RESEARCH ARTICLE

Preparation and Characterization of Nanostructure Akermanite Powder by Mechanical Activation Method

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ARTICLE INFO

Article History:
Received 20 May 2016
Accepted 13 July 2016
Published 30 July 2016

Keywords:
Mechanical activation
Akermanite
Nano powder

ABSTRACT

Objective(s): So far, extensive research has been conducted on the preparation and characterization of nano ceramics based on Ca-Si by sol- gel method and bioactivity was evaluated, but, a few researches have paid attention to the preparation of materials by mechanical activation (MA). The aim of this study was the preparation of akermanite nano powder by mechanical activation method and bioactivity evaluation.

Methods: Akermanite was prepared by MA method and subsequent heat treatment. Samples were mixed of calcium oxide (CaO), silicon dioxide (SiO_2) and magnesium oxide (MgO) with molar ratio of 2:2:1, respectively. These were milled for 6 h, 8 h, and 10 h with ball-to- powder ratio 10:1 and rotation speed of 300 rpm. After synthesis, the samples were pressed under 25 MPa and heated at 1100 ºC for 3 h. X-ray diffraction (XRD), transmission electron microscopy (TEM) and energy-dispersive x-ray spectrum (EDX-mapping) analysis were performed to characterize three kinds of powder. Bioactivity evaluation of the akermanite ceramics was investigated by being immersed in the simulated body fluid (SBF).

Results: According to XRD pattern, the sample which was milled for 10 h at heat treatment at 1100 °C only indicated the pure akermanite phase. The crystalline size of nano powder indicated that with ball milling time increase, the sizes of crystalline were decreased. Also, SEM images showed that, apatite nucleation happened and it grew on the sample surface.

Conclusions: In the present investigation, the nanostructure akermanite powder can be prepared by mechanical activation (MA).

How to cite this article
Marzban K. Preparation and Characterization of Nanostructure Akermanite Powder by Mechanical Activation Method. Nanomed Res J, 2016; 1(2):79-83. DOI: 10.7508/nmrj.2016.02.003

INTRODUCTION

For repairing bone damage caused by trauma, surgical resection and congenital deformity corrections, bone tissue engineering is useful [1]. As new biomaterials, bioinorganics and the use of metal ions in the synthesis of new materials have received substantial attention [2]. Bioactive materials are qualified by their close connection with living bone via hydroxyapatite formation [3]. The results indicated that a very important group of materials is based on CaO-Mg-SiO_2 system that might be used as novel bioactive materials for bone regeneration [4]. Previous studies have shown that akermanite ($Ca_2MgSi_2O_7$) ceramics had more mechanical strength and bioactivity than Ca-P ceramics [5]. Common methods for synthesizing CaO-SiO_2-base ceramics are sol-gel, spray drying, and evaporative decomposition of solutions and the solid state reaction of oxides at high temperature. Mechanical activation is a versatile method used for the preparation of nano powders and composites. Various types of materials, including metallic, ionic, and oxides (rare earth oxides, zirconia,

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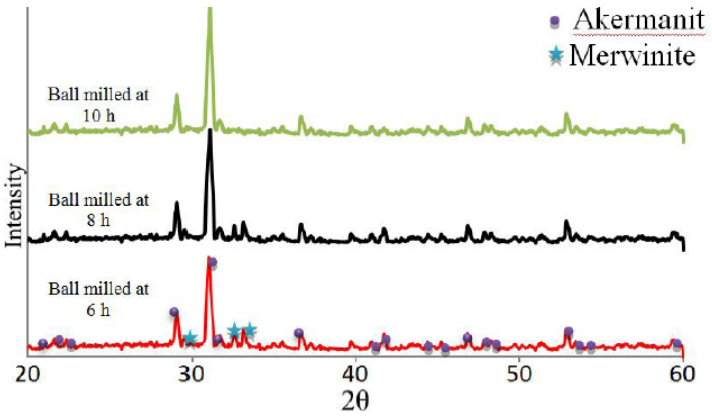

Fig. 1. XRD pattern of akermanite powders milled for 6, 8, and 10h and sintered at 1100°C for 3h

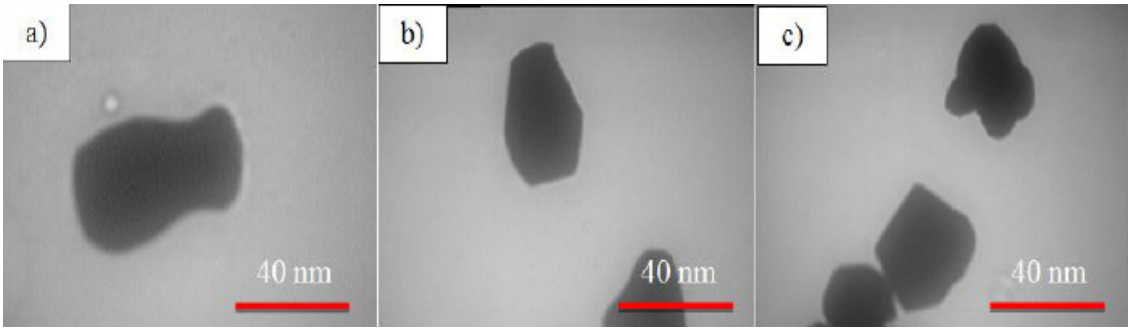

Fig. 2. TEM micrographs of the synthesized akermanite nanopowder milled for 6h (a), 8h (b), and 10h (c)

titanium oxide, etc.) have been prepared using this method [6]. The method has many advantages such as simplicity and low cost. Pressure can be applied by conventional milling equipment such as attrition, planetary, or vibratory mills ranging from low-energy ball mills to high energy stirred mills [7]. So far, extensive research has been conducted on the preparation and characterization of nano ceramics based on Ca-Si such as Ca-Si-Mg [8-11], / Zn [12, 13], / Ti [14-16] by sol-gel method and bioactivity was evaluated, but, a few researches have paid attention to the preparation of materials by mechanical activation (MA). Hence, it is necessary to investigate the characteristics of akermanite synthesis by MA. Tissue connection rate depends on the velocity of HA formation, surface and porosity properties, and ionic change in the measurement solution. The apatite layer formation on the surfaces of glasses and glass-ceramics is an important factor in their bioactivity which plays key role in the tissue–biomaterial interface [17]. Thus the aim of the study was the investigation of the synthesis and characterization of akermanit nano powder by MA method and the bioactivity evaluation in the SBF.

MATERIALS AND METHODS

Preparation and Characterization of Powders

Akermanite was prepared by mechanical activation method and subsequent heat treatment. Samples were mixed of calcium oxide (CaO), (98% purity, Merck), silicon dioxide (SiO_2) (98% purity, Merck) and magnesium oxide (MgO) (98% purity, Merck) with molar ratio of 2:2:1, respectively. These kinds of powders were ball milled for 6, 8, and 10 h with ball-to- powder ratio 10:1 and rotation speed of 300 rpm. After synthesis of akermanite, the powders were pressed under 25 MPa and heated at 1100 °C for 3 h. X-ray diffraction (XRD; Philips PW 3710), transmission electron microscopy (TEM; EM208S) and distribution of particles akermanite were shown by the Energy-Dispersive X-ray spectrum (EDX-mapping; Philips XL30). According to Scherrer equation [16], the crystalline size of nano powder was calculated by:

$$t = 0.89\,\lambda / \beta \cos\theta$$

Where t is the particle size, λ is the wave length, β is peak width chosen at half height in radians and θ is the Bragg angle.

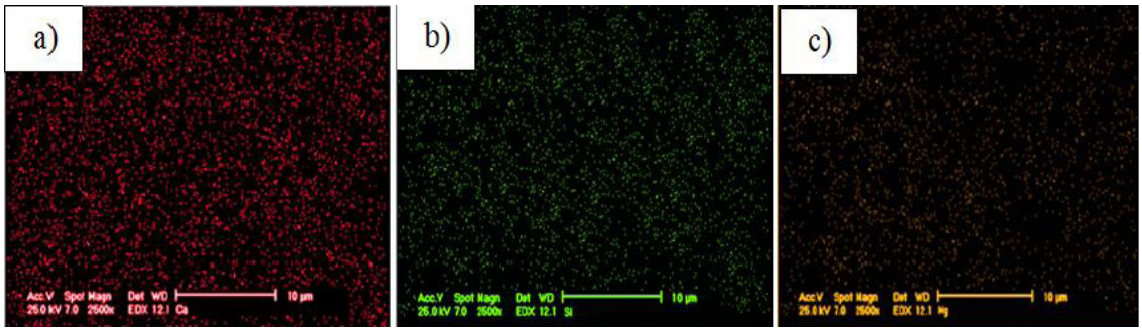

Fig. 3. EDX mapping results of chemical elements distribution Ca (a), Si (b) and Mg (c) on samples for 10h

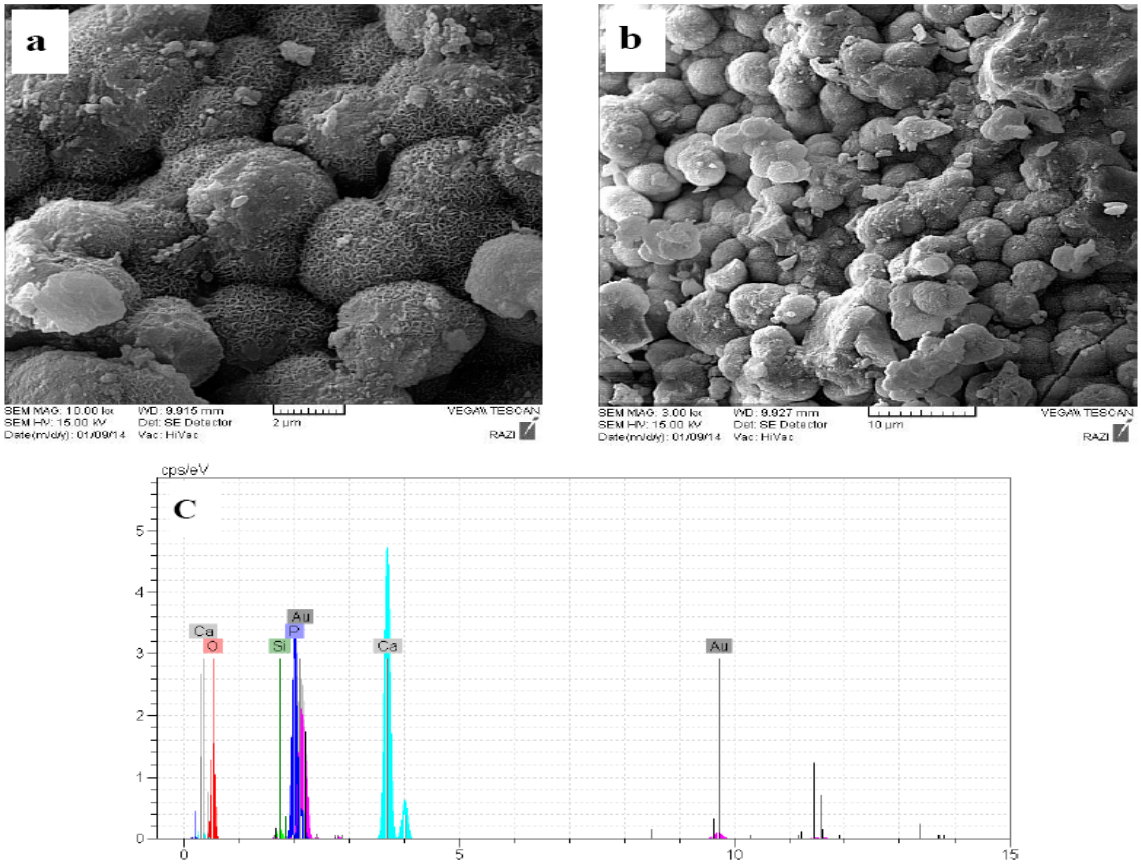

Fig. 4. SEM micrographs (a), Under higher magnification (b) and EDS patterns of akermanite powder (c) soaked in SBF for 30 days after 10 h of milling

Preparation of Samples for Analysis

The nano-powders were mounted rigidly on a specimen holder called specimen stub and coated with gold for 3 min using a sputter coater (Eiko IB3, Tokyo, Japan). The microscope was operated to visualize the samples at 15 kV.

X-ray powder diffraction (XRD) at a scan rate of 0.02◦/min with Cu Kα radiation was used for the crystallographic structural analysis of the sample.

Samples preparation has been done by obtaining a suspension from ultrasonification of the powder in ethanol on a foil surface followed by dropping on a cupper grid and finally dried to capture the images by means of TEM.

Preparation of samples has been done by obtaining a suspension of the powder in ethanol, then, dropping on a cupper mesh and eventually dried to capture the Pictures by instruments of TEM.

Soaking in SBF

Bioactivity evaluation of the akermanite ceramics was determined by immersion in the SBF. The SBF ion concentrations were prepared according to the method described by Kokubo [18]. After that maintenance into SBF for 30 days, the sample was removed from solution and dried at 30 °C for 24 h and finally characterized. Scanning electron microscopy – energy dispersive spectroscopy (SEM-EDS; VEGA\\TESCAN) was employed to study the apatite- formation ability of nanostructure.

RESULT AND DISCUSSION

The XRD pattern of akermanite powder obtained by the mechanical activation method at three ball mill times (6h, 8h, and 10h) and heat treatment at 1100 for 3 h (Fig. 1). According to the XRD patterns, sharp peak at 31.2° attributable to an akermanite phase (corresponding to JCPDS card no. 00-076-0841).
The patterns belong to the samples that milled for 6 h and 8 h have a little impurity in comparison with the other pattern (Fig. 1). However, for the samples milled after 6 h and 8 h, the powders contained a few impurities of merwinite ($Ca_3MgSi_2O_8$). The results obtained from Scherrer equation revealed that if the ball milling time increases the size of crystalline decreases [19]. The range of particle sizes at three kinds of powder is shown (as in Table 1). Therefore, the sample ball milled for 10 h is the best sample containing pure akermanite phase and crystal size.
TEM micrographs of the ball milled powder after 6 h, 8 h, and 10 h and being heated at 1100°C for 3 h (Fig. 2). TEM observations (for 10 h) showed that akermanite particle was composing of spherical and sharp points; however, the particle akermanite was nano-sized. As it is observed in TEM images, the nanostructure of powder demonstrated that, the grain size was about 40-60 nm (Table. 1).

EDX mapping analysis was used for investigating the Ca, Si, and Mg elements disperse in the ball milled sample for 10 h (Fig. 3). The EDX cartography of the samples showed that all akermanite elements (Ca, Si, and Mg) have been preserved and well dispersed all over the surface. Presence of the elements in the surface can induce more bioactivity, which significantly promotes the formation of hydroxyapatite [17]. Two methods are used to investigate the bioceramics in vitro bioactivity. First method is used to evaluate the apatite-formation potential in SBF. The other method is to investigate in culture medium. The SBF method is mainly utilized to test the in vitro bioactivity of bioceramics [20]. Fig. 4 (a) and (b) shows SEM micrographs of the surface morphology of the prepared akermanite after being soaked in SBF after 30 days for the ball milled sample after 10 h. SEM images showed that, apatite nucleation happened and it grew on the sample surface. Fig. 4c describes that Ca and P are exist on the surface. Since the akermanite composition is p-free; the presence of p in the EDS analysis can related to the formation of apatite after 30 days. The previous report has revealed that CaO and SiO2 release is necessary for the formation of bone-like apatite on the surface [21].

Table. 1 Comparing the particle size at three kinds of powder

Samples	particle size (nm)
Ball milled at 6 h	58.8 nm
Ball milled at 8 h	50.3 nm
Ball milled at 10 h	43.2 nm

CONCLUSIONS

In the present investigation, the nanostructure akermanite powder can be prepared by mechanical activation (MA). This result indicates that, the sample ball milled for 10 h is the best sample containing pure akermanite phase and the smallest particle size. The study shows that the powder obtained by mechanical activation method possesses apatite-formation potential. It can be used as a new biomaterial for bone tissue engineering purposes.

ACKNOWLEDGEMENTS

This research was supported by the Department of Biomedical Engineering, Amirkabir University of Technology.

CONFLICTS OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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