Synthesis and Characterization of Forsterite Refractory by Doping with Kaolin

A. Bhargavi Rani[1]*, A. Raja Annamalai[1,2] M. R. Majhi[3], A. Harish Kumar[1]

[1]Department of Metallurgical and Material Science Engineering , RGUKT-NUZVID, Nuzvid – 521 202 AP, India.

[2]Presently as Associate Professor, School of Mechanical and Building Science, VIT University, Vellore – 632 014, Tamilnadu, India.

[3]Departmentof Ceramic Engineering, IIT-BHU, Varanasi-221005, UP, India.

*Corres.author: bhargaviranianne@gmail.com

Abstract: The present study investigates the synthesis of forsterite by adding kaolin to improve properties of refractory. Forsterite was synthesized by solid-solid reaction of MgO and SiO_2 and kaolin added as additive and sintered Up to 1450^0C for 2hr soaking period. Synthesized forsterite was characterized by physical properties measurements (%Apparent Porosity, Bulk Density, %Water Absorption, Apparent Specific Gravity, %Weight Loss, %Linear Shrinkage) and XRD analysis was done to the sintered forsterite. Properties (%A.P, B.D, %W.A, T, %W.L, %L.S) were improved by addition of kaolin due to formation of cordierite phase which melts at sintering temperature. 3.0% kaolin was more effective in promoting densification.
Keywods: Forsterite, Cordierite, densification, kaolin, sintering, XRD.

1. Introduction

Forsterite is one of the most efficient materials for critical production stages of modern iron and steel industry, namely, steel making and casting or ladle metallurgy [1-6]. It has low thermal expansion coefficient and extremely used for low electrical conductivity that makes it an ideal substrate material for electronics. Also, it shows good refractoriness, good chemical stability and excellent insulation properties even at high temperatures [7-10]. It is a naturally occurring mineral. Natural Forsterite occurs with the end member of its isomorphous series Fayalite ($2Fe.SiO_2$), which melts at 1205^0C and hence the melting point of natural mineral is lower. Forsterite is a mineral of magnesium orthosilicate having formula Mg_2SiO_4 and having melting point at around 1890^0C [11-14]. Bulk specific gravity of Forsterite refractories is 2.50 which is less than that of magnesite bricks. Porosity range is 24-28%. They fuse at a temperature of 1890^0C and having cold crushing strength of more than 2200 lbs per square inch [1, 14]. Forsterite together with high alumina refractory material is used for lining regenerator checkers without using a neutral chrome-magnesia layer [15]. Forsterite refractory nature is suitable for use in thermal insulation. Forsterite insulating bricks still offer a challenge to refractory producers and users due to non-reactivity with magnesia bricks, relatively low thermal conductivity and relatively cheap raw materials. Finally concluded that can be used in combination with magnesia and chrome-

magnesite bricks up to 1550^0C, with high Alumina up to 1500^0C and with dolomite up to 1400^0C. The evolution and growth of Forsterite ceramics was manifested over the last few decades to improve the physic-mechanical properties; strength, dielectric constant, thermal expansion, thermal conductivity and mechanical performance. As per earlier studies, properties improved through addition of alumina, mullite, magnesium aluminate spinel and partially stabilized Zirconia. To fire in an oxidizing atmosphere and to withstand high temperature mechanical stress and holding time, Alumina- Forsterite mixtures are most preferable [1,14]. The previous investigations were done to know the effective densification by kaolin addition; i.e. up to 3% Kaolin densification is effective[16]. The present research aims at preparation of Forsterite refractory by adding Kaolin (0-6% by wt) to improve properties and up to what extent kaolin can be used to improve properties without effecting any property, and to compare the physical properties of Forsterite refractory with different compositions of kaolin i.e. 0%, 5% and 6%.

2. Experimental Procedure

In present work Forsterite was synthesized by solid-solid reaction between Magnesium Oxide (MgO) and Quartz (SiO_2) in proportion of 56% and 44% respectively and fired at 1450^0C for different soaking periods(1hr and 2hr). After firing of material was tested for further studies of properties. Raw materials proportions are tabulated in Table I. Pellets were made by mixing compositions of powders of MgO, SiO_2 and kaolin in the weight proportions of 56%, 44% and (0%, 5%, 6%) respectively. Magnesia as pure grade (MgO) and Silica as Quartz AR grade (SiO_2) and proper mixing of powders for 30min by adding binder of PVA (5g in H_2O).Steel die (1.5cm in dia) was used for cold pressing and 15 tons/sq.inch pressure is applied in a hydraulic press (uniaxial).Pressure applied at const rate of loading for all pellets and applied for 1min. For efficient pellet making Acetone was used for cleaning. Initially sintering of pure Forsterite (without kaolin addition) was done and later with addition of kaolin was done. Before sintering pre measurements were taken. Sintering was done at temperatures of 1350^0C and 1450^0C in Silicon Carbide resistance heating furnace by the help of high Alumina crucibles. At each temperature sintering was done for two soaking periods i.e. 1hr and 2hr. The average heating rate was 5^0-7^0C/min. After sintering in every instance properties were measured i.e. Apparent Porosity, Bulk Density, Apparent Specific Gravity, Water Absorption and the relationships between these properties and percentage of kaolin added were analyzed. In present work, boiling water method was used to find properties; this method is only applicable to burnt bricks. Before putting in water bath, dry the sample at 10^0C in Hot Air Oven and taken dry weight of sample and was placed sample in water bath and boiled for 2hrs.During boiling, ensure that sample should not in contact with either heating coil or bottom of water bath. After boiling and cooling taken the weight of sample while suspended in water (S), immediately after taking of suspended weight, gently wiped bottom of sample with tissue paper and taken soaked weight (W).Apparent porosity, Water Absorption, Apparent Specific Gravity and Bulk density were calculated by using following formulas.

Apparent porosity (P) = (W-D)/ (W-S) *100

Water Absorption (A) = (W-D)/ D *100
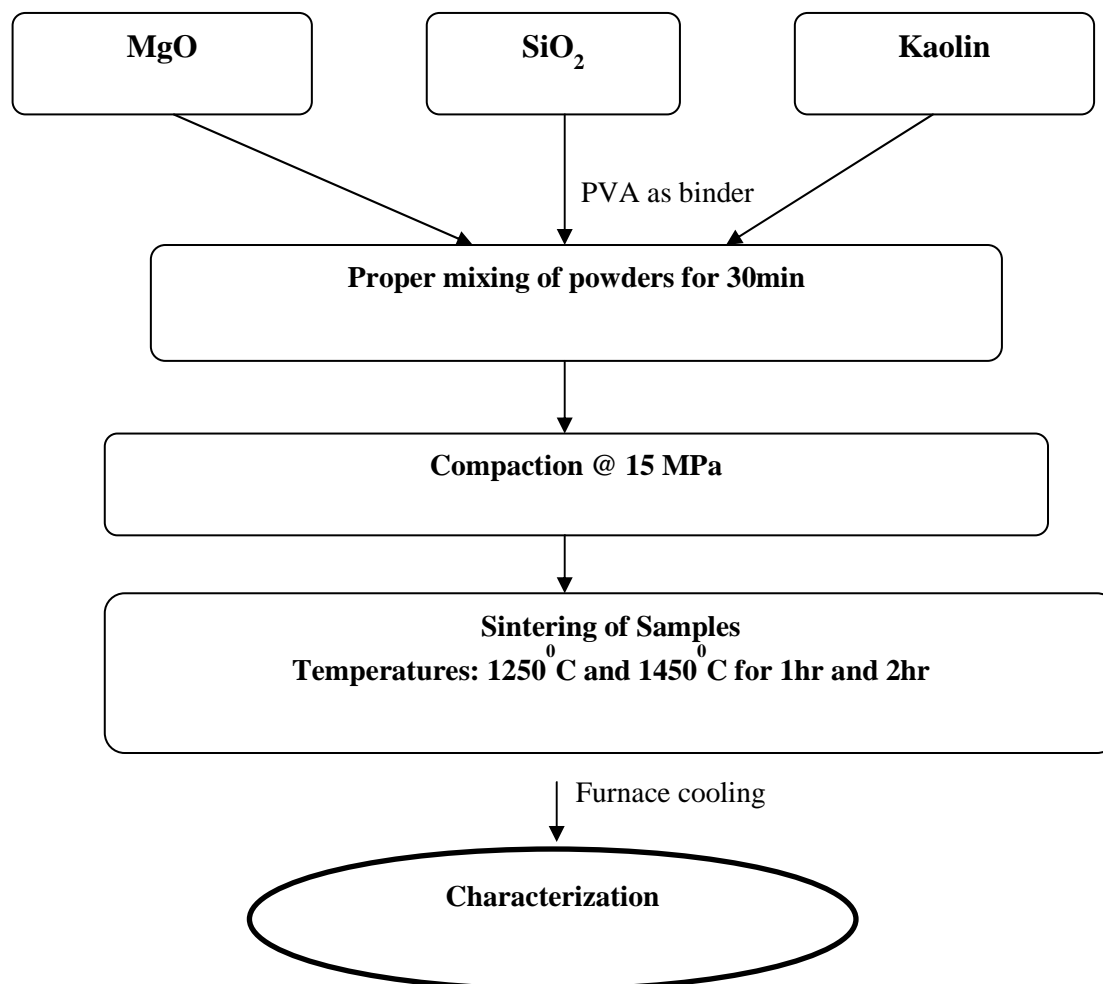
Apparent Specific Gravity (T) = D/ (D-S) *100

Bulk Density (B) = D / (W-S) *100

Where,

S= Suspended weight of sample in water,
W= Soaked weight of sample in air,
D= Dry weight of sample.

Experimental Flow Sheet

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┌──────────────┐      ┌──────────────┐      ┌──────────────┐
│     MgO      │      │     SiO₂     │      │    Kaolin    │
└──────────────┘      └──────────────┘      └──────────────┘
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PVA as binder

Proper mixing of powders for 30min

Compaction @ 15 MPa

Sintering of Samples
Temperatures: 1250^{0}C and 1450^{0}C for 1hr and 2hr

Furnace cooling

Characterization

Fig I: Experimental Flow sheet.

Table I: Raw materials used for the experiment.

| Raw Materials	Percentage used (by wt)
Magnesium Oxide (MgO)	56%
Quartz (SiO$_2$)	44%
Kaolin (Additive)	(0-6)%
PVA(Binder)	5gm in 100ml H$_2$O

Results and Discussions

Magnesium Oxide (56%) and Quartz (44%) were mixed and fired up to 1450^0C for 2hrs. Peaks characterizing small amount of forsterite along with other phases like free silica, clinoenstatite, enstatite and magnesium silicate, are detected. The result indicates incomplete interaction of MgO and SiO_2 to form forsterite completely. To confirm forsterite phase XRD was done and forsterite, MgO and SiO_2 phases were formed (Fig 2). Sintering was done for forsterite at 1250 and 1450^0C and soaking period was varied up to 2hr by the addition of 0, 5.0 and 6.0 percent of Kaolin. After sintering, %A.P, B.D, App. Sp. Gravity, %W.A, %W.L, %L.S were calculated and the relationships between percentage of Kaolin addition and these properties were plotted (Fig-III – Fig-VIII) and tabulated (Table II-III).

From all results (Table II-III and Fig III-VIII) it clearly indicates that the densification of forsterite is influenced by the minor additions of Kaolin. Even though up to 5.0% of kaolin can be used to forsterite, the optimum value for maximum densification is 3.0% (by wt) of kaolin. At 1400^0C, the maximum density attained in 200min soaking time with 3.0% kaolin is 2.78 g/cc (87% of theoretical density) [16]. 5.0% kaolin also increasing properties and feasible composition to forsterite but 3.0% kaolin is more effective than 5.0% kaolin. Kaolin gives more densification due to formation of Cordierite phase which melts at sintering temperature by forming a glassy phase of low viscosity. Increase in properties at higher temperature (1450^0C) than lower temperature (1250^0C) is due to melting of cordierite gives rise to viscosity of liquid due to rearrangement of grains of liquid flow process [17]. It can be proved that densification achieved with 3.0% kaolin at 1400^0C. As kaolin content exceeds 3.0%, the predominant mechanism to cause densification is liquid flow process followed by diffusion at the contact in the solid and liquid phase giving increase in densification by solution re-precipitation. The figures shown that (%apparent porosity, %water absorption vs percentage addition of kaolin) Fig (III-IV) for both 1hr and 2hr soaking period, %A.P and %W.A decreased from 0% to 5.0% and gradually increased after 5.0% i.e. 6.0%. Decrease in %A.P and %W.A were needed; it should be low to acquire better properties like high strength and high thermal conductivity. Individually also for 0% and 5.0% as soaking period, temperature increases %A.P and %W.A decreased (Table II and III) and for 6.0% as soaking period, temperature increases %A.P and %W.A increased which was not feasible. The figures shown that (bulk density, app.sp.gravity, % weight loss and % linear shrinkage vs percentage addition of kaolin) Fig (V-VIII) for both 1hr and 2hr soaking periods, B.D, App.sp.G, %W.L and %L.S increased from 0% to 5.0% and gradually decreased after 5.0% of kaolin i.e. 6.0%, increase in this properties were needed, refractory with high bulk density will be better in quality by this can get better resistance to chemical attack, better abrasive resistance. Increase in bulk density increases volume stability and heat capacity. The bricks with high specific gravity are more cost but strength is also for high specific gravity. Individually also for 0% and 5.0% as soaking period, temperature increases these properties decreased (Table II and III) which was not feasible. Although there is no kaolin in forsterite properties increased because of sintering. After heat treatment the properties will surely increase depending on composition. Sintering was done by atomic diffusion, diffusion occurs above absolute zero but it occurs much faster at higher temperatures, because of sintering without kaolin also properties were in progressive order.

Even though for without kaolin addition also properties were improved. It is not effective than with addition of kaolin. In without kaolin properties improved because of heat treatment. The values of properties were effective in composition along with kaolin addition with forsterite. Cordierite phase melted before forsterite while sintering, due to this reason properties were much more improved. By considering thesis effective[16], comparing with 3.0% kaolin values with 5.0%, 3.0% was more effective in promoting densification. After 3.0% it is not much effective. Improving in properties was only up to 5.0% kaolin, above that for 6.0% kaolin properties were disgusted. In forsterite kaolin was added as additive, the effect of additives varies according to sintering temperature and optimum amount of addition, 6.0% kaolin was not feasible to forsterite because after 5.0% kaolin, it was not forming cordierite phase with forsterite, due to this properties were disgusted.
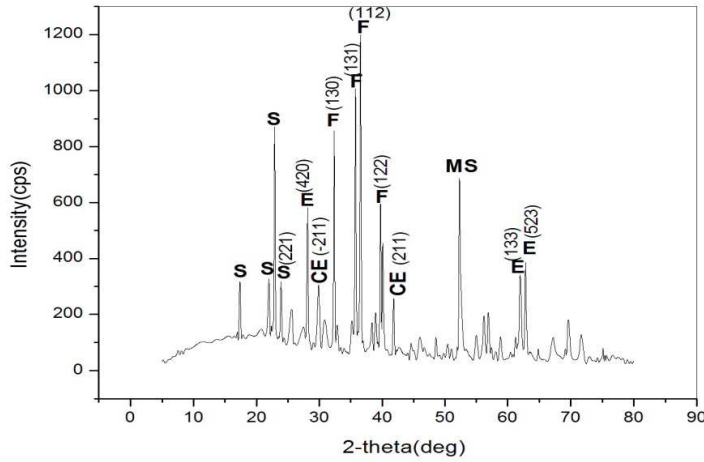
Fig II: X-ray diffraction pattern of 56wt% of MgO +44wt%of SiO$_2$+0% of kaolin sintered at 1450^{0}C for 2hrs,F,forsterite;S,free silica; CE, clinoenstatite; MS, magnesium silicate; E,enstatite.
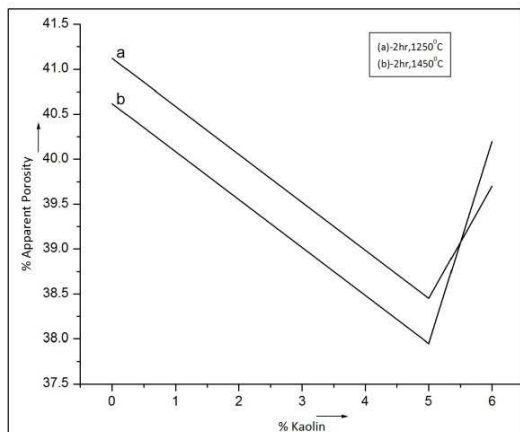
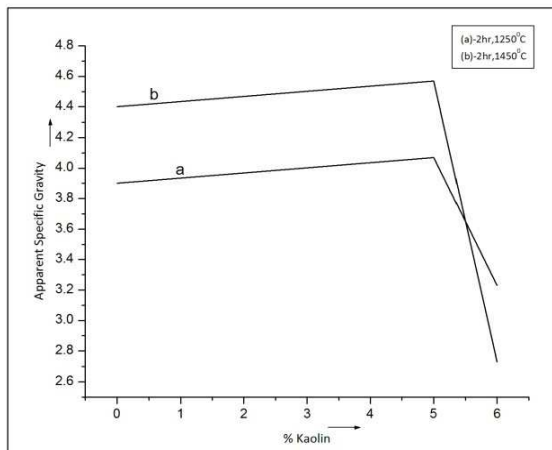

Figure III: Relationship between %Apparent Porosity and %Kaolin for forsterite with 2hr soaking period at different temperatures.
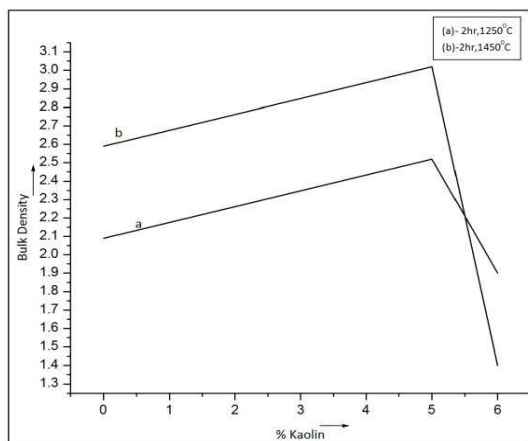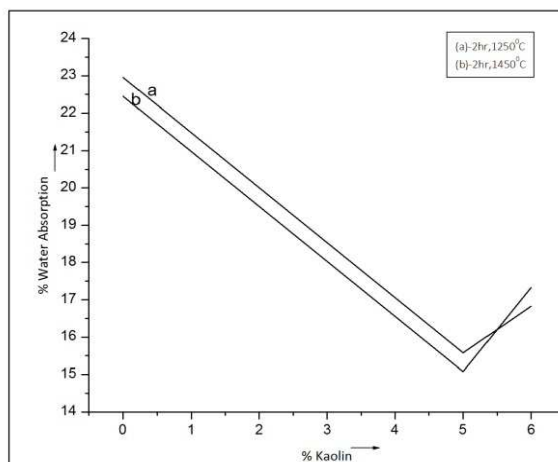

Figure IV: Relationship between Bulk Density and %Kaolin for forsterite with 2hr soaking period at different temperatures.
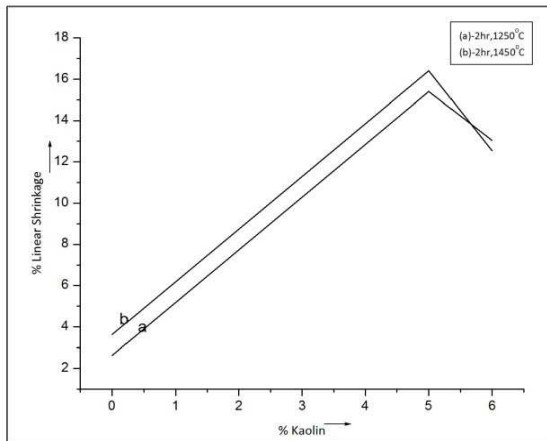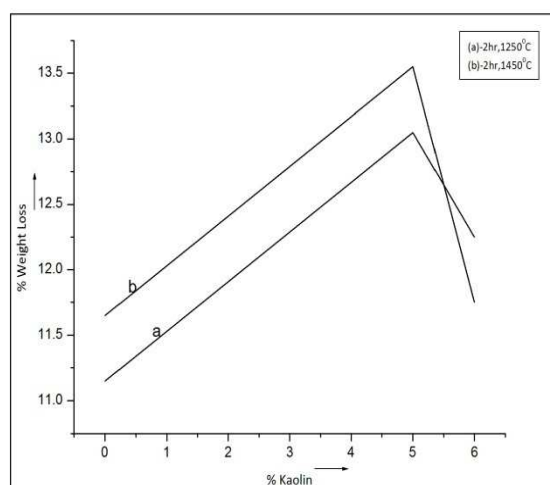
Figure V: Relationship between %Linear Shrinkage and %Kaolin for forsterite with 2hr soaking period at different temperatures.

Figure VI: Relationship between Apparent Specific Gravity and %Kaolin for forsterite with 2hr soaking period at different temperatures.

Figure VII: Relationship between %Water Absorption and %Kaolin for forsterite with 2hr soaking period at different temperatures.

A. Bhargavi Rani *et al* /Int.J. ChemTech Res.2014,6(2),pp 1390-1397.

1396

Figure VIII: Relationship between %Weight Loss and %Kaolin for forsterite with 2hr soaking period at different temperatures.

Table II: Properties of Forsterite at sintering temperatures 1250^0C for different soaking periods (S.P) (1hr and 2hr) by the addition of Kaolin (K.A) (0, 5.0, 6.0% by wt).

K.A (by wt %)	S.P	%A.P	%W.A	B.D	T	%W.L	%L.S
0%	1hr	41.37	23.2	1.84	3.65	10.9	2.13
	2hr	41.12	22.95	2.09	3.90	11.15	2.63
5%	1hr	38.70	15.83	2.27	3.82	12.8	14.9
	2hr	38.45	15.58	2.52	4.07	13.05	15.4
6%	1hr	39.45	16.58	2.15	3.48	12.5	13.3
	2hr	39.70	16.83	1.90	3.23	12.25	13.05

Table III: Properties of Forsterite at sintering temperatures 1450^0C for different soaking periods (1hr and 2hr) by the addition of Kaolin (0, 5.0, 6.0% by wt).

K.A (by wt %)	S.P	%A.P	%W.A	B.D	T	%W.L	%L.S
0%	1hr	40.8	22.70	2.34	4.15	11.40	3.13
	2hr	41.1	22.95	2.09	3.90	11.15	2.63
5%	1hr	38.7	15.83	2.27	3.82	12.8	14.9
	2hr	38.4	15.58	2.52	4.07	13.05	15.4
6%	1hr	39.4	16.58	2.15	3.48	12.5	13.3
	2hr	39.7	16.83	1.90	3.23	12.25	13.05

Summary

Forsterite was synthesized by solid-solid reaction between MgO and SiO_2.

1. As per investigations done so far, it clearly indicated that properties were increased for kaolin-forsterite system.

    a. For both 3.0% and 5.0% addition of kaolin properties were improved but as compared to 5.0%, 3.0% was more effective.

    b. For 6.0% kaolin properties were disgusted, so 6.0% kaolin was not suitable for forsterite.

    c. Due to formation of cordierite which melts at sintering temperature before forsterite and forming a glassy phase of low viscosity, kaolin gives more densification.

2. Up to 5.0% kaolin was feasible to forsterite, but up to 3.0% kaolin the properties were more effective.

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