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Research Article

A Novel Bone Cement: Approaching to Human Bone Mechanical Properties by Using LHDs

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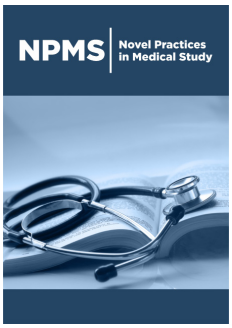

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Abstract

A composite based on three components, including PMMA (Polymethyl Methacrylate), HA (Hydroxyl Apatite), and LDH (Layered Double Hydroxide), was prepared, characterized, and applied as bone cement. Bone cement is a biomaterial used for hip and knee joint replacements in clinical applications. Four types of bone cements are used in dentistry and orthopedics. Including a) acrylic cements based on PMMA and b) PPF (Polypropyl Fumarate) cements. For decades, polymerized methacrylates have played a pivotal role in the construction of denture bases, serving as a reliable and widely used material in dentistry. PMMA polymer is synthesized by free radical polymerization. LDH with the general formula of $[M(II)_{1-x}M(III)_x(OH)_2]^{x+}.(A^{n-})_{X/n}\cdot mH_2O$ has M(II) and M(III) cations and A^{n-} inter-layer anion. The importance of LDH is for its planar layered structure. HA is a thermodynamically stable calcium phosphate similar to the human hard tissues in morphology and composition. $[M(II)_{1-x}M(III)_x(OH)_2]^{x+}.(A^{n-})_{X/n}\cdot mH_2O$ has M(II) and M(III) cations and an A^{n-} inter-layer anion. The importance of LDH is its planar, layered structure. HA is a thermodynamically stable calcium phosphate similar in morphology and composition to human hard tissues. In this work, we made a green Zr-Ni LDH and combined it with PMMA and HA in a specific ratio. The composite is characterized using XRD, FTIR, and SEM analyses.

Keywords: PMMA; HA; LDH; Bone cement; Mechanical properties

Introduction

Poly Methyl Methacrylate (PMMA) has received significant attention in recent years and is regarded as one of the most efficient and promising polymers, with applications in diverse fields [1]. Polymerized methacrylates have been commonly used to make denture bases for many decades. PMMA polymer is synthesized by free radical polymerization and exhibits low batch variation compared to other natural or raw polymers. Due to its easy maintenance and low manufacturing costs, PMMA has been used in numerous applications [1]. Prior to the use of PMMA as a biomaterial, there was a lot of chemical research done. According to legend, the discovery of "acide acrylique" occurred in 1843. This is derived from 'acreolan', the latin word for vinegary, acid or acrid and refers to the penetrating smell of the monomer. It was discovered in 1936 that combining finely ground PMMA powder with a liquid monomer produced a doughy substance. The PMMA monomer's partial dissolution is to blame for this. Polymer chains from the PMMA become available for free radical polymerization, and entanglements of these chains with newly formed chains lead to an intimate connection between the newly formed chains [2,3]Jeremy M</author><author>Ahn, Dongchan</author><author>Eldred, Donald V</author><author>Fielitz, Alyssa J</author><author>Heyl, Tyler R</author><author>Lee, Myoungbae</author><author>Mangold, Shane</author><author>Pearce, Eric Z</author><author>Reinhardt, Carl W</author><author>Roggenbuck, Cheryl %J Macromolecules</author></authors></contributors><titles><title>Photocured Simultaneous and Sequential PDMS/PMMA Interpenetrating Polymer Networks</title></titles><pages>5826-5839</pages><volume>55</volume><number>13</number><dates><year>2022</year></dates><isbn>0024-9297</isbn><urls></urls></record></Cite></EndNote>.

Layered Double Hydroxides (LDH), which are one type of layered materials and are also known as anionic clays, are promising layered materials due to some of their interesting

properties, such as ease of synthesis, unique structure, uniform distribution of different metal cations in the brucite layer, surface hydroxyl groups, flexible tunability, intercalated anions with interlayer spaces, swelling properties, oxo-bridged linkage, high chemical and thermal stability, and ability to intercalate different types of anions [4,5]. The general formula for these LDHs is $(Mg_{1-x}Al_x(OH)_2)^{x+}(A-)^{x-}nH_2O$, where "A" represents the interlamellar anion that restores the electroneutrality of the compound. We call these the II-III LDHs. We recently reported that the preparation of Zn-Ti LDH and Co-Ti LDH consisting of di- and tetra-valent cations is possible [6,7]. The present work examines the possibility of preparing another example consisting of bi- and tetra-valent cations, LDH, by a mechanochemical route that has been known as a green method.

Experimental Methods

Synthesis of Ni-Zr LDH in a mortar

Ni-Zr LDH were prepared by a mechanochemical route with a simple and green mechanical grinding method [8]. NaOH pellets (4.5g, 112.5mmol) were added to a powder mixture of nickel (II) nitrate hexahydrate (9.16g, 31.5mmol) and zirconium nitrate (5.79g, 13.5mmol) and manually ground to a paste. The paste was washed four times with deionized water (20mL), dried under vacuum at 40 °C, powdered, and analyzed [9].

Synthesis of three-component composite

In this case samples were prepared with 3%(w/w) of hydroxyapatite and 0,1,3,5,7,10%(w/w) LDH. In the first step hydroxyapatite, PMMA and LDH powder were thoroughly combined at the designated weight percentage. The MMA liquid was then added to the mixture. The mixture was kept in a vacuum state for 30 seconds to let gases escape. To prepare composie tablets, the mixture was added to the cast and waited for it to become hard.

Result and Discussion

Ni/Zr LDH characterization

Figure 1 shows XRD pattern which indicates that the Ni-Zr LDH was directly synthesized. Two sharp peaks in different 2θ 10.31, 20.33, 34.02, 38.08, 44.80, 60.49 and 61.50 were referred to (003), (006), (012), (015), (018), (110) and (113) plates, respectively [10]. The FTIR spectrum of Ni-Zr LDH was shown in Figure 2. The widespread and intense band in the 3446cm^{-1} area is due to the stretch vibrations of O-H groups present in the interlayer and water molecules, which are in layers. 1625cm^{-1} is related to water bending vibrations. The sharp bands in 1379cm^{-1} and 827cm^{-1} are related to stretch and bending vibrations of interlayer nitrate anion, respectively. However, the band seen in 1357cm^{-1} is related to CO_3^{2-} which is caused by existing CO_2 in deionized water [6,8]. Figure 3 shows the infrared spectrum of the virgin sample; the C=O stretching vibration of the ester group appears around 1725cm^{-1}, the two doublet bands at (1140, 1190cm^{-1}) and (1240, 1265cm^{-1}) correspond to the C-O stretching vibrations of ester groups. The absorptions around 1440 and 1480cm^{-1} characterize, respectively, the asymmetric bending vibrations of the (C-CH_3) and (C–CH_2) bonds [11].

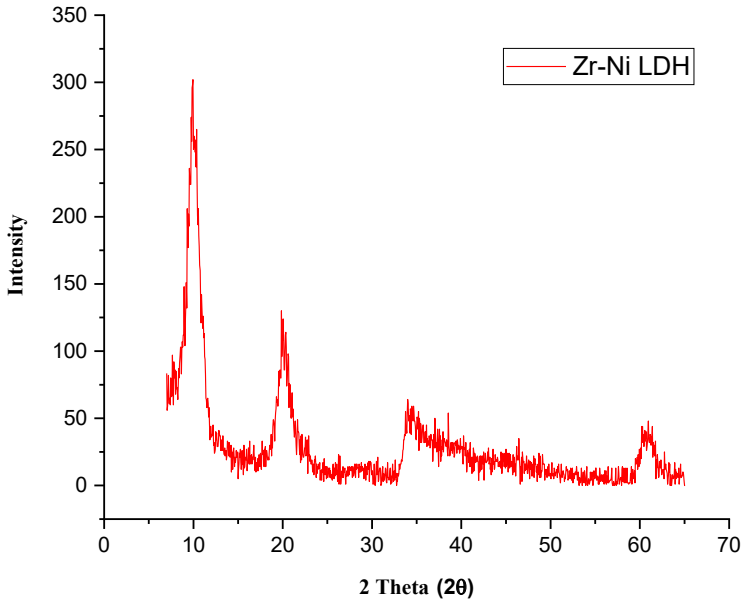

Figure 1: The XRD pattern of Ni-Zr LDH.

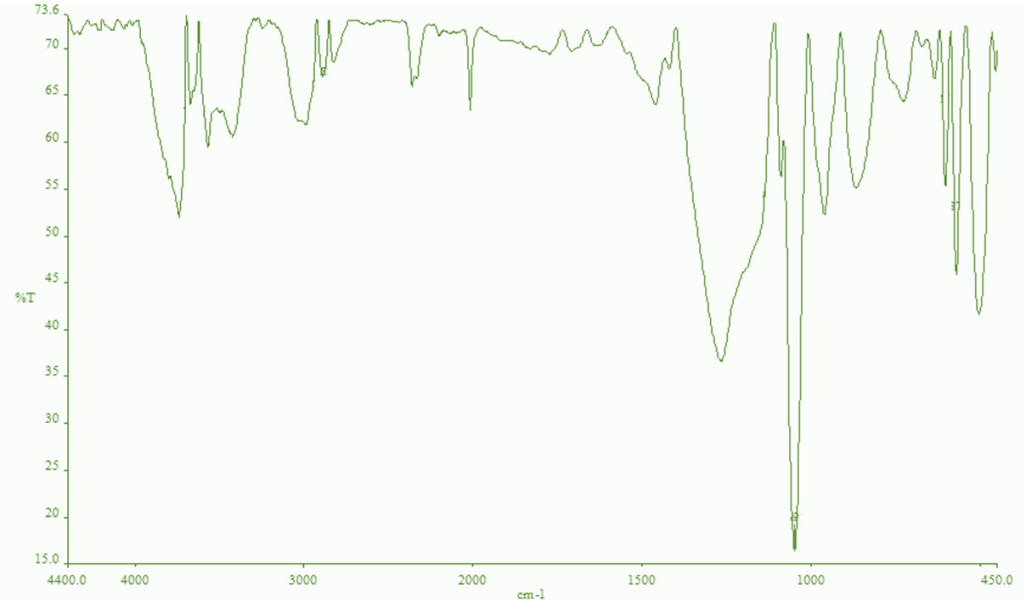

Figure 2: FTIR spectrum of Ni-Zr LDH.

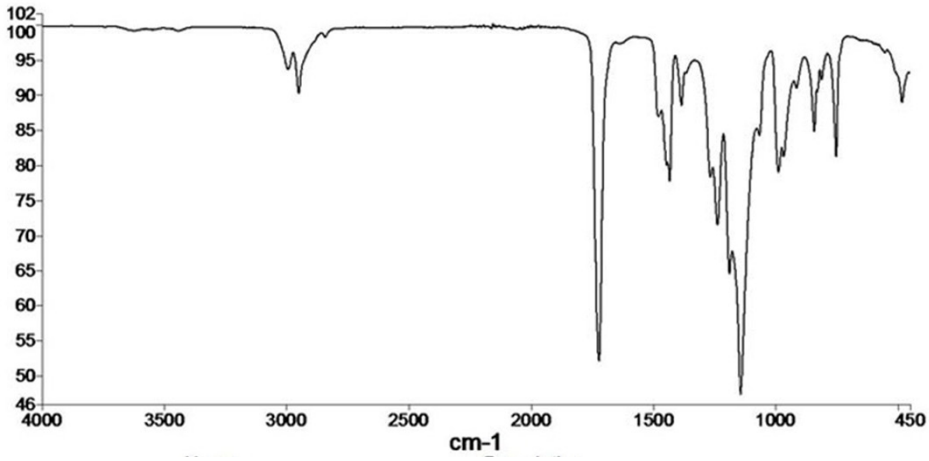

Figure 3: FTIR spectrum of PMMA.

Scanning electron microscopy

The method was selected to investigate the morphology and particle size of Ni-Zr LDH. Figure 4 shows FE-SEM images, which indicate that the Ni-Zr LDH plates were directly synthesized.

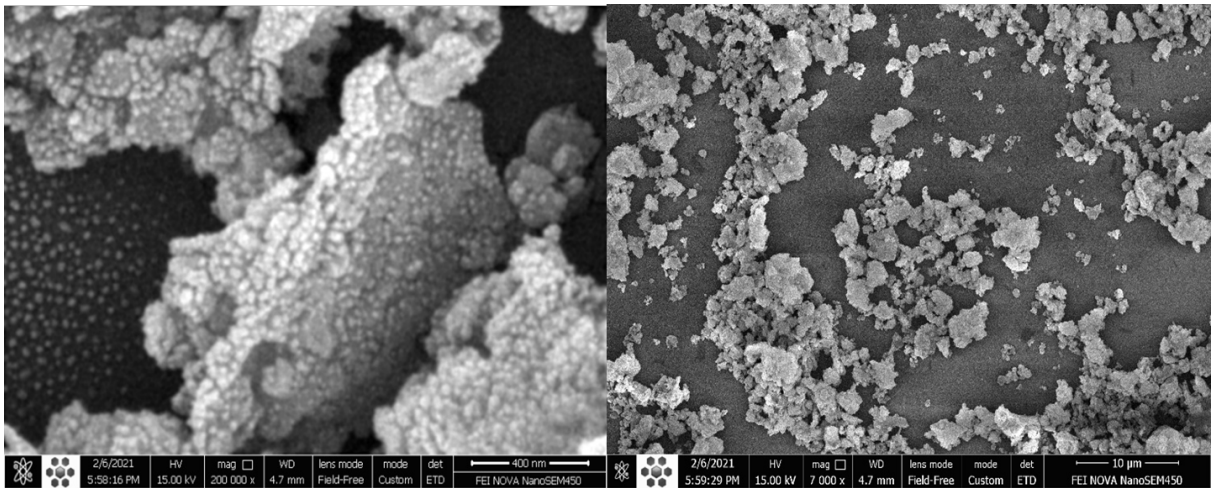

Figure 4: The LDH FE-SEM images.

Three component composite images

Figure 5 shows images of three component composite with 0, 1,3,5,7 and 10%(w/w) LDH, respectively. Pores were not observed for 1 and 3% (w/w) of LDH. Figure 6 shows an image of a three-component composite with 5, 7, and 10% (w/w) LDH, respectively. That indicates pores will increase with an increasing percentage of LDH. This phenomenon causes an increase in the concentration of tension and a decrease in the strength of the composite. Due to the increase in LDH percentage and hydroxyl groups, the active sites of polymer will be occupied, and the porosity will increase. Figure 7 shows the relationship between the force applied to the composite and its displacement, which is the method to calculate the composite modulus. This figure Shows good repeatability of the nanoindentation test of the sample with 3% (w/w) LDH, which indicates the test is reliable. Table 1 explains that three-component composites have the best elastic modulus with 5% of LDH. With 3% hydroxyapatite and 5% LDH, PMMA's elastic modulus rises to 15.3%, approaching the mudolus of human bone.

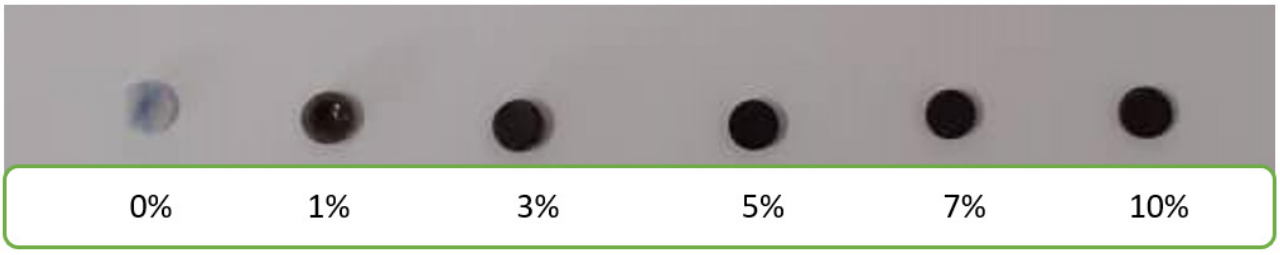

Figure 5: Images of three component composite with 0, 1,3,5,7 and 10% (w/w) LDH.

Figure 6: Images of a three-component composite with 5, 7, and 10% (w/w) LDH.

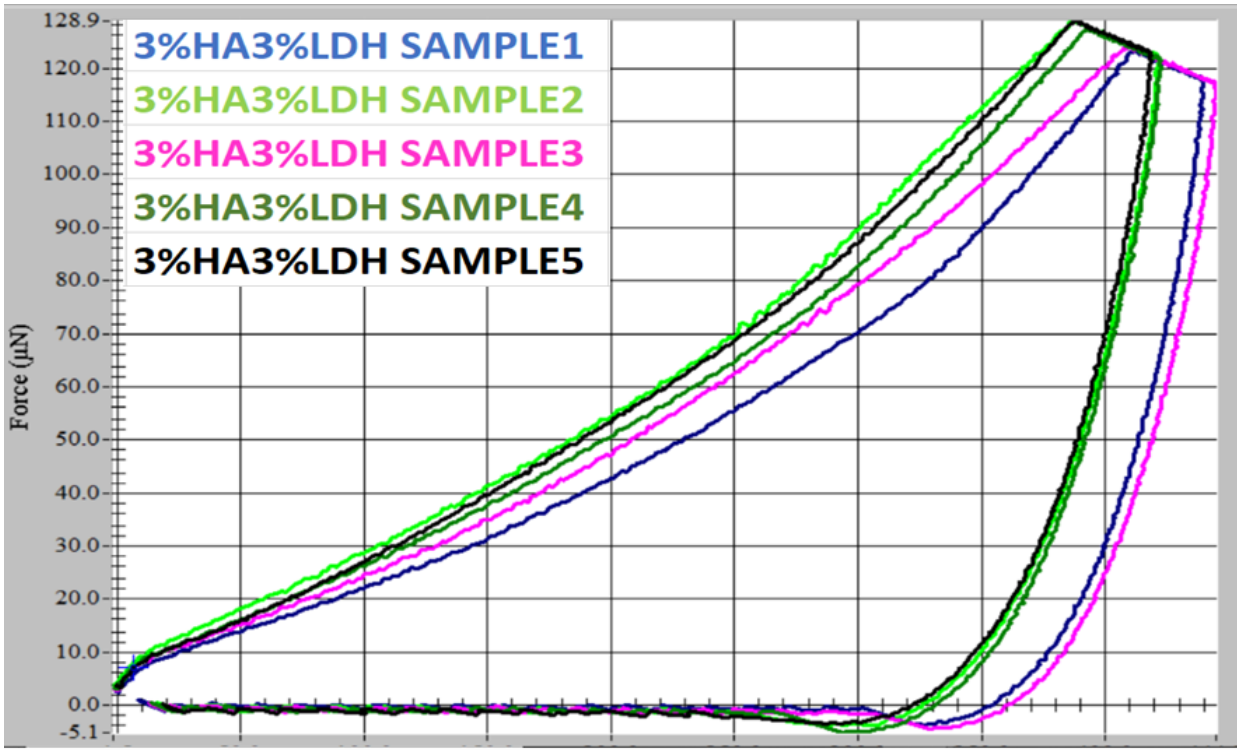

Figure 7: Force-displacement diagram of 5 sample with 3% LDH.

Table 1: Comparison of 0%, 1%, 3%, 5%, 7% and 10%(w/w) LDH elastic modulus.

Ni-Zr LDH.%w/w	HA 3%-Ni-Zr LDH-
%0	6.52
%1	7.1
%3	6.04
%5	7.52
%7	5.52
%10	7.12

Conclusion

Ni-Zr LDH were proposed to compose with cement bone because of micro sized particle of LDH and Zr mechanical properties. Micro size of LDH will prevent from concentration of tension and decreasing strength of composite. But pores will increase in macro size by increasing of percentage of LDH. This phenomena cause increasing of concentration of tension and decrease of strength of composite. In addition 5% of LDH will increase the elastic modulus 15%, which approaches human bone elasticity modulus.

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