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# NOVEL ANTI-DECAY SELF-SETTING PASTE OF HYDROXYAPATITE/COLLAGEN NANOCOMPOSITE UTILIZING GPTMS

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Key Words: Hydroxyapatite, collagen, bone-like nanocomposite, 3-glycidoxypropyl)methyldiethoxysilane, anti-decay, self-setting paste

Bone is a typical inorganic/organic nanocomposite mainly composed of hydroxyapatite (HAp) nanocrystals and collagen molecules. The composition and nanostructure is closely related to bone's biomechanical and biochemical properties. One of the most important things for bone is a bone remodeling process that maintains mechanical strength of bone to allow walking and running as well as homeostasis of calcium in our body. In fact, sintered HAp composed of HAp crystals approximately 1  $\mu\text{m}$  in particle size is considered as non-bioresorbable; however HAp nanocrystals easily resorbed by osteoclasts. A hydroxyapatite/collagen bone-like nanocomposite (HAp/Col) was successfully synthesized by the authors via self-organizing process. [1] The HAp/Col is incorporated into bone remodeling process completely as the same as the autologous bone transplanted and substituted with new bone in 3 months. Porous type HAp/Col was also developed by the authors and shows sponge-like viscoelasticity in wet condition. [2] The porous HAp/Col had been tested clinically and has been being sold in Japan from April, 2013 as ReFit<sup>®</sup>. According to the clinical test [3], the HAp/Col shows higher rate of remarkable efficiency in comparison to Osferion<sup>®</sup>, porous  $\beta$ -tricalcium phosphate, for substitution with newly formed bone. In addition, sponge-like deformability allows to fit irregular shape of bone defects as well as to press to pack the porous HAp/Col in cavity created by removal of benign bone tumor. On the other hand, recent need from surgeon for bone filler is a self-setting bone paste with bioresorbability. Previously, the HAp/Col anti-decay self-setting paste was prepared successfully with the use of sodium alginate and calcium compounds. It showed tolerant paste properties; however, the paste contained 30 % in mass of calcium compounds that could interfere excellent biological activity of the HAp/Col. In this study, the HAp/Col paste with injectability, self-setting and anti-decay abilities was prepared using the HAp/Col powder and 1 % in volume of 3-glycidoxypropyl)methyldiethoxysilane (GPTMS) aqueous solution. The paste obtained was evaluated by viscosity, hardening behavior and anti-decay property tests.

The HAp/Col was synthesized according to ref 1. The HAp/Col was compacted with specially designed mold by uniaxially press squeezing of water from the HAp/Col. The HAp/Col compact was then crushed and ball-milled to obtain the HAp/Col particles of 100  $\mu\text{m}$  or less in size. The GPTMS was dissolved in pure water at 1 % in volume and steadily placed in 25 °C for 1 h to allow hydrolysis of the GPTMS to form silanol groups. The HAp/Col powder (powder phase, P) and the GPTMS aqueous solution (liquid phase, L) were mixed at 0.20-2.00 of P/L ratio in  $\text{g}/\text{cm}^3$  to obtain a HAp/Col paste. A viscosity of the HAp/Col paste obtained was measured according to ref 1. Briefly, the paste obtained was shaped to cylinder at 5 mm in diameter and 5.1 mm in height and start pressing at 10 min after mixing by 2 kg weight for 10 min. A spread area of the paste was measured from digital photo with Image-J. Hardening behavior was measured using the viscosity test as a function of time, because softness of the paste did not allow to apply conventional the needle method. Anti-decay property was tested according to JIS T 0330-4:2012 [5]; briefly, the paste shaped in cylinder 4.8 mm in diameter and 16.5 mm in height was placed on wire mesh and, at 5 min after mixing, soaked in phosphate buffered saline for 72 h. Debris were then collected and measured their mass, and decay ratio was calculated as debris/original masses. The paste with P/L ratio of 0.2 could not shaped by its high fluidity and that of 2.0 could not shaped because of its aggregation. The paste with P/L ratio of 0.33 could injected through 18G needle and others could injected through syringe with 1.8 mm in inner diameter. The viscosity of the paste increased with increasing in P/L ratio and did not depend on the amounts of GPTMS. The initial hardening was observed first 30-40 min from mixing and gradually hardened. The paste with P/L ratio of 1.5 showed mechanical strength more than 1 MPa with viscoelastic property. No significant decay was observed for all pastes. The HAp/Col-GPTMS paste can be good candidate for high performance injectable bone filler as well as a raw material for 3D printing.

[1] M. Kikuchi *et al.*, *Biomater.*, 22, 1705-11, (2001); [2] M. Kikuchi, *et al.*, *Key Eng. Mater.*, 254-256, 561-4 (2004); [3] K. Shinomiya *et al.*, *Seikei-geka*, 63, 921-6, (2012), *in Japanese*; [4] K. Ishikawa, *et al.*, *J. Biomed. Mater.*, 36, 393-399 (1997). [5] JIS T 0330-4:2012, Characterization of calcium phosphate paste.