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Investigation of Ceramic-Metal Composites Produced by Electroless Ni Plating of AlN- Astaloy Cr-M

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Abstract**—**The microstructure, mechanical properties and metalgraphic characteristics of Ni plated AlN-Astaloy Cr-M powders were investigated using specimens produced by tube furnace sintering at 1000-1400 °C temperature. A uniform nickel layer on AlN powders was deposited prior to sintering using electroless plating technique. A composite consisting of ternary additions, metallic phase, Ni and ceramic phase AlN within a matrix of Astaloy Cr-M had been prepared under Ar shroud and then tube furnace sintered. The experimental results carried out by using XRD (X-Ray Diffraction) and SEM (Scanning Electron Microscope) for composition (10% AlN-Astaloy Cr-M) 10% Ni at 1400 °C suggest that the best properties as 132.45HB and permittivity were obtained at 1400 °C.

Keywords**—**Composite, Electroless, Nickel plating, Powder metallurgy, Sintering.

I. INTRODUCTION

A good combination of high strength and ductility of the aluminum based metal matrix composites (MMCs) have introduced the material to a wide area of possible advanced applications [1]. Metal–matrix composites are of high interest for the applications in aerospace, automotive and electronics industries [2]. Porous materials have been widely applied in many industries such as aeronautics and aerospace, medical, metallurgy, construction, and mechanics etc. [3]. Among these materials, the Fe-Al intermetallic compounds play the dual role of both structural and functional materials because this porous material not only has superior high temperature oxidation and corrosion resistance of the intermetallic compound [4] The powder metallurgy process has already been used to fabricate Al–AlN composites with a low volume fraction of AlN (< 30 vol.%) [5], [6]. Aluminum Nitride (AlN), a covalently-bonded ceramic, does not occur naturally, and it is synthesized from the abundant elements aluminum and nitrogen. This unusual combination of properties makes AlN a critical advanced material for many future applications in optics, lighting, electronics and renewable energy [7].

AlN powder filler plays an important role in producing next-generation, high-performance resins (e.g., those with high thermal conductivity of around 10 W/m-K, as well as electric insulation). These materials are in demand in order to satisfy the strong need for heat dissipation in modern high-power consumption electronic devices, such as electric vehicles (EVs) in the automobile industry, high-power LED lamps, and other thermal sheets/greases for chip packages [8]. Composite electroplating is a method that involves codepositing fine particles of metallic, non-metallic compounds or polymers in the plated layer to improve material properties such as wear resistance, lubrication and corrosion resistance [9]. The composite coatings of Ni–AlN exhibit higher hardness, less friction and better wear resistance than that of pure nickel coatings which were prepared under the same electrodeposition conditions. The incorporation of AlN particles decreases the thermal expansion and improve the oxidation resistance of the composite coatings [10].

Electroless nickel composite coatings containing submicron aluminum nitride particles. To prepare the composite coating, the selection of second phase particles is also equally important. Aluminum nitride is well known for its superior wear resistance, low coefficient of friction, higher hot hardness, good resistance to high temperature oxidation as well as to aqueous corrosion. Hence, systematic studies were carried out to prepare Astaloy CrM-AlN composite coatings by electroless deposition method [10]. Plain Ni coatings were also prepared for comparison. Deposits were characterized for their structure, phase transformation behavior and hardness at various heat treatment temperatures. At crystallization temperatures deposits were heat treated and analysis was carried out to find out the phases formed. Grain size has also been calculated for as-deposited and heat treated coatings at crystallization temperatures.

II. EXPERIMENTAL METHOD

Starting powders employed in this study were as follows: the purity of 99.8% for Si_3N_4 and Astaloy Cr-M powders with a particle size lower than 50 µm, and AlN ceramic-metal composite was produced by electroless Ni plating of Astaloy Cr-M and AlN powders.

Ni plating was achieved by suspending the starting Astaloy Cr-M and AlN powders in a Ni containing solution ($NiCl_2.6H_2O$) at 90-95 °C and by adding Hydrazine Hydrate ($N_2H_4.H_2O$) and 35 vol.% Ammonia solution while keeping the pH at 9-10. With increasing temperature, ammonia evaporation rate increased rapidly; therefore, a dripper was used to add more ammonia for adjusting pH of the plating

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solution. In the meantime, the solution was continuously stirred and the pH was constantly monitored by using a Philips PW 9413 Ion-Activity Meter. The reaction was allowed to continue until sufficient Ni was added for plating all the Astaloy Cr-M and AlN powders, then, Ni plated Astaloy Cr-M and AlN powders were filtered out of the solution by using a paper filter and repeatedly washed off by distilled water and then oven dried at 105°C. and then followed by sintering at 1000-1400 ºC for 2h using a tube furnace.

Astaloy Cr-M powder composition is given in Table I. From XRD results of specimens sintered at 1400 ºC, Fe_2Si intermetallic phase appears to be coexisting as two separate constituents.

TABLE I
CHEMICAL COMPOSITION OF ASTALOYCR-M (%)

Powder	Al_2O_3	F_3O	SiO_2	CaO	MgO	Na_2O	K_2O	TiO_2	LOI
AstaloyCr-M	55-60.35	19.95-22.85	3.15-6.95	0.27-3.90	0.02-0.04	0.06-0.16	0.12-0.27	2.66-3.27	9.7-11.58

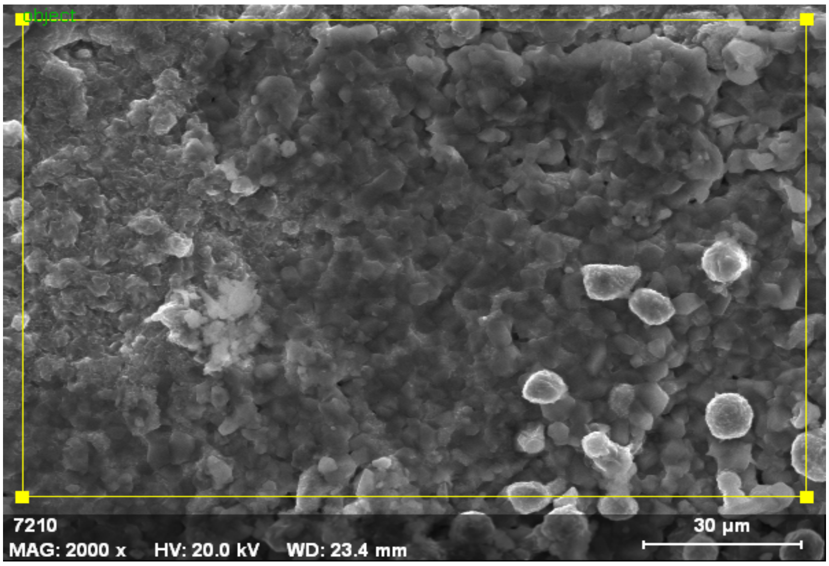

Fig. 1 SEM micrographs of as-received Ni Plated 80% Astaloy Cr-M + 10% AlN Mag.2kX 1400 ºC

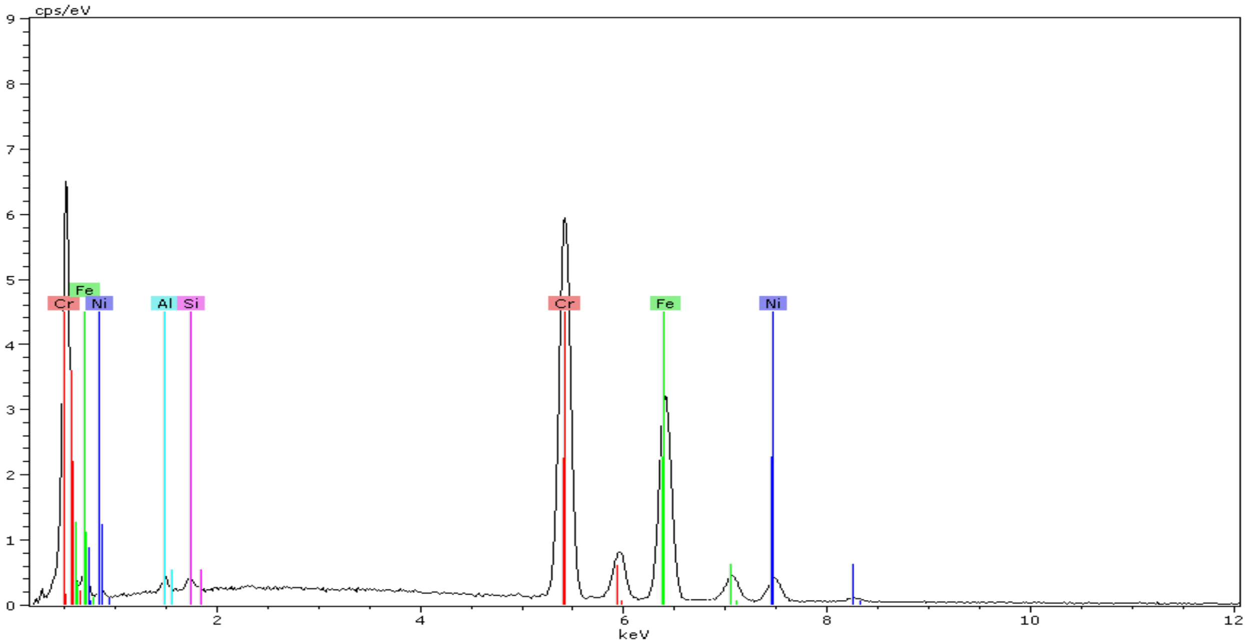

Fig. 2 The EDX elemental analyses with respect to sintering temperature

In the experimental study, the samples were prepared in AlN-Astaloy-Cr-M powders were plated by using the electroless nickel plating techniques. After that powders shaped in hydraulic press under 300 bar pressure. The shaped samples were sintered for two hours within the temperature range 1000, 1100, 1200, 1300 and 1400 ºC under argon gas atmosphere in conventional furnace. The sintered samples were prepared for the mechanical and metallographic analyses. The contents of the plating bath are given in Table II.

TABLE II
THE CHEMICALS OF NICKEL PLATING BATH AND THEIR RATIOS.

Chemicals	Conditions
AlN powders	10g
Astaloy Cr-M	80g
Nickel Chloride ($NiCl_2.6H_2O$)	40g
Hydrazine Hydrate ($N_2H_4.H_2O$)	20%
Distile Water	80%
Temperature (°C)	90-95
pH Value	9-10

The purity of 99.9% for AlN powders with a particle size is lower than 40μm. The composition of was calculated according to formula (10% AlN +80% Astaloy Cr-M)Ni, that is, (10%AlN+80%Astaloy Cr-M)Ni, specimens were prepared in 5 g cylindrical compressed pre-form. The mixture was shaped by single axis cold hydraulic pressing using high strength steel die. A pressure of 300 bar was used for the compacting all the powder mixtures. The cold pressed samples underwent for a sintering at 1000-1400 °C for 2 hours in a tube furnace using argon gas atmosphere. The specimens were cooled in the furnace after sintering and their hardness and shear strengths measurements were carried out using METTEST-HT (Brinell) hardness tester and Shimadzu Autograph AG-IS 100KN universal tensile tester machine, standard metallurgical specimen preparation was made to reveal the microstructure of the joints and powder compacts.

Shimadzu XRD-6000 X-Ray Diffraction analyzer was operated with Cu K-alpha radiation at the scanning rate of 2 degrees per minute. LEO 1430 VP model Scanning Electron Microscope fitted with Oxford EDX analyzer was used for the microstructural, and EDX compositional analysis was used for measuring metallographic properties.

The volumetric changes of (10% AlN+80% Astaloy Cr-M)Ni composite material after sintering were calculated by using (d=m/V) formula (Fig. 3). The volume of pre-sintered and post-sintered samples was measured with Archimedes principle. All the percentages and ratios are given in weight percent unless stated otherwise.

III. RESULTS AND DISCUSSION

A. Characterization of Specimens

In the study, the prepared and shaped (pressed) samples were sintered at temperatures ranging at 1000-1400 ºC in tube furnace and they were ready for the physical, mechanical and metallographic analyses. Density-composition change curve is shown in Fig. 3. The highest sintered density was achieved at 1400°C as 6.52 g/cm^3.

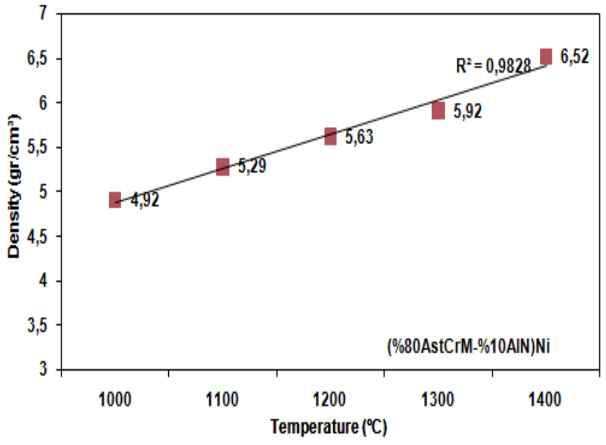

Fig. 3 The density change with respect to sintering temperature

The hardness-composition change diagram is shown in Fig. 4. The hardness values of the composite samples produced using tube furnace sintering technique within the temperature range 1400 °C from powders obtained as a result of plating (10% AlN+80% Astaloy Cr-M) Ni powders through electroless Ni plating method in tube furnace were given. According to this, the highest hardness value in the composite samples produced using electroless plating method was observed to be 132,45HB at 1400 °C.

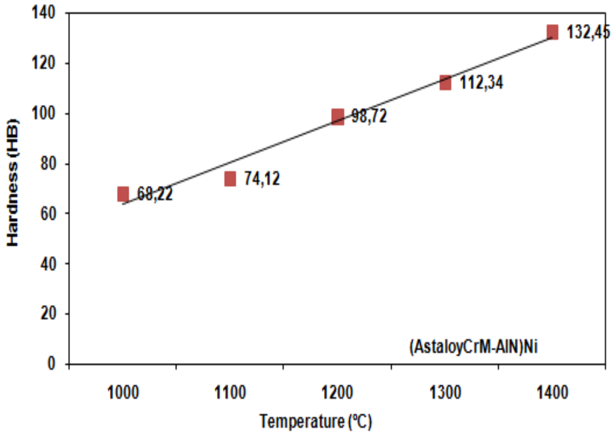

Fig. 4 The hardness change with respect to sintering temperature

B. Structural Analysis

After nickel plating process, whether the plating was achieved in (80% Astaloy Cr-M + 10% AlN)Ni powders or not was examined through SEM analysis. It was observed that grains were bonded to each other and the particles grew larger. In addition, there were pores exhibiting homogeneous dispersion among the grains.

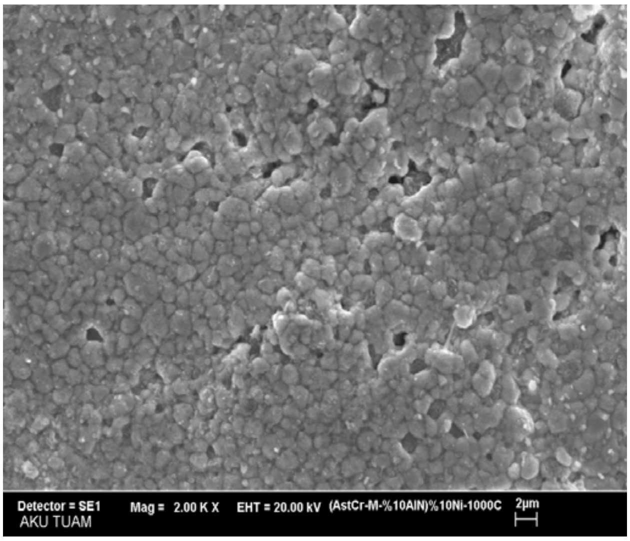

Fig. 5 SEM micrographs of as-received Ni Plated 80% Astaloy Cr-M + 10% AlN Mag.2kX 1000ºC

SEM and XRD analyses were carried out on the specimens to reveal the effect of Ni plating and characterize the phases present within the specimen. Fig. 6 shows Ni plated particles having a layer of Ni with low density of porosity before sintering process.

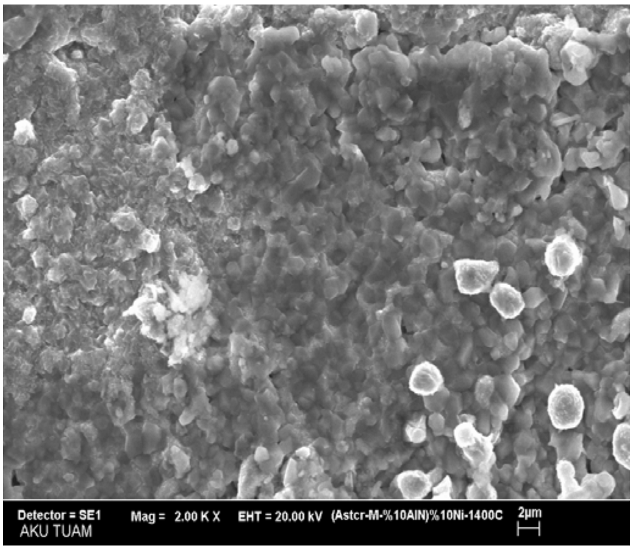

Fig. 6 SEM micrographs of as-received Ni Plated 80%Astaloy Cr-M + 10% AlN Mag.2kX 1400ºC

In Fig. 7, Fe_2Si, AIN, Ni, FeNi and FeC peaks can be seen in the XRD analysis from Ni Plated 80% Astaloy Cr-M+10% AlN at 1000 °C and Ni Plated 80% Astaloy Cr-M+10% AlN composite sintered in tube furnace at 1000 °C.

In Fig. 8, Fe_2Si, AIN, $FeSi_2$, Ni, FeNi and FeC peaks can be seen in the XRD analysis from Ni Plated 80% Astaloy Cr-M+10% AlN at 1400°C and Ni Plated 80% Astaloy Cr-M+10% AlN composite sintered in tube furnace at 1400°C.

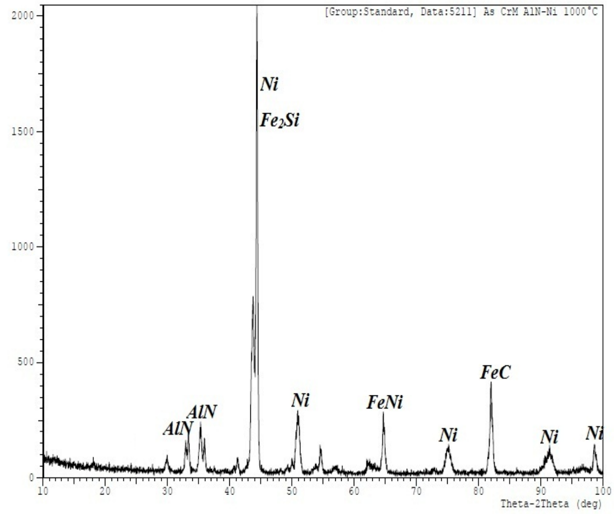

Fig. 7 X-ray diffraction patterns of the (80% Astaloy Cr-M+10% AlN) 10% Ni sintered at 1000ºC

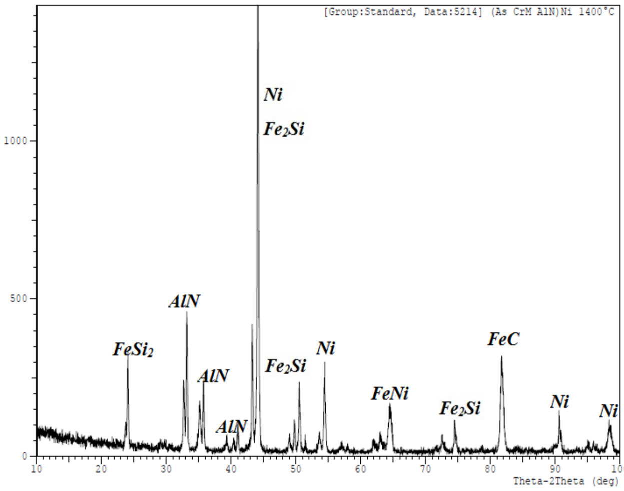

Fig. 8 X-ray diffraction patterns of the (80% Astaloy Cr-M+10% AlN) 10% Ni sintered at 1400ºC

80% Astaloy Cr-M+10% AlN powders were plated 10% Ni and then sintered in a conventional furnace within the temperature range 1000 ºC, 1100 °C, 1200 °C, 1300 °C, and 1400 ºC. After sintering, a considerable drop in the mechanical properties of specimens was observed. It was concluded that (80% Astaloy Cr-M+10% AlN) 10% Ni particles were occured by Fe_2Si, AlN, $FeSi_2$, Ni, FeNi and FeC phases at 1400 °C and Hardness test results suggest that (80% Astaloy Cr-M+10% AlN) %10Ni composite sintered at 1400°C shows Brinell hardness values respectively.

IV. CONCLUSION

The following results were concluded from the experimental findings:

- The highest density in composite made from (80% Astaloy Cr-M+10% AlN) 10% Ni powders sintered at different temperatures was obtained at 1400 °C. The highest density sample was found as 6.52 g/cm^3 at 1400°C.

- The highest hardness in (80% Astaloy Cr-M+10% AlN) 10% Ni composite samples fabricated using powder metallurgy method was found as 132,45 HB at 1400 °C.
- It was seen that best mechanical properties were observed (See Figs. 3 and 4) at 1400 °C for 80% Astaloy Cr-M+10% AlN) 10% Ni.

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