706 721 6252.

E-mail address: ftay@georgiahealth.edu (F.R. Tay).

¹ These two authors contributed equally to this work.

[1]. Because of the presence of reactive silanol groups generated during hydrolysis, quaternary ammonium silanes can attach covalently to substrate surfaces via Si–O linkages to exert nonmigrating microbiocidal functions [4]. Due to its low toxicity, 3-(trimethoxysilyl)propyl dimethyloctadecyl ammonium chloride (SiQAC), an organofunctional trialkoxysilane, has been used in antimicrobial coatings of medical devices [5,6]. The antimicrobial activity of this compound is attributed to the long, lipophilic $-C_{18}H_{37}$ alkyl chain that penetrates bacterial cell membranes to produce leakage, autolysis and cell death of bacteria that come into direct contact [7]. The disadvantage of a surface attachment/grafting approach is that antimicrobial activity is lost after the surface layer is worn off. This disadvantage can be eliminated by incorporating SiQAC into the bulk resin rather than using it as a coating.

Although silane-based sol-gel chemistry has been used to prepare quaternary ammonium silane-based antimicrobial polymers, such processes do not result in macromonomers with polymerizable