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Fabrication and antibacterial properties of hydroxyapatite-based housings of vibration humidifiers

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Housings of vibration humidifiers were fabricated using hydroxyapatite modified by different concentrations of polyethylene glycol, a plasticizer. The influence of the plasticizer concentration on the compressive and bending strengths of the housings was analysed. The highest compressive and bending strengths were 423.9 MPa and 620 MPa for samples containing 5 wt.% and 7 wt.% of plasticizer, respectively. The highest hardness was 480 HV for the sample containing 7 wt.% of plasticizer. After being submerged in the humidifier water for 7 days, an antibacterial test confirmed that the water in the humidifier containing a hydroxyapatite-based housing had antibacterial properties.

Key words: Hydroxyapatite, polyethylene glycol, antibacterial, vibration humidifier, mechanical strength.

Introduction

In South Korea, the housings for vibration humidifiers are fabricated by processing metals and polymers. However, during use, housings often undergo severe corrosion because they come in direct contact with water, which consequently exposes them to various bacteria and fungi. These microorganisms breed in the water stored in humidifiers and are released into the environment during humidification, thus spreading diseases and may ultimately lead to epidemics. Therefore, improper management of humidifiers can cause serious health consequences to humans, such as lung disease, due to the exposure and proliferation of germs such as bacteria [1-4]. Conventional methods to prevent this problem include thoroughly cleaning and chemically treating the humidifiers. However, these methods are inconvenient; moreover, chemical treatment has been discontinued because of its role in causing respiratory diseases. Therefore, it is imperative to use ceramics in the fabrication of housings for vibration humidifiers because they are non-corrosive and have anti-bacterial and anti-fungal properties. To achieve this objective, we investigated ceramic hydroxyapatite as a housing material for vibration humidifiers.

Hydroxyapatite is known for its excellent in vivo stability; its chemical formula, $Ca_5(PO_4)_3(OH)$, is similar to the molecular makeup of bone material [5-7]. Several reports have shown that hydroxyapatite adsorb bacteria and viruses [8-12]. However, despite being mechanically strong, hydroxyapatite ceramics are more difficult to process by techniques such as machining, forming, and heat treatment as compared to metals. In this study, we investigated the effect of adding a plasticizer on the processing, sinterability, and antimicrobial properties of hydroxyapatite.

Experimental Procedure

Samples, materials, and characterization

We synthesized a powder of hydroxyapatite using calcium carbonate ($CaCO_3$) and phosphoric acid (H_3PO_4) by a wet chemical method. The Ca : P ratio of the powder was fixed at 1.67. First, $CaCO_3$ was mixed with distilled water and stirred for 5 hrs. Then, H_3PO_4 was slowly added to the $CaCO_3$ solution and stirred for 12 hrs. After the chemical reaction, the solution was dried at 100 °C, and the resultant dry powder was sintered at 1350 °C. The structure of the sintered powder was analysed using X-ray diffraction (XRD, MP-XRD). The plasticizer polyethylene glycol 400 (PEG-400, $H(OCH_2CH_2)_nOH$) was added to hydroxyapatite. The plasticizer concentration in hydroxyapatite was varied between 1 and 9 wt.%. The housing of the vibration humidifier was produced by pressing. For sintering, the pressed housing was heated to 1350 °C at a rate of 5 °C/min for 1 hr. The sintered plasticizer was then cooled to the ambient temperature at a rate of 4 °C/min.

Measurement of compressive strength

In order to measure the compressive strength, 1, 3, 5, 7, and 9 wt.% of plasticizer was added to hydroxyapatite. The composite samples were moulded into HAp cylinders of 10 mm diameter. Then, the samples were pressed under a pressure of 1 ton/cm^2 for 20 sec. The pressed samples were then heated to

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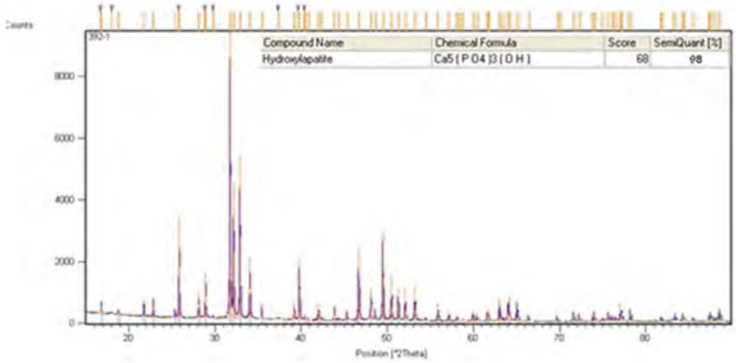

Fig. 1. XRD pattern of sintered hydroxyapatite.

1350 °C at a heating rate of 5 °C/min and sintered for 1 hr.

Measurement of bending strength

Again, 1, 3, 5, 7, and 9 wt.% of the plasticizer was added to hydroxyapatite. Now, the composite samples were moulded into HAp plates of dimension 4.70 mm × 25.0 mm and were subjected to a pressure of 1.51 ton/cm^2 for 20 sec. Next, the pressed samples were heated to 1350 °C at a heating rate of 5 °C/min and sintered for 1 hr.

Measurement of Hardness

To measure hardness, 1, 3, 5, 7, and 9 wt.% of the plasticizer was added to hydroxyapatite and the composite samples were moulded into HAp plates of dimension 4.7 mm × 25 mm. Then, the samples were consecutively polished with 800 grid, 1200 grid, and 2400 grid sanding discs, as well as with alumina powder having an average particle size of 1.00 µm. Next, they were washed in a sonicator and dried at 100 °C before carrying out the hardness test.

Antibacterial test

The sintered hydroxyapatite was moulded into doughnut-shaped samples. The samples were placed in a humidifier containing city water. The humidifier was used for 7 days. After 3 d, the water in the humidifier was collected for analysis. Standard methods Agar was cast on Petri plates evenly inoculated with city water. Each plate was incubated at 30 °C for 48 hrs, before counting the amount of bacteria.

Results and Discussion

Characterization of hydroxyapatite

The structure of the hydroxyapatite powder used in this study was determined using XRD data, as shown in Fig. 1. The peaks representing hydroxyapatite were found at the scattering angles (2θ) of 31.8 °, 32.27 °, and 32.92 °. The degree of crystallinity was determined as 98%.

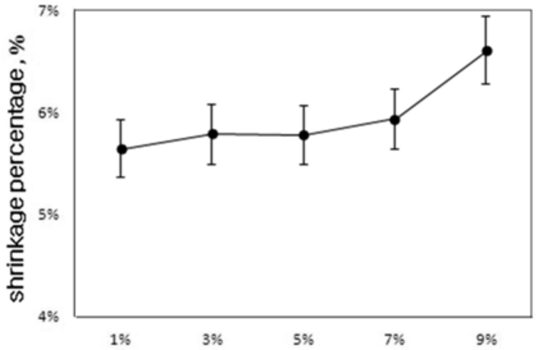

Fig. 2. Shrinkage versus amount of plasticizer.

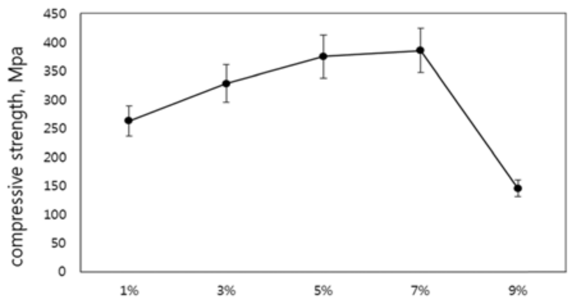

Fig. 3. Compressive strength versus amount of plasticizer.

Shrinkage due to plasticizer

Fig. 2 shows the dependence of the shrinkage of hydroxyapatite housing on the amount of plasticizer added. The percentage shrinkage of the samples varied between 5.62% and 6.59%. The shrinkage increased only slightly with increasing the amount of plasticizer. This result confirms the use of plasticizer to facilitate the heat treatment of hydroxyapatite mouldings.

Mechanical properties

As shown in Fig. 2, compressive strength differed for each sample;the highest compressive strength of 385.6 MPa was measured for the sample containing 7 wt.% of plasticizer. As shown in Fig. 3, the bending strength did not exhibit much change with the amount

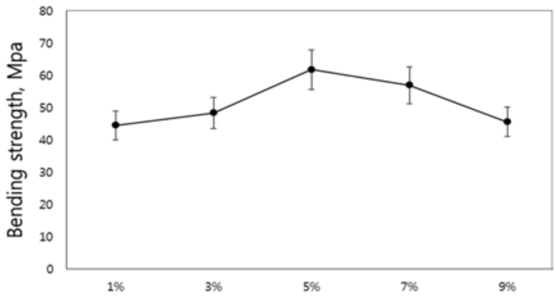

Fig. 4. Bending strength versus amount of plasticizer.

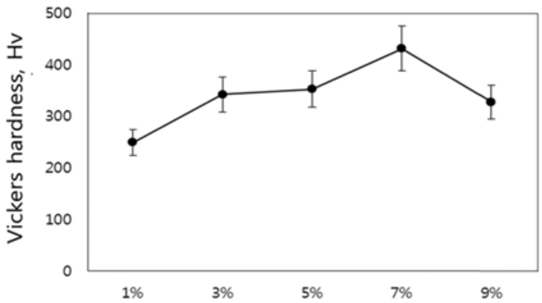

Fig. 5. Vickers hardness versus amount of plasticizer.

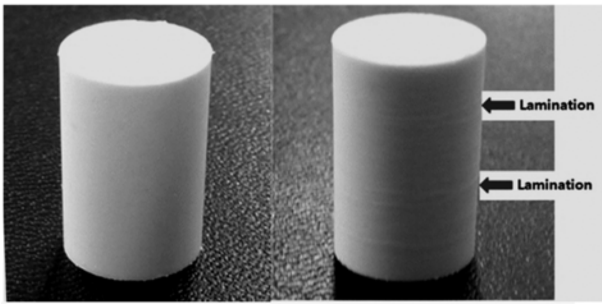

Fig. 6. Hydroxyapatite sample with 7 wt.% plasticizer (left-hand-side image) and 9 wt.% plasticizer (right-hand-side image).

of plasticizer; however, in the sample containing 5 wt.% of plasticizer, a highest bending strength of 61.8 MPa was measured. As shown in Fig. 4, the highest hardness was 430.6 HV for the sample containing 7 wt.% of plasticizer. With an increase in the amount of the plasticizer added, the compressive strength, bending strength, and hardness also increased proportionately. However, at 9 wt.% of plasticizer, a decrease in mechanical property was observed. Therefore, the ideal amount of plasticizer is between 5 and 7 wt.% (see Fig. 5). If more than 9% of the amount of the plasticizer, laminationsare produced because of excessive shrinkage of the sintered hydroxyapatite, which weakens the mechanical properties.

Antibacterial test

The samples containing plasticizer in the range 5-7 wt.% were moulded in a doughnut-shaped structure (internal diameter: 18.70 mm;external diameter: 37. 10 mm) and placed in transducer housing mounted on

Fig. 7. Standard housing (left-hand-side image) and hydroxyapatite housing (right-hand-side image).

Fig. 8. Results of antibacterial test (after 3 d).

an existing humidifier, which was used for 7 d. The antibacterial test was conducted using water that remained in the humidifier after 7 d of operation. The pH of this remaining water was 9.8, confirming that it was alkaline and bacterial growth did not occur. As a result, we believe that housings of vibration humidifiers fabricated with hydroxyapatite show antimicrobial activity.

Conclusion

Hydroxyapatite powder modified with different concentrations of polyethylene glycol plasticizer was used to fabricate housings of vibration humidifiers. The influence of the amount of plasticizer on the mechanical properties of the housing was investigated in order to prepare the optimum housing. The highest compressive strength, bending strength, and hardness were measured for the hydroxyapatite sample containing 7, 5, and 7 wt.% of plasticizer, respectively. On increasing the amount of plasticizer, the mechanical properties of the housing improved. However, at 9 wt.% of plasticizer, a decrease in the mechanical properties was observed because of the formation of laminations. Housing treated by heating was prepared with hydroxyapatite powder and plasticizer. After 3 d of being submerged in a vibrator containing city water, the hydroxyapatite samples imparted antibacterial properties to the water, irrespective of the amount of plasticizer added. Therefore, the housing containing 5-7 wt.% of plasticizer shows the optimum combination of antibacterial effects and mechanical properties for vibration humidifiers.

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