DEPENDENCE OF MECHANICAL ALLOYING OF THE Al-Ti POWDER ON THE PROCESS VARIABLE

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ABSTRACT

MA process can provide as attractive method of preparing Al based alloys having larger volume fraction of submicron sized high melting point intermetallic phase of Al_3Ti, which might offer weight saving over conventional high temperature Al-alloys. The aim of the present study is therefore to investigate the dependence of mechanical alloying on the process variables for the Al rich composition in Al-Ti system in order to determine the optimal MA process condition.

Homogeneity of Al-Ti composite particles can be obtained by the following processing variables; addition of 1.5wt.% stearic acid as PCA, the milling speed of 400rpm and MA milling time more than 10 hours. The mechanically alloyed powder prepared by the above condition reveals the achievement of a steady state processing, i.e., equiaxed powder particles, constant powder size distribution and a saturation hardness. The intermetallic compound phase of Al_3Ti in the matrix was identified by X-ray diffraction analysis.

INTRODUCTION

Aluminum alloys have been largely used in the component parts of aircraft, automobile and ship due to its favorable characteristics such as low cost, high specific strength and excellent corrosion resistance[1,2]. However, it is difficult to use the conventional aluminum alloys in the high performance structural parts, such as aerospace skin and the jet engine components, due to the their low mechanical properties at

somewhat higher temperatures[3,4].

Recently there have been introduced several new approaches for the development of the high performance and high temperature Al-alloys, which must exhibit a significantly superior combination of elevated temperature strength, stiffness and thermal stability in comparison with conventional aluminum alloys[5-7]. Mechanical alloying process (MA) is also one of these approaches to providing some advanced aluminum alloys with high strength and good thermal stability at elevated temperature[7].

Mechanical alloying (MA) is a dry, high energy ball milling process for producing composite metallic powders with a controlled, fine microstructure[8]. It is carried out in a highly agitated ball mill by the repeated cold welding and fracturing of a mixture of metal powder. Since no liquid is present at any time during processing, it can be expected that the microstructural features obtained by MA process are superior to those obtained by RSP or MMC. Thus, MA provides the possibility to produce the ultrafinely dispersed and homogeneously alloyed phases only through solid state processing. Furthermore, a great advantage of MA is that all the following strengthening mechanisms can be superimposed; oxide and carbon dispersion strengthening, fine grain size strengthening and solid solution strengthening[4].

Therefore, some Ti based alloys, which are mainly used in structural parts at temperature range of 300-400℃, seems to be satisfactorily replaced with by Al-Ti alloys prepared by MA process.

Among the Al-Ti intermetallic compound, the Al-rich compound Al_3Ti has a low density (3.3g/cm^3), a relatively high melting point (1350℃) and expected good oxidation resistance[9,10]. Accordingly the formation of fine Al_3Ti dispersoids in Al matrix offers considerable potential for elevated temperature structural applications. These fine Al_3Ti dispersoids should be prepared by MA method through only a solid state reaction, because the formation of fine dispersoid by other method such as RSP or conventional ingot metallurgy was restricted from a thermodynamic point of view[7].

In mechanical alloying, the processing variables are very various. They are the charge ratios of balls to powder, the type and amount of process control agent, milling speed, milling atmosphere and milling time. The optimal processing variables could be determined by observing their effects on the characteristics of MA powders.

Therefore, in the present study, the dependence of mechanical alloying on the process variables for the Al rich composition in Al-Ti system was investigated in order to determine the optimal MA processing conditions.

EXPERIMENTAL

Aluminum powders used for the experiment were supplied by Korea Nonferrous Metal Powder Company. They had an average particle size of 59μm, with the purity of 99.5% and a morphology of granular shape. Titanium powder was irregular particle shape with a minimum purity of 99%, supplied by Alfa product. 8wt.%Ti powder was added for MA Al-Ti alloy.

Mechanical alloying of aluminum usually requires the use of process control agent(PCA) to prevent excessive welding of aluminum to itself, the ball charge, the attritor tank, and impellers. The amount of process control agent added during mechanical alloying of aluminum must be: a) enough to prevent excessive welding of the aluminum, but b) not so much as to prohibit completely that welding. In the present study, the powdered organic surfactant of 1.5wt.% stearic acid were charged into the attritor prior to processing , which has the chemical formula $CH_3(CH_2)_{16}COOH$.

Mechanical alloying was accomplished by using a batch attritor of 750cc capacity, a high energy ball mill, with 3/16 inch stainless steel balls under argon atmosphere to avoid oxidation. The powders were processed for various times ranging from 30 minutes to 900 minutes without any intermediate opening of the grinding container. The ratio of balls to powder was kept 65 to 1 and the milling speed was 300rpm and 400rpm respectively.

The particle size variation, depending on the different MA processing times, were analyzed by conventional sieving method. Microhardness measurements of powder particle were restricted in the powder size of -80+100mesh in order to insure consistency and reproducibility in the hardness measurements. In order to obtain reliable statistical data, 12 indentations were made on the large particle of each sample and their average value was taken. Powder specimens were prepared by a mounting technique for microstructural observation.

The mechanically alloyed Al-8wt.%Ti powders were hot consolidated in vacuum. Prior to vacuum hot pressing, the MA powders were firstly cold compacted into a shape of disk under a pressure of 100MPa. Then, this cold compacted powders were further hot pressed under a pressure of 200MPa at 430℃ in a vacuum of 1×10^{-2}torr for 60 minutes. The structural characteristics of hot pressed specimens were investigated by X-ray diffraction analysis and microstructural analysis.

EXPERIMENTAL RESULTS AND DISCUSSION

Microstructure Evolution During Mechanical Alloying

Fig.1 shows SEM morphologies(a,c) and optical micrographs(b,d) of the as-received Al(a,b) and Ti(c,d) powders. As shown in this figure, Al powders can be classified as granular shape, while Ti powders rather as irregular shape.

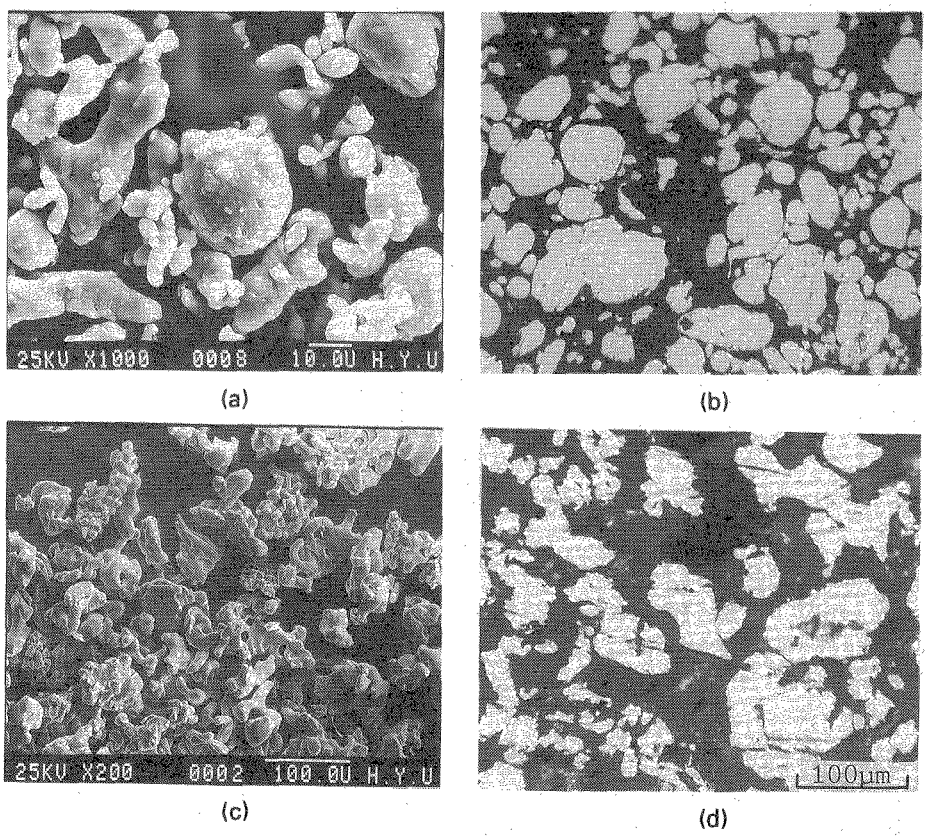


Figure 1. SEM morphologies(a,c) and optical micrographs(b,d) of elemental powder used in the present study; Al powder(a,b) and Ti powder(c,d)

The progress on the mechanical alloying of Al-8%Ti powders was successively controlled by examining the dependence of MA powder morphology on attrition times, i.e., MA processing times. Fig.2 shows the cross-sectional microstructure of Al-Ti powder particles after MA processing in various milling times up to 900 minutes with milling speed

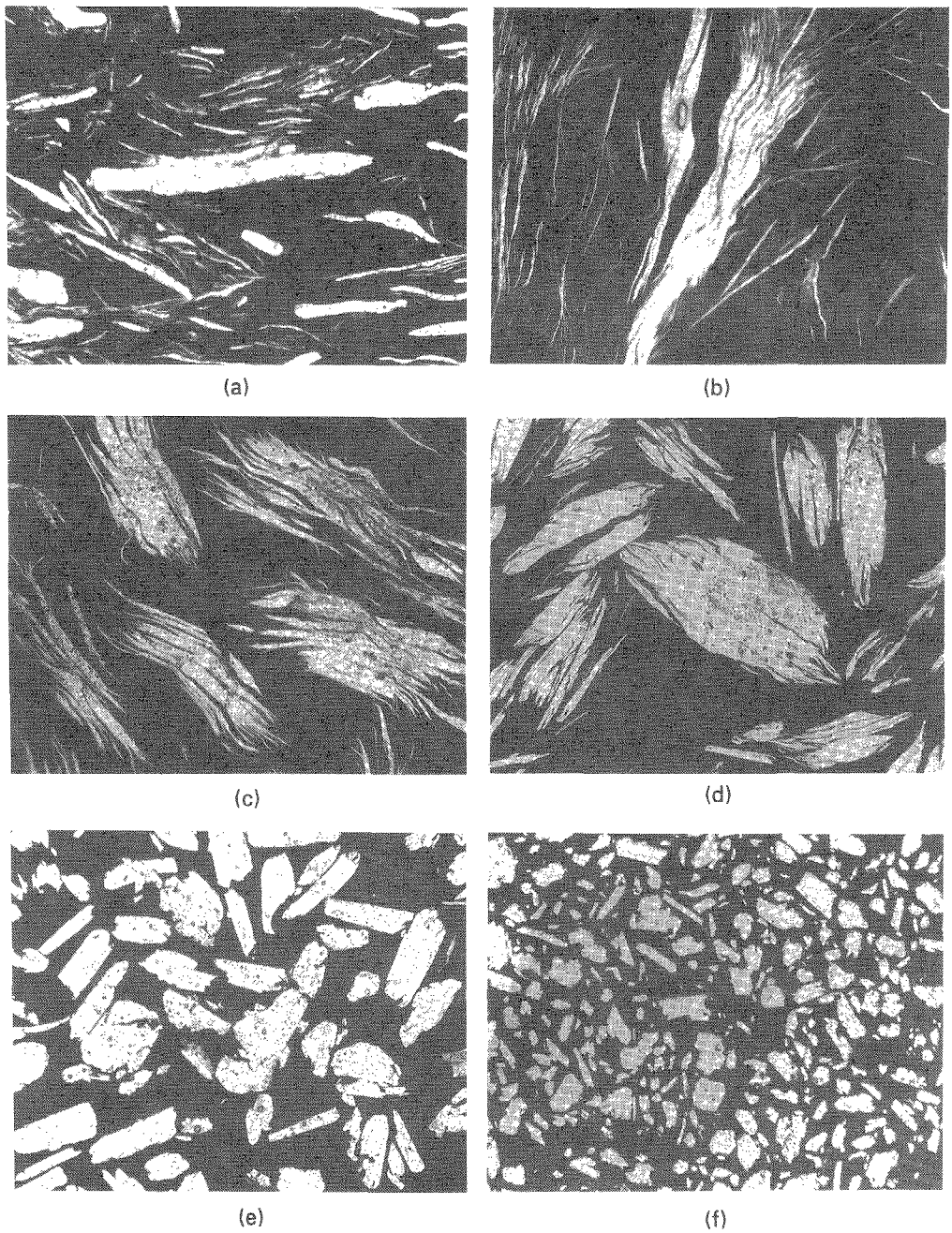


Figure 2. Optical micrographs of mechanically alloyed powder processed for various times; (a) 30min., (b) 1hr., (c) 2hr., (d) 3hr., (e) 7hr. and (f) 15hr.

of 400rpm. After 30 minutes of MA processing, as shown in Fig.2(a), the initial period of mechanical alloying has been reached. This stage is characterized by the flattening of the starting powder charge due to the large impact force between the colliding balls. After 60 and 120 minutes of processing, as shown in Fig.2(b),(c), a period of welding predominance has been reached and multilayered composite lamellae can be observed in the flaked particles. As milling proceeds, the flaked particles appear to be somewhat rounded as shown in Fig.2(d). Toward the end of the milling cycle, Fig.2(f), the MA Al-Ti powders appear to be equiaxed and are also more rounded in their morphology, as a consequence of repeated cold welding and fracturing. At this stage the powder is relatively flowable and can be readily transferred for further processing.

Fig.3 illustrates the composite microstructure of MA Al-Ti powder particle observed by SEM. As shown in this figure, Ti particles, being indicated by an arrow, are sandwiched between layers of Al in early stage of milling process and this also marks the beginning of the formation of the characteristic "lamellar" microstructure. With continued milling the sandwiched Ti particles will be present as very thin sheets within the Al particles. Furthermore, the Ti sheets will be eventually broken-up and dispersed within the Al particles.

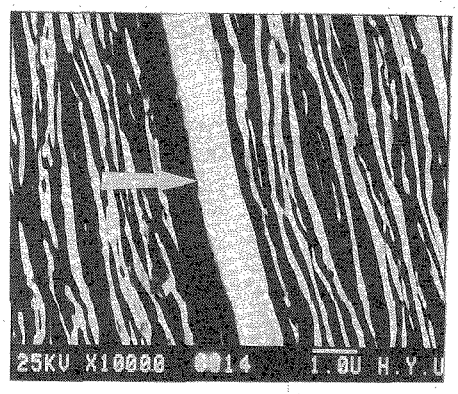


Figure 3. The SEM micrographs of MA Al-Ti powder in composite structure after 120 min. of milling with the milling speed of 400rpm. The arrow indicates Ti element.

Structure Refinement Rate and Energy Consideration

During the initial period of processing a lamellar structure exist in the mechanically alloyed powders. Fig.4 shows the relation between the MA

processing time and the average lamellar thickness of MA powder particles in the early stages of processing with milling speed of 300rpm. The above relation can be also interpreted as the relationship between structure refinement rate and input energy, because input energy is proportional to MA processing time. As shown in this figure, the measured lamellar thickness decreased with increasing processing time and the value of 1.92μm was obtained in 180 minutes of processing.

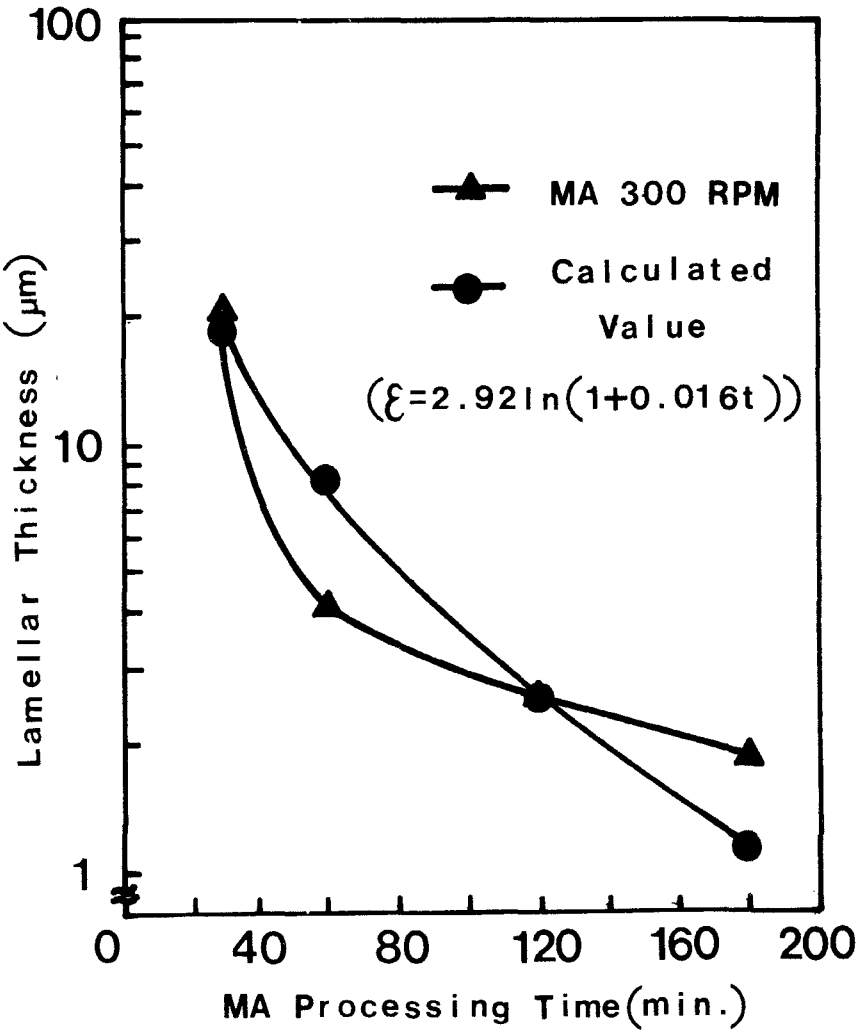


Figure 4. Variation of lamellar thickness as a function of MA processing time.

The average rate of change of lamellar thickness can be calculated if certain assumptions are made concerning the energetics and statistics of

the process[11]. First, assume that the rate at which material is trapped between colliding balls is independent of time. Therefore, the energy input rate to the process is constant, as given in Eq.(1)

$$\frac{dE}{dt} = K1 \qquad (1)$$

where E = energy, t = time, K_1 = const.

Next assume that the energy required per unit strain for a constant volume of material is a linear function of the instantaneous Vickers' hardness of the powder, as given in Eq (2)

$$\frac{dE}{d\varepsilon} = K_2 Hv \qquad (2)$$

where ε = true strain = $\ln\frac{L_0}{L}$, L = lamellar thickness,

During the first 300 min. of processing the Vickers hardness measured can be approximated by the following relation.

$$Hv = 0.2t + 12.2 \qquad (3)$$

where t = time, $30 < t < 420$

Therefore, combining Eq. (1), (2) and (3) and combining constants K_1 and K_2 we obtain :

$$\frac{d\varepsilon}{dt} = \frac{K_3}{Hv} \quad (K_3\text{:const.})$$

The strain, ε, as function of time is obtained by integrating over time :

$$\varepsilon = \frac{K_3}{0.2}\ln(1+0.016t)$$

The value of the system constant K_3 is calculated from the data at 120 min. where Lo=59.69μm and L=2.61μm. This value, 0.584, was used to calculate the lamellar thickness at other times shown in Fig.4. The measured lamellar thickness is in reasonably good agreement with the predicted lamellar thickness.

Particle Size and Hardness Measurement of Mechanically Alloyed Powders

The relationship between mean particle size and MA processing time with milling speed of 400rpm are shown in Fig.5. At the early stages of processing up to 120 minutes , the particle size becomes much larger than initial powder size due to the flattening of starting powders. The particle size decreases with further processing due to the effects of the roundness by milling and the powder fragmentation by work hardening. After 600 minutes of processing, the powder particle size has become more uniform in the size range of 15-20μm and has reached a steady state processing.

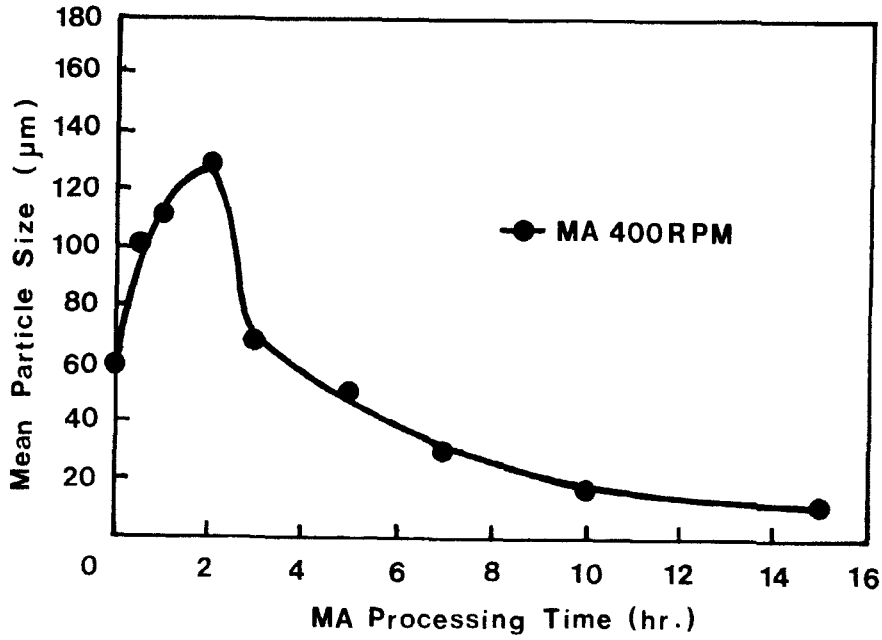


Figure 5. The relation between mean particle size and MA processing time with milling speed of 400rpm.

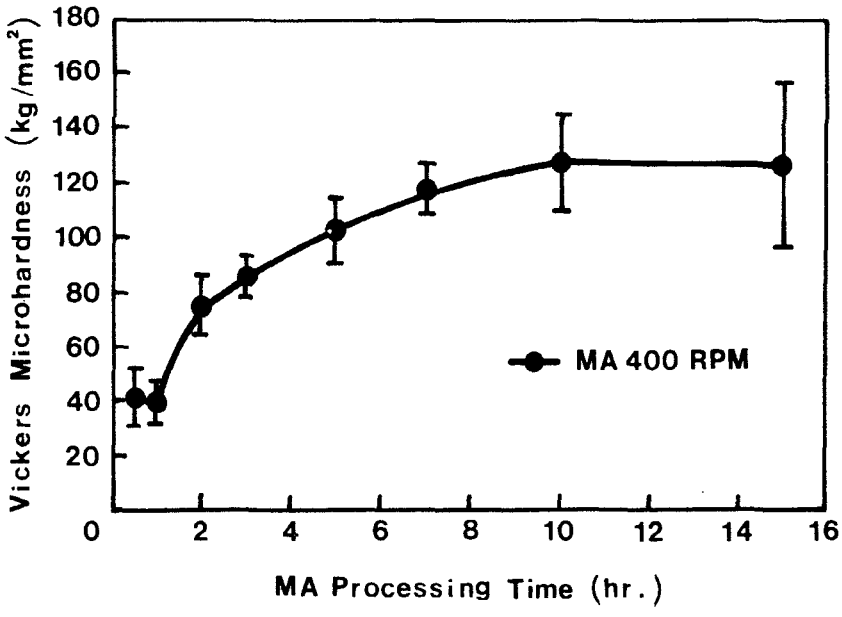


Figure 6. Variation of microhardness as a function of milling times in mechanical alloying of the Al-8wt.%Ti powder.

Microhardness measurement of individual particles provides a suitable measure of the effect of milling on metal powders, because hardness is a measure of cold work and internal defects produced during mechanical alloying.

The variation in microhardness with processing time is shown in Fig.6. A slight hardness drop at 60 minutes is due to the incomplete cold welding of the flaked particles. The microhardness increase almost linearly with increase of the processing time, reaching a saturation hardness value of 128kg/mm^2 after 600 minutes of processing. Such a saturation hardness value is supposed to be derived from the work softening such as dynamic recovery effect which balances further cold work.

From the morphological evaluation as well as particle size and hardness measurement, the achievement of a steady state processing can be identified at milling time more than 10 hours[11].

X-ray Diffraction Analysis and Chemical Homogeneity

Fig.7 illustrates the X-ray diffraction patterns of Al-8wt%Ti powders after mechanical alloying for (a) just blended, (b) as-milled for 10hr. and (c) for 15hr. with 300rpm of milling speed. As milling proceeds, the patterns clearly show the decrease of peak intensity and line broadening. Significant line-broadening is observed presumably due to the refinement and nonuniform strain of particles[12]. Lines corresponding to Ti are almost disappeared at milling time of 15hr. such a disappearance seems to be due to the formation of a pseudo solid solution, which is possibly formed by accumulation of defects, local heating and shortened diffusion distance during milling[8].

SEM micrograph, EDS intensity pattern and X-ray image analysis of the mechanically alloyed Al-8wt.%Ti powder are shown in Fig.8. The mechanical alloying was treated with the milling speed of 300rpm for 7hr. Fig.8(a) shows EDS intensity pattern on the polished single particle surface shown in (b) and Fig.8(c) is shows X-ray image of Ti element on the particle shown in (b). Fig.8(b) reveals that locally white region is Ti element introduced by incomplete processing. From this microstructural observation and the fact that the titanium has not uniformly dispersed, it may be concluded that the powders are not yet fully mechanically alloyed in these processing variables.

Fig.9. shows SEM micrograph(a) and X-ray image analysis(b) of the mechanically alloyed Al-8wt.%Ti powder. It was mechanically alloyed with milling speed of 400rpm for 10hr. As shown in this figure, the titanium elements have dispersed very homogeneously with atomic scale within the single powder particles.

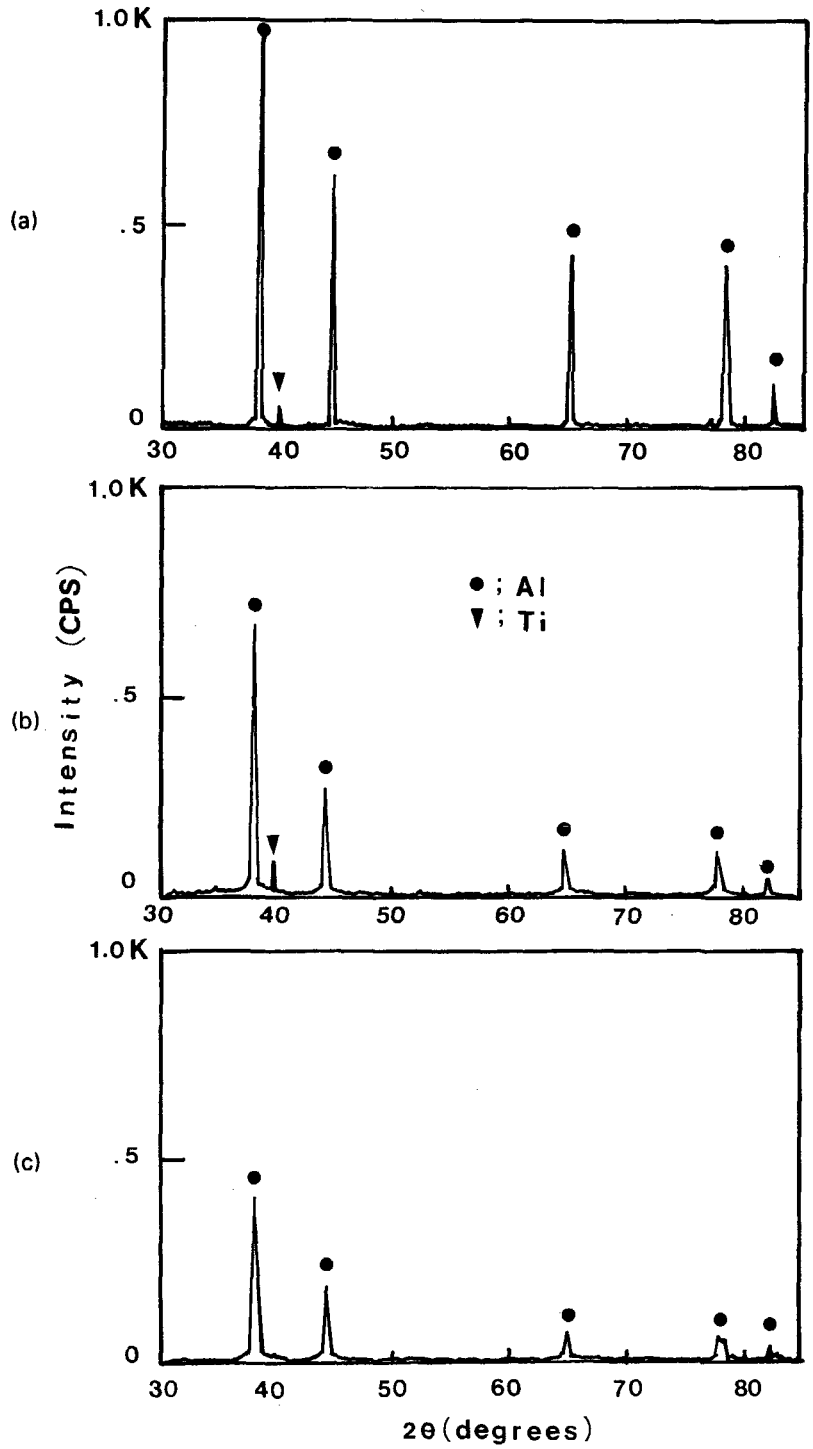
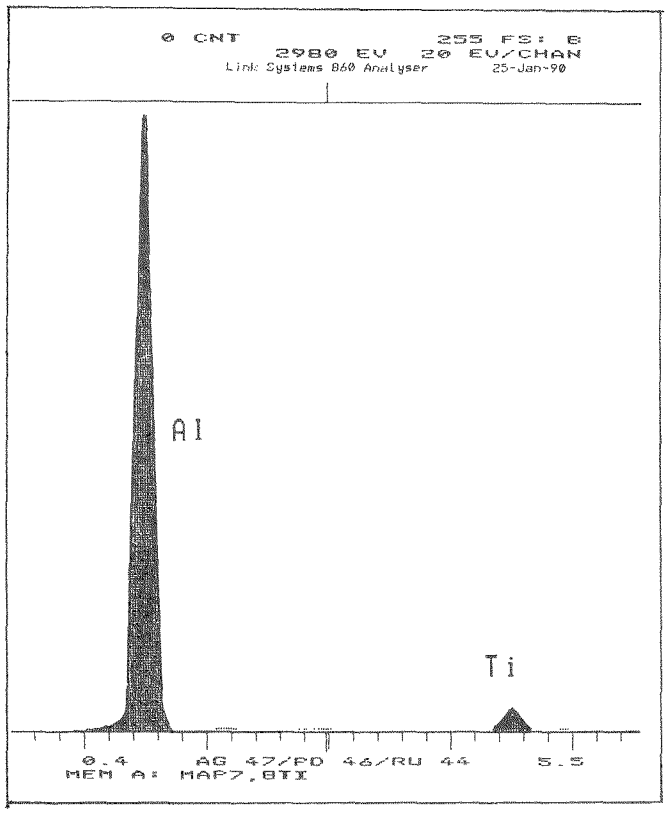
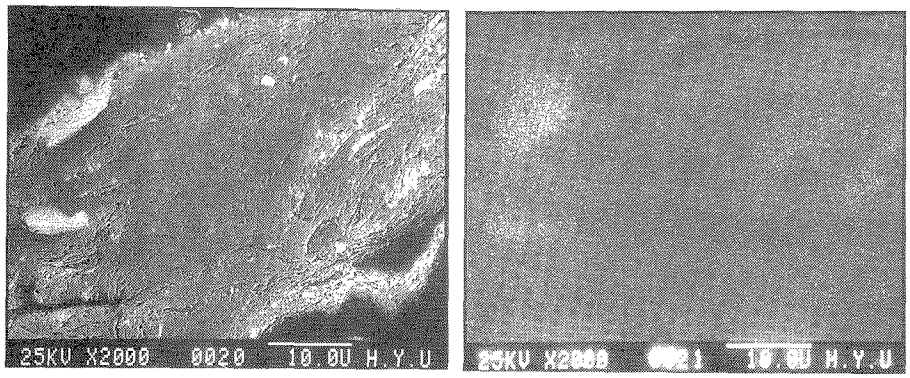


Figure 7. X-ray diffraction patterns of MA Al-8wt.%Ti powders after mechanical alloying for (a) just blended, (b) as-attrited for 10hr. and (c) for 15 hr.

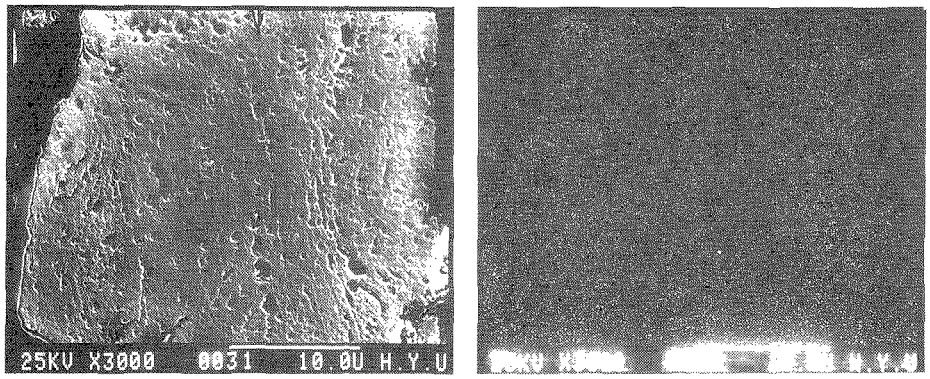


(a)



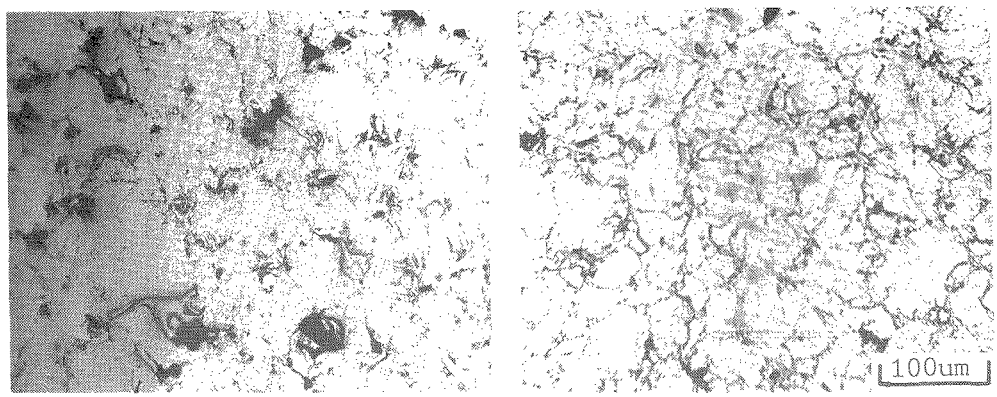
(b) (c)

Figure 8. SEM micrograph, EDS intensity pattern and X-ray image analysis of Al-8wt.%Ti powder after mechanical alloying for 7hr. with milling speed of 300rpm.; (a) EDS intensity pattern on the polished single particle surface shown in (b), and (c) X-ray image of Ti element on the particle shown in (b).



(a) (b)

Figure 9. SEM micrograph and X-ray image analysis of Al-8wt.%Ti powder after mechanical alloying for 10hr. with milling speed of 400rpm.; (a) the polished single powder particle and (b) X-ray image of Ti element on the particle shown in (a)



(a) (b)

Figure 10. Optical micrographs of MA Al-8wt.%Ti powder compact; (a) sintered at 600℃ in N_2 atm. for 1hr. after cold pressing (88%T.D.) and (b) hot pressed in a vacuum of 1×10^{-2}torr at 430℃ for 1 hr.(98%T.D.)

Characteristics of Vacuum Hot Pressed Specimen

The mechanically alloyed Al-Ti powders were too hard to uniaxially cold compact to green densities greater than 90% of theoretical density. This is due to the severe cold working of the powder particles during processing. However, near theoretical densities were successfully achieved by vacuum hot pressing.

Fig.10 shows the optical microstructures of MA Al-8wt.%Ti powder compact; (a) sintered at 600℃ in N_2 atm. for 1hr. after cold pressing and (b) hot pressed in a vacuum of 1×10^{-2}torr at 430℃ for 60 minutes. The microstructure of sintered specimen shows many large pores, having the sintered density equivalent only to 88% of theoretical density. However, the vacuum hot pressed specimen shows very densified microstructure, having the relative density of more than 98%.

Fig.11 illustrates the X-ray diffraction pattern of MA Al-8wt.%Ti specimen after it was hot pressed in a vacuum of 1×10^{-2}torr at 430℃ for 60 minutes. As shown in this figure, some Al_3Ti intermetallic

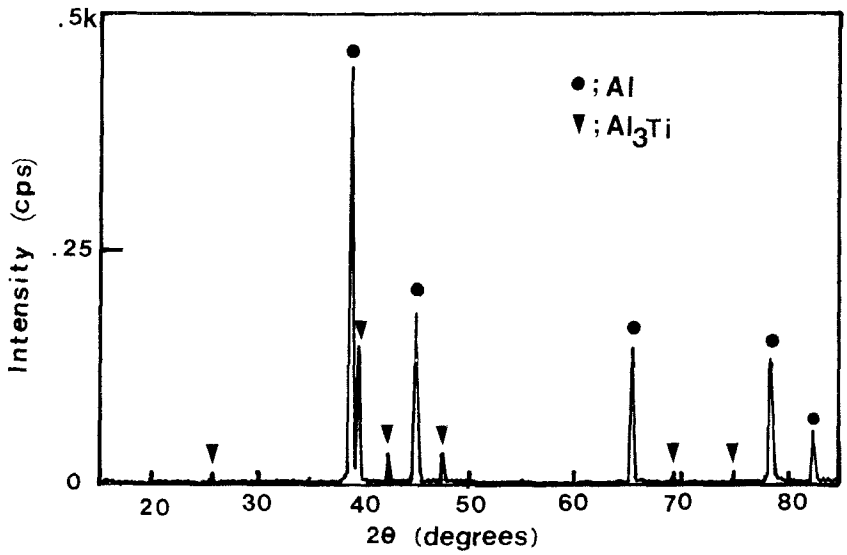


Figure 11. X-ray diffraction pattern of MA Al-8wt.%Ti powder compact, hot pressed in a vacuum of 1×10^{-2}torr at 430℃ for 1hr.

compound peaks are now detectable besides Al peaks. The heating of powders during hot pressing causes the formation of the equilibrium structure, i.e., a mixture of Al and Al_3Ti. The attainment of equilibrium is

due not only to the effectiveness of the MA process in creating an ultrafine Ti dispersion, but also to the presence of a high concentration of defects introduced during milling process[13].

CONCLUSION

1. Homogeneity of Al-Ti composite particles can be obtained by the following processing variables; addition of 1.5wt.% stearic acid as PCA, the milling speed of 400rpm and MA milling time more than 10 hours.
2. The mechanically alloyed powder prepared by the above condition reveals the achievement of a steady state processing, i.e., equiaxed powder particles, constant powder size distribution and a saturation hardness.
3. The intermetallic compound phase of Al_3Ti in the matrix was identified by X-ray diffraction analysis.

ACKNOWLEDGEMENT

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