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DEVELOPMENT AND CHARACTERIZATION OF ALUMINA/ALUMINUM CO-CONTINUOUS COMPOSITE BY REACTIVE MELT INFILTRATION TECHNIQUE

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ABSTRACT

Present research focuses on the development of Alumina/Aluminum Composite. The intended composites are a new class of materials having wide ranging applications in automotive and aerospace industries. From the extensive reviews it was considered appropriate to employ reactive melt infiltration technique to develop these composites. The composites can be developed in open and chosen reaction environments. Effect of temperature and composition of aluminum were mainly considered in their development. The composites so developed were characterized using analytical facilities such as XRD and SEM. It has been observed that experimental findings are in line with the technical information already published in research articles.

Keywords: *Co-Continuous Composites, Diatomite Earth, Silica Refractory Brick, Aluminum-8% Magnesium Alloy, Reactive-melt Infiltration*

INTRODUCTION

Alumina/Aluminum co-continuous composites are the class of composites which is also known as co-continuous ceramic composites or C4. This is a new class of advanced composite materials. These composites are used in the manufacturing of automotive and aerospace components. These composites have high class properties like light in weight, higher value of stiffness, three dimensional properties, electrical conductivity and strength.

Co-continuous ceramic composites were originally developed (Breslin et al. 1995). These materials were initially produced by dipping porous silica preform in molten aluminum without the application of external pressure. These composites have the capability of near net shape. When two materials (silica and molten aluminum) come in close contact, the liquid aluminum reduces the silica to silicon and converts itself to alumina and aluminum network with silicon which is reduced from the silica. The new born material contains 70% alumina and 30% aluminum. The ceramic phase provides 'stiffness and strength' and the aluminum provides toughness to the material (Zhang et al 1994). In order to fabricate these composites many methods have been developed like melt infiltration, casting, vacuum infiltration and Primax process (Ray 1993, Urquhart 1991). For the purpose of mass production the liquid melt infiltration of porous ceramic is best route as compared to methods. Various reinforcements like Al_2O_3, TiC, AlN, SiC have been used to study the effect of infiltration at various temperatures. Likewise the intended composite is produced by using a porous ceramic sample with natural porosity (Xi and Yang 1996, Chen and Chung 1996, Toy and Scott 1990, Han and Feng 2000, Han et al 1997).

In the fabrication process of this composite, the silica porous preform is immersed in the liquid aluminum for certain period of time and after cooling characterized for its mechanical properties like compression test, tensile test, fracture toughness, hardness and modulus of rupture etc. The silica glass reacts with the molten aluminum and forms $Al_2O_3 \cdot Al(Si)$ which has the ceramic phase as high as 60%. The chemical reaction is found to be very slow below 800°C. Above this temperature the metal infiltrates the porous silica preform and makes co-continuous composite of alumina and silicon. The infiltration rate increases noticeably around 800°C at the rate of 3 mm/hour. It was previously shown that it is because of the formation of γ–alumina at this temperature rather than α-alumina which is formed at 1100°C (Raj and Thompson 1994). And it is quite strange that at the end of infiltration at 800°C, the phase formed is α-alumina. It is so because γ–alumina is an intermediate phase which enhances infiltration. The speed of infiltration also depends upon the volume of molten aluminum available at that moment (La Vecchia et al 2003, Aghajanian et al 1991).

In the current work a porous ceramic body with two different porosity levels and capillary sizes were infiltrated with pure liquid aluminum and

Al-8%Mg alloy. One of the ceramic bodies was the insulating porous refractory brick and the other was sintered diatomaceous earth. The infiltration was carried out at different elevated temperatures in reducing and open atmospheres by employing reactive melt-in technique. Thereafter the composites were characterized for hardness, microstructure, depth of infiltration and continuity of metal.

Experimental

The composite samples were produced at different temperatures and the temperature, at which the metal was melted and impregnated the porous silica refractory samples to produce the composite, was also noted. The raw materials used in this process were: pure aluminum, aluminum–8% magnesium alloy, porous silica brick, diatomite, pure graphite, coal (powder), coke. The process was carried out under open and reducing atmospheres. X-ray diffraction (XRD) was carried out to analyze the chemical composition of the ceramic samples.

Reactive melt infiltration method was used to develop the C-4 composite. Initially the Al-8% Mg alloy was produced by melting and mixing pure aluminum with 8% magnesium. Pure aluminum and Al-8% Mg alloy were cast into a cylinder of suitable diameter to make its melting and infiltration possible in the current process. The pieces of one inch length were cut out from both cylinders. Then a circular cavity (of the same diameter and height as of the ceramic samples) of 12 mm diameter and 6mm depth was cut in each 1-inch sliced cylindrical length so that the ceramic samples can be fit into it. One kind of ceramic sample was prepared by cutting from the porous brick and the other by pressing and sintering the diatomite powder at 900^{o}C. These ceramic samples were placed into the metallic sample's cavity, one in each metallic sample, and were covered with the same metallic sliced plates as shown in Figure-1. The physical data of the sample to be infiltrated is mentioned in Table-1. Thereafter these assemblies were placed in graphite crucibles and heated in furnaces to different temperatures in reducing and open atmospheres. The temperature profiles at which infiltration was carried out is given in Table-2. Physical data of insulating refractory brick is given in Tables-3 and for diatomite is mentioned in Table-4.

During heating at different temperature regimes as melting of the metal progressed and the very same infiltrated into porous refractories.

Thereafter the samples were taken out to cool and rates and depths of infiltration were investigated. Scanning electron microscopy (SEM) was used for the microstructural investigation of these samples.

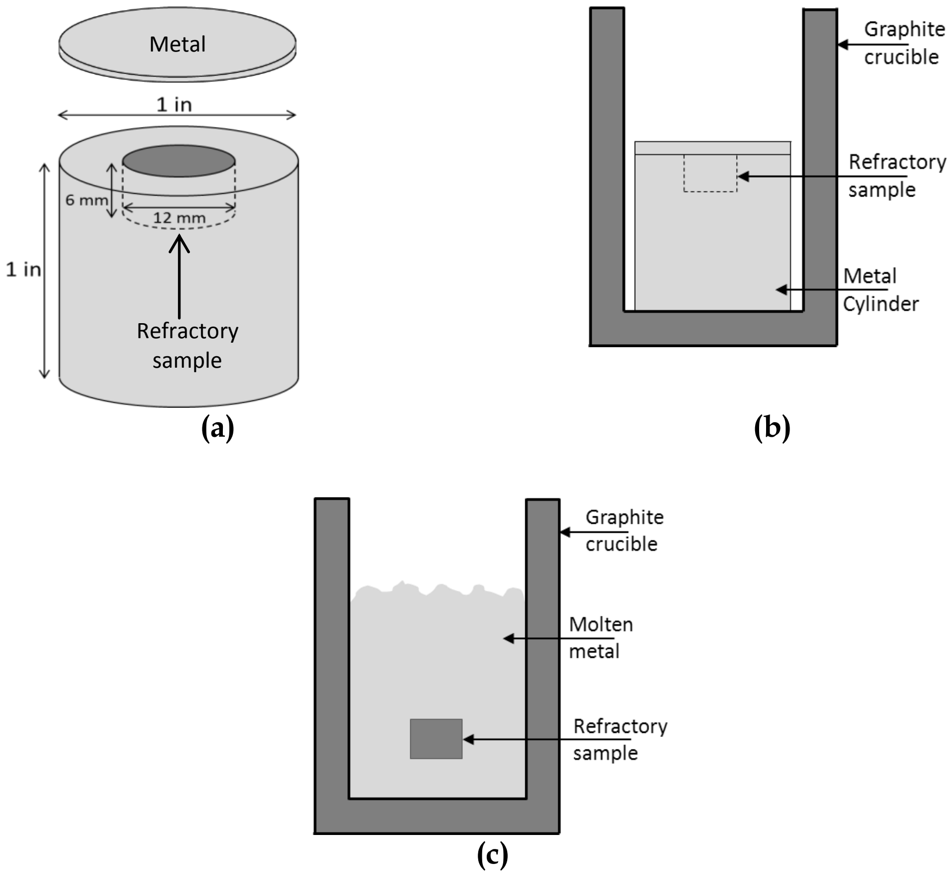


Fig-1: (a) Schematics of the sample's assembly used in the process, (b) The sample assembly in a graphite crucible, (c) The refractory sample being infiltrated by the molten metal.

Table-1: Physical data for the samples to be infiltrated

	Refractory Brick	Diatomite
Diameter	12 mm	12 mm
Height	6 mm	6 mm
Sintering Temp	900°C	900°C
Sintered Density	0.688 g/cm^3	1.06 g/cm^3

Table-2: Heating temperatures for porous refractory brick and diatomite for reactive melt-in infiltration

Heating temperature for porous refractory brick for infiltration with pure Al and Al-8% Mg alloy	950°C	1150°C	1300°C	1450°C
Heating temperature for sintered diatomite sample for infiltration with pure Al and Al-8% Mg alloy	750°C	850°C	950°C	1000°C

Table-3: Physical data of insulating refractory brick

Composition	80% silica, 20% alumina
Bulk Density	0.688 g/cm^3
True Density	1.292 g/cm^3
%age porosity	46%

Table-4: Physical data of diatomite earth

Composition	Silica 89.70%, Alumina 3.80%, Red Iron Oxide 1.3%, CaO 0.4%, TiO_2 0.2%, MgO 0.43%, Na_2O+K_2O 2.5%
Bulk Density	0.35 g/cm^3
True Density	2.2 g/cm^3
%age Porosity	84.1%

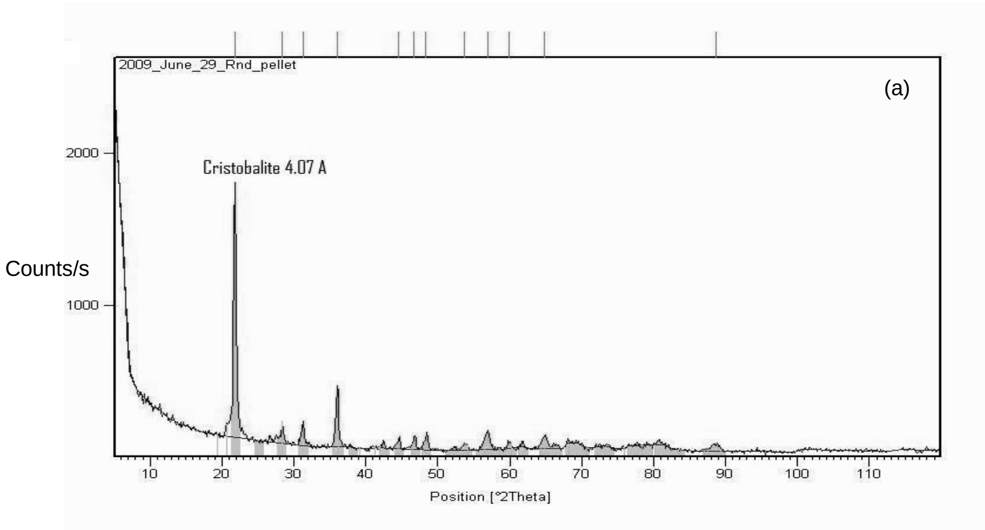
RESULTS AND DISCUSSION

XRD Analysis of Refractory Samples

Figure 2 shows the XRD analysis of diatomite and porous silica refractory brick samples. It is clear from XRD patterns that the major content is silica which is in the form of cristobalite. It is a common and stable form of silica with d = 4.07Å.

Infiltration of Pure Aluminum

No infiltration evidence was found uptil 1450°C in both refractory samples. This was due to aluminum which readily oxidized even in reducing atmosphere created by the fine carbon dust over the liquid surface of aluminum. The ceramic as well as aluminum cylinder plus lid remained uncreated with each other even at such a high temperature.



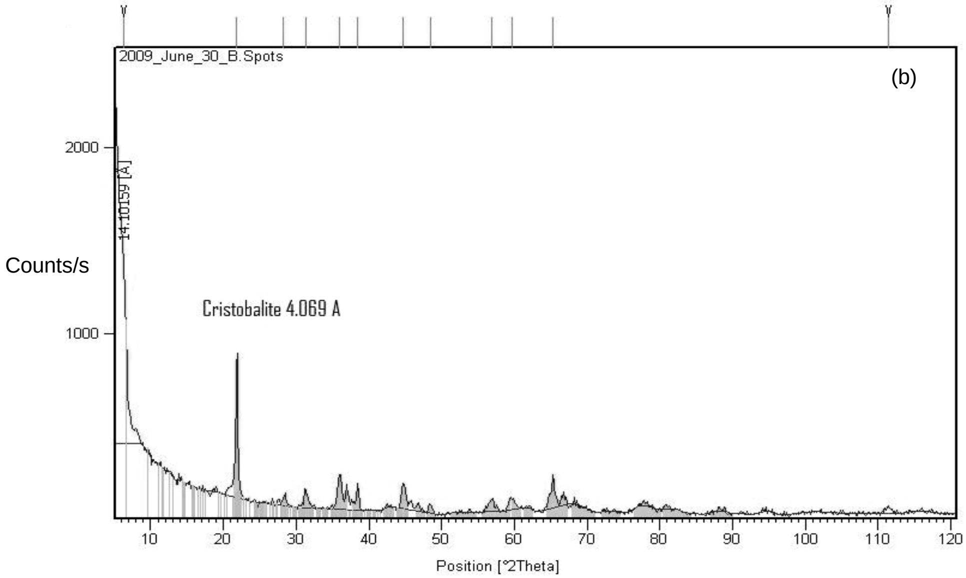


Fig-2: XRD Patterns for Diatomite round pallet (a) and round porous silica brick (b) showing major content silica in the form of cristobalite at d = 4.07Å.

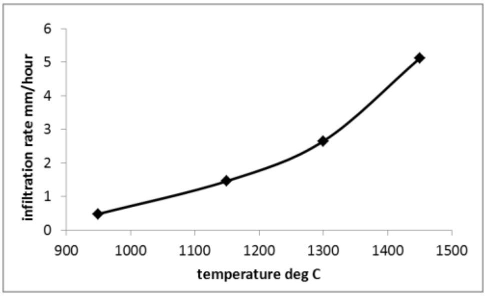
INFILTRATION OF Al-8% Mg ALLOY

In Refractory Brick

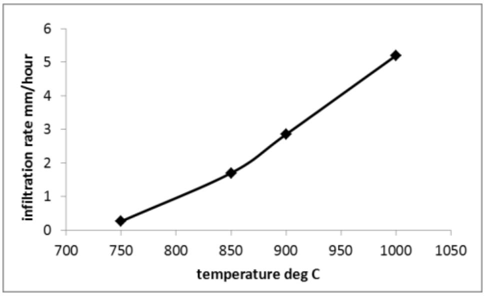
When the samples of porous silica refractory brick were infiltrated, the infiltration rate was different at different temperatures as shown in Figure 3(a). The results show that the infiltration rate increases with the increase in temperature. No infiltration was observed without lid. Results were better in the open atmosphere rather than in reducing atmosphere. The average hardness value was noted to be 350 HV.

In Diatomite Earth

The infiltration rates in diatomite earth were determined and presented graphically in Figure 3(b). There is clear indication that infiltration rate is fairly faster in diatomite as compared to the refractory preforms. No infiltration was observed without lid. The average hardness was noted to be 503 Vickers. The results were better in open atmosphere.



(a)



(b)

Fig-3: Effect of temperature on infiltration rate (a) in porous silica refractory brick samples, (b) in diatomite samples.

The Ceramic Network

Figure 4 shows the SEM micrograph of the ceramic network. The dense mass seen in the micrograph is alumina in nature formed by the chemical reaction of aluminum alloy and the ceramic network. The grey area is the dense alumina and the white spot is the silica particle that remained unreacted during the infiltration process.

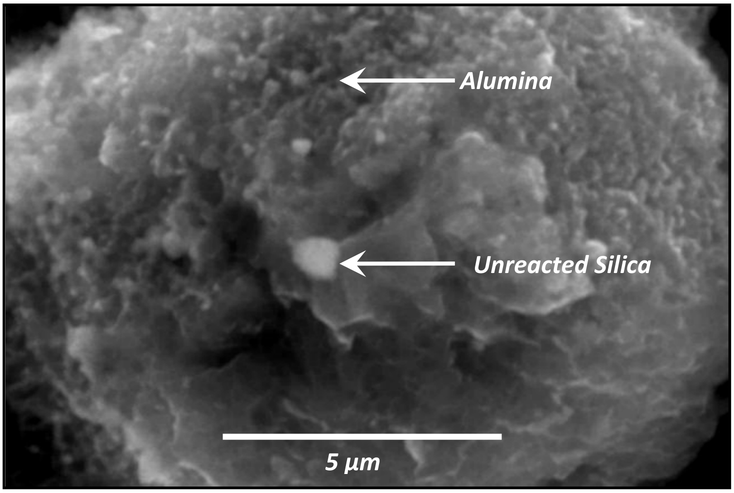


Fig-4: Scanning electron micrograph of the ceramic network, showing dense alumina (grey phase) and white unreacted silica.

Metallic Network

Figure-5 shows the dense mass which is aluminum composite in nature formed by the chemical reaction of aluminum alloy and the ceramic network. Initially it was Al-Mg alloy but after reaction, the silicon coming from the reduction of the silica also entered into the metallic phase changed its composition by the supply of silicon.

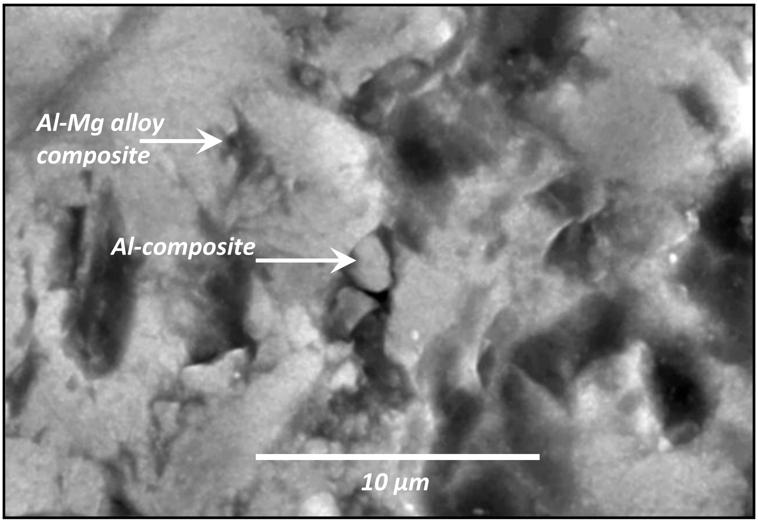


Fig-5: Scanning electron micrograph of the metallic network, showing Al-Mg alloy and Al-composites.

Interface of Metal and Ceramic

Figure 6 shows the interface of the two network regions. The lighter area is the metallic phase and the darker area is the ceramic phase. Both regions are quite compacted and interpenetrated as it is clear from the micrograph.

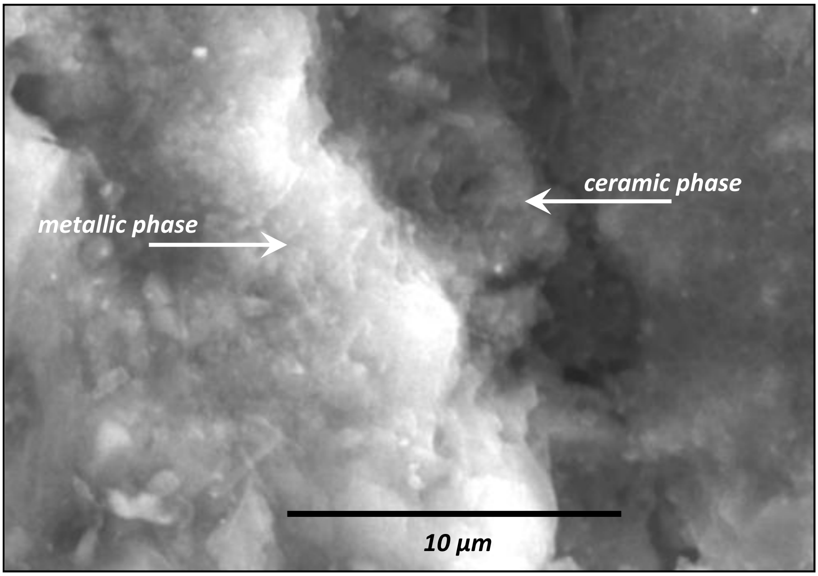


Fig-6: Metal–Ceramic interface. The light area is metallic phase and the dark area is ceramic phase.

Infiltration of Pure Aluminum to Porous Silica Refractory

In case of pure aluminum there was no observation of aluminum metal infiltration into the porous refractory preforms even at elevated temperatures. This is due to the high affinity of aluminum towards oxygen. When aluminum is melted in the open atmosphere, it readily gets oxidized and forms a thin protective layer on its surface, which not only protects it from further reaction with the atmosphere but also with the silica refractory. Because both of these (alumina and silica) are ceramics in nature and their parent metallic substances, aluminum and silicon, are already in their stabilized form in the form of oxides, therefore infiltration was not observed in this case. Secondly the reaction of aluminum under the protective surface of coke dust was also impossible because aluminum has much more chemical affinity towards oxygen.

Infiltration of pure Aluminum to Diatomite

The infiltration is dependent on the physical adherence, chemical reactivity, and capillary size of the porous ceramic mass. Again there was no infiltration in this porous mass. The pore size was very fine in this case because these are naturally occurring pores made by the living unicellular diatom millions of years ago under the sea water bed and then buried in the form of huge lumpy mass. The pores size is in nanometers so infiltration was difficult to proceed.

Infiltration of Aluminum-Magnesium Alloy to Porous Silica Refractory

As previously discussed the infiltration depends upon the physical, chemical adherence and capillary action. So in order to increase the chemical reactivity of the metallic phase it was alloyed with 8% magnesium. Magnesium was chosen because of its high chemical reactivity and good alloying nature with aluminum. This process showed very good infiltration. The infiltration in porous silica refractory preforms can be attributed to lower viscosity of the alloy, higher chemical reactivity of the alloy, lower melting point, fine capillary action, temperature and pressure of the metal being infiltrated. The exact mechanism is that first magnesium attacks the refractory and creates some potential surface area to be further attacked because of its high reactivity. As evidenced in the published literature the exact mechanism is that first magnesium attacks the refractory and creates some potential surface area which is to be further attacked because of its high reactivity (Han and Feng 2000, Han et al 1997, La Vecchia eta al 2003).

Infiltration of Aluminum-Magnesium Alloy to Porous

Diatomaceous Earth

The infiltration rate was higher than the previous case because of fine capillaries. The diatomaceous earth has very fine capillaries upto nano level. Under the simple optical microscope the microstructure was not visible because of the fineness of the capillaries. That's why its hardness was taken as average. In the micrographs if we look at the interface of the metal and ceramic it is quite compacted to each other.

CONCLUSIONS

- The co-continuous ceramic composites can be produced in the open atmosphere.
- Diatomaceous earth as the ceramic porous phase has proven to be a better option for the development of C-4 composites.
- The infiltration rate can be enhanced by increasing the temperature of the process.
- Reducing atmosphere is not required for the production of C-4 composites.
- The cost of the production of C-4 composites is less by using reactive-melt infiltration technique.
- The equipment used for the development of C-4 composites is simple.
- Technology development and transfer is fairly easy.

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