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# Fabrication of Bioactive Organic Polymer by Using Apatite Nuclei-Contained Inorganic Binder

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**Keywords:** Apatite nucleus, Inorganic binder, Organic polymer, Bioactivity, Adhesive strength.

**Abstract.** Apatite nuclei were dispersed in inorganic binder and the apatite nuclei-contained inorganic binder was coated on polyethyleneterephthalate plate. Hydroxyapatite was induced by the apatite nuclei dispersed in the binder and hydroxyapatite layer was formed on the surface of the substrate by soaking in SBF. The hydroxyapatite layer showed high adhesive strength to the substrate.

## Introduction

Organic polymers are easily processed in various shapes and combined with various materials. If organic polymers acquire bioactivity, development of hard and soft tissue implants possessing various mechanical properties with high bioactivity and bioaffinity is expected.

When pH of simulated body fluid (SBF) with ion concentrations similar to those of human blood plasma is raised, fine particles of calcium phosphate are precipitated. The fine calcium phosphate particle is very effective for forming hydroxyapatite from SBF and we named it apatite nucleus [1].

In the previous studies, we precipitated apatite nuclei in the pores of porous polyethylene by using SBF [2-4] and we deposited apatite nuclei in the pores of porous polyethylene by electrophoretic deposition [5]. Either composite showed high bioactivity and the formed hydroxyapatite showed high adhesive strength to the composite.

In the present study, apatite nuclei were mixed with an inorganic binder named Binder B-10 produced by JAPAN NANO COAT CO.,LTD, Japan and the apatite nuclei-contained inorganic binder was coated on polyethyleneterephthalate (PET) plate. Bioactivity was investigated by soaking in SBF. The adhesive strength between the formed hydroxyapatite and the PET plate was also measured.

## Materials and Methods

**Preparation of Bioactive Organic Polymer by Using Apatite Nuclei-Contained Inorganic Binder.** We prepared SBF [6,7] by dissolving reagent-grade NaCl, $NaHCO_3$, KCl, $K_2HPO_4 \cdot 3H_2O$, $MgCl_2 \cdot 6H_2O$, $CaCl_2$, and $Na_2SO_4$ in ultrapure water with the composition as shown in Table 1 in ultrapure water. The pH of SBF was raised to pH 8.5 by using trishydroxymethylaminomethane at 25.0 °C, and precipitated apatite nuclei in the SBF, which were collected by filtration using a 50 nm polytetrafluoroethylene membrane filter and washed with distilled water. The apatite nuclei were dispersed in ethanol with ultrasonic vibration. The apatite nuclei-dispersed ethanol was mixed with the above mentioned Binder B-10. Thus obtained apatite nuclei-contained inorganic binder was coated on the PET plate ($10 \times 10 \times 1$ mm$^3$) and dried at room temperature.

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Table 1. Ion concentrations of simulated body fluid and human blood plasma

| | Ion Concentrations [mmol·dm$^{-3}$] | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $Na^+$ | $K^+$ | $Mg^{2+}$ | $Ca^{2+}$ | $Cl^-$ | $HCO_3^-$ | $HPO_4^{2-}$ | $SO_4^{2-}$ |
| Human blood plasma | 142.0 | 5.0 | 2.5 | 1.5 | 103.0 | 27.0 | 1.0 | 0.5 |
| SBF | 142.0 | 5.0 | 2.5 | 1.5 | 147.8 | 4.2 | 1.0 | 0.5 |

**Test of Bioactivity.** Bioactivity of the substrate was evaluated by soaking in SBF at pH 7.4 at 36.5 °C for up to 14 d. The surface of the substrate was analyzed by thin film X-ray diffraction (TF-XRD; Model Rint 2500, Rigaku, Japan), scanning electron microscopy (SEM; Model ESEM-2700, Nicon, Japan) and energy dispersive X-ray analysis (EDX; Model DX-4, EDAX International, USA)

**Adhesive Strength Measurement.** The adhesive strength between the substrate and formed hydroxyapatite layer was measured by a modified ASTM C-633 method [8]. Both sides of the substrate were attached to SUS jigs (10×10 mm$^2$) by Araldite® glue and tensile load was applied with universal testing machine (Model AGS-H Autograph, Shimadzu, Japan) until fracture occurred.

## Results and Discussion

**PET Plate after the Coat of Apatite Nuclei-Contained Inorganic Binder.** In Fig. 1 (a), SEM micrograph of the surface of the PET substrate and in Fig. 1 (b), EDX profile of the surface of the PET substrate is shown. In Fig. 2 (a), SEM micrograph of the surface of the substrate after the coat of the apatite nuclei contained-inorganic binder is shown. Many cracks of the binder were observed in the SEM micrograph. In Fig. 2 (b), EDX profile of the surface of the substrate after the coat of the apatite nuclei contained-inorganic binder is shown. A peak of Si, constituent of Binder B-10, was detected.

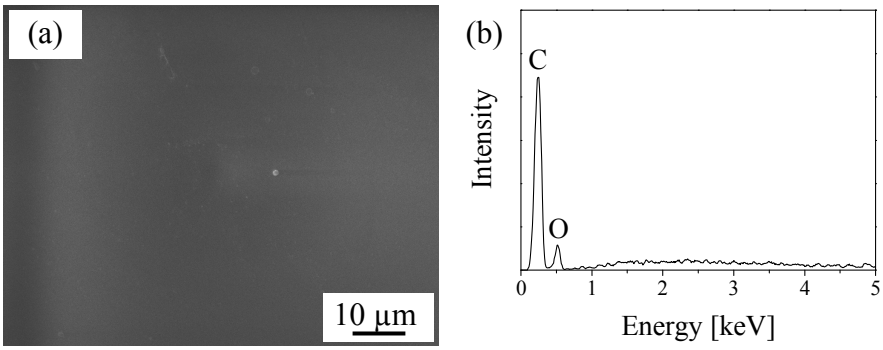


Fig. 1 (a) SEM micrograph and (b) EDX profile of the surface of the PET substrate.

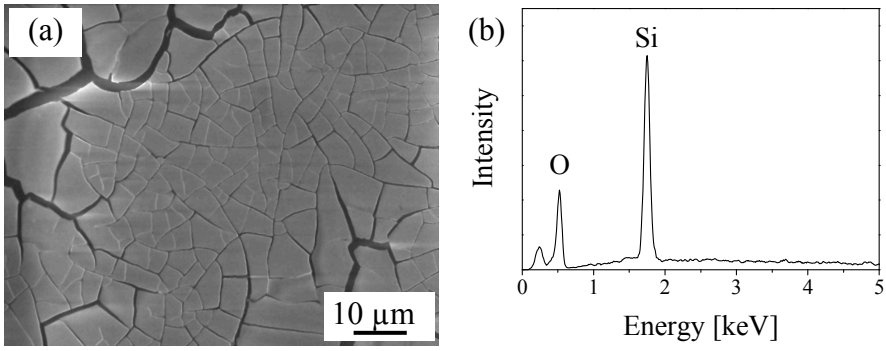


Fig. 2 (a) SEM micrograph and (b) EDX profile of the surface of the substrate after the coat of the apatite nuclei-contained inorganic binder.

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**Bioactivity of the Binder Coated PET Plate.**

**TF-XRD Measurement.** Fig. 3 shows the TF-XRD profiles of the surface of the binder coated PET substrate, the substrate after the soak in SBF for 3 d and for 7 d. After the soak for 3 d, a diffraction peak of hydroxyapatite was detected, and after the soak for 7 d, two diffraction peaks of hydroxyapatite were detected. This result indicates that apatite nuclei dispersed in the binder induced hydroxyapatite within 3 d.

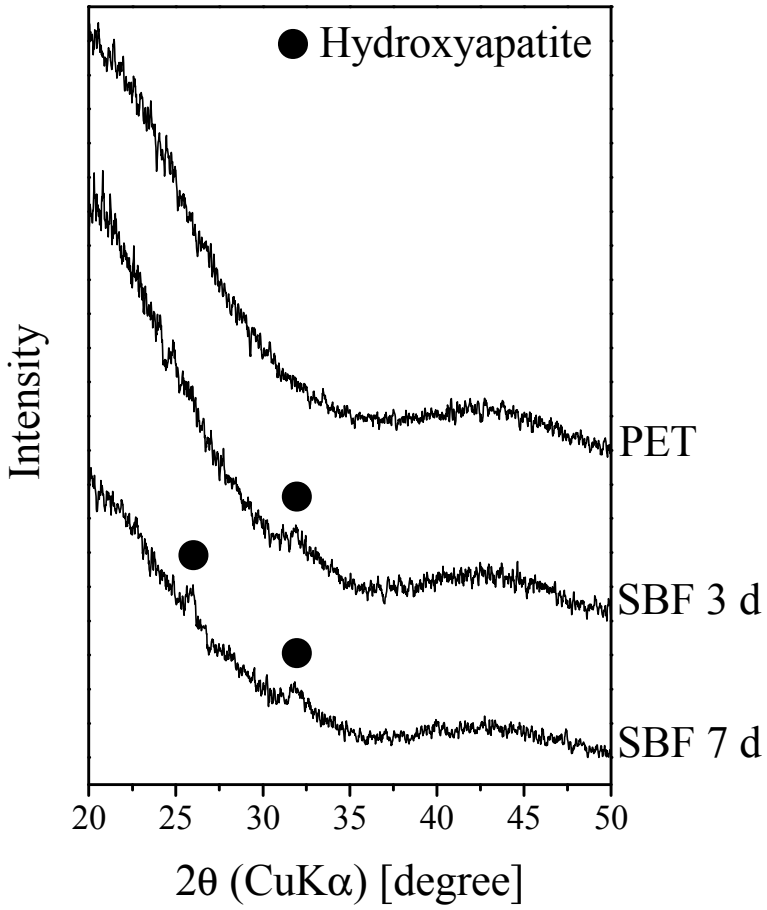


Fig. 3 TF-XRD profiles of the surface of the PET substrate, the substrate after the soak in SBF for 3 d and for 7 d.

**SEM Observation and EDX Analysis.** In Fig. 4 (a), SEM micrograph of the surface of the substrate after the soak in SBF for 3 d is shown. Hydroxyapatite was observed on the whole surface of the substrate. In Fig. 4 (b), SEM micrograph of high magnification is shown. Needle like crystals characteristic to hydroxyapatite were observed. In Fig. 4 (c), EDX profile of the surface of the substrate after the soak in SBF for 3 d is shown. Peaks of Ca and P, constituents of hydroxyapatite, were detected. These results indicate that apatite nuclei dispersed in the binder induced hydroxyapatite on the whole surface of the substrate within 3 d. In Fig. 5 (a) and (b), SEM micrograph and EDX profile of the surface of the substrate after the soak in SBF for 7 d is shown. After the soak for 7 d, hydroxyapatite grew and covered whole surface of the substrate.

**Adhesive Strength of Hydroxyapatite Layer.** The adhesive strength between the formed hydroxyapatite layer and the substrate after the soak in SBF for 14 d was 6.5 ± 1.1 MPa for 5 samples. By EDX analysis, a peak of Si, constituent of the binder, was detected on the fractured surface of hydroxyapatite layer side and not detected on that of the substrate side. It is considered that the above adhesive strength is due to that of the binder to the PET substrate.

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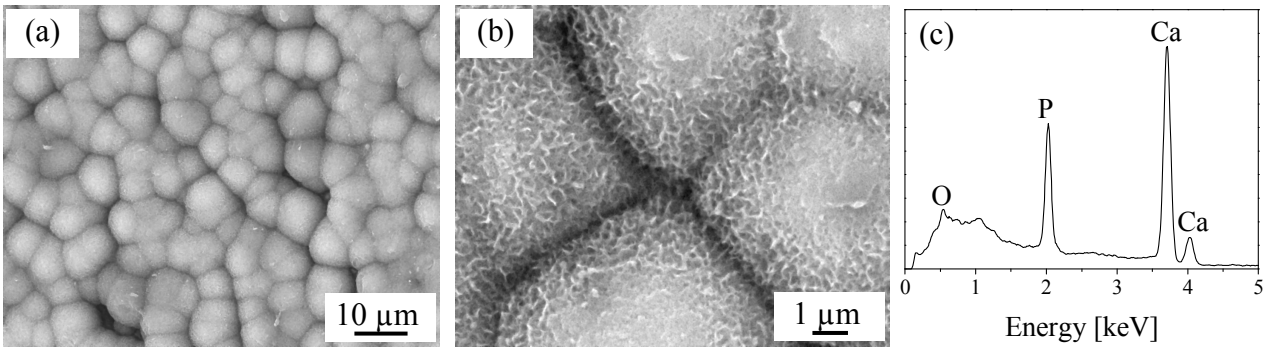


Fig. 4 (a) SEM micrograph, (b) high magnification of (a), and (c) EDX profile of the surface of the substrate after the soak in SBF for 3 d.

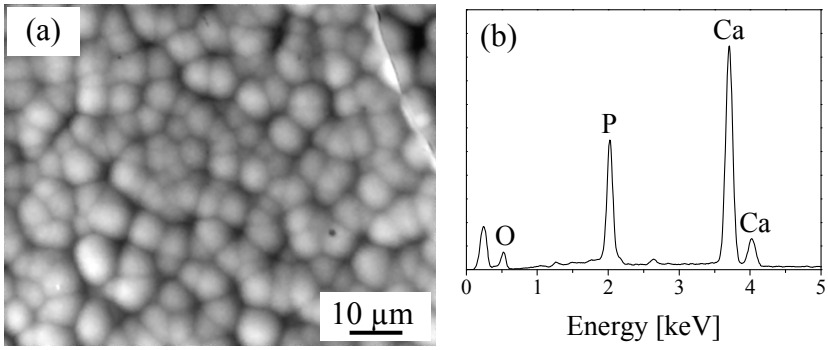


Fig. 5 (a) SEM micrograph and (b) EDX profile of the surface of the substrate after the soak in SBF for 7 d.

**Summary**

Bioactive Organic Polymer was fabricated by coating apatite nuclei-contained inorganic binder on PET plate. High bioactivity of this material was confirmed by soaking in SBF. High adhesive strength between the formed hydroxyapatite and the substrate was obtained. This material is promising excellent implant with high bioactivity and bioaffinity.

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