The Journal of CPRI,
Vol. 10, No. 2, June 2014 pp. 385-394

A Study on the Physical and Morphological Characteristics of Aluminum Cenosphere Composite Sintered at High Temperature in Microwave

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Aluminium Metal matrix Composites (AMC) have been fabricated through powder metallurgy route comprising of Aluminum as matrix and reinforced with Cenosphere, a low density material in the form of hollow and porous spheres. The densification of the composite has been carried out in an advanced processing technique called the microwave sintering,which is rapid and economical. AMC with Cenosphere addition of 40 volume % has been prepared and sintered at various temperatures. The sintered composites have been studied for the mineralogical phases by XRD, morphology by SEM and physical properties such as Density, Apparent Porosity and Hardness (BHN). The results obtained have been compared with AMCs that were sintered conventionally. The microwave sintered samples showed better properties in terms of Porosity, Bulk Density and Brinell hardness values compared to the conventionally sintered ones.

Keywords: *Cenosphere, microwave sintering, Aluminum metal matrix composite, Powder Metallurgy.*

1.0 INTRODUCTION

Metal Matrix Composites (MMCs) have enhanced properties including higher strength, lower thermal expansion, high fatigue life and higher wear properties as compared to those of other matrix alloys [1]. Among the metallurgical processes, Powder Metallurgy (PM) is an attractive processing technique to produce near net shape products and is commonly used for the fabrication of engineering components and particulate reinforced metal matrix composites. The PM process usually involves mixing of powders of the matrix alloy with the reinforcing particles, followed by compaction and sintering. With the increasing demand for light weight materials due to rising costs of fuel and demand for higher efficiency, materials selectors are actively looking into diversifying the use of Aluminium based materials in lightweight applications [2]. Aluminium alloys are cheap, have low density, excellent strength, ductility and corrosion behavior [3]. Hence it is important to develop innovative and cost effective sintering techniques to process Aluminium components through PM route.

The filler addition is the key factor which enhances the physical and mechanical properties of the composites. Fly-ash based 'Cenosphere' in the form of hollow and porous structure produced during coal combustion in thermal power plants, possesses good mechanical and tribological characteristics, is used as a filler material. Cenosphere are unique in the way that they are low density, non-toxic andnon metallic hollow micro particles, that are light in weight and has high melting points[4,5].

One of the necessary steps in the PM approach is that of sintering. The main purpose of sintering is to improve bonding between the powders and to minimize porosity to realize an overall

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improvement in the properties such as improved density, reduced porosity and development of metal-metal bond between metallic powders, and metal-ceramic bonds between matrix and reinforcement [6]. In conventional processing, MMCs have certain disadvantages like higher processing time and energy intense, and hence, this calls for adoption of advance processing technique such as microwave sintering. Microwave (MW) energy is a form of electromagnetic energy with the frequency range of 300 MHz to 300 GHz and having wave length between 1mm to 1m. Microwave heating is a process in which the materials couple with microwaves, absorb the electromagnetic energy volumetrically, and transform into heat. This heating mechanism enhances diffusion process, reduces energy consumption, has rapid heating rates and reduces processing time. MW sintering improves physical and mechanical properties of the material [7, 8].

Metal matrix composites comprising of Aluminium metal as matrix, embedded with Flyash Cenosphere particulates, and fabrication through the powder metallurgy route is being studied. The densification of powder compacts of these materials are carried out by microwave assisted sintering at high temperatures.Aluminium metal matrix composites when reinforced with suitable particles, are known to possess higher specific strength and low thermal expansion co-efficient leading to development of cheaper compositeswith good engineering properties.

2.0 EXPERIMENTAL PROCEDURE

2.1 Materials

In the present study, atomized Aluminium metal powder of 99.5 % purity from M/s. NICE Chemicals, with a particle size range of ASTM 200 mesh (75 μm) was used as the matrix material for the composite. The Cenosphere obtained from fly-ash collected from M/s. NTPC Simhadri Thermal Power Station was used as the filler in the composite. Cenosphere with an average particle size of 10- 100 μm was used in this experiment.

2.2 Processing

A batch consisting of mix of the above materials was prepared which comprised of Aluminium powder: Cenosphere in the ratio of 60-40 volume % respectively. The materials were mixed thoroughly in laboratory make mechanical mixer. 5% polyvinyl alcohol solution was used as a binder while mixing to obtain the green strength of the composite. The mix was then pressed into cylindrical pellets of size 12 mm height x 12.5 mm diameter at a pressure of 5 MPa in a laboratory pellet press. The green composites were then thoroughly dried in oven at 108°C for 2 hours to remove the moisture. Two sets of pellets were prepared for sintering each set in different sintering routes for the purpose of study and comparison.

2.3 Sintering

One set of the dried samples were sintered in microwave sintering facility at various temperatures ranging from 650 to 1050°C in steps of 100°C separately. The sintering of the pellets was carried out at in a BHEL make Microwave Sintering Facility rated at 1.1 kW power, microwave frequency of 2.45 GHz, at a power level of 100 %. The sintering time comprised of 90 minutes for each temperature sintering cycle with soaking time of 42 minutes at maximum temperature.

The other set of samples were sintered separately in the same temperature range in a conventional laboratory muffle furnace resistance heated with kanthal element. The sintering cycle comprised of 7 hours with soaking of the sample for 2 hours at the peak temperature.

The sintered samples were later taken up for characterization.

2.4 Characterization

The mineralogical phase analysis of the sintered samples have been carried out in PANalytical make XPert PRO Model X-ray Diffractometer using Cu-Kά radiation.

The morphological study of the composites were carried out in Leica make Q500MC Model Scanning Electron Microscopic (SEM) with Energy Dispersive X-ray spectroscopy (EDAX) operating at an accelerating voltage of 15 kV.

The physical properties of the composite such as Bulk Density and Apparent Porosity were carried out by Archimedes principle and as per ASTM C830 standard respectively. The Brinell Hardness test was carried out as per ASTM E10 using Zwick 3212 Hardness testing machine.

3.0 RESULTS AND DISCUSSIONS

Initially, a set of composite containing Aluminum 60%, Cenosphere-40% powder pressed samples was sintered in microwave near the Aluminum melting temperature at 650°C and the measured properties of this composite is appended below.

TABLE 1

PROPERTIES OF ALUMINUM- CENOSPHERE PELLET SINTERED IN MICROWAVE

Temperature deg C	Apparent Porosity (%)	Brinell Hardness Number [HB]	Bulk Density [g/cc]
Aluminum- 60% Cenosphere-40% sample			
650	26.4	14	1.43

The microstructure of this microwave sintered sample (Figure 1) sintered at 650°C shows Cenosphere dispersed in the Aluminum matrix with Alumium powder appearing as flakes surrounding the Cenopshere particle. A sizeable amount of fine pores that are open to the surface are also observed which contribute to the apparent porosity of the composite.

The XRD graph (Figure 1a) of the sample microwave sintered sample shows Aluminum peaks confirming the presence of pure metallic Aluminum in the bulk of the material without formation of an oxide phase. The Aluminum (Al) is present is the metallic form. The Mullite phase ($Al_6Si_2O_{13}$) which is an Alumino-silicate is found in the matrix and this is contributed by the Cenosphere which is basically an Alumino-silicate material. Silicon(Si)which has been reduced from the Silica present in Cenosphere is also depicted in the XRD pattern.

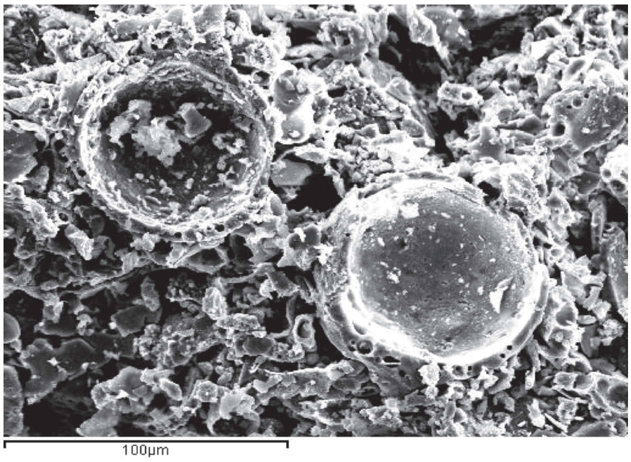

FIG. 1 MICROSTRUCTURE OF MICROWAVE SINTERED COMPOSITE AT 650°C

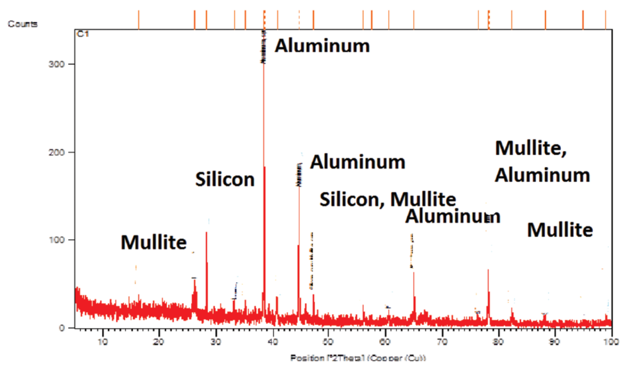

FIG. 1A XRD OF MICROWAVE SINTERED COMPOSITE AT 650°C

The physical properties measured for the microwave sintered sample at 650°C showed Apparent porosity of about 26.4 % contributed by the pores concealed in the bulk and the pores that are open to the surface. The bulk density of this sample was 1.43 g/cc and had a BHN value of 14 and all the above values are depicted in Table 1.

Further, to study the effect of high temperature sinteringcarried out by conventional method and by the microwave sintering process, a set of Aluminum Cenosphere composites samples were sintered beyond 650°C at temperatures of 650, 850, 950 and 1050°C separately. These high temperature sintered samples have been studied for their microstructure and phase analysis which are discussed below.

3.1 SEM and XRD Analysis of Conventionally Sintered composites:

The SEM image of the conventionally sintered composite at 650°C (Figure 2), shows flakes of Aluminium metal powder surrounding the cenosphere. The sphericity of the cenosphereis intact with certain pores appearing on the cenosphere surface. The corresponding XRD peaks (Figure 2a) show Aluminum (Al) as predominant phase with Silicon (Si). The Aluminum powder at this stage is in metallic form with out undergoing oxidation or forming oxide.Sintering the composite at 850°C (Figure 3), it can be seen that spherical shape of the cenosphere particle starts crumbling. The increase in the temperature has diminished the sphericity of the cenosphere. The corresponding XRD peak (Figure 3a) shows Silica (SiO_2) peaks, along with Alumina (Al_2O_3) in the composite matrix, contributed from Cenosphere which is basically an alumino-silicate material with presence of small amounts of Alpha Quartz as free Silica.

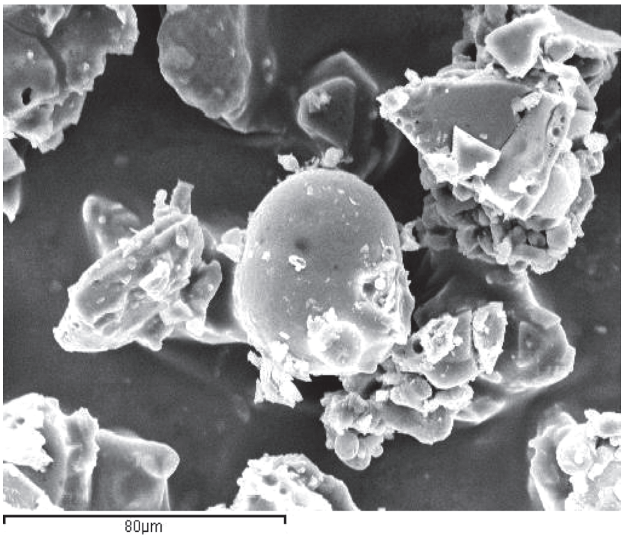

FIG. 2 MICROSTRUCTURE OF CONVENTIONALLY SINTERED COMPOSITE AT 650°C

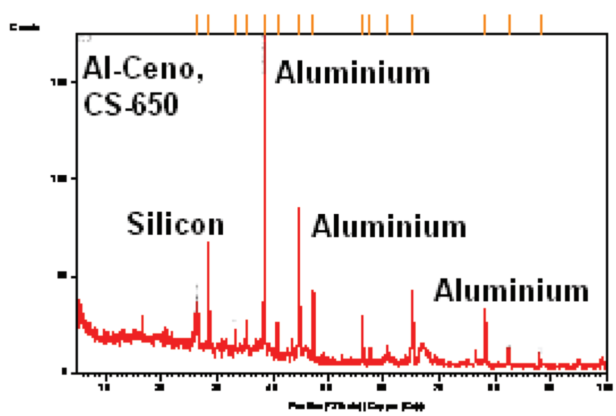

FIG. 2A XRD OF CONVENTIONALLY SINTERED COMPOSITE AT 650°C

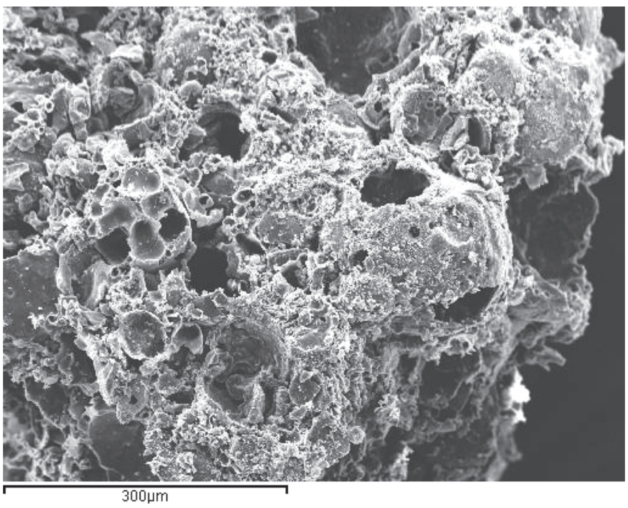

FIG. 3 MICROSTRUCTURE OF CONVENTIONALLY SINTERED COMPOSITE AT 850°C

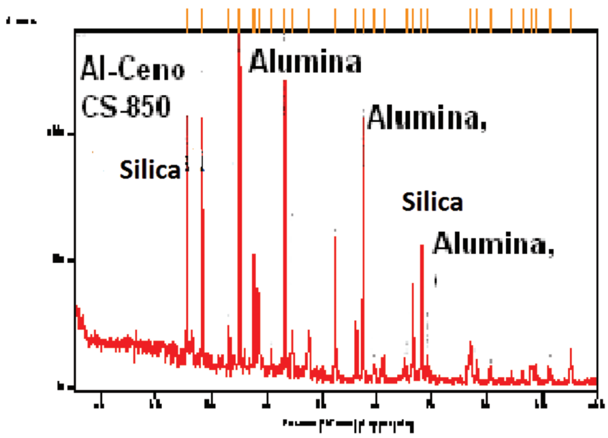

FIG. 3A XRD OF CONVENTIONALLY SINTERED COMPOSITE AT 850°C

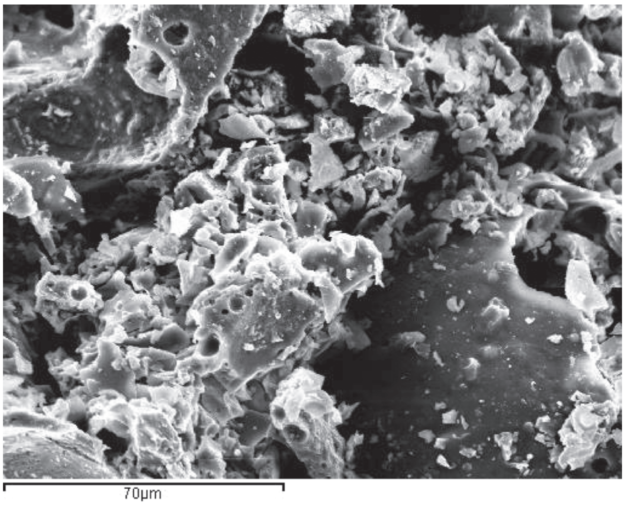

FIG. 4 MICROSTRUCTURE OF CONVENTIONALLY SINTERED COMPOSITE AT 950°C

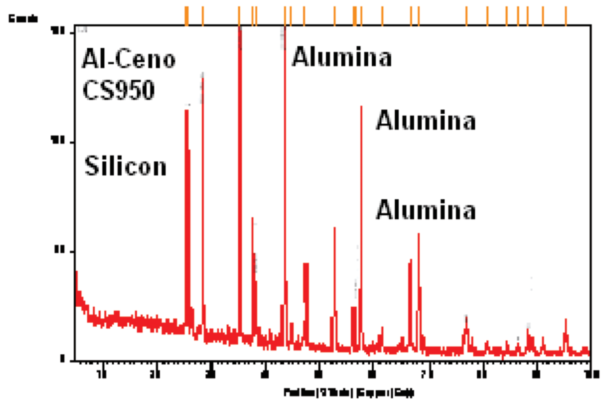

FIG. 4A XRD OF CONVENTIONALLY SINTERED COMPOSITE AT 650°C

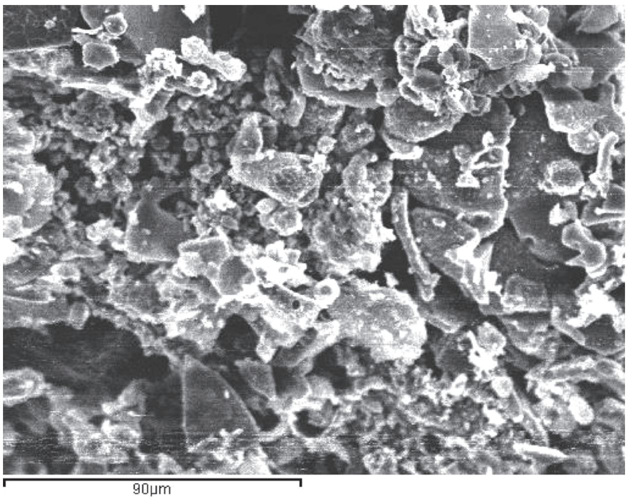

FIG. 5 MICROSTRUCTURE OF CONVENTIONALLY SINTERED COMPOSITE AT 1050°C

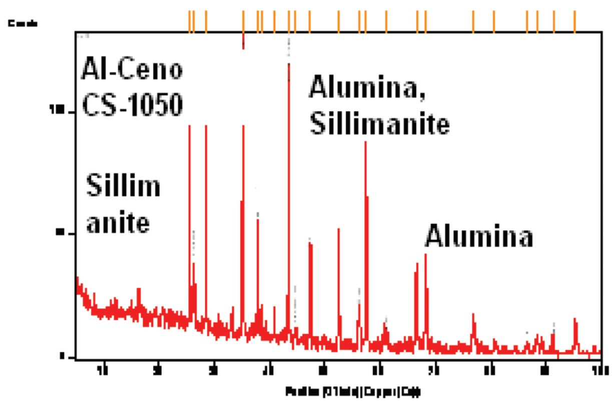

FIG. 5A XRD OF CONVENTIONALLY SINTERED COMPOSITE AT 1050°C

Further, sintering the composite to 950°C (Figure 4) shows that the cenosphere particle almost losing its spherical shape. The corresponding XRD graph (Figure 4a) shows the formation of Alumina as the major phases. There is shift in the 2θ values from 38-2θ degrees to 27-2θ degree values with formation of more crystalline phases which are represented by the peaks in the XRD pattern. On further sinteringthe composite to 1050˚C (Figure 5) the SEM picture shows fragments and chunks of sintered composite mass which appears porous. The XRD graph shows (Figure 5a) the formation of Sillimanite (Al_2SiO_5), an Alumino-Silicate, and Alumina(Al_2O_3) as the major phases which have formed at high temperature during sintering of the composite.

Microstructure & corresponding XRD graphs of Conventionally Sintered composites Figure 2) 650°C, Figure 3) 850°C, Figure 4) 950°C and Figure 5) 1050°C

3.2 SEM and XRD Analysis of Microwave Sintered composite:

The SEM and the XRD image of the microwave sintered composite at 650°C has already been discussed (Figure 1) which showed sphericity of the cenosphere particle which is intact, with pores on the cenosphere surface and the surrounding

Aluminium powder appearing as flakes. From the corresponding XRD graph (Figure 1a) it showed that the predominant compounds present are Aluminium (Al) and Mullite ($Al_6Si_2O_{13}$) and Silicon(Si).

Further sintering of the composites at 850°C (Figure 6) shows the beginning of the crumbling of the shape of the cenosphere particle with laminations appearing on its surface. The increased temperature has diminished the spherical surface of the cenosphere. The corresponding XRD graph shows (Figure 6a) the transformation of Corundum and Mullite ($Al_6Si_2O_{13}$) phases into Alumina (Al_2O_3) and recrystallized Silicon (Si) phases.

On further sintering of the composite at 960°C (Figure 7), it can be seen that the shape of the cenosphere particle occur as chunks and shrunk porous mass. The further increase in the temperature has continued to diminish the sphericity of the cenosphere. The XRD graph shows (Figure 7a) reveals the formation of Alumina and Corundum as the predominant phases. There is a shift in the 2θ values from 38-2θ degrees to 18-2θ values. More crystalline peaks are observed predominantly throughout the XRD pattern.

Further the SEM micrograph of the aluminum-cenosphere pellet sintered at 1050°C (Figure 8), shows the shape of the cenosphere particle occuring as chunks of crumbled mass which has been further reduced to fragments. The corresponding XRD graph (Figure 8a) shows the formation of Corundum a high temperature form of Alumina, appearing as a major phase in the composite structure.

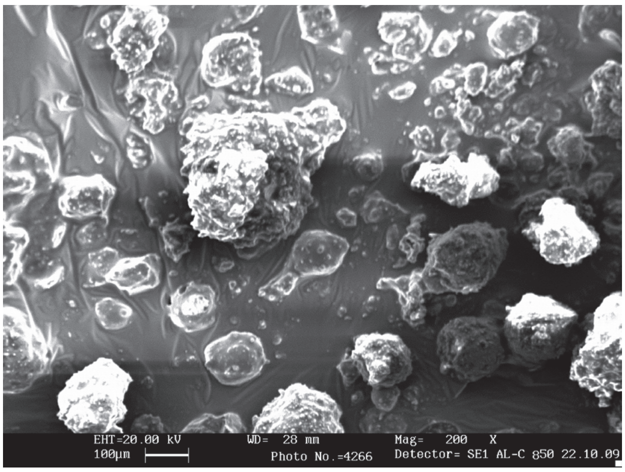

FIG. 6

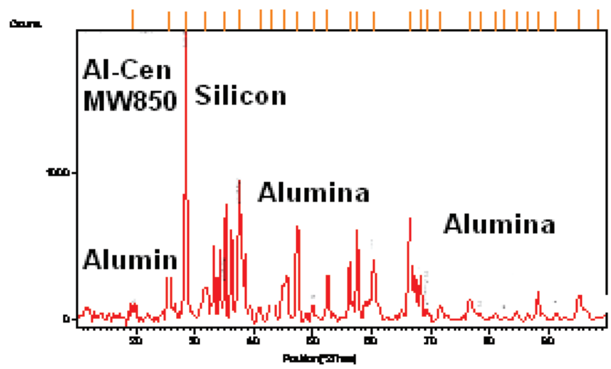

FIG. 6A

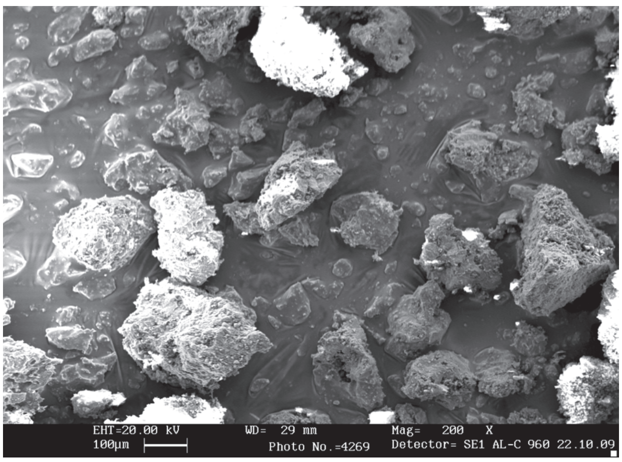

FIG. 7

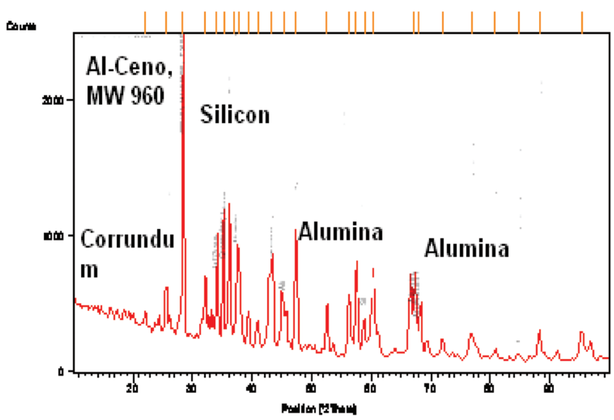

FIG. 7A

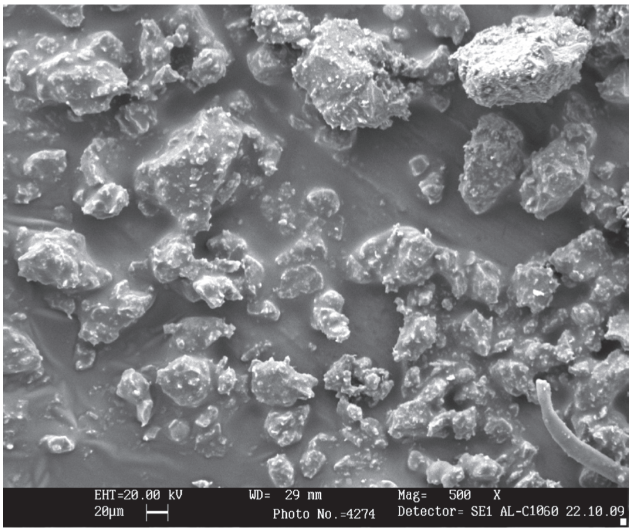

FIG. 8

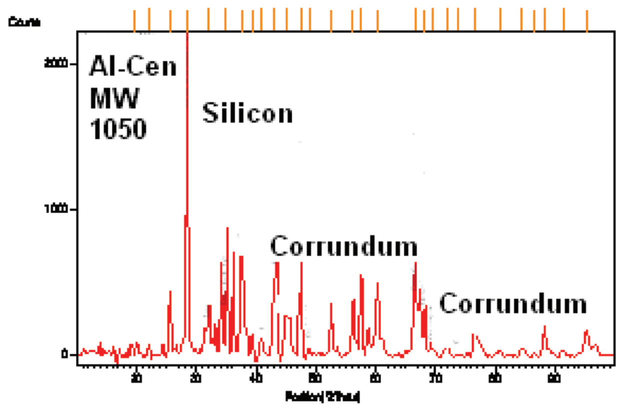

FIG. 8A

Microstructure & corresponding XRD graphs of Microwave Sintered composites at 6) 850°C, 7) 960°C and 8) 1050°C

3.3 Properties of conventionally and Microwave sintered composite samples:

The results of the Bulk Density(g/cc),Apparent Porosity (%), and Brinell Hardness Number [BHN] of the conventional and microwave sintered samples are depicted in Table 2.

TABLE 2

COMPARISON OF PROPERTIES OF MICROWAVE AND CONVENTIONAL SINTERED COMPOSITES SAMPLES

Temperature [°C]	**Apparent Porosity (%)**	**Brinell Hardness Number [HB]**	**Bulk Density [g/cc]**
Microwave sintered composites			
650	26.4	14	1.43
Conventionally sintered composites			
650	38.0	12	1.10

The Brinell Hardness Number (BHN) was calculated as follows:

$$BHN = 2\,P / (\pi\, D\, (D - (D^2 - d^2)^{1/2})) \qquad ....(1)$$

where P = load on the indenting tool (kg) on the sample, D = diameter of steel ball (mm), d = measure diameter at the rim of the impression (mm)

The Apparent Porosity (P_a) of the samples was calculated by water immersion method using Archimedes's principle.

$$P_a\,\% = \frac{(m_3 - m_1)}{(m_3 - m_2)} \times 100 \qquad ....(2)$$

Where m1 is the mass of a dried sample in air (g); m2 is the mass of the saturated suspended weight of sample in water (g); m3 is the mass of the sample saturated with water.

4.0 OBSERVATIONS

It is observed from Table 2 that as the temperature of sintering increases, there is increase in bulk density and hardness with reduction in apparent porosity in the sintered samples.

The Porosity (%) values of micro waves inter edcomposites are lower when compared to conventionally sintered composites. The Brinell Hardness Number of microwave sintered composites is higher compared to conventionally sintered composites.

At higher temperatures of sintering, pores become more closed because micro pores vanish during the sintering process of the material. The sintering temperature and the process influence the bulk density, mechanical strength, thermal stability, porosity and shrinkage of the samples [9]. The development of physical and mechanical properties is related to the phases formed due to reaction sintering between alumina and alumino-silicates and formation of compact microstructure [10].

Material density is an important characteristic for predicting mechanical properties and permeability. As the density increases (and porosity decreases), the mechanical properties also increase with decrease in permeability[11].

It is observed that there is formation of complex oxides in the composite matrix when sintering at higher temperatures beyond 650°C. The same has been observed by other authors, who have reported that the thermodynamic analysis of the Aluminum Cenosphere composite indicate a possibility of chemical reaction between the Alumium melt and Cenosphere particles leading to reduction of alumina, silica and iron oxide during their contact with the melt. The elemental Si formed by the reduction reaction would alloy with the matrix and that the Gibbs free energy and the heats of reaction of this reaction are highly exothermic in nature. As a result greater amount of eutectic silicon is seen in the composite and the chemical reaction indicates the increase in silicon level in the matrix [12]. This may be possible since the XRDgraphs of this study also indicate the presence of Silicon and Alumina peaks in the high temperature sintered samples.

5.0 CONCLUSION

1. Aluminum metal matrix composites can be fabricated at a temperature of about 650˚C. The matrix formed is that of pure Aluminum metal without undergoing oxide formation. The filler Cenospheres are evenly dispersed throughout the matrix with its shape intact.
2. It is observed that the aluminium metal powder reacts with Cenosphere to form alumino-silicates and alumina (Al_2O_3) at temperatures beyond 650˚C and above in the oxidizing atmosphere. Beyond this temperature the Cenosphere has undergone self-sintering to become complex oxides the matrix also seizes to be metallic. The matrix formed is a complex mixture of various alumino-silicates and high temperature oxides.
3. Microwave sintering can be an adoptive & effective rapid sintering method for development of Aluminium- Cenosphere composites fabricated through powder metallurgy route at lower temperatures.
4. The microwave sintered Aluminum Cenosphere metal matrix composite samples showed better physical and morphological properties when compared with the conventionally sintered ones. The Microwave sintered samples showed decrease in Apparent Porosity by 44%, increase in Hardness by 17%, and increase in Bulk Density by about 30% compared to the conventionally sintered samples.
5. The microwave sintering has also shown that the sintering takes place uniformly throughout the bulk of the material. The sintering process is rapid, has high heating rates, reduced processing times, uniform temperature throughout with minimal thermal gradients.

ACKNOWLEDGEMENT

The authors thankfully acknowledge Sri. N. Murugesan, Director General, Central Power Research Institute, Bangalore for according permission to publish this paper. Thanks are also to Mr. S. Vynatheya, Scientific Officer and Ms. Lakshmi, GOI Trainee for their help in sample preparation and materials characterization.

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