Al-Li/SiCp Composites and Ti-Al Alloy Powders and Coatings Prepared by a Plasma Spray Atomization (PSA) Technique

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There has been increasing use of Al-Li alloys in the aerospace industry, due mainly to the low density and high elastic modulus of this material. However, the problem of low ductility and fracture toughness of this material has limited its present application to only weight- and stiffness-critical components. Development of Al-Li/ceramic composites is currently being investigated to enhance the service capabilities of this material. The Ti-Al alloy is also of interest to aerospace-type applications, engine components in particular, due to its attractive high-temperature properties. Preparation of fine powders by plasma melting of composite feedstock and coatings formed by plasma spraying was carried out to examine the effect of spray parameters on the microstructure and properties of these materials. Characterization of the powders and coatings was performed using the scanning electron microscope and image analyzer.

Examination of the plasma-sprayed powders and coatings has shown that in the Al-Li/SiC composite there is melting of both materials to form a single composite particle. The SiC reinforcement was in the submicron range and contributed to additional strengthening of the composite body, which was formed by a cold isostatic press and consolidated by hot extrusion or hot forging processes. The plasma-sprayed Ti-Al powder showed four categories of microstructures: featureless, dendritic, cellular, and martensite-like.

1. Introduction

ALUMINUM-LITHIUM and titanium-aluminum alloys are of tech-nological importance to the aerospace industry. Both materials offer the advantage of lower weight and enhanced mechanical properties. Aluminum-lithium alloys have been developed as aircraft structural materials (Ref 1, 2), and, when used to replace conventional high-strength alloys in existing aircraft, could provide an 8 to 10% saving of the aluminum structural weight (Ref 3). Other advantages of Al-Li over conventional high-strength aluminum alloys include increased stiffness, better fatigue resistance, and broader temperature capability. The addition of ceramic particulates as reinforcement to the Al-Li alloys enhances their high-temperature stability, strength, elastic modulus, and higher service-temperature capabilities. Reinforcement materials include carbides, borides, nitrides, and oxides (Ref 4, 5). Several studies have been made in recent years on the processing and properties of aluminum alloy/ceramic composites (Ref 6-8). Among the processes used are spray atomization and codeposition, powder metallurgy, and plasma spray codeposition (Ref 9-11).

Titanium aluminides have the advantage of low density (3.76 g/cm³), high-temperature oxidation resistance, and a relatively high modulus of elasticity (Ref 12). There has been renewed interest in this material in recent years due to the improvement made in

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fracture toughness and creep properties (Ref 13, 14). The processing of this material has been inherently difficult. However, with the advent of powder metallurgy routes in recent years these difficulties have been reduced significantly (Ref 15).

At present, there is increased use of rapidly solidified powders in powder metallurgy. This is due mainly to the advantages of extended solid solution by means of metastable phases, improved chemical homogeneity, and smaller grain size. Among the most popular methods of producing rapidly solidified powder is the atomization method, which involves the breaking up of a liquid metal stream into an aerosol of molten droplets. Subsequent solidification of these droplets takes place in flight. The atomization of the liquid can be achieved using gas, ultrasound, centrifuge, or water (Ref 16, 17). In gas atomization, the molten alloys initially induction melted in vacuum are atomized using an annular gas flow nozzle. Argon or helium can be used to atomize the liquid metal. However, argon is preferred for economic reasons. The atomized powder is collected by a cyclone collector.

Plasma spraying is a widely applied technique in the production of fine powders and surface coatings. The powders can be prepared by feeding the raw feedstock into the high-temperature flame where the powder melts and solidifies as spherical particles. There can also be a small degree of atomization, due likely to the turbulence of the jet that shears off material from the surface of the liquid droplets to form very fine particles, which then collide and fragment. Another method of producing fine particles using the plasma jet is to direct the molten particles at a solid rotating substrate, where the molten droplets fragment into many smaller spherical particles upon impact.

The present study investigates the plasma spraying and plasma spray atomization of (PSA) of Al-Li/SiC and TiAl to form coatings and fine powders, respectively. In the PSA process, metal and/or ceramic powder(s) are injected into a high-
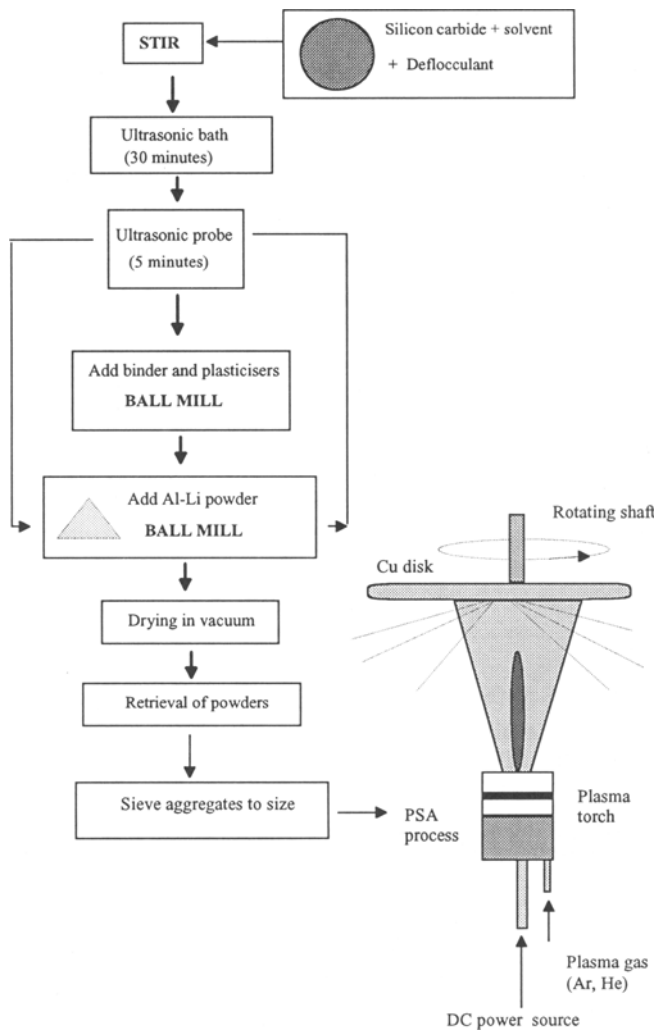
Fig. 1 Steps involved in preparation of Al-Li/SiC aggregates by the ceramic slurry method, followed by plasma spray atomization

Fig. 2 Ti-Al powder, prepared by the plasma rotating electrode process that was used as feedstock in plasma spray atomization

temperature plasma jet where the particles become molten. These molten droplets are then accelerated toward a solid rotating disk where they disintegrate into many finer fragments. This method has been applied to the production of fine stainless steel (AISI 316), yttria-stabilized zirconia (ZrO_2-7wt%Y_2O_3), and Al-Si and Al-Ni powder (Ref 18, 19). The Al-Li/SiC composite powders for the PSA process are prepared from a novel micro-granulation technique that results in a largely agglomerated feedstock. The TiAl powder is produced by the plasma rotating electrode process. A helium plasma is used to melt a rapidly rotating titanium alloy bar, and the molten droplets that spin off are rapidly solidified in the inert atmosphere. The particles are spherical and generally have excellent flow properties. Their size ranges from ~50 to 500 µm. Control of chemical composition and interstitial impurity level is important because ductility is very sensitive to these elements. Interstitial impurities such as oxygen, carbon, nitrogen, and boron can significantly affect the ductility when concentrations exceed ~0.1 wt%. The plasma-processed materials are examined using metallographic techniques, scanning electron microscopy (SEM), and energy dispersive x-ray analysis (EDXA).
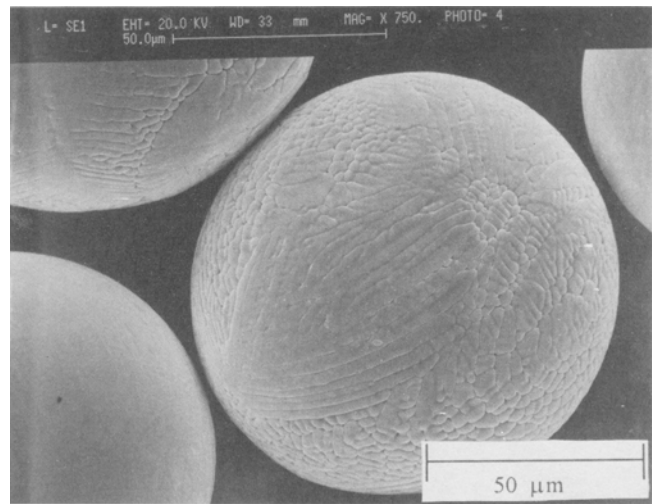
2. Experimental Procedure

The Al-Li alloy powder used in this study was the gas-atomized 8090 series prepared at Sumitomo Light Metals, Japan. The chemical composition of the powder, in weight percent, was Al-2.54Li-1.49Cu-0.91Mg-0.13Zr. Only powders that were <105 µm were used. The reinforcement was submicron SiC powder (average particle size ~0.8 µm). A major problem associated with the submicron ceramic powder is agglomeration during mixing, which results in an inhomogeneous microstructure in the metal-matrix composite and poor mechanical properties due to pores and poor interfacial properties with the matrix. Several mixing methods have been used to prepare well-mixed aggregates of the Al-Li alloy powder with the ceramic reinforcement: the use of a fluidized bed, spray drying, and ball milling (Ref 20, 21). Ball milling, together with appropriate treatment of the ceramic with ultrasonic energy, dispersed the ceramic with the metal powder satisfactorily. Figure 1 outlines the preparatory steps involved in the Al-Li/SiC feedstock.

The Ti-Al powder, from Osaka Titanium Co., Japan, had a nominal composition, in weight percent, of Ti-36Al-0.14Fe-0.051O. The particle size was <250 µm. SEM observation showed that the particles were spherical in shape and exhibited a dendrite-like structure on the particle surface (Fig. 2).

The production system for the fine powder using the PSA method consisted of a 40 kW direct current (dc) plasma spray system and a double-wall stainless steel chamber that housed the rotating substrate. The plasma torch used was the SG-100 from Miller Thermal Inc., and the powders were fed to the plasma torch by means of a closed-loop computerized rotor wheel powder feeder. The driving mechanism for the rotating substrate was mounted on top of the chamber, while the plasma gun was fixed onto a steel bar holder welded to the bottom of the baseplate of the chamber (Fig. 3). A more detailed description of the powder production system is reported in an earlier work (Ref 22). The plasma spraying of Al-Li/SiC composite powder was carried out under low-pressure conditions, while Ti-Al was sprayed under atmospheric conditions. A modified system was fabricated to
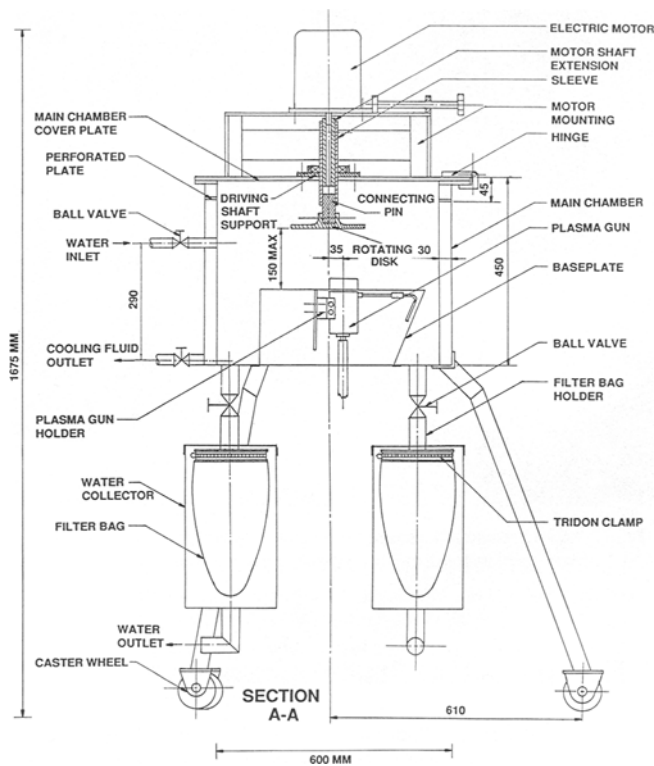
Fig. 3 Schematic diagram of the plasma spray atomization system

Fig. 4 Polished cross section of a ball milled Al-Li particle coated with SiC

Table 1 Operating parameters for direct current plasma and process parameters for plasma spray atomization

Component	Parameter or subcomponent	Condition or value
Plasma gun	Gun model	SG 100, 40 kW
	Anode	No. 145, forward injection
	Cathode	No. 129, subsonic mode
	Power	570 A, 30 V
	Arc gas	Argon, 340-370 kPa (50-53 psi)
	Auxiliary gas	Helium, 340 kPa (50 psi)
Powder feeder	Feeder	Fine powder wheel
	Powder gas	Argon, 280 kPa (40 psi)
Rotating substrate	Material	Copper
	Speed	1410 rpm
	Quenching medium	Air

enable the PSA process to be operated under low-pressure conditions (Ref 23). The chamber was evacuated to <50 mbar, flushed with argon, and evacuated again before the plasma torch was ignited. The pressure during spraying ranged from 100 to 400 mbar. Table 1 lists the operating parameters used in the preparation of the powders and coatings.

2.1 Characterization of Powders and Coatings

The microstructure of the fine powders collected from the chamber was examined by optical microscopy and SEM with EDXA. Particle size analysis was carried out using Sedigraph 5100 equipment, based on the x-ray sedimentation method, and Fritsch Analysette 22 equipment, based on laser diffraction of
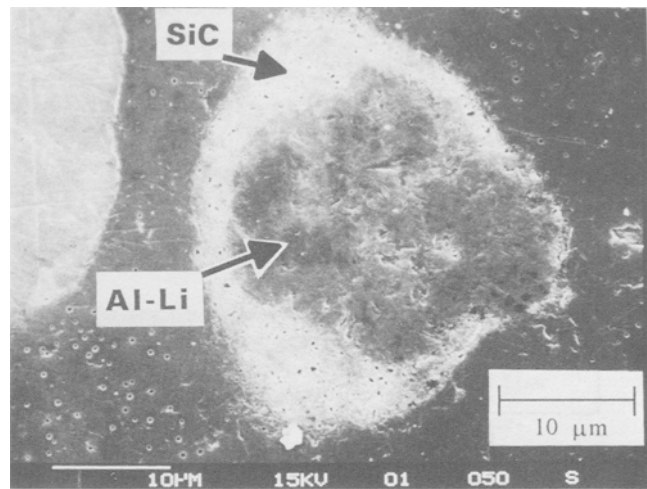
particles. Phase analysis was carried out with a Philips MPD 1880 x-ray diffractometer system. The operating parameters were CuKα radiation (45 kV/30 mA), divergence slit of 1°, receiving slit width of 0.1 mm, and a 2θ scan rate of 1°/min.

3. Results and Discussion

3.1 Al-Li/SiC

The feedstock for this composite was prepared by ball milling of ultrasonically treated ceramic powder with Al-Li powder. This method yielded well-distributed SiC among the Al-Li particles (Ref 20, 21). The aggregates contained ~20 wt% SiC. Aggregates of <106 μm were sieved out for the PSA process. Figure 4 shows the polished cross section of such particles. The submicron SiC particles evenly coat the Al-Li.

Table 2 lists the parameters used in the preparation of the Al-Li/SiC composite powder and coatings. Sample 1 was prepared under normal atmospheric pressure, while samples 2 to 4 were prepared under low-pressure (100 to 400 mbar) conditions. The coatings exhibited a dispersion of SiC particles in the Al-Li matrix. The microstructure contained some interlamellar voids and pores among the rapidly quenched layers of Al-Li and SiC. Some unmelted or partially melted particles were also observed, and most of the voids were found around these particles. Measurement of the porosity in the coatings using a combination of SEM and the image analyzer showed the porosity level to be in the range of 4 to 12%. The coating in sample 1 was found to contain many unmelted or partially melted particles (Fig. 5 and 6). This is perhaps indicative of insufficient arc power (15 kW) to melt all of the powder feedstock in the plasma. On the other hand, the coatings of samples 2 to 4 did not reveal such characteristics (Fig. 7). The SiC in the coatings was in the form of individual spherical particles or thin films wedged between successive Al-Li lamellae. The size of the SiC particles varied between ~2 and 4 μm.

The plasma-atomized particles contained a fine dispersion of SiC within the individual particles (Fig. 8). The presence of SiC

was confirmed by EDXA, which showed a silicon peak in the detection pattern in addition to an aluminum peak. There were, however, some particles in sample 1 that exhibited a molten layer of SiC on the outer circumference (Fig. 9), and in addition, individual SiC particles (submicron to ~2 μm) were observed. The SiC particles probably arose from particles that detached from the Al-Li powder and coalesced with each other in the liquid phase to form larger SiC. The SiC particles could also have been formed through the melting of SiC aggregates from the feedstock.

The SEM examination also revealed individual Al-Li particles without any SiC dispersion. It is likely that the Al-Li/SiC composite particles were formed through different mechanisms during the plasma spray process, such as in-flight reaction within a single aggregate, in-flight collision between several molten droplets, and mixing of the molten droplets at the substrate surface prior to ejection from the rotating disk. Both SiC and Al-Li must melt simultaneously for the composite powder to form. The SiC has a higher melting point than Al-Li; therefore, the arc power must first melt the SiC "coat" before successful formation of the composite powder can take place.

The majority of Al-Li/SiC composite alloy particles ranged from ~1 to 20 μm, and the average size in most samples was ~6 μm.

3.2 Ti-Al

Five experiments were carried out for the Ti-Al powders. The PSA parameters, such as plasma arc power, substrate rotating speed, distance between the plasma torch and the substrate, and the plasma gas composition were changed in each run to examine the effect they have on the production of powders. The changes were made relative to the process parameters listed in Table 1. Two types of powder characteristics were observed in the plasma-processed Ti-Al powders. The powders collected from the base of the stainless steel chamber (see Fig. 3) where
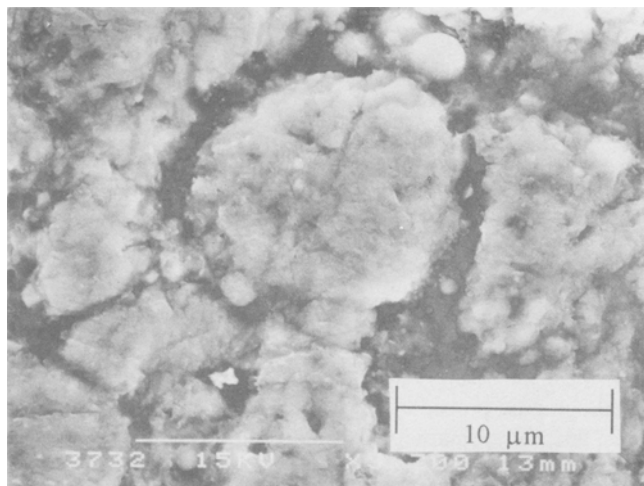

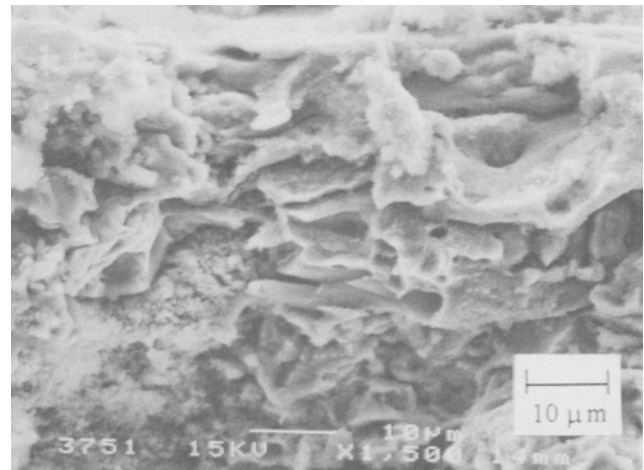**Fig. 6** Fractured surface of Al-Li/SiC sample 1 coating

Fig. 5 SEM micrograph of Al-Li/SiC sample 1 coating
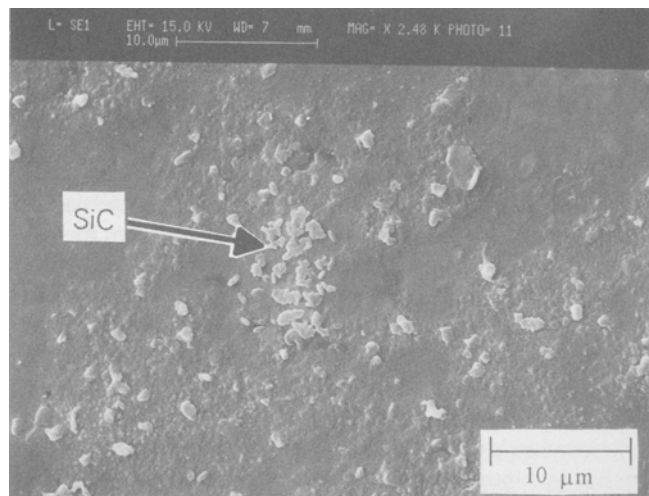

Fig. 7 Polished cross section of Al-Li/SiC sample 3 coating

Table 2 Changes in process parameters for preparation of Al-Li/SiC composite

Sample	Arc power	Plasma gas (Ar/He), kPa	Powder feedrate, rpm	Rotating speed, rpm	Distance between plasma gun and substrate, mm
1	500 A, 30 V	340/340	3.5	300	95
2	800 A, 32 V	340/340	3.5	2100	95
3	800 A, 32 V	340/550	3.5	2100	240
4	800 A, 32 V	340/550	3.5	2100	175

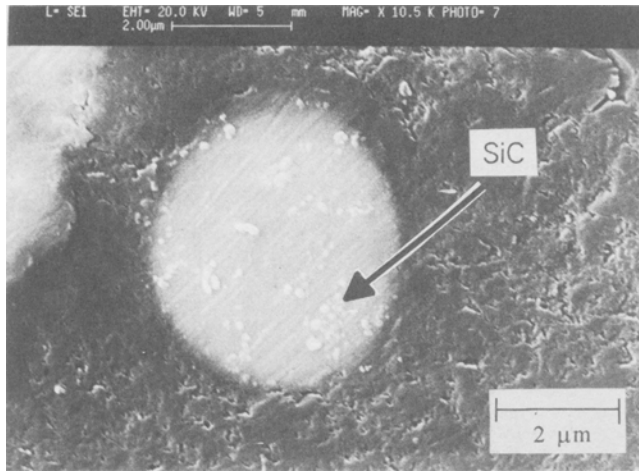**Note:** 50 psi = 3.4×10^5 Pa; 80 psi = 5.5×10^5 Pa.

Fig. 8 SEM micrograph of polished cross section of Al-Li/SiC sample 3 particle showing a dispersion of SiC within the Al-Li particle
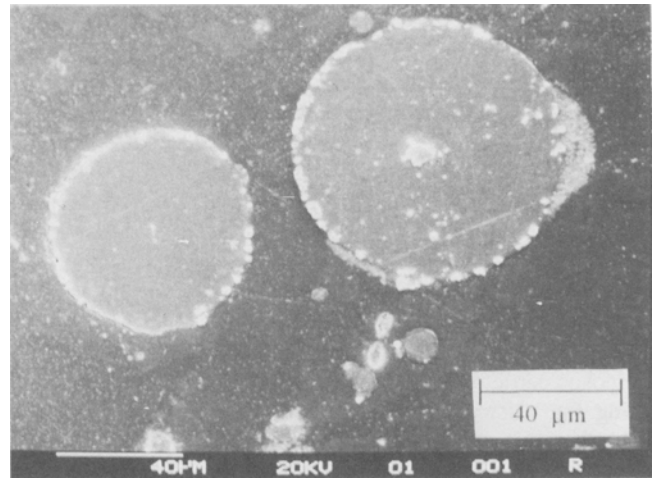

Fig. 9 SEM micrograph showing a melted layer of SiC on Al-Li

Table 3 Changes of process parameters for plasma spray atomization of Ti-Al from those in Table 1

Sample	Process variable	Change From	To
1	No change (similar to Table 1)	...	...
2	Plasma current	570 A	800 A
	Substrate rotating speed	1410 rpm	2010 rpm
3	Substrate speed	2010 rpm	1410 rpm
	Distance between plasma gun and substrate	130 mm	80 mm
4	Substrate rotating speed	1410 rpm	2010 rpm
5	Auxiliary gas pressure	340 kPa (50 psi)	690 kPa (100 psi)

Table 4 Summary of particle size distribution for plasma-processed Ti-Al powders

Sample	Powder collected from base <20 μm	Average size, μm	Powder collected from side <20 μm	Average size, μm
1	22.5%	200	...	...
2	15.1%	210.8	...	...
3	7.5%	205.6	27.5%	38.4
4	8.7%	215.7	23.8%	38.4
5	49.0%	105.4	26.25%	49.2

the PSA process takes place had nearly the size and shape of the as-received powder, while the powder collected from the side wall of the chamber was significantly finer (~5 to 80 μm). Tables 3 and 4 list the changes in the process parameters and the particle size distributions of the Ti-Al powder collected from the base and the side wall of the chamber, respectively.

Table 4 shows that sample 5 exhibited the most appreciable change in size. In the other Ti-Al samples, apart from the existence of fines, the particle size of the powder collected from the base did not differ significantly from that of the as-received powder. This reduction in powder size can be attributed largely to the increase in the secondary plasma gas flow (helium), which resulted in a more efficient heat transfer in the plasma and increased velocity of the plasma jet. The other process parameters, such as disk rotating speed, distance between the plasma gun and the disk, and plasma current level, did not influence the production of fine particles. This is consistent with the mechanism involved in microatomization, whereby the particles from the plasma must remain in the molten state for effective fragmentation to occur. Figure 10 shows the cumulative particle size distribution for the as-received powder and plasma-processed powder from samples 2 and 5. A significant reduction in particle size below approximately 212 μm occurred. This was more marked in sample 2, where nearly 40% of the plasma-processed powder had particles greater than 212 μm in size. Other samples also showed a similar trend, with the apparent limiting size rang-

ing from 211 to 220 μm. Hence, in the absence of high auxiliary gas flow, the reduction in particle size is very much limited to particles less than approximately 211 to 220 μm.

Phase analysis using the x-ray diffractometer showed that there was negligible change in the phase composition of the powder collected from the base. On the other hand, powder that deposited on the side wall of the reaction chamber revealed a high amount of the α_2 phase and titanium dioxide in the form of rutile. Rutile probably forms from the oxidation of titanium in the powder when it is in-flight and immediately prior to final solidification.

Optical microscopy observation of powder using standard metallographic procedures revealed four types of microstructures: featureless, martensitic, dendritic, and cellular. These microstructures were studied in greater detail under the SEM. The plasma-processed Ti-Al powder showed the presence of a fine dendritic structure radiating from a single phase area and a fine martensite-like structure of the two phases, namely α_2 and γ-TiAl (Fig. 11). A study of this two-phase Ti-Al using transmission electron microscopy by Feng et al. showed that the two-phase material composition promotes the formation of alternating lamellae of α_2 and corresponding twin-related laths. The microstructural result indicates that deformation that occurs via the formation of α_2 and twin-related laths plays the primary role in the capability of two-phase materials for continuous deformation (Ref 24).

Figure 12 shows the fractured surface of a Ti-Al coating where the lamellar structure is observed. Figure 13 shows the so-
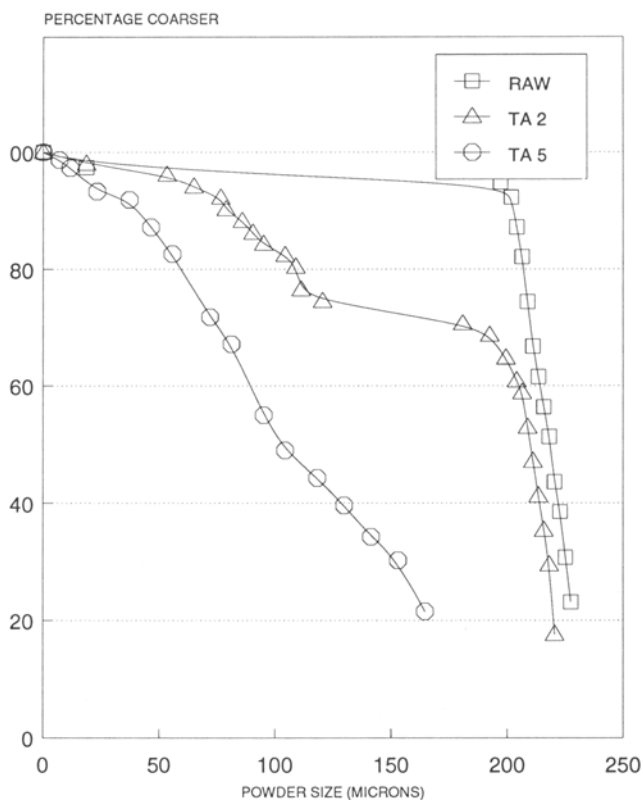
Fig. 10 Cumulative particle size distribution for as-received powder and plasma-processed powders
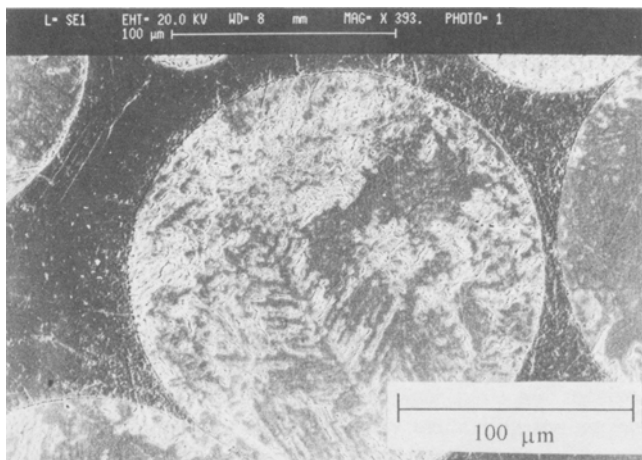

Fig. 11 SEM micrograph of polished cross section of plasma-atomized Ti-Al particle

Fig. 12 Fractured surface of Ti-Al coating

Fig. 13 Solidification structure in a plasma-sprayed single Ti-Al lamella

lidification structure of Ti-Al within a single lamella where a columnar or granular structure forms.

4. Conclusion

Fine powders and coatings of Al-Li/SiC composite and Ti-Al have been produced using a PSA technique. The results show that plasma arc power of >1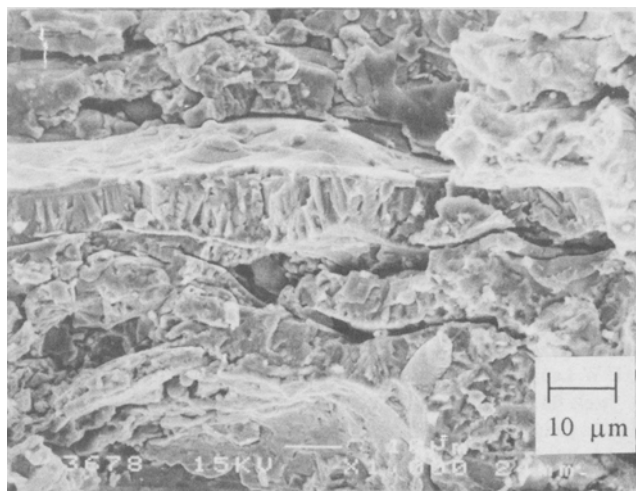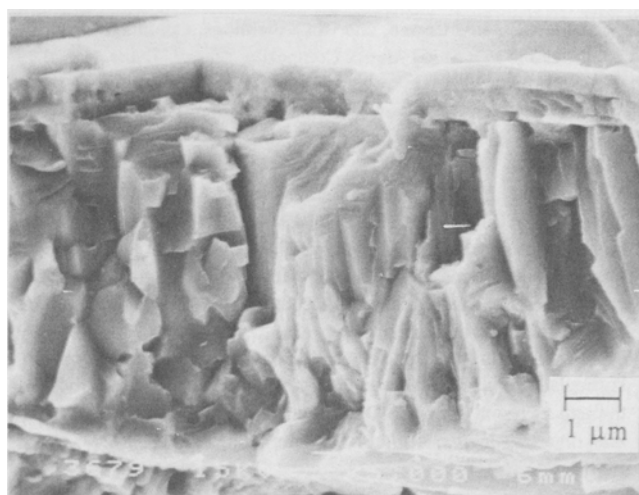5 kW is needed for complete melting of the Al-Li/SiC feedstock to ensure formation of an acceptable coating. The plasma spray parameters used have produced particles containing a fine dispersion of SiC within the Al-Li matrix material. There were, however, individual particles of SiC and Al-Li among the powder collected. These arose from detachment of the SiC from the Al-Li in the plasma jet.

The plasma spraying of Ti-Al was hindered by the use of large particles as the feedstock (~250 μm). The coatings examined showed unmelted or partially melted particles and poor adhesion between the lamellae. The PSA operation to form the fine powder was found to depend on the plasma gas composition. It was found that increasing the pressure of the secondary plasma gas phase (helium) to 690 kPa (100 psi) improved the production of fine powders.

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