Electro-Discharge Consolidation of Atomized High Strength Aluminum Powders

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Electro-discharge consolidation (EDC) was carried out for atomized Al-based alloy powders produced at YKK. Input energies of 2.5 to 3.8 kJ/g produced compacts of 88 to 99% in theoretical density. The densification can be described by a simple relation of $\Delta\rho = 11.8[ER_s/(R_s + 2)]^{0.36}$ in % where R_s is the specimen resistance, and it predominantly depends on input energy, E . A comparison of XRD spectra before and after EDC with respect to the peaks' position and intensity indicates that no microstructural change occurred. Oxide film removal by EDC was confirmed by TEM observations. Compression tests of EDC compacts at room temperature produced under the present conditions (specimen resistance $R_s = 9.4\text{--}15.8\text{ m}\Omega$) yielded 800 MPa and 24% elongation for true ultimate compressive stress and true compressive strain, respectively. The annealing treatment at 573 K for 1.2 ks raises compressive strength by at expense of ductility due to precipitation of solute atoms, indicating that a super-saturated solid solution is maintained in the EDC state.

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I. Introduction

Powder atomization often produces a supersaturated solid solution due to extremely high quenching rates. The conventional consolidation, for example by hot-pressing, however, inevitably results in formation of various compounds of undesirable amounts and sizes due to uncontrollable precipitation of excessive solute atoms. Also the removal of oxide films on the prior particle surface cannot easily be done and requires additional, severe hot working such as extrusion with a very high reduction ratio. The existence of prematurely broken oxide films in the solid bulk drastically reduces the formability. Accordingly, the preservation of microstructure inherent in rapidly solidified materials and removal of oxide films have been an everlasting theme in powder metallurgy of aluminum alloys.

Suppression of excessive precipitation requires low temperature and short time sintering for rapidly solidified powders. These conditions cannot, however, guarantee high-density consolidation. Among a few available consolidation techniques, electro-discharge consolidation (EDC) utilizes a high-voltage (up to 30 kV), high-density current (10^{16} A/m^2) pulse (300–500 μs) discharged from a capacitor bank. In fact, recent work on rapidly solidified aluminum alloys⁽¹⁾⁽²⁾ have shown that the EDC technique is characterized by four major advantages over the conventional consolidation techniques. These are: (1) the preservation of original microstructure after consolidation, (2) densification of up to 20–25%

from green density, (3) oxide film removal during a discharge and (4) improved mechanical properties resulted from items 1–3.

Atomized powders of an Al alloy containing Ni, Cu, Ti, Zr and Mm (Misch metal) were developed at YKK. They were subjected to EDC at ambient temperature to produce a high-density solid bulk with no microstructural change as well as no oxide film on the prior particle boundaries, for which it is expected that the resultant mechanical properties are grossly improved compared to those obtained by conventional consolidation processes.

II. Material and Experimental Procedure

Atomized Al alloy powders containing Ni, Cu, Ti, Zr and Mm, with a particle size distribution of 30–50 μm , were produced by YKK Co., Japan. X-ray diffraction was performed on the as-received powders using a Rigaku diffractometer with a Cu target for 2θ from 30–90° at a scan speed of 2°/min and a collection interval of 0.05°. Powders weighing 2.00 g were placed into an open ceramic die of 50^L × 8^W mm. An external pressure up to 450 MPa was applied to the powders on a hydraulic MTS press. During pressing, the specimen resistance was measured using a 4-point resistance apparatus and the green density was monitored from a change in the upper punch position as a function of applied pressure. Upon reaching the desired green density, the powder specimen under external pressure was discharged from a 500 μF capacitor with input energy ranging from 2.0–3.5 kJ/g. XRD was repeated on the consolidated specimens, followed by

density measurements using the Archimedes principle. Vicker's microhardness was also measured with a 0.98 N load and a 12 s holding time. The specimens having a dimension of $1.5 \times 1.5 \times 5$ mm in size were cut using a water-cooled diamond saw out of EDC compacts. Compression tests were performed on a screw-driven Instron testing machine at strain rates of $1.8\text{--}4.7 \times 10^{-4}/\text{s}$ at room temperature. From load-elongation curves, the 0.2% proof stress (yield stress), compressive stress at maximum load and strain to fracture are measured.

III. Results and Discussion

1. Densification by EDC

EDC applies a high-voltage, high-density current pulse to the powder under external pressure for a very short period of time. Joule heating occurs at the particle-particle contact points to instantaneously weld powder particles, resulting in densification. Accordingly it depends on the green density ρ_g and specimen resistance R . Let us now consider how these parameters affect EDC. The achieved density, ρ_a may be described as a function of many parameters,

$$\rho_a(\%) = f[\rho_g(P), R_g(P), E(C, V), t_d(C)] \quad (1)$$

where both ρ_g and R_g depend on applied pressure P , input energy E that depends on the capacitance C and applied voltage V through $CV^2/2$, and the discharge time t_d depends on C . Here t_d is fixed once C is fixed. Accordingly, the major parameters affecting the achieved density are input energy E and applied pressure P that affects both ρ_g and R_s .

Figure 1 shows the effect of applied pressure on the green compact density and its resistance at ambient temperature. Shown here is the logarithm of relative green density (%) plotted against applied pressure. Since the full density of this material is not known precisely, it is calculated to be 3.16 Mg/m^3 using a mixture rule as the first approximation. In this format, the densification behavior during cold compaction can be apparently described by a three-stage process. In region (a), rearrangement of the powder particles occurs when pressure is initially applied to the specimen. The relative green density of the powders increased from 52 to 62% within this region. Upon further increasing the applied pressure, the powders experience yielding, resulting in an increase in green density (region (b)). The density in this region increased up to about 71% at an applied pressure of 180 MPa. Meanwhile, the resistance decreased from 35 to 16 m Ω due to an increase in the particle-particle contact area. Region (c) can be described as a work hardening stage where the green density increases slowly up to a maximum value of 81% under a 430 MPa pressure. On the other hand, the specimen resistance saturates down to a minimum value of 9 m Ω at the same maximum pressure, which is getting to the same order of the circuit resistance of 2 m Ω . Since the maximum densification by EDC is known to be 22% at most from the past ex-

Fig. 1 Relationship among pressure, resistance and green density.

Fig. 2 Relationship between compact density and resistance-normalized input energy as a function of specimen resistance.

perience⁽⁴⁾, the present system has a limited condition between the lowest green density of 78% (at 350 MPa) and the highest green density of 81% at 430 MPa. Accordingly, the applied pressure must be adjusted within this range so that the expected specimen resistance would be greater than 9 m Ω . Input energy ranging from 2.5 to 3.8 kJ/g was chosen. The resultant bulk densities after EDC are illustrated in Fig. 2. Here the relative achieved density is plotted against the specimen-resistance-normalized input energy (input energy times the specimen resistance normalized with the circuit resistance of 2 m Ω , $E[R_s/(R_s+2)]$) a function of the initial specimen resistance. Since this material produces a very low resistance under applied pressure, the effectiveness of producing heat for welding powder particles is considered to be proportionally reduced. The term $R_s/(R_s+2)$ refers to an efficiency of a discharge. Nevertheless, the achieved densities of these compacts range from 88–99% of theoretical and exhibit a strong dependence on input energy. This dependence appears to be parabolic. It is also observed that the initial green density is very influential on the achieved density. This is reflected in the parallel nature of the curves, in-

dicating that densification, $\Delta\rho$ ($=\rho_{\text{achieved}}-\rho_g$) may not depend on specimen resistance but solely depends upon input energy.

Densification $\Delta\rho$ is depicted in Fig. 3(a), where it is plotted against the resistance-normalized input energy. Regardless of the initial specimen resistance, all the data points fall on a single curve, indicating that input energy has been normalized. Densification starts at 16% with a normalized input energy of 2.1 kJ/g and parabolically saturates to about 20% at a maximum energy of 3.3 kJ/g. This leads to a conclusion that this powder must be applied pressures to give the green density of up to 80% in order to obtain a fully densified compact. The alignments of all the data on a single curve in Fig. 3(a) infers the densification mechanism during EDC must be the same regardless of the initial compaction condition.

Depicted in Fig. 3(b) is replotting in a log-log scale of the data shown in Fig. 3(a). Now all the data points fall on a straight line and confirms a single densification mechanism. Densification $\Delta\rho$ can be represented empirically as

$$\Delta\rho = A [E_{\text{norm}} R_s / (R_s + 2)]^N$$

in (%) for $R_s < 20 \text{ m}\Omega$ (2)

where A is a material constant based on powder particle characteristics and an exponent N an EDC setup parameter. E_{norm} is input energy E divided by the mass of powders. From the current experiment, $A=11.8$ and $N=0.36$ are obtained.

It has been shown in EDC of aluminum-base alloy powders⁽¹⁾⁽²⁾ that this index N was 0.36 for the specimen resistance of less than 20 m Ω and 0.62 for the specimen resistance of greater than 20 m Ω . In a similar EDC experiment for nanocrystalline niobium-base alloy powders⁽³⁾, the same N value of 0.62 was obtained for R_s of 80 to 130 m Ω . The same value of N for these completely different materials indicates that it can be characteristically related to the current EDC setup. An index N of less than unity implies that input energy is partially consumed at the ion switch that closes the circuit by creating plasma produced by imposing a high voltage. But it also depends on the specimen resistance that dictates the distribution efficiency of current to the powder through the ratio $[R_s / (R_s + R_c)]$ where R_c is the circuit resistance. It appears that a transition from high to low efficiencies exists at R_s of about 20 m Ω (the ratio is $\sim 91\%$). At this moment the real meaning of this critical specimen resistance is not well understood. Because of very low resistances under pressure for this atomized powder, some alteration of powder characteristics will be needed for improved EDC efficiency.

2. Preservation of original microstructure after EDC

One of the prominent characteristics in EDC processing is the ability to preserve the unique microstructure inherent in powders. One way of confirming this characteristics is to check XRD before and after EDC. A representative example is portrayed in Fig. 4. Here, the XRD spectrum of as-received atomized powders is compared to that of an EDC compact that was discharged with the maximum input energy (3.8 kJ/g). First, the intensity of Al(111) in an EDC compact is not increased much, indicating even grain growth does not occur. Secondly the minor peaks for various compounds appear unchanged by a discharge with respect to peak positions and their intensities, indicating that precipitated phases existing in as-received powders does not grow. The majority of alloying elements still remained in solid solution.

Some data concerning the peak position 2θ and its half width B for identifiable compounds, except for the major peak of Al(111), are compiled in Table 1 to compare before and after EDC. As seen from the table, changes in peak position and half-width are minimal. Especially it should be noted that the half-width B is an indicative parameter of precipitate size which turns out to be almost the same as that of the as-atomized powder. Therefore, it can be concluded that the microstructure of the original atomized powder has been preserved in EDC bulk solids with densities approaching 99% of theoretical. TEM ob-

Fig. 3 Densification by EDC versus resistance-normalized input energy.

Fig. 4 X-ray spectrum for As-received powder and an EDC compact.

servations are made for an EDC compact produced by discharging an input energy of 3.8 kJ/g. A typical microstructure is shown in Fig. 5, where the prior particle boundaries are shown by arrows. First, powder particles are completely welded together with no trace of oxide films. Secondly, the size of precipitated phases is very small in the powder particle matrix, indicating the as-atomized state has been well preserved. In addition, there are solid-solutionized zones along the prior particle boundaries. It indicates that the Joule heat generated by a discharge has been dissipated to the matrix very quickly to leave the precipitation-free, super saturated solid solution zone by forcing any pre-existing precipitate going back to the solid solution. Also recrystallization might

Fig. 5 Microstructure of EDC compact.

have proceeded while the heat was dissipated into the matrix but in a manner not to extend its recrystallization beyond this interface region. Accordingly, it may be said that the microstructure in the interface region has been modified due to a very high quenching rate. This has been previously observed in EDC of Al-8Cr-2Ti atomized powders⁽¹⁾⁽⁴⁾ and Al-11Fe-1V powders⁽⁵⁾.

3. Mechanical properties of EDC compacts

The strength of EDC compacts can be estimated roughly by a hardness test. Figure 6 shows the results of Vicker's microhardness testing on three EDC compacts which have densities between 93–99% produced under three different discharge conditions with respect to applied pressure and input energy. It is clearly seen that the hardness remains constant regardless of discharge condition, indicating any significant microstructural changes have occurred. Judging from the hardness of about 200

Table 1 Peak positions (2θ) and peak widths (B) at half of the maximum intensity of different precipitations before and after EDC.

	Al ₃ Ni (211)		AlNi ₃ (111)		Al ₃ Zr (008)		Al ₂ Zr (112)		AlTi ₂ (201)	
	2θ	B	2θ	B	2θ	B	2θ	B	2θ	B
As-received powder	35.15	0.33	36.15	0.42	41.70	0.33	40.25	0.33	40.70	0.33
EDC: 3.4 kJ/g, 14 mΩ	35.12	0.30	36.15	0.41	41.69	0.30	40.22	0.33	40.73	0.34
EDC: 3.8 kJ/g, 16 mΩ	35.15	0.30	36.18	0.41	41.72	0.32	40.26	0.40	40.72	0.35
EDC: 3.8 kJ/g, 14 mΩ	35.09	0.34	36.11	0.47	41.68	0.34	40.20	0.36	40.69	0.34
EDC: 3.8 kJ/g, 12 mΩ	35.21	0.33	36.17	0.44	41.72	0.33	40.26	0.37	40.75	0.35

Fig. 6 Vickers hardnesses at different input energies.

(D.P.N.), the compressive strength σ of about 700 MPa is expected according to an empirical relation of $H_v = 3\sigma$ where H_v is hardness. However, the strength is very sensitively influenced not only by microstructure but also by the achieved density (in other words the porosity).

Compression tests were carried out for EDC specimens having a dimension of $1.5 \times 1.5 \times 5$ mm at strain rates of 1.8 to $4.7 \times 10^{-4} \text{ s}^{-1}$ and room temperature. Shown in Fig. 7 is a typical compressive stress-strain curve obtained for an EDC compact of 95% density. Clearly seen is a true plastic strain of approximately 20% exhibited even by a 95% density bulk. This suggests that the oxide film on the prior particle surface might have been removed during a discharge. From a series of compression tests, the true compressive yield stress (taken at a 0.2% strain), the true ultimate compressive stress (at the maximum load) and true plastic strain (%) were calculated for EDC compacts produced under various discharge conditions.

P/M products inevitably contain voids or pores. It is expected that these pores are interconnected if the achieved density is less than 92% or so, and the amount of these pores adversely affect the mechanical properties. Normally the compact strength will vary with density as follows⁽⁶⁾:

$$\sigma = \sigma_0 K (\rho_a / \rho_t)^m \quad (3)$$

where σ_0 is the wrought strength, K is a geometric and processing constant similar to a stress concentration factor, m gives the exponential dependence on density ρ_a and ρ_t is theoretical density. The ratio of ρ_a / ρ_t is the fractional (relative) density. The influence of the fractional density on the compressive yield stress, ultimate compressive stress and total plastic strain is illustrated in Fig. 8. Here, these three parameters are plotted against relative compact density on a full logarithmic scale. As expected, they increase linearly with increasing the compact density. The exponent m for the yield and ultimate strengths is 6.8 and 3.8 respectively, which are in the expected range.

Fig. 7 A typical true strain-true stress curve obtained from the compressive testing of one EDC compact.

Fig. 8 Mechanical properties of the powder after EDC.

It is amazing to observe about 10% strain and a significant amount of work hardening from the yield to the ultimate even for a porous compact of 84% relative density. This again suggests the EDC compact is ductilized as a result of the oxide film removal. When the density reaches 99%, the true compressive yield stress is increased to 470 MPa, the true ultimate compressive stress exceeds 800 MPa and the true total strain at fracture is 24%. It should also be noted that the as-EDC compacts are not fully hardened yet compared to conventionally produced compacts that have inevitably experienced extensive heats to temperatures at which massive precipitation is expected.

Since EDC preserved the as-atomized microstructure and partially improved its microstructure by creating a super-saturated solid solution, it can be expected that annealing at elevated temperatures enhances precipitation hardening. An EDC compact having a relative density of 95%, produced by discharging 3.0 kJ/g to powders of 10.8 mΩ, was annealed at 573 and 713 K for 0.6 to 3.6 ks in air. The compressive tests were carried out at room

Fig. 9 True strain-true stress curves of one EDC compact under different heat treating conditions.

temperature with a strain rate of $4.7 \times 10^{-4} \text{ s}^{-1}$. The results are shown in Fig. 9. Low temperature anneals for less than 1.8 ks increased not only the yield strength (from 452 to 613 MPa) and the maximum strength (from 707 to 841 MPa) at the expense of ductility (from 21.3 to 12.5%). This is clearly due to precipitation hardening from the super-saturated solid solution. On the other hand, anneals at 573 K for 1.8 ks and 713 K for 3.6 ks decreased both the yield strength but increased ductility, exhibiting an over-ageing behavior. Table 2 summarizes the mechanical properties of annealed materials compared to those for as-EDC material. These results clearly imply that EDC can not only preserve the original microstructure inherent in atomization but also partially produce a super-saturated solid solution. The mechanical properties can be tailored by a post-EDC heat treatment.

Table 2 Mechanical properties of one EDC compact before and after heat treating.

	$\sigma_{0.2}$ /MPa	$\sigma_{\max}$ /MPa	$\epsilon_{\text{fracture}}$ (%)
As-EDC	452.2	706.9	21.3
573 K $\times$ 0.6 ks	470.8	724.9	17.2
573 K $\times$ 1.2 ks	613.2	840.9	12.5
573 K $\times$ 1.8 ks	404.4	665.9	17.5
713 K $\times$ 3.6 ks	336.7	563.7	25.6

IV. Conclusions

(1) EDC of an atomized Al alloy produced bulk solids with densities approaching 99% in theoretical density.

(2) The EDC efficiency of these powders was greatly influenced by the very low green specimen resistance under pressure.

(3) The microstructure inherent in the atomized powders was preserved in the EDC compacts and partially became a super-saturated solid solution.

(4) The oxide film on the powder surface was removed, resulting in the enhanced mechanical properties exceeding 800 MPa and 24% for true ultimate compressive stress and true compressive strain, respectively.

(5) Annealing at 573 K for less than 3.6 ks increased both yield strengths by precipitation hardening.

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