Effects of plasma parameters and collection region on synthesis of iron and nickel aluminide composite particles during thermal plasma processing

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Abstract. Iron and Nickel aluminide composite particles were synthesized by non-transferred DC plasma spray torch at atmospheric pressure. Irregular shaped ball milled, micron sized powders were fed in to the plasma flame using argon as carrier gas. The particles got molten and vaporized. The vapour condensed on the walls of the reaction chamber and nanoparticles were formed. The molten particles got spheroidized due to surface tension forces. Powders as formed were collected in the plasma reactor at three different sections (Section A, B and C). These powder particles were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The particle size and morphology of the composite particles strongly varied depending on the processing parameters and collection region. The results were discussed.

1. Introduction
Intermetallic composites are made of two or more metals or of a metal and a non metal. Aluminides of transition metals such as iron, nickel, titanium, niobium and cobalt are a few examples. Intermetallic bulk materials are used for a variety of high-temperature structural applications. Moreover, they offer properties useful in many applications; their low density, good electrical and heat conductivity and, in some cases, they are magnetic. There is particular interest in the production of intermetallic nanoparticles that could find successful applications in fields like metallurgy, aerospace industry, magnetic for bar coding, magnetic ink, cancer therapy, bio-physics, environmental pollution control and gas storage etc [1-3].

There are various methods of synthesizing composite particles such as mechanical alloying, hydrogen plasma metal reaction (HPMR), laser vaporization with controlled condensation (LVCC) etc [4-6]. Thermal plasma method has been effective method for synthesizing nano and spheroidized particles, because of very high temperature, high density and high quench rates possible in thermal plasma processing. In this paper deals with the preparation of iron and nickel aluminide composite particles.

2. Experimental Setup and Methodology
A dc non transferred plasma spray torch is used for generating thermal plasma. The torch is energized by a power supply with a maximum capacity of 40 kW DC power. It has an open circuit voltage of 150 V with 100% duty cycle. The maximum current drawn could be 800 A DC and it can be cooled by forced air. Plasma reactor consists of a stainless steel double walled water cooled cylindrical chamber.
A non transferred plasma spray torch is fixed on the top of the reaction chamber and the torch nozzle goes inside the chamber coinciding with its axis. The nozzle part for powder feeding is just outside the cylinder and the nozzle exit is inside the cylinder. The bottom of the reaction chamber sits vertically through a flange on a collection chamber cylindrical in shape and kept horizontally. The reaction chamber and collection chamber are double walled and cooled separately. The schematic of the experimental set up is shown in figure 1.

Argon and argon-nitrogen were used as plasma forming gases with argon as carrier gas. Plasma was generated by striking an arc between the electrodes. Plasma is pushed out through the nozzle due to high velocity of the gas. The plasma flame outside the nozzle is called the plasma jet. The dimensions depend on power and gas flow rate. For a stable plasma jet the input power was adjusted. Typical operating parameters are given in table 1. Using argon as carrier gas, the powders were fed into the plasma flame. The particles heated by the plasma get accelerated. The powder particles when gain thermal energy during in-flight, start melting. The individual particles of the starting powder have different trajectories in the plasma flame and different temperature histories. When the temperature increases the melting percolates from of the surface to the deeper layers of the particle.

In the formation of nano powders, however, evaporation of small grains followed by homogeneous nucleation from the vapour phase and grain growth. Quenching is seemed to be the dominant process during the same. In this process the particles are partially or fully molten. The smaller size particles get completely evaporated, whereas surface evaporation occurs in the case of bigger particles. Due to the cooling of the chamber the vapour condenses on the chamber walls as nanoparticles. The nano powders were collected from two different regions in the chamber walls i.e. section A and section B which are 30 cm to 60 cm from the nozzle exit.

Large particles are partially or fully melt. These particles get accelerated through the plasma flame and leave with a high velocity. Inside the chamber, they transverse through the atmospheric air. The particles are quenched and get solidified. It is then collected in the bowl at the bottom of the chamber (section C). All the particles were not uniformly molten while passing through the plasma flame, apparently because different particles take different paths through the flame. It is necessary to subject the particles to an enhanced water-quenching treatment to ensure complete melting and homogenization. The schematic of plasma water quenching process is shown in figure 2.

Commercially available Iron, Nickel and Aluminium powders in the size range between 40 to 100µm were used as the precursors. The particles were in various irregular shapes. The precursor powders were prepared by ball milling with suitable ratios of composition (Fe / Ni 85 % and Al 15 %) by weight by FRITSCH- Planetary ball mill. The planetary ball mill has zirconia balls of 20 gm (4 No.) and 2 gm (20 No.). Ball milling results in the formation of composite of powders. X-ray powder diffraction (XRD) analysis was carried out on a Rigaku B/max 2500 diffractometer using CuKα radiation. Particle size and morphology were determined by scanning electron microscopy (SEM) on a Philips XL 30 ESEM model.

| Table 1. Typical operating parameters |
|--------------------------------------|
| Power Input (kW) | 10, 15 |
| Plasma gas, argon (l/min) | 20 |
| Pressure (MPa) | 0.59 |
| Secondary gas, nitrogen (l/min) | 2 |
| Pressure (MPa) | 0.20 |
| Carrier gas, argon (l/min) | 3 |
| Anode cooling water flow rate (l/min) | 12 |
| Cathode cooling water flow rate (l/min) | 10 |
| Reaction chamber water flow rate (l/min) | 16 |
| Powder feed rate (g/min) | 14 |
| Quenching Medium | Air, Water |
3. Results and Discussion

3.1 Iron Aluminide

3.1.1. X-Ray diffraction analysis (XRD). Powders as formed were collected from different sections of the plasma reactor (Section A, B and C). The XRD spectral patterns of the powders processed at power levels of 10 and 15 kW are shown in figures 3 and 4 (sections A and B). The pattern vividly elucidates the presence of iron aluminide of different compositions Fe₃Al and FeAl. Fe₃Al and FeAl are used as bond coats in plasma spray applications, as the formation of the intermetallic compound during the spray process leads to metallurgical bonding. It is due to the high exothermicity of the formation reaction [7].

There was no significant difference of phase formation in the XRD pattern of the powders collected from sections A and B. However the peak intensity was found to vary indicating that the powders collected from section B were more crystalline than those from section A at both power levels (10 and 15 kW). At the same time, the XRD pattern shows the presence of minor amounts of iron oxide and alumina peaks. This is due to the fact that the plasma jet acts as an aspirator drawing in air from the surrounding atmosphere, which reacts with the molten metallic particles forming. It results in the formation of Fe₃O₄ and Al₂O₃ (oxidation). However, iron aluminide peak intensity was found to increase with the increase in input power levels. This is because, at higher input power levels, the jet velocity and therefore particle velocities are higher consequently decreasing the interaction time with oxygen. This result in the lower concentration of oxides at higher temperatures.

The average crystallite size can be calculated by using Scherrer’s equation [8], \( D = \frac{0.94 \lambda}{\beta \cos \theta} \) where \( D \) is the crystallite size, \( \lambda \) is wavelength of the X-rays, \( \beta \) is the full width at half maximum (FWHM) of the corresponding peak and \( \theta \) is the scattering angle of the diffraction peak. The crystallite sizes calculated at collection regimes sections A and B for both the power levels 10 and 15 kW are shown in table 2.
3.1.2. Scanning electron microscope analysis (SEM). SEM images of the Fe-Al ball milled powder are shown in figure 5. The particles were in different shapes, mostly in micron sizes. The morphology of the powders after plasma synthesis is shown in the figures 6 to 9. It reveals remarkable material transformation. The change in material morphology and chemical reaction are evident from the figures. The individual particles of the starting powder have different trajectories in the plasma flame, during the plasma process and hence different temperature histories. Particles, which pass through the hot inner region, can melt or evaporate depending on their size. If the fed particles were too large, they leave the plasma flame region without melting, because of the short residence time in the hot zone. However, there can be appreciable surface evaporation from finer particles, which easily form nanoparticles of the corresponding oxide [9].
Figure 6. SEM images of Fe-Al powder processed at 10 kW (a) section A, (b) section B

Figure 7. SEM images of Fe-Al powder processed at 10 kW (a) section C, (b) section C (Water quenching)

Figure 8. SEM images of Fe-Al powder processed at 15 kW (a) section A, (b) section B
Under different operating conditions employed, the particles seemed to be nearly spherical in shape and some of the particles agglomerated and mixed with unconverted particles. The extent of conversion to the aluminide was found to be a strong function of the operating parameters. It is seen that the powders processed at higher power levels (15 kW, section B) have more number of single spherical nanoparticles than powders processed at lower power levels (10 kW, section B).

Most of the particles were found to be agglomerated at 10 kW (section A) power level. The particle agglomeration at section A may be caused due to the following reasons: i.e. (i) Due to shorter distance from the plasma jet, section A was found to be hotter than the section B. (ii) The travelling time of the evaporated species between the plasma jet and the reactor wall is shorter in section A compared to section B. Hence the condensation time of the evaporated species in section A is lower than in section B.

At 15 kW (section B) power level particles were distributed in the size range between 30-70 nm. A good fraction of about 10% nanosized particles could be collected at Section A and B. The most significant parameters affecting the particles size and distribution were the size of the precursor powder, the input power level, gas flow rate and quenching rate. Hence, the high cooling rate of the thermal plasma process controlled the tendency of nano particle synthesis of the plasma processed powders.

On the other hand, the micron sized spherical particles were produced by melting of particles followed by quenching with proper cooling arrangement. Such particles were collected at the bottom of the reaction chamber (section C). The surface morphology of the spherical powders produced at 10 and 15 kW is shown in figures 7(a) and 9(a) respectively.

The presence of spherical powder particles observed in section C is attributed to the fragmentation of the molten particles moving at high speed that became smaller micron sized spherical powders after solidification. It can be noticed that some of the powder particles were non spherical in shape. This is because of the uneven thermal treatment of the powder particles during the interaction within the plasma. The uneven thermal treatment results due to the incongruity of plasma zone temperature and the disparity in the interaction time of particles with the plasma.

The powder particles collected in the water bath was kept under the bottom of the reaction chamber. The morphology of the processed powders collected from section C with water quenching is shown in the figures 7(b) and 9(b). From the SEM results it was observed that water quenched powders were more spherical in nature.
3.2 Nickel Aluminide

3.2.1. X-Ray diffraction analysis (XRD). Figures 10 (a) and (b) shows the XRD pattern of 10 kW plasma processed powders collected on sections A and B respectively. The XRD pattern showed predominant Ni$_3$Al peaks along with NiAl and small amount of NiO and Al$_2$O$_3$ phases in both the collection sections. However the intensity of nickel aluminide peak was found to be slightly higher in section B which may be attributed to an increase in fraction of formation of the nickel aluminide compound. The results show that the flight distance strongly affects the phase composition of the powder with respect to a given power level.

![Figure 10](image1.png)

**Figure 10.** XRD Spectra of Ni-Al powder processed at 10kW (a) section A, (b) section B

![Figure 11](image2.png)

**Figure 11.** XRD Spectra of Ni-Al powder processed at 15kW (a) section A, (b) section B

Figures 11 (a) and (b) shows the XRD pattern of powder processed at 15 kW power level and collected from sections A and B respectively. The results indicated that at 15 kW power level, the powder from sections A and B show lesser NiO and Al$_2$O$_3$ phases which may be attributed due to higher processing power level.

Generally plasma jet acts as an aspirator drawing in air from the surrounding atmosphere, which reacts with the molten metallic particles formed. Subsequently it results in the formation of NiO. Since plasma processing is in atmospheric air, the extent of oxidation of the powder particle is a function of the specific torch design, operating parameters, and powder collection region. The oxide formation can be controlled by adjusting the flow rates of the powders and the power levels and increasing the in-flight residence time. At higher input power levels, the jet velocity and therefore the particle velocities are higher, decreasing the interaction time with oxygen. Consequently, the concentration of the oxides gets lowered at higher temperatures.
The crystallite sizes calculated at collection regimes, sections A and B for both the power levels 10 and 15 kW are shown in table 2. The width of the XRD peaks is related to particles size through Scherrer’s equation.

| Collection region | FeAl Powder   | NiAl Powder   |
|-------------------|---------------|---------------|
|                   | 10 kW         | 15 kW         | 10 kW         | 15 kW         |
| Section A         | 31.58         | 33.43         | 32.43         | 34.54         |
| Section B         | 33.43         | 34.54         | 30.52         | 36.66         |

From the above results it is clearly understood that, both the collection regime (flight distance) and the input power level seem to have strong influence on crystallite size. The factors such as power input to the torch, flame temperature of plasma, gas flow rate and quenching distance affected the phase composition of nanosized powder. The particle size distribution in the feed stock powder has to be in a narrow range to get high proportion of ultra fine powders. The relative particle size of nickel and aluminium powder is another important factor; the size ratio is properly chosen to ensure that the different powders melt under a given set of operating conditions. The density and thermal properties of specific heat, melting point and heat of fusion decides the ratio of different particle size [7].

3.2.2. Scanning electron microscope analysis (SEM). The SEM photograph of ball-milled NiAl powders is shown in the figure 12. It is clearly seen that the particle size ranged from 30 to 100 µm and exhibited wide array of different morphological shapes.

![Figure 12. SEM images of Ni-Al ball milled powder before processing](image)

![Figure 13. SEM images of Ni-Al powder processed at 10 kW (a) section A, (b) section B](image)
Figures 13 (a) and (b) show the SEM photographs of nanosized nickel aluminide powder processed at 10 kW power level collected from sections A and B respectively. It is seen that there is a wide distribution in particle size ranging from few nanometers to about 70 nm. Smaller particles melt and evaporate during processing and condense on the walls of the reaction chamber forming nanosized particles. Bigger particles may partially or fully melt or surface evaporates. These particles during flight solidify and get collected. Particles during in-flight may also agglomerate to form larger particles. Individual particles are not discernible, but seen as agglomerates. Particles collected from section A (figure 13 (a)) were more agglomerated than those collected from section B (figure 13 (b)). The morphology of powders which were collected from section A at 15 kW power level (figure 14 (a)) shows that the particles were similar agglomerates. However the powders collected from section B at 15 kW (figure 14 (b)) were not much agglomerated. A good fraction of about 10% nanosized particles could be collected at Section A and B.

Figures 15 (a) and (b) show the SEM pictures of typical particles of NiAl powder processed at 10 and 15 kW power level collected at section C. Majority of the powder particles was spherical in shape, yet a few exhibited doughnut geometry. The difference in particle morphology is because that all the particles passing through the plasma stream are not subjected to the same thermal treatment. The important causative factors for the same are the spread in particle size of the feed material. The temperature and enthalpy of the plasma and the subsequent interaction of molten, partially molten and vapour particles in the plasma jet also results in the difference of particle morphology. Since the residence time of the particles in the plasma is of the order of a few tens of milliseconds, smaller particles vaporize and larger particles get partially or fully melt. Near spherical morphology is also obtained by the interaction of partially molten particles with vapour particles [10].
4. Conclusion

Synthesis of aluminides of iron and nickel was made using a non transferred plasma torch. The result shows that a good fraction of about 10% aluminide nanosized particles could be collected at Section A and B. In addition, the synthesis exhibited that the torch input power, feedstock particle size, plasma gas flow rate and particle collection region are the important parameters in deciding the size and formation of aluminides.

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