OUKA
The University of Osaka
Institutional Knowledge Archive

| | |
|---|---|
| Title | Effect of Relatively Insoluble Compounds on β Phase Precipitation in 5083 Aluminum Alloy |
| Author(s) | Enjyo, Toshio; Kuroda, Toshio; Shinonaga, Hideyuki |
| Citation | Transactions of JWRI. 1978, 7(2), p. 173-180 |
| Version Type | VoR |
| URL | https://doi.org/10.18910/10543 |
| rights | |
| Note | |

*The University of Osaka Institutional Knowledge Archive : OUKA*

https://ir.library.osaka-u.ac.jp/

The University of Osaka

# Effect of Relatively Insoluble Compounds on $\beta$ Phase Precipitation in 5083 Aluminum Alloy†

Toshio ENJYO*, Toshio KURODA** and Hideyuki SHINONAGA***

**Abstract**

*An investigation has been made into the effect of relatively insoluble compounds on $\beta$ phase precipitation in Al-Mg series 5083 alloy. Their compounds are usually introduced by additive iron, silicon, manganese, chromium and titanium to the basic alloy in order to improve stress corrosion cracking and to inhibit recrystallization. During solution treatment of $\beta$ phase, the solution behavior of these insoluble compounds above 500°C and precipitation behavior of them at 300°C-500°C are concluded by means of electrical resistivity measurement. There are various insoluble compounds in the aluminum alloy used in this investigation, and of them, C phase-Al (Fe, Mn) Si, $\alpha$ phase-$Al_{12}Fe_3Si$ and E phase-$Al_{18}Cr_2Mg_3$ were identified by means of X ray diffraction analysis. The precipitation of $\beta$ phase by aging at 120°C and 250°C is affected by the precipitation of these insoluble compounds, and then $\beta$ phase precipitation is accelerated with decreasing of the amount of these compounds. Consequently, for the welding of 5083 alloy, the distribution, the quantity and the size of $\beta$ phase and these insoluble compounds are affected by welding procedure and weld heat input. And namely the difference of these microstructures by various thermal cyclings due to welding seems to influence upon sensitivity to stress corrosion cracking of weld joint of this alloy.*

## 1. Introduction

Al-Mg series 5083 aluminum alloy has characterized by ease of welding, good weld strength and toughness, and excellent resistance to corrosion.[1)2)] This alloy has been used for cryogenic vessels and piping such as LNG tankage and marrine applications. Recently, Al-Mg alloy has also been employed for structural and chemical applications at elevated temperatures such as large storage tank for hot (120°C) ammonium nitride solution.

Generally, this alloy has contained additive iron, silicone, manganese, chromium and titanium to the basic alloy in order to improve stress corrosion cracking resistance and to inhibit recrystallization. Their additive elements have formed various relatively insoluble compounds by prolonged homogenized heating after casting.[3)] And their compounds are useful for their improvements[4)]. For the welding, excellent properties due to additive elements are disappeared in heat affected zone (HAZ), especially in the neighbour of weld bond line, because the relatively insoluble compounds are considered to show the solution or precipitation behavior at the weld bond line.

Generally, precipitation of $\beta$ phase ($Mg_2Al_3$) is high susceptible to stress corrosion cracking[5)6)], and then, precipitation of $\beta$ phase is considered to be affected by their insoluble compounds.

In this paper, the relationship between solution or precipitation behavior of relatively insoluble compounds at various solution temperatures and precipitation behavior of $\beta$ phase was investigated.

## 2. Experimental Procedures

**Table 1** shows the chemical compositions of 5083-0 material used.

**Table 1** Chemical compositions (wt%) of material used and the plate thickness

| | Cu | Si | Fe | Mn | Mg | Zn | Cr | Ti | Zr | Al | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 5083 | 0.02 | 0.15 | 0.20 | 0.64 | 4.7 | 0.01 | 0.11 | 0.01 | – | Bal. | 62[t] |

The heat treatments of the specimen were carried out in silicone oil bath in the range up to 200°C and in salt bath in the range 200-560°C. Temperature necessary to

† Received on October 23rd, 1978
* Professor
** Research Instructor
*** Graduate Student (Present, Tokyo Shibaura Electric Co. Ltd.)

measurement has been holded in ±1°C. Microstructures were made by etching with 20% phospholic acid solution. Thin foils for transmission electron microscope were prepared using jet polishing technique in electropolishing solution in **table 2** after the specimen was cut up into

**Table 2** Chemical compositions of electropolising solution for transmission electron microscopic specimen

| | |
|---|---|
| 10 % | Perchloric acid |
| 10 % | water |
| 70 % | ethanol |
| 10 % | glycerol |

strips about 0.3 mm thick without strain by electric discharge machine.

$\beta$ phase and relatively insoluble compounds were identified by X-ray diffraction analysis. $Cu\ K_\alpha$ and $FeK_\alpha$ were employed in X-ray analysis as shown in **table 3**.

**Table 3** Conditions in X-ray diffraction method

| Target | Cu | Fe |
|---|---|---|
| Filter | Ni | Mn |
| Tube voltage | 35 kV | 30 kV |
| Filament current | 10 mA | 10 mA |
| Count full scale | 200-1000 c/s | 2000 c/s |
| Time constant | 2-5 sec | 1 sec |
| Scanning speed | 1°/min. | 2°/min. |
| Chart speed | 1 cm/min. | 2 cm/min. |
| Divergency slit | 1° | 1° |
| Receiving slit | 0.15 mm | 0.30 mm |
| Detector | Scintillation Counter | Proportional Counter |

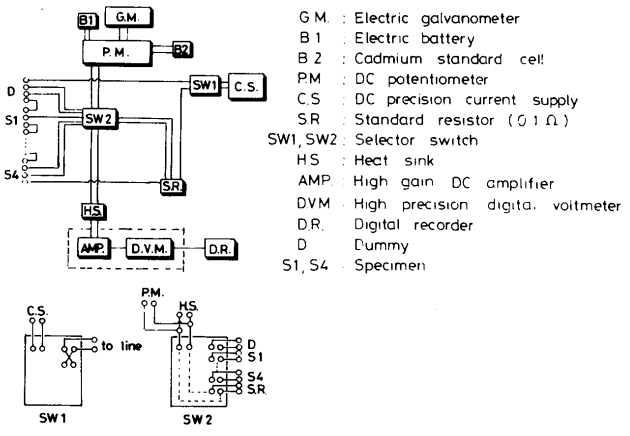



**Fig. 1** Block diagram for the circuit in electrical resistivity.

Solution or precipitation behavior of relatively insoluble compounds and $\beta$ phase was mainly investigated by electrical resistivity measurement. The specimen dimensions is 0.3 mm thick and 1.5 mm wide. Distance between the terminals is about 30 mm. Pure aluminum was used as lead wire and which was connected with the specimen by spot welding. **Fig. 1** shows the block diagram for the circuits in electrical resistivity measurement. The resistivity was derived from the measurement with a potentiometer of the voltage across the specimen when a constant current of 100 mA, stabilized to ±10 $\mu$A, was passed through it. Potential difference was measured to an accuracy of 0.02 $\mu$V. The measuring is carried out at liquid nitrogen temperature.

## 3. Results and discussions

### 3.1. Identification of relatively insoluble compounds and their solution and precipitation behaviors during solution treatment of $\beta$ phase.

Relatively insoluble compounds formed by impurities and additive elements have been proposed as $Al_{18}Cr_2Mg_3$, $Al_{12}Fe_3Si$, Al (Mn, Fe) Si, $Al_{12}$ (Fe, Mn)$_3$ Si, $Al_{12}Mn_3Si$ and $Al_{12}$ (Cr, Mn) by many investigators.[6),7)] It is called that their compounds can be distinguished with the size, for example, Fe-, Si-series compounds are relatively large size and Cr-, Mn-, Zr-series compounds are intermediate size.[6)] **Photo. 1** shows the transmission electron micrographs of thin foils for the relatively insoluble compounds.

The large compounds of 5000 Å and the compounds of relatively small size of 400 Å-1500 Å exist in the alloy, and their compounds are generally considered to inhibit recrystallization and growth of crystal. But their compounds were not able to be identified by electron diffraction analysis. Then, the identification of relatively insoluble compounds was carried out using X-ray diffraction analysis. **Fig. 2** shows the X-ray diffraction patterns of aluminum alloy used on the rolling direction surface and short transvers direction surface. In comparison of Al (111) and Al (200), the relative intensity (111) : (200) is generally 100:47. But their intensities show the same value on the ST direction surface, and then X-ray intensity of Al (111) is lower than that of Al (200) on the L direction surface, because it is considered that 5083-0 specimen used in this investigation has rolling texture structure. **Fig. 3** shows the X-ray diffraction patterns on the heat-treated 5083 aluminum alloy. **Fig. 3-(a)** shows the diffraction pattern of deposit metal by electron beam welding (EBW), **Fig. 3-(b)** shows that of base metal heat-treated at 560°C for 72 hours and **Fig. 3-(c)** shows that of base metal heat-treated at 400°C for 113 hours. Though

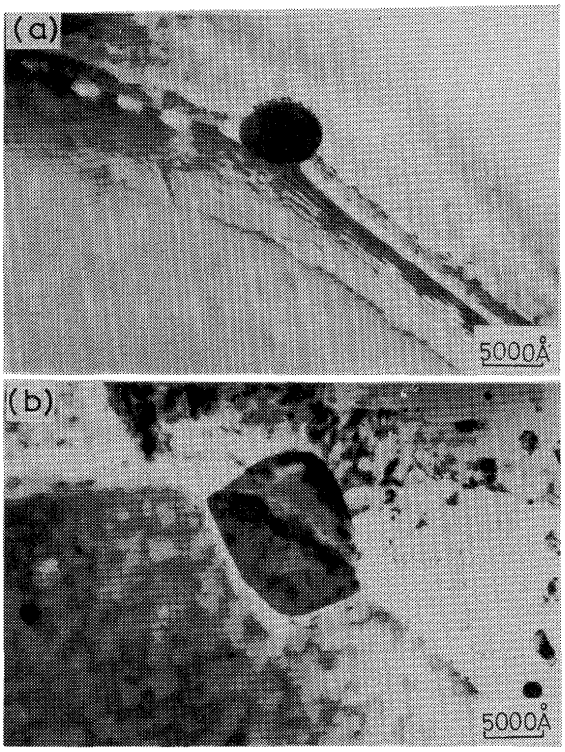



**Photo. 1** Transmission electron micrographs of relatively insoluble compounds.

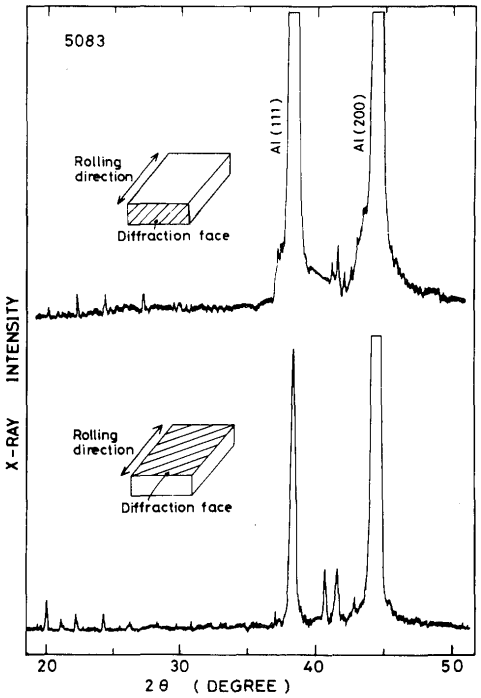



**Fig. 2** X-ray diffraction patterns on rolling direction surface and short transverse surface for 5083 alloy used.

the alloy includes various kinds of relatively insoluble compounds, we identified them by evaluating the peak at low angle side by means of calculations. As the results,

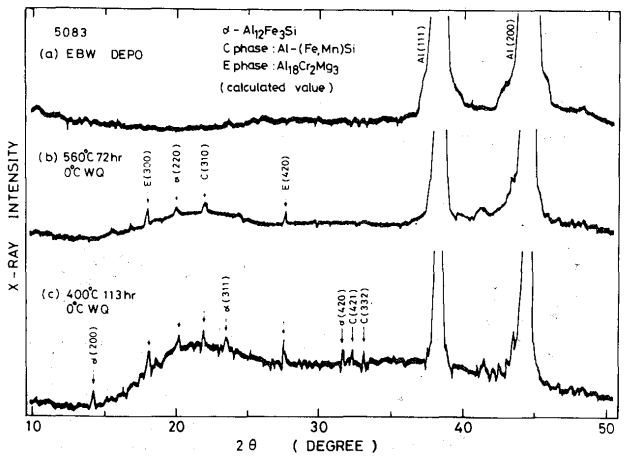
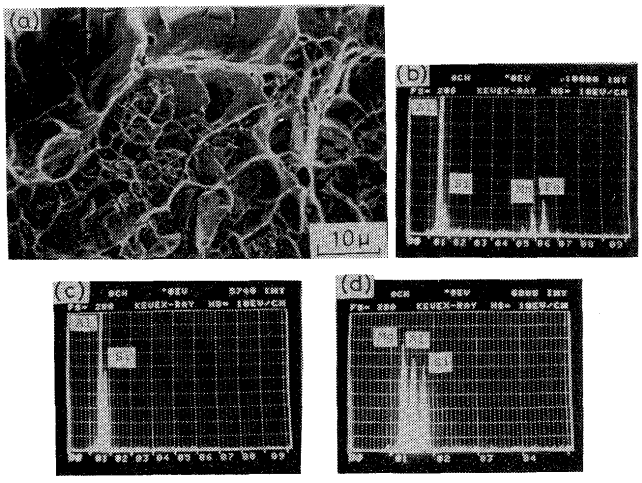



**Fig. 3** X-ray diffraction patterns on various heat-treated 5083 aluminum alloys.

we also identified three kinds of the compounds, namely $\alpha$ phase-$Al_{12}Fe_3Si$ (a=12.52 Å, cubic), C phase-Al (Fe, Mn) Si (a=12.63 Å, cubic) and E phase-$Al_{18}Cr_2Mg_3$ (a=14.55 Å, cubic). According to the results of Fig. 3, the peak owing to relatively insoluble compounds were hardely recognized for deposit metal of EBW welds. E-phase, $\alpha$-phase and C-phase were recognized for 560°C heat-treated specimen, but their intensities were low, therefore relatively insoluble compounds were considered to show considerably solution behavior.

The other side, for 400°C heat-treated specimen, many plane's peaks were recognized and their intensities were higher than that of Fig. 3-(b), therefore $\alpha$ phase and C phase were considered to have been precipitated considerably by this heat treatment.

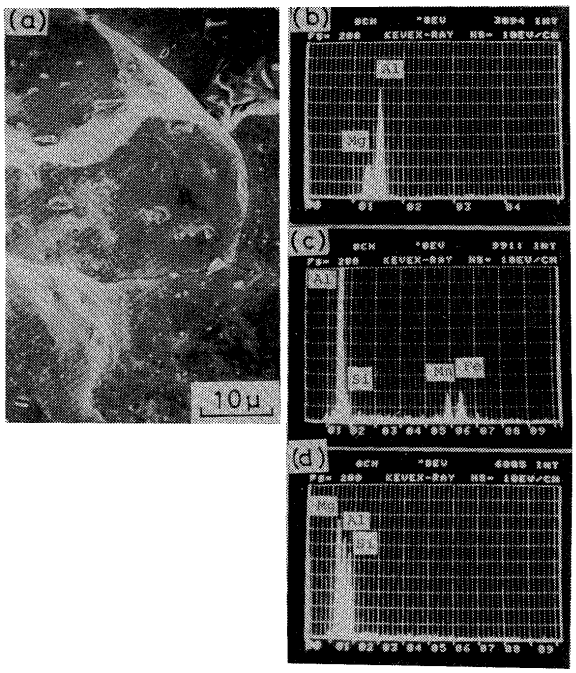
**Photo.** 2 shows the analysis of relatively insoluble




**5083 ST DIRECTION**

**Photo. 2** Analysis of relatively insoluble compounds on the fracture surface of ST direction specimen for 5083-0 material by X-ray energy dispersion method.

compounds on the fracture surface of ST direction specimen for 5083-O material by X-ray energy dispersion method. The fracture morphology was transgranular fracture, and the compounds and inclusions are present in dimple bottom. **Photo. 3** shows the analysis of relatively




SCC
5083 (560°C 2hr, 0°C WQ
120°C 1month)

**Photo. 3** Analysis of relatively insoluble compounds on the fracture surface of stress corrosion cracking in 3.5% NaCl + 0.5% $H_2O_2$ solution for ST direction heat-treated at 120°C for 1 month.

insoluble compounds on the fracture surface of stress corrosion cracking in 3.5% NaCl + 0.5% $H_2O_2$ solution for ST direction specimen heat-treated at 120°C for 1 month. The fracture morphology was completely intergranular fracture, and on the surface, Al (Fe, Mn) Si series compounds were often recognized.

**Fig. 4** shows the isothermal aging curves of electrical resistivity at various solution temperatures for 5083 base metal. Generally, it is said that electrical resistivity changes due to soluto atom's concentration in the matrix, therefore, the resistivity becomes higher as the precipitates and the compounds show the solution behavior, reversely the resistivity becomes lower as they show the precipitation behavior. The resistivity decreases as aging time is prolonged for 400°C and 460°C aging, therefore the precipitation of relatively insoluble compounds occurs as aging time is prolonged, namely the precipitation of the compounds is predominant at their aging temperatures. The other side, for aging temperature 560°C, the

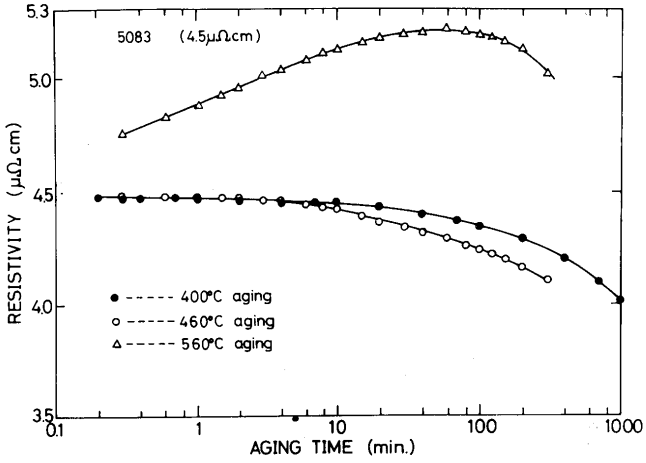



**Fig. 4** Isothermal aging curves of electrical resistivity at various solution temperatures for 5083 base metal.

resistivity became 0.3 μΩ cm higher than that of as-received specimen (4.5 μΩ cm) by being aged for only 18 seconds at 560°C, namely relatively insoluble compounds show the solution behavior by being aged for a few times at 560°C. As aging exceeds nearly 100 minutes, the resistivity reversely falls. It is suggested that the relatively insoluble compounds which precipitate even at 560°C are present in various kinds of the compounds.

**Fig. 5** shows the isothermal aging curves of electrical

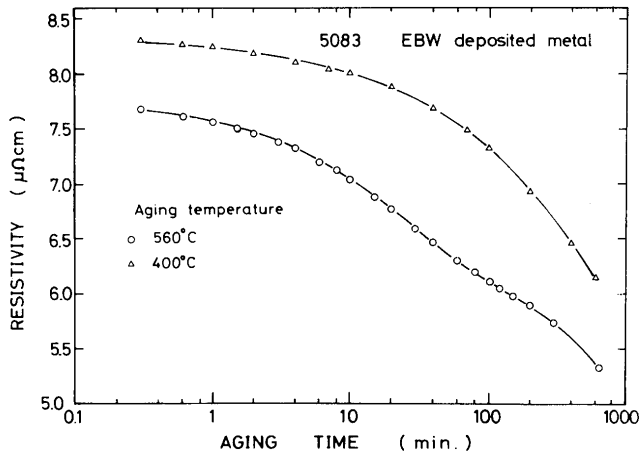



**Fig. 5** Isothermal aging curves of electrical resistivity for EBW welds deposit metal of 5083 alloy.

resistivity obtained from EBW welds deposit metal. The insoluble compounds precipitate at any aging temperature, for the reason that the compounds are solutionized by force in the matrix for EBW welds.

The resistivity fluctuates after 100 minutes exceeded for 560°C aging, and which is suggested that at least two kinds of the compounds exist.

### 3.2 Effect of relatively insoluble compounds on $\beta$ phase precipitation

Fig. 6 shows the isochronal annealing curves of

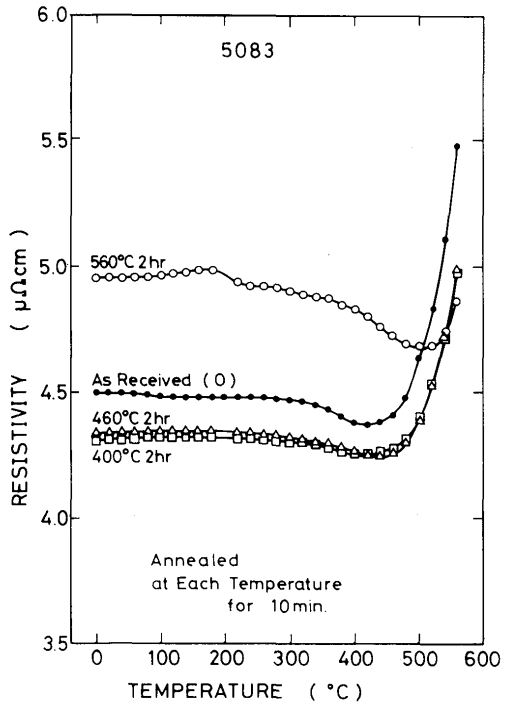



Fig. 6 Isochronal annealing curves of electrical resistivity for 5083 base metal after heat treatment at various temperatures.

electrical resistivity for base metal. The resistivity at 0°C varried with solution temperatures, namely the resistivity of 560°C heat-treated specimen was higher than that of 400°C and 460°C heat-treated specimens as shown in Fig. 4.

It is generally said that $\beta$ phase precipitates at the grain boundary by about 120°C aging and in the grain and at the grain boundary by about 250°C aging.[9)10)] But for the isochronal annealing method shown in Fig. 6, the resistivity change due to $\beta$ phase precipitation has hardely recognized at 120°C and 250°C.

Fig. 7 shows the isochronal annealing curve of EBW welds deposit metal. The resistivity at 0°C shows the high value above 8 $\mu\Omega$cm as shown in Fig. 7, for the reason that the compounds are solutionized by force in the matrix. In the isochronal annealing curve, the resistivity suddenly falls at the temperature 350°C to 500°C, it is suggested that previously solutionized relatively insoluble compounds precipitate at their temperatures again.

The other side, if relatively insoluble compounds had already precipitated such as the 400°C heat-treated specimen in Fig. 6, they hardely precipitate by reheating of 350°C to 500°C.

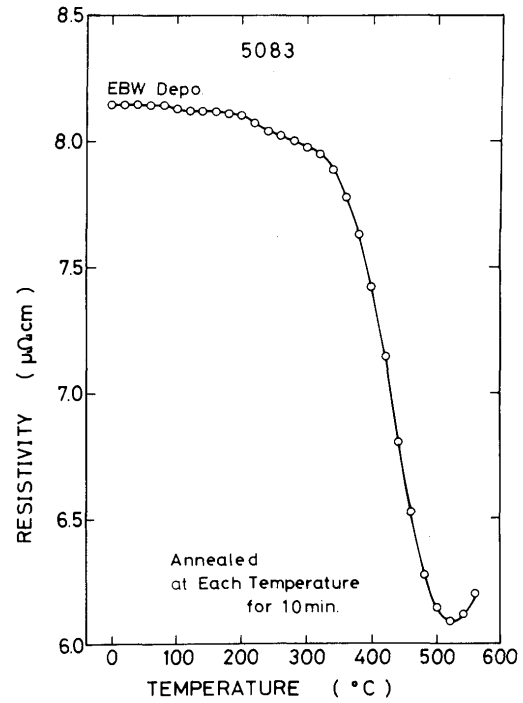



Fig. 7 Isochronal annealing curve of electrical resistivity for EBW welds deposit metal of 5083 alloy.

The resistivity changes at 120°C and 250°C annealing temperature were clearly recognized as shown in Fig. 7, namely it means that $\beta$ phase precipitates at their temperatures. Consequently, it is suggested that $\beta$ phase precipitation is accelerated by solution of relatively isoluble compounds.

On the phenomena, isothermal aging curves were evaluated in detail. Fig. 8 and Fig. 9 show the isothermal

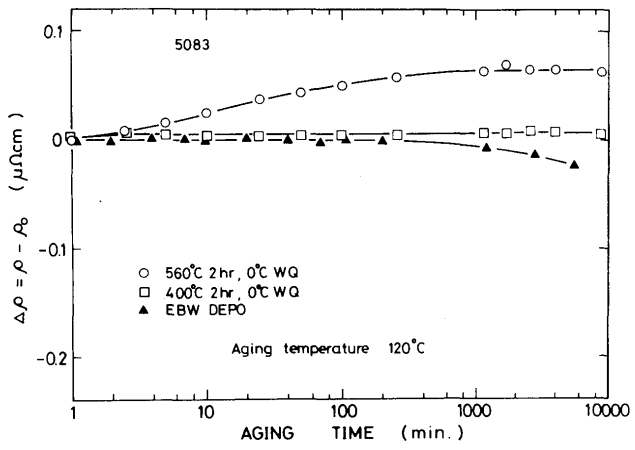



Fig. 8 Isothermal aging curves of electrical resistivity at 120°C for the specimens heat-treated at various temperatures.

aging curves of electrical resistivity at 120°C and 250°C respectively for the base metals heat-treated at 560°C and 400°C, and EBW welds deposit metal.

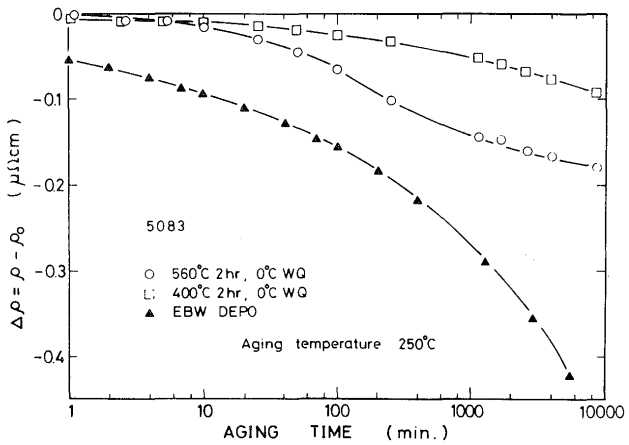



Fig. 9 Isothermal aging curves of electrical resistivity at 250°C for the specimens heat-treated at various temperatures.

Vertical axis ($\Delta\rho = \rho - \rho_0$) shows the difference between value ($\rho_0$) immediately measured after quenching from solution temperature and value ($\rho$) at each aging time. But it was not able to measure the electrical resistivity immediatelly after welding for EBW welds deposit metal, therefore the value after being aged at each temperature for 0.3 minutes was proposed as $\rho_0$.

The electrical resistivity at 120°C aging increases in increasing of aging time for 560°C solution-treated specimen.

The results considered to produce for the reason that the decrease of the resistivity due to $\beta$ phase precipitation was superimposed to the increase of resistivity value due to G.P. zone precipitation. Therefore, measuring of the electrical resistivity was unsuitable for study of $\beta$ phase precipitation at 120°C aging. The other side, the electrical resistivity at 250 °C aging decreases as aging time increases for various treated specimens as shown in Fig. 9. It is said that G.P. zone hardely precipitate at 250°C aging, therefore their resistivity changes will be occured by $\beta$ phase precipitation. Namely, their results show that $\beta$ phase precipitation at 250°C aging is accelerated with decreasing of the amount of these compounds.

As shown in **Photo. 4**, the microstructure variety owing to grain boundary etching for specimens aged at 120°C after water quenching from 400°C and 500°C was observed in order to evaluate $\beta$ phase precipitation at the grain boundary. The grain boundary etching was not yet occured at any solution temperature for 2 hours aging.

The grain boundary of 400°C heat-treated specimen was etched discontinuously, but that of 560°C heat-treated specimen was etched continously for 2 days aging. For 1 month aging, the grain boundary of 400°C heat-treated specimen was yet etched discontinuously, but that of 560°C heat-treated specimen was etched severely and continuously. Namely, as relatively insoluble compounds are solutionized, following $\beta$ phase precipitation is accelerated, based on the micrograph's evidence. Furthermore, according to the microstructure observation shown in **Phoro. 4**, the structure for the specimen heat-treated at 560°C changes from rolling texture to recrystallized structure, namely it was confirmed that the action to

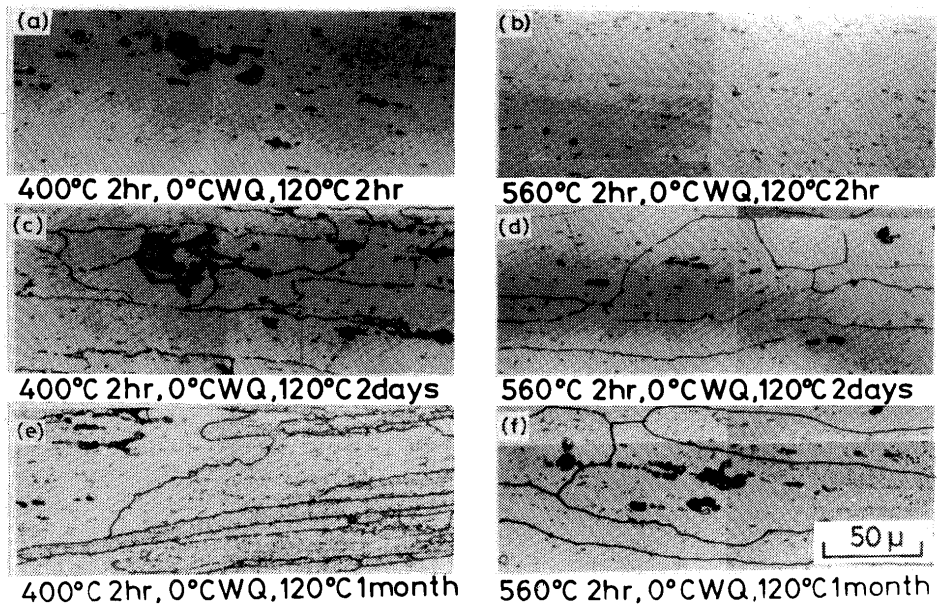



**Photo. 4** Microstructure variety owing to grain boundary etching for specimens aged at 120°C after quenching from 400°C and 560°C.

inhibit recrystallization disappears as the relatively insoluble compounds are solutionized.

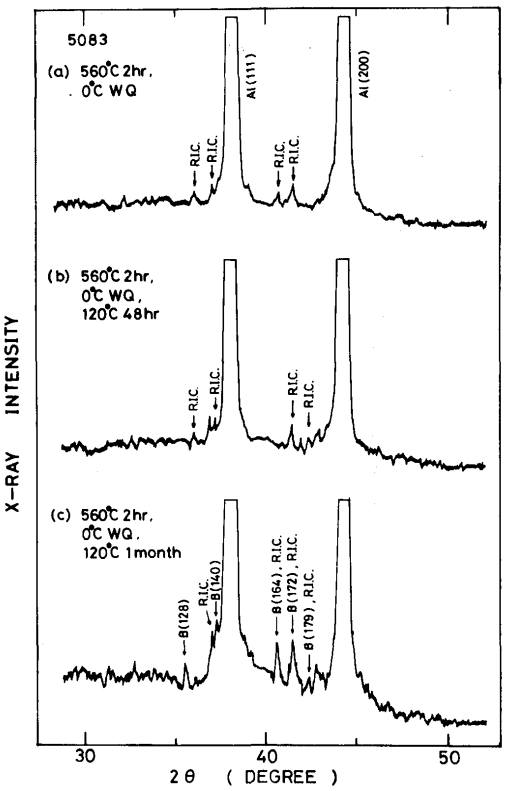



Fig. 10 Variation of X-ray diffraction pattern with aging time at 120°C for specimens solution-treated at 560°C. (RIC: Relatively insoluble compounds)

Identification of $\beta$ phase was carried out using X-ray diffraction analysis. **Fig. 10** shows the change in diffraction patterns when the specimens heat-treated at 560°C were aged at 120°C. Identification of $\beta$ phase was evaluated by calculation. The lattice parameter of $\beta$ phase, which is fcc, is a=28.28 Å[8] The diffraction line of $\beta$ phase exists at the same angle as that of relatively insoluble comounds as shown in Fig. 10. The diffraction line of $\beta$(128) has the strongest intensity, and that of $\beta$(164), $\beta$(172) and $\beta$(179) fall on that of relatively insoluble compounds. But the intensity of diffraction line due to $\beta$ phase increases with increasing of the aging time at 120°C. Therefore, it is unquestionable that $\beta$ phase precipitates on the aging process at 120°C.

**Photo. 5** shows the transmission electron micrographs showing the $\beta$ phase precipitation at the grain boundary and relatively insoluble compounds for various heat-treated base metals and EBW welds deposit metal. 5083-O materials (as-received) has the rod-type compounds of length 3000 Å and the round-type compounds in the size range 1000-2000 Å as shown in Photo. 5-(b) The relatively insoluble compounds are hardely observed for EBW welds deposit metal as shown in **Photo. 5-(a)** and dislocations produced by quench strain due to rapid quenching are observed. Therefore it is considered that the dislocation density is high for EBW welds deposit metal. The reason that the resistivity is higher than that of base metal shown in Fig. 7 is considered to be due to the phenomena as

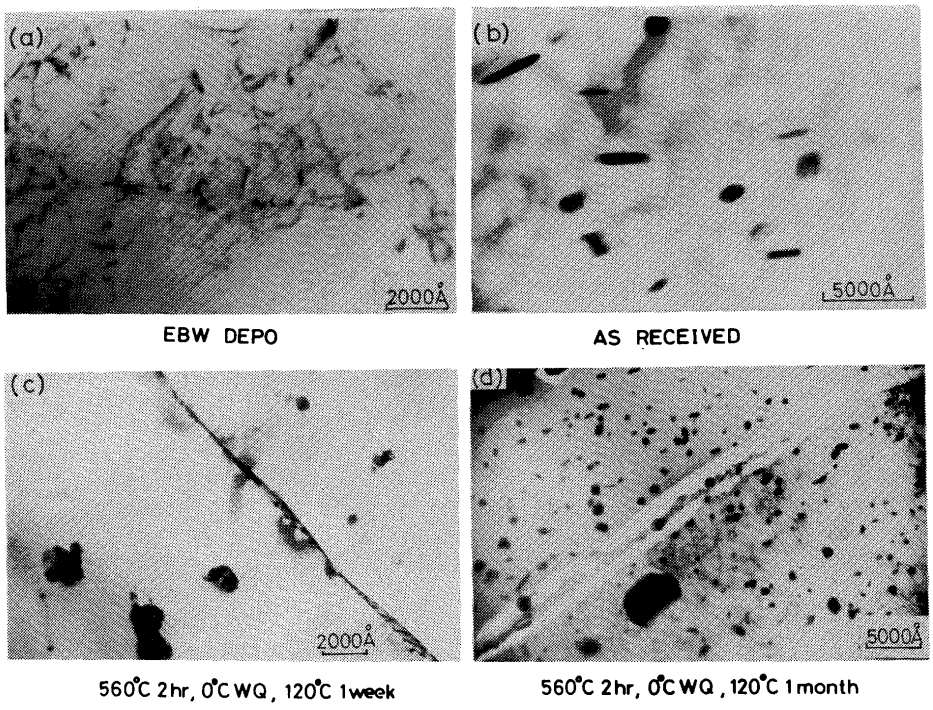



**Photo. 5** Transmission electron micrographs showing $\beta$ phase precipitation at the grain boundary and relatively insoluble compounds for various heat-treated base metals and EBW welds deposit metal.

shown in Photo. 5-(a).

The relatively insoluble compounds are present a few and the size is larger than that of as-received material for the specimen after 560°C solutionizing followed by 1 week aging at 120°C as shown in **Photo. 5-(c)**, and the $\beta$ phase precipitates discontinuously at the grain boundary. In the specimen aged at 120°C for 1 month as shown in **Photo. 5-(d)**, $\beta$ phase precipitates both in the grain and at the grain boundary. Especially the grain boundary is observed as the white band due to etching of $\beta$ phase during preparation of thin foil.

Generally, G.P. zone at room temperature or the low temperature in Al-Mg alloy can be detected only the resistivity measurement[11], and G.P. zone is thermally unstable and hardely contribute to age hardening, on the contrary to Al-Zn-Mg alloy and Al-Cu alloy.

The resistivity in the range 0°C to 200°C increases slightly for the isochronal annealing curves shown in Fig 6 and for isothermal aging curve of 560°C heat-treated specimen shown in Fig. 8. The resistivity decreases owing to $\beta$ phase precipitation, but in this investigation, the resistivity increases reversely in spite of $\beta$ phase precipitation. It is suggested to be due to the reason that G.P. zone containing Cu atom[12] exists even at above 100°C for the material used. For high temperature aging such as 250°C aging, $\beta'$ precipitates and $\beta$ phase precipitations are generally accelerated by containing the element of Cr, Ag and Be. But it is interesting that $\beta$ phase precipitation is retarded by the precipitation of relatively insoluble compounds in spite of containing Cr element for 5083 material used. In this paper, it is not clear whether any series relatively insoluble compounds affects the $\beta$ phase precipitation. The fact that $\beta$ phase precipitation is affected by the solutionizing or precipitation of relatively insoluble compounds is important in order to evaluate the microstructure, and mechanical and chemical properties on welds.

Namely it is suggested that $\beta$ phase precipitation at weld bond area for large heat-input welding is much more than that of base metal which the compounds had precipitated homogeneously, and which has a bad effect in the mechanical and chemical properties on welds.

## 4. Summary

An investigation has been made into the effect of relatively insoluble compounds on $\beta$ phase precipitation in Al-Mg series 5083 aluminum alloy. The results obtained in this investigation are summerized as follows.

(1) 5083 aluminum alloy used contains many kinds of the relatively insoluble compounds due to impurities and additive elements of Fe, Si, Cr, Mn, so on, and of them, C phase-Al (Fe, Mn) Si, $\beta$ phase-$Al_{12}Fe_3Si$ and E phase-$Al_{18}Cr_2Mg_3$ were identified by means of X-ray diffraction analysis.

(2) When the alloy are heated above solution temperature of $\beta$ phase such as welds, the relatively insoluble compounds show the solution or precipitation behavior. By means of electrical resistivity measurement, the compounds show the solution behavior above 500°C, and precipitation behavior in the temperature range 340°C to 500°C, respectively.

(3) The precipitation of $\beta$ phase for aging at 120°C and 250°C is affected by the precipitation or solution of these relatively insoluble compounds, and then $\beta$ phase precipitation is accelerated with decreasing of the amount of these compounds

(4) Therefore, it is suggested that $\beta$ phase precipitation at weld bond area for large heat-input welding produces much more than that of base metal, and which has a bad effect in the mechanical and chemical properties on welds.

### Acknowlegements

The authors wish to thank Mr. M. Nakamura and Mr. T. Horinouchi for their variable contributions in this work. The support of the Light Metal Educational Foundation is grately acknowledged.

### References

1) W. L. Fink and D. W. Smith: Trans. AIME tech. pub. 760 (1936) 12
2) A. Eikum and G. Thomas: Acta Met. 12 (1964) 537
3) R. B. Niederberger, J. L. Basil and G. T. Bedford: Corrosion 22 (1966) 68
4) E. C. W. Perryman and S. H. Hadden: J. Inst. Metals 77 (1950) 207
5) P. Brenner and G. J. Metcalfe: J. Inst. Metals 81 (1952-53)
6) G. T. Hahn and A. R. Rosenfield: Met. Trans. 6A (1975) 653
7) B. J. Punwoody and D. M. Moore: J. Inst. Metals 101 (1973) 172
8) A. Saulnier and P. Mirand: Mem. Sci. Rev. Met. 57 (1960) 91
9) P. T. Gilbert and S. E. Hadden: J. Inst. Metals 77 (1950) 237
10) E. H. Dix, W. A. Anderson and M. B. Shumaker: Corrosion 15 (1959) 55t
11) C. Panseri, T. Federighi and S. Ceresara: Trans. AIME 227 (1963) 1122
12) Y. Baba: Trans. Japan Inst. Metals 13 (1972) 76