Effect of forming conditions of poly-lactic acid/hydroxyapatite to tensile strength of canine bone fixation plate using full factorial experimental design

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Abstract. Problems of using metallic bone fixation plates for canine lead to the idea of the replacement with biocomposite materials. In this research, polylactic acid (PLA) blends with hydroxyapatite (HA) powder 5-15% was used as biocomposite material. The specimens were formed by hot compression molding and the experiment in investigating suitable forming condition was based on the 2^3 full factorials with the center point design of experiment. There are many types of research attempting to develop a prototype of bone fixation plates from a substitute material including forming conditions, in order to obtain the effective bone fixation plate to use. This paper presents the tensile strength for the proposed biocomposite bone fixation plates. After the mechanical testing on various conditions, the tensile strength results ranged from 40 to 60 MPa and it showed that the higher HA ratio had significant effect to the decrease of the tensile strength. This preliminary experiment reveals the tensile strength of forming conditions of PLA/HA composite, which may indicate a direction for improving the better mechanical properties of the bone fixation plates.

1 Introduction

The metallic implants have been widely used in orthopedic and dental applications. The advantages of these materials such as good mechanical strength, easy forming and biocompatibility. However, many problems such as like stress shielding or stress protection atrophy, ionic release and allergic or toxic reactions have been introduce [1]. These risks related to the use of implants have led to the requirement of an additional operation to remove the implant after fracture healing [2]. To overcome these problems such as using noble metals coated with bioinert or bioactive materials. Because of these disadvantages, research into other materials became very significant. These disadvantages show obviously a need for non-metallic biodegradable devices for osteosynthesis of sufficient strength during the healing period and without causing side effects [3]. These would take away the necessity for an unpopular second operation to remove plates and screws used for internal fixation of fractures or after orthognathic surgery [4]. Furthermore, there is also significant importance in developing biodegradable replacements for metallic implants to eliminate the requisite to remove the implant after the bone has healed. Human bone is a highly ordered and complex structure consisted in organic part is hydroxyapatite (HA) along with collagen type II. Properties of HA are bioactive and good mechanical properties using for coated-metallic implant and bone repair [5]. Nevertheless, mechanical properties of HA is insufficient load bearing applications. Hence, synthetic, as well as some natural polymers have been being studied as appropriate materials for biomimicking the fibrous component in natural bone. There is a very short list of synthetic biodegradable polymers that are components in medical device implants. Polylactic acid (PLA) was the primary reported resorbable polymer to be used for fracture fixation [7]. Currently, PLA is still subjects of ongoing research into the development of biodegradable orthopedic biomaterials.

In a current animal study, PLA/HA plate for fixations of mandibular fractures was tested in comparison with stainless-steel fixation plate. These fixation plates were used to stabilize transverse mandible shaft fractures in canine.

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Therefore, this work aims to investigate and identify appropriate forming conditions for a PLA/HA composite using the experimental design based on the full factorial method. The optimal conditions were evaluated based on tensile strength. Finally, the actual mechanical properties of the composite specimen were used to apply to the forming composite bone fixation plate for the future.

2 Materials and methods

2.1 Material

2.1.1 Preparation of poly-L-lactic acid (PLA)

PLA was synthesized from lactic acid monomer through ring opening polymerization at 120 °C for 72 h using Tn (II). butoxide 0.01% mol per 500 g lactic acid to PLA powders (Molecular weight = 100,000–150,000 and viscosity = 2.17 ± 0.059 cP)[6, 8, 9].

2.1.2 Preparation of hydroxyapatite (HA)

HA powder prepared from removing tissue from the bovine bone and boiled the bones. Then, soaked the bones in oxidizing solution for 72 h and calcined at 950 °C for 12 h [8]. Finally, milling and sieving to be powders [6].

2.1.3 Preparation of biocomposite material

Biocomposite materials can be prepared from mixing the poly-L-lactic acid (PLA) and hydroxyapatite (HA) 5-15% of PLA using alumina balls to shaker about 15 min in the cylindrical Teflon mold to homogeneous material [10].

2.2 Specimens forming

Add biocomposite materials to hot compression mold to form the specimens using a custom-made mold. The applied molding temperature at 140-160 °C [11], molding pressure at 3-7 MPa and set holding time for 30 second [12]. every condition. Then, specimens are tested mechanical properties and statistical.

2.3 The experimental design and optimization

The experimental design for optimization study compression conditions of biocomposite materials according to 2^3 full-factorial design with the center point using Minitab16 software. The three factors include HA ratio, molding temperature and molding pressure as shown in table 1. Total of 20 experimental consists of 8 factorial designs, 2 center points and all of the design experiments were 2 replicates. The experimental design considers responses as tensile strength and bending strength of specimens.

2.4 Mechanical testing

The specimens are tested tensile strength from ASTM D 368 specimens by the universal testing machine (Instron5566). The cross-head speed was set at 0.1 mm/min.

Table 1. Parameters for experimental design.

Factors	Variation Levels		
	Low (-1)	Medium (0)	High (+1)
HA (%)	5	10	15
Molding Temperature (◦C)	140	150	160
Molding Pressure (MPa)	3	5	7

3 Results and discussion

3.1 Microstructural characterization

3.1.1 Particle size and x-ray diffraction (XRD) analysis of materials

The particle size analysis of PLA and HA powders were characterized by laser diffraction method in ethanol. The mean of particle size of PLA and HA were 231.29 μm and 8.11 μm, respectively. The HA powder was verified by x-ray diffraction analysis because the synthesized HA must be confirmed their chemical composition with JCPDS-ICDD No.09-0432, the comparison of synthesis HA and standard HA composition pattern show in Fig. 1.

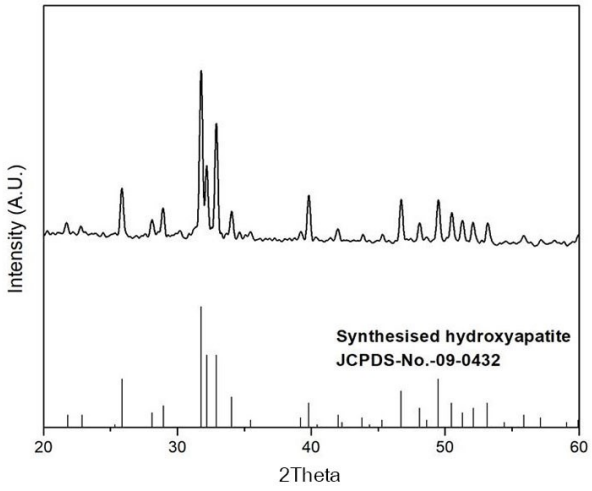

Fig. 1. XRD pattern of synthesized HA powder.

3.1.2 Differential scanning calorimetry (DSC) analysis of PLA

From the DSC curves of PLA showed the glass transition temperature (Tg), onset, endset and peak of endothermic temperature were 55 ◦C, 145 ◦C, 160 ◦C and 152 ◦C respectively. Because the DSC information describes the thermal behavior of matrix material, these data lead to control the range of forming temperature in experimental at 140-160 ◦C.

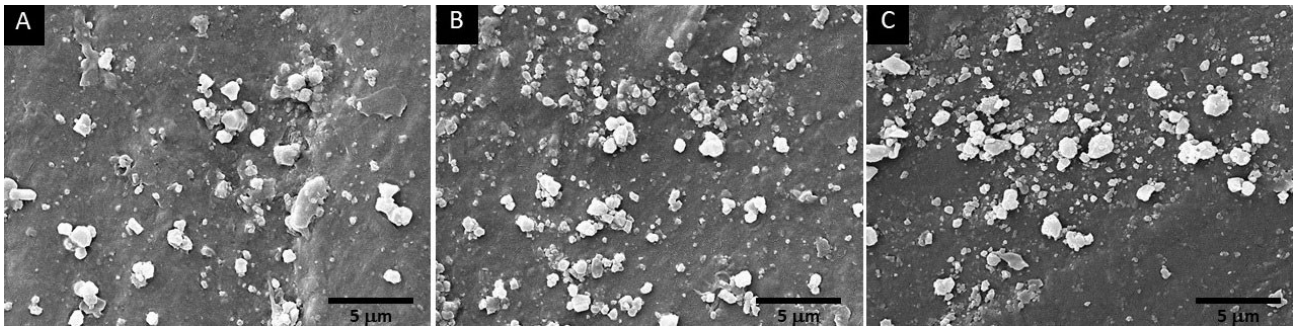

Fig. 2. SEM images of biocomposite material at 5 (A), 10 (B) and 15 wt% HA (C).

3.1.3 Scanning electron micrographs (SEM) of biocomposite material

The Scanning electron micrograph of biocomposite material at 5000x magnification for 5-10 wt% HA shows in Fig. 2. It shows the HA particle distribution was found to be successfully integrated into PLA matrix.

3.2 Mechanical properties

The results of mechanical testing for different molding condition specimens. All three factors are HA ratio, molding temperature and molding pressure. The values obtained from the mechanical tested statistically analyzed using Minitb16 software. Table 2 shows the R-Sq and R-Sq (adj) in the experiment are 88.03% and 79.32%, respectively. In addition, it presents the HA ratio had a significant effect on tensile strength because p-value of HA ratio is less than 0.05. The highest tensile strength of specimen was found at 5 wt% HA for molding temperature and molding pressure were 160 ◦C and 7 MPa, respectively. The tensile strength of various ratios of PLA/HA are varied from 40-60 MPa. Hence, this tensile strength value could be compared with cortical bone that shows in table 3. The result in table 4 shows the tensile strength dropped when HA particle contains higher than 5 wt% HA. Probably more loading HA particle that effect to ductility of PLA matrix. Consequently, brittle of the composite with 10 and 15 wt% HA will be increased and decreased tensile strength as well. Nevertheless, both of molding temperature and molding pressure had no significant effect to tensile strength. Due to molding temperature was not exceed the melting point of PLA, there, the micro structure of PLA matrix was not difference with similar cooling rate. In addition, the molding pressure used in the experimental at each level are not sufficiently different to affect to tensile strength because of the limitation of molding machine. Finally, Fig. 3 shows that the residual plots for tensile strength to indicating normally distribution data and normal probability plot, it can be predicted the trend of data to occur.

Table 2. The estimated effects and coefficients tensile strength.

Term	Coef	SE Coef	T	P
Constant	49.443	0.6670	74.12	0.000
HA ratio	-5.469	0.6670	-8.20	0.000
Temperature	1.394	0.6670	2.09	0.061
Pressure	1.334	0.6670	2.00	0.071
HA ratio*Temp	-0.745	0.6670	-1.12	0.288
HA ratio*Pressure	-0.330	0.6670	-0.49	0.631
Temp*Pressure	0.650	0.6670	0.97	0.351
HA *Temp*Pressure	-1.111	0.6670	-1.67	0.124
Ct Pt	-0.408	1.4916	-0.27	0.790
R-Sq = 88.03%		R-Sq (adj) = 79.32%		

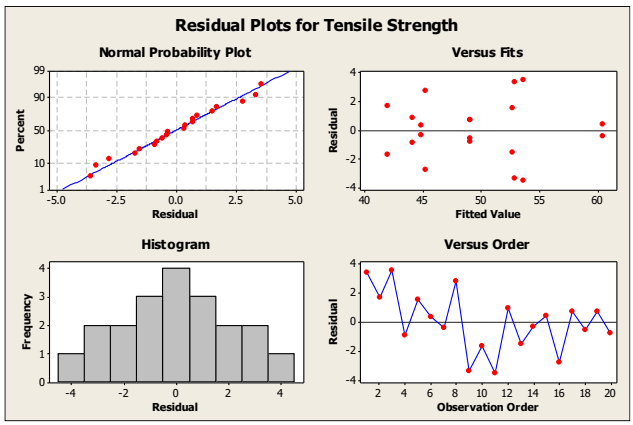

Fig. 3. The residual plots for tensile strength.

Table 3. The tensile strength of cortical bone and materials.

	Tensile Strength (MPa)
Cortical Bone (transverse direction)	52 [13]
Stainless Steel (316L)	586 [13]
Polylactic Acid	50 [14]
Hydroxyapatite	40 [15]

4 Conclusions

The study of the forming conditions of composite PLA/HA by hot compression molding using 2^3 full-factorial design with the center point. All factor in this study consist of the HA ratio, molding temperature, and molding pressure, but the HA ratio only had a significant effect on tensile strength. The range of tensile strength at 5-15 wt% HA about 40-60 MPa on average but, this

value decreases when increasing of HA ratio. The highest tensile strength is using 5 wt% HA for molding temperature at 160 ◦C and molding pressure at 7 MPa, this condition gives the tensile strength about 60 MPa. For future work, the mechanical values are input into the finite element analysis to determine the probability of actual use on transverse mandible shaft fractures in canine.

Table 4. Tensile strength of the specimens.

Orders	HA ratio (wt%)	Temperature (◦C)	Pressure (MPa)	Tensile Strength (MPa)
1	5	140	3	56.23
2	15	140	3	43.57
3	5	160	3	57.19
4	15	160	3	43.18
5	5	140	7	54.20
6	15	140	7	45.11
7	5	160	7	60.09
8	15	160	7	47.97
9	5	140	3	49.51
10	15	140	3	40.15
11	5	160	3	50.06
12	15	160	3	44.98
13	5	140	7	51.15
14	15	140	7	44.47
15	5	160	7	60.86
16	15	160	7	42.36
17	10	150	5	49.73
18	10	150	5	48.45
19	10	150	5	49.73
20	10	150	5	48.23

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