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Citation: [AIP Conference Proceedings](#) **1710**, 030037 (2016); doi: 10.1063/1.4941503

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Synthesis of Nano-forsterite Powder by Making Use of Natural Silica Sand

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Abstract. Nano-forsterite powder with natural silica sand and magnesium powder as the raw materials have been successfully synthesized. The silica sand was purified followed by a coprecipitation process to obtain colloidal silica. The magnesium powder was dissolved in a chloric acid solution to obtain MgCl₂ solution. The nanoforsterite powder was synthesised using a sol-gel method which included the mixing the colloidal silica and the MgCl₂ solution with various aging and filtering processes. The samples were dried at 100 °C using a hot plate and then the dried powders were calcinated at 900 °C for 2 hours. The samples were characterised for their elements and phase compositions using X-ray Fluorescence (XRF) and X-ray Diffraction (XRD) methods, respectively. The diffraction data were qualitatively analyzed using *Match!2* software and quantitatively using *Rietica* software. The crystallite size was verified using Transmission Electron Microscopy (TEM). Results of XRD data analysis showed that the forsterite content reached up to 90.5% wt. The TEM average crystallite size was approximately 53(6) nm.

Keywords: Nano-forsterit, natural silica sand, MgCl₂, aging, sol-gel process

INTRODUCTION

Forsterite (Mg₂SiO₄) is a crystalline magnesium silicate and derived from mineral olivine group (1), which has an orthorhombic crystal structure. Forsterite has a high biocompatibility and good mechanical properties that are appropriate for uses in the field of health (2). With a melting point at around 1890°C, forsterite is also commonly used as a high-temperature refractory material (3). Nano-forsterite powders are known to have better bioactivity than forsterite with micron size (4) as well as that they are biodegradable (5).

One source of SiO₂ is silica sand which can be found in many parts of the world. Use of silica sand as the direct source of silica for synthesis of forsterite has not been optimally studied. Various techniques were used to obtained high-purity forsterite (6–15) but none used silica sand as the raw material. This research was designed to adding the value of the potential for natural silica sand from Tanah Laut regency, South Kalimantan, as the primary material in the synthesis of nanoforsterite.

EXPERIMENTAL

This research aimed to synthesize high-purity nanometric forsterite by a wet method by making use of silica sand as the silica source. The first important process in the preparation of starting material was to clean natural silica sand from non-Si elements. The purification steps were cleaning and sieving, magnetic separation, milling and soaking with hydrochloric acid. The product of the step was denoted as silica powder. The gel solution was prepared by dissolving silica powder in a strong base solution followed by a coprecipitation process. The gel solution were aged before it was washed for 15 times using distilled water to remove the salts that are formed during the coprecipitation process. The gel was maintained in the distilled water prior to be used as precursors - it was called colloidal silica. Magnesium powder is dissolved in a strong acid to obtain a precursor of magnesium chloride (MgCl_2).

The nanoforsterite powders were synthesized by mixing the precursors using two volume ratio of silica and magnesium precursors 1: 2.5 (NFTL SA 1) and 1: 3 (NFTL SA 2). PVA was also added into the solution mixtures as a catalyst, and then the mixture was stirred at 80°C for 2 hours. The samples were aged for 24 hours and filtered with filter paper to produce mixture gels. Next, the gels were tested for DSC/TGA to determine the calcination temperature. The gel samples were dried at 100°C before calcined in a Carbolite RHF 1400 Furnace at 900°C for 2 hours. XRD measurements were conducted to reveal the phase characteristics. Qualitative analysis of XRD data was done by software *Match!2*, while the quantitative phase analysis was done using *Rietica*. A TEM image was taken to reveal the size of crystalline forsterite.

RESULTS AND DISCUSSION

Several XRF measurement have been managed to clarify the steps of purification of the natural silica sand from Tanah Laut into silica powder. The Si contents of the natural sand, after sieving and washing, after milling and after coprecipitation were respectively 94.7(2)%, 95.6(2)%, 97.7(1)% and 98.0(2)% by weight. The final coprecipitation product was maintained in the form of colloidal silica which was then mixed with MgCl_2 solution using a magnetic stirrer. To determine the calcination temperature for forsterite formation, a dried mixed sample was thermally examined using TGA/DSC system (Figure 1). Both thermal and mass change effect were measured concurrently on the same sample during the firing from 0 to 1200°C . The dashed line shows the TGA result which is representative of the thermal effects to display the phase transitions. An endothermic peak appears at about 125°C which can be related to the removal of physically-adsorbed water within the system. The fluctuative exothermic peak between 125 and 600°C indicates the de-chlorination of MgCl_2 , crystallization of SiO_2 and possible reactions between SiO_2 and MgO precursors. A weak exothermic peak at approximately 870°C indicates the formation of forsterite as point out by the arrow. According to this result, the calcination temperature was then set at 900°C , in order to avoid excessive crystal growth, since the purpose of our synthesis was to form a nanocrystalline forsterite. This temperature calcination was also selected by Kamalian *et al* (16) but different in the heating rate. Kamalian applied a continue

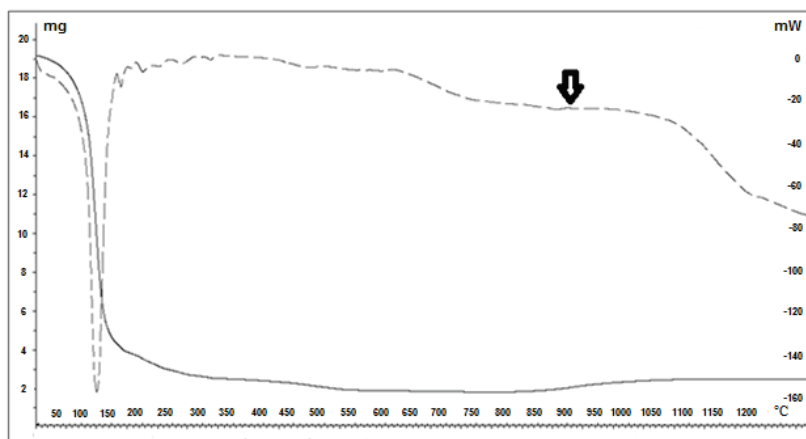


FIGURE 1. DSC-TGA thermal history pattern for the silica gel- MgCl_2 mixture.

heating rate in 10 °C/min, meanwhile, we divided the heating rate into several steps. From 0 to 250 °C the heating rate was applied at 29 °C/min. It was then set at 23°C/min up to 370 °C. Next , it further slowed down to 500° C at 16 °C/min and then to 780 °C at 9 °C/min . The last heating rate was set at 6 °C/min to reach 900 °C.

Forsterite formed in both samples and its formation reaction is assumed to follow Equation:

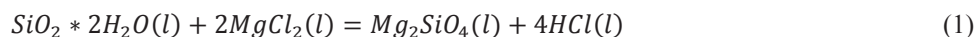


Figure 2 illustrates the XRD patterns for samples calcined at 900 °C for 2 hours. The qualitative analysis indicated that the dominating phase of all samples is forsterite (AMCSD No. 0000328), while periclase (AMCSD No. 0000501), cristobalite SiO₂ (AMCSD No. 0001629) and protoenstatite MgSiO₃ (AMCSD No. 0004955) are found as secondary phases. These results agree with those from Tavangarian (14) who claimed that it was very difficult to avoid the presence of secondary phase. The XRD pattern for NFTL SA 1 shows that the sample is more crystallized than the other, although the secondary phases are also present. Therefore, in the synthesis of forsterite by this method, it is important to control the global stoichiometry 2:1 for Mg:Si, as proven by Andre (3).

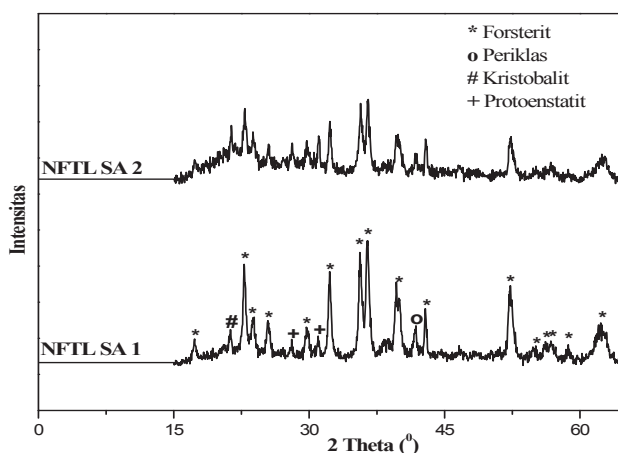


FIGURE 2. XRD patterns (CuK α radiation) for samples calcined at 900°C for 2 hours.

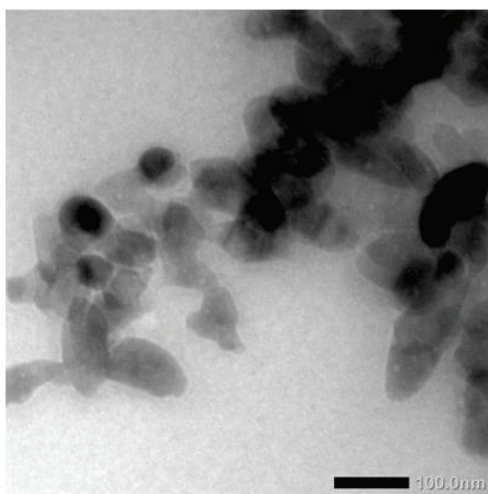


FIGURE 3 TEM images of the NFTL SA 1 calcined at 900 °C for 2 hours

The diffraction peak width of forsterite is relatively broad ($ca > 0.2^\circ 2\theta$) for all samples, which indicates that the crystallite size of forsterite could reach nanometric scale (17). The nanometric size of the NFTL SA1 forsterite powder was confirmed by a TEM image (Figure 3). The average size was 53(6) nm after calculation using the cross-

TABLE 1. Rietveld relative weight fractions in the nano forsterite powders and their standard deviations using *Rietica*. The least significant digits of the standard deviation are given in the parentheses.

Sample	Quantity(wt%)			
	Forsterite e(F)	Periclas(P)	Cristobalite(C)	Protoenstatite(Pr)
NFTL SA 1	90.5(9)	4.1(3)	1.7(1)	3.8(4)
NFTL SA 2	75.6(3)	9.1(6)	10.1(1)	5.2(6)

section method with standard deviation error (value in the bracket) was calculated using $S_x = \frac{\sqrt{\sum_{i=1}^k (\delta x_i)^2}}{k(k-1)}$, where δx is deviation and k is number measurement. The universal shape of nanoforsterite was tapering, it is accordance with the structure data. Forsterite has a space group *Pbnm* with orthorhombic structure and crystal lattice parameters $a = 4.756$, $b = 10.207$ and $c = 5.980$ Å (18). From the structure is known that the ratio $a/c \approx 1$ and $b/a \approx 2$. Thus, the crystallites of forsterite as seen from the TEM image elongated in the y direction.

Table 1 presented the results of Rietveld-based *Rietica* quantitative analysis for both samples. The NFTL SA 1 sample exhibits a higher nanoforsterite relative weight fraction, ie 90.5 ± 0.9 wt%. Consequently, the selected wet method can produce nanoforsterite with fair purity.

CONCLUSIONS

Nanoforsterite powders have been successfully synthesized through a solution reaction containing a colloidal silica from Tanah Laut and $MgCl_2$ solution at calcination temperature of $900^\circ C$ for 2 hours. In spite of the result showed secondary phases of periclas, cristobalite and protoenstatite, a fair purity can be gained by the method. The TEM image revealed that the average size of the forsterite particles was 53(6) nm.

ACKNOWLEDGMENTS

The research work was supported by “Hibah Unggulan Perguruan Tinggi (PUPTN)” ITS Research Grant, No. 003246.105/IT2.11/PN.08/2015. We indebted with BPPDN DIKTI for the doctoral fellowship programs.

REFERENCES

1. Ando M, K. Himura K, Tsunooka T, Kagomiya I, Ohsato H. Synthesis of High-Quality Forsterite. *Jpn J Appl Phys* Vol 46 No 10S P. 2007;46(10B):7112.
2. Fathi MH, Kharaziha M. Mechanochemical synthesis and characterization of nanostructure forsterite bioceramics. *Int J Mod Phys B*. 2008;22(18):3082–91.
3. Andre D. Aqueous Synthesis of Forsterite (Mg_2SiO_4) and Enstatite ($MgSiO_3$). *J Sol-Gel Sci Technol*. 2002;24:221–8.
4. Kharaziha M, Fathi MH. Synthesis and characterization of bioactive forsterite nanopowder. *Ceram Int*. 2009 Aug;35(6):2449–54.
5. Tavangarian F, Emadi R. Nanostructure effects on the bioactivity of forsterite bioceramic. *Mater Lett*. 2011 Feb 28;65(4):740–3.
6. Dachs E, Geiger CA, von Seckendorff V, Grodzicki M. A low-temperature calorimetric study of synthetic (forsterite + fayalite) $\{(Mg_2SiO_4 + Fe_2SiO_4)\}$ solid solutions: An analysis of vibrational, magnetic, and electronic contributions to the molar heat capacity and entropy of mixing. *J Chem Thermodyn*. 2007 Jun;39(6):906–33.

7. Ghomi H, Jaberzadeh M, Fathi MH. Novel fabrication of forsterite scaffold with improved mechanical properties. *J Alloys Compd*. 2011 Feb 3;509(5):L63–8.
8. Kiss SJ, Kostić E, Djurović D, Bošković S. Influence of mechanical activation and fluorine ion on forsterite formation. *Powder Technol*. 2001 Jan 15;114(1–3):84–8.
9. El Hadri M, Ahamdane H, El Idrissi Raghni MA. Sol gel synthesis of forsterite, M-doped forsterite (M = Ni, Co) solid solutions and their use as ceramic pigments. *J Eur Ceram Soc*. 2015 Feb;35(2):765–77.
10. Fathi MH, Kharaziha M. Two-step sintering of dense, nanostructural forsterite. *Mater Lett*. 2009 Jul 15;63(17):1455–8.
11. Naveed Hosseini S, Salimi Jazi H, Fathi M. Novel electrophoretic deposited nanostructured forsterite coating on 316L stainless steel implants for biocompatibility improvement. *Mater Lett*. 2015 Mar 15;143:16–9.
12. Sanosh KP, Balakrishnan A, Francis L, Kim TN. Sol–gel synthesis of forsterite nanopowders with narrow particle size distribution. *J Alloys Compd*. 2010 Apr 9;495(1):113–5.
13. Saberi A, Alinejad B, Negahdari Z, Kazemi F, Almasi A. A novel method to low temperature synthesis of nanocrystalline forsterite. *Mater Res Bull*. 2007 Apr 12;42(4):666–73.
14. Tavangarian F, Emadi R, Shafyei A. Influence of mechanical activation and thermal treatment time on nanoparticle forsterite formation mechanism. *Powder Technol*. 2010 Mar 25;198(3):412–6.
15. Kosanović C, Stubičar N, Tomašić N, Bermanec V, Stubičar M. Synthesis of a forsterite powder by combined ball milling and thermal treatment. *J Alloys Compd*. 2005 Mar 8;389(1–2):306–9.
16. Kamalian R, Yazdanpanah A, Moztarzadeh F, Mozafar M, Ravarian R, Moztarzadeh Z, et al. Synthesis And Characterization Of Bioactive Glass/Forsterite Nanocomposites For Bone And Dental Implants. *Ceram-Slikaty*. 2012;56(4):331–40.
17. Pratapa S, Hartono B. X-Ray Diffraction Microstructural Analysis of Bimodal Size Distribution MgO Nanopowders. *AIP Conf Proc*. 2010;
18. Smith J, Hazen R. The Crystal Structures of Forsterite and Hortonolite at Several Temperature Up to 900°C. *Am Mineral*. 1973;58:588–93.