Early and Recent Research on Aluminum in Japan

Y.Murakami, OSTEC and Kyoto University

ABSTRACT: In order to introduce the situation of research on aluminum in Japan, I would like to talk on some subjects from the early years of research on aluminum in this country and also to refer to some recent researches.   I will also note the activities of the two Japanese organizations and some others, which have been contributing greatly to the promotion of R&D of aluminum materials in academic circles.

The total consumption of aluminum in Japan was 4.12 million metric tons in 1997, and research and development of aluminum is rather active; Japan has grown to be one of the advanced countries for the aluminum industry as well as for research and development in aluminum. However, in the 1930s and 1940s, Japan was in her earliest infancy because the commencement of production of primary aluminum in Japan was in 1934, although the production reached a peak, as it often does, during war time with nearly 145,000 metric tons, being produced because of the demand for military use. As for research and development, much effort had been exerted to catch up with the leading countries in Europe and USA.. In this presentation, how the researches on aluminum in Japan progressed from an early state to the present will be shown by means of various examples. Firstly, I shall begin to talk with three studies which were carried out in the early days and are particularly worth mentioning.

1-1   Development of EDS (Extra-Super Duralumin) [1, 2, 3]

A high strength Al-Zn-Mg- Cu alloy, named " ESD " (Extra-Super Duralmin), was developed originally in Japan , and adopted officially as a structural alloy for the "Zero-type " navy fighter in 1938 . ESD has a similar composition to that of 7075 alloy registered later in 1943 in the USA.

Al-Zn-Mg-Cu alloys, such as AA 7075 provide high strength and stiffness in the T6 temper, but are prone to stress-corrosion cracking. Recently, considerable efforts have been directed towards a better understanding of the mechanism of SCC, and the important features have been clarified to improve the resistance to SCC in high-strength aluminum alloys from the results of experiments using various kinds of advanced experimental techniques. However, in the early days of the 1930s and 1940s, alloy developments were carried out mainly with the help of the "Trial ad Error" method.   In war time , the Japanese navy needed an aircraft structural material with 10 kgf/mm² higher tensile strength than that of the Super-Duralmin, which had the highest tensile strength at that time. To meet this demand, Drs Isamu Igarashi and Goroh Kitahara [1,2,3], Japanese metallurgists, pursued intensive studies on the three kinds of alloys, Super-Duralmin(D), Sanders Alloy (S) and E Alloy (E), the compositions of which are shown in Table 1 .   They found firstly an alloy with tensile strength of 58 - 70 kgf/mm², but the problem of SCC remained. The composition of this alloy is shown also as Cr-free ESD in Table 1. Then they continued to study extensively effects of small amounts of alloying elements such as Cr, Mn, Ti, Mo, Co, Be, Ni, Ag, Ca, Si, Sn on the susceptibility to SCC of the above alloys, and found that addition of Cr was most benificial and the crucial

factor for prevention of SCC. From these extensive examinations, they obtained a high-strength alloy containing small amounts of Cr, named ESD (Extra-Super Duralmin), and the related alloys with improved resistance to SCC, as shown in the lower part of Table 1 and Table 2. The nominal

Table 1   Composition on Alloys Studied Initally, and Composition of ESD No. 1, No. 2 and AA 7075

Alloys	Chemical Composition (wt %)					
	Cu	Zn	Mg	Mn	Cr	Al
Super-Duralmin	4	–	1.5	0.5	–	Remainder
Sander Alloy	–	8	1.5	0.5	–	Remainder
E (Zinc Duralmin)	2.5	20	1.5	0.5	–	Remainder
Cr-Free ESD	2-2.5	8-10	1.5	0.5	–	Remainder
	Zn	Mg	Cu	Mn	Cr	
ESD NO. 1	7-9	1.2-1.8	0.5-2.5	0.3-1.0	0.1-0.4	Remainder
ESD No. 2	6-7	1.6-2.0	1.5-2.5	0.3-1.0	0.1-0.4	Remainder
AA 7075	5.1-6.1	2.1-2.9	1.2-2.0	0.3	0.18-0.28	Remainder

Table 2   Japanese Patents of ESD and Related Alloys

Patent No.	Alloys	Patent Applied Date		Compositions (wt%)					
		Applied	Registered	Zn	Mg	Cu	Cr	Mn	Ti
135,036	High Strength Alloy (ESD)	1936/6/6	1940/2/28	3 - 20	1-10	1-3	0.1-2	0.1-0.4	0.01-0.5
113,183	High Strength Alloy	1936/9/9	1940/11/13	4 - 20	1-3	1-3	–	1.2 - 3	0.01-0.5
155,806	Clad Alloy	1939/10/14	1943/4/2	0.5-1.5	–	–	0.1-0.3	–	–— Sb ——
161,892	High Strength Alloy	1942/5/29	1944/5/29	3 - 20	1-10	1-3	0.1-0.3	0.1-0.2	0.01-0.2

composition of ESD is Zn 3 - 20%, Mg 1 - 10%, Cu 1-3%, Cr 0.1 - 2%, Mn 0.1 - 2%. However, ESD No.1 used practically had the composition of Zn 7 - 9%, Mg 1.2 - 1.8%, Cu 0.5 - 2.5%, Mn 0.3 -1.0%, Cr 0.1 - 0.4, and the mechanical properties of UTS > 570MPa, $\sigma_{0.2}$ >490MPa, δ >5%. Later, in order to meet the requirement for improved fatigue properties, a modified version (ESD No.2) had been developed. This alloy, as extruded, had a fatigue strength of more than 196 MPa in spite of the same UTS. The composition of ESD No.2 was Zn 6 - 7%, Mg 1.6 - 2.0%, Cu 1.5-2.5%, Mn 0.3 - 1.0%, Cr 0.1 - 0.4%. They had recognized, as mentioned above, that a small addition of Cr did play an important part in SCC due to effective grain refinement. The development of ESD in the early days was a major achievement, although the research was carried out only with the accumulated practical experience of the metallurgists.

1-2   The First Establishment of the Phase Diagram of the Al-Cu-Mg Alloy System and Designation of "S" Compound [4,5]

The phenomenon of age-hardening and the first age-hardenable alloy originated from the discovery by Alfred Wilm in 1906, when he produced " Duralmin ", of which the nominal composition is Cu 3.5%, Mg 0.5%, Mn 0.5% and Al remainder. The discovery was accidental and dramatic. No one knew the reason why the alloy hardened, and thus there existed no fundamental ideas to guide research toward better age-hardening alloys. Decisive progress was made by an American metallurgist, Paul Merica in 1919, who showed that, in the equilibrium state, the solubility of the alloying element increased with temperature in the Al-Cu binary alloy system. Nevertheless, early metallographic investigation showed that the microstructural changes during the initial stages of aging

were on too fine a scale to be resolved by the light microscope, yet it was in these early stages that the most profound changes in properties were found. In 1938, decisive evidence was afforded by A. Guinier and G.D.Preston, who found the so-called "Guinier - Preston Zones or G. P. Zones " independently by using the X-ray Laue method.

An addition of Mg to the binary Al - Cu alloy offered a high response to both natural and artificial aging. Therefore, there was a strong need for the ternary phase diagram of the Al-Cu-Mg system. The first complete constitution of the Al rich-corner of the Al-Cu-Mg system was reported in 1937 by Prof. Hideo Nishimura, Kyoto University (my former teacher) [4] in the Journal of the Japan Institute of Metals, 1937, Vol. 1, p.8, although the ternary diagram established by G, G, Urazov and M. S. Mirgalovskaya in 1949 [5] (*Izvest. Sekt. Fiziko-Khimich. Analiza, 1949, 18, 117*) is now well known due to be being included in the Metals Handbook edited by ASM.
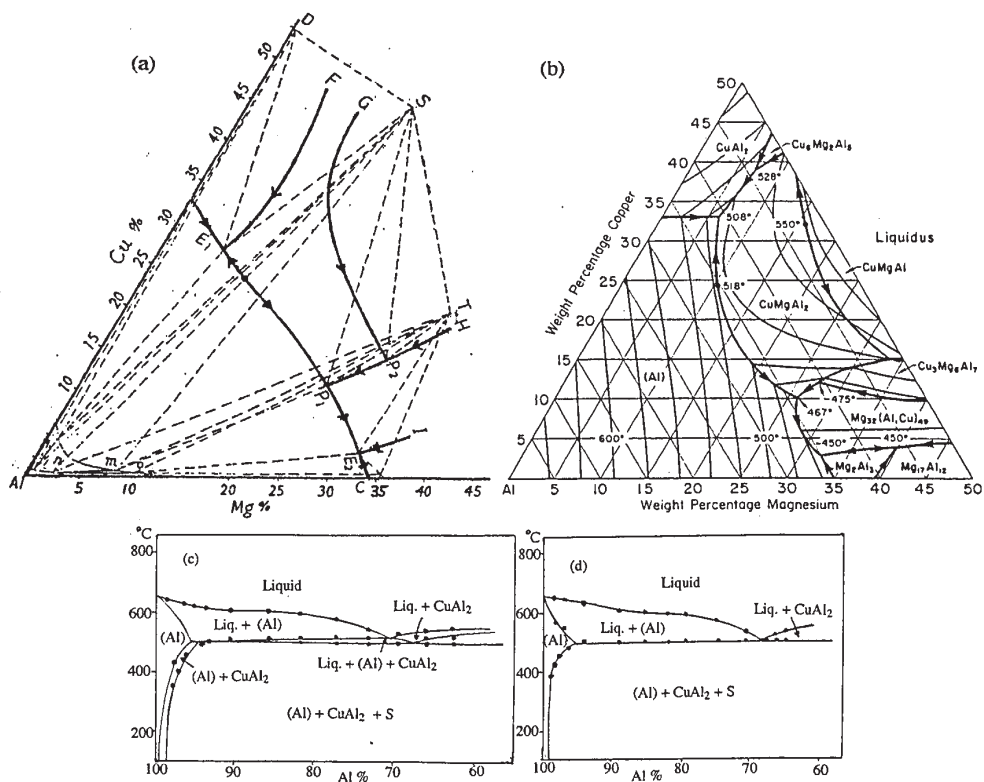

Fig, 1 The Phase Diagrams of the Al-Cu-Mg System by (a) Prof. H. Nishimura [4], (b) Urazov and Mirgalvskaya [5] ; and The Two Vertical Sections of the Al-Cu-Mg System [4], (d) Cu : Mg = 7 : 1 (Duralmin), (d) Cu : Mg = 3 : 1 (Super-Duralmin).

The phase diagram and two vertical sections with Cu:Mg ratio 7:1 (Dulalmin) and 3:1 (Super-Duralmin) studied by Prof. Nishimura [4] is shown, together with Urazov and Mirgalvskaya's published in the Metals Handbook [5] (Metals Handbook, 8th edition, 8(1973), 386), in Fig. 1 (a) to (d) respectively. These two diagrams are very similar to each other, and they contain two kind of the ternary intermetallic compound designated firstly as 'S' and 'T' by Prof. Nishimura, and thus a

priority should be given to Prof. Nishimura's work. His academic achievement was two-fold; one was to establish the phase equilibrium in the Al-rich corner of the Al-Cu-Mg system, and the other was to obtain the conclusion that aging of Duralmin and 2024 type Super-Duralmin is a direct result of the change in solid solubility with temperature of the S-compound, as clearly shown from the two vertical sections in Fig. 1.

1-3 Invention of the " Riken Alumite Process " - A Sealing Process by Pressured Water Vapor [6]

Aluminum should have a low corrosion resistance principally due to the fact that it is very active thermodynamically. However, most aluminum alloys have good corrosion resistance due to the presence of a thin, adherent naturally occuring film of Al_2O_3 on the surface. At present, even thicker , e. g. 10 - 20 μm, surface films are produced by the more commonly used treatment of anodizing. In this case, the component is made the anode in an electrolyte, such as an aqueous solution containing 1 - 3% oxalic or 10 - 18% sulphulic acid, which produces a porous Al_2O_3 film that is subsequently sealed, i. e. rendered non-porous, by pressured water vapor. Moreover, the anodic coating can be dyed to give attractive colors and, colored materials are widely used in architectural as well as ornamental applications.

The sealing process by pressured water vapor was originally invented in 1932 by Dr. Akira Miyata, a scientist of Riken (The Institute for Physics and Chemistry Research) in Tokyo. This process of anodizing and sealing by pressured water vapor is well-known, particularly in Japan, as the " Riken Alumite Process ". ([6] Akira Miyata and Shoji Setoh, Science Papers of the Institute for Physics and Chemistry Research, Tokyo, **17** (1932), 189). The discovery of this process was accidental. When he was washing off remaining oxalic acid electrolyte from the surface of anodized articles in boiling water, he accidentally found out that oxide film on one particular article was turned transparent. The surface of this article had been unintentionally put on another and thus was not exposed to boiling water. From this suggestive hint, he proceeded further with work using the treatment in pressured water vapor of 4 - 5 atmosphere, and reached the conclusion that the porous γ-Al_2O_3 film was able to be sealed due to volume expansion of γ-$Al_2O_3 \cdot H_2O$ produced by combining of one molecule of H_2O by pressured water vapor, and became to be transparent. Moreover, this film was proved to be very hard and corrosion-resistant even in a strong alkaline solution. This invention can be seen to be an outstanding achievement of early aluminum research in Japan.

2-1 The Japan Institute of Light Metals (JILM)

The Japan Institute of Light Metals (JILM) was established on 26th May 1965 through the reorganization of its precessor, the Light Metal Research Conference which started in November 1951. JILM has since then greatly expanded as a venue for close communication and cooperation among researchers and engineers in academic, administrative and industrial circles. In October 1991, the International Conference " RASELM '91' " was held as a commemorative event of the 40th anniversary of its foundation, and many partcipants came to our country from 30 countries throughout the world. " ICAA - 6 " is now being held here in Toyohashi with the support of JILM.

At present, the membership has risen to 2234 (30th September, 1997), comprising 64.0% private enterprises, 22.1% universities and colleges, 4.8% public institutes and 9.1% students. JILM's activities are varied, ranging from the publication of the Journal of JILM, issued monthly, the spring and autumn technical meetings , the technical committees of the various divisions, seminars and symposia,to the sponsorship of international conferences.

In particular, the technical meetings, in spring and autumn, provide opportunities to exchange up-to-date research information and to network with colleagues working in academic and industrial circles. The numbers of oral presentations in the meeting increase year by year and, recently, more than 150 technical presentations focus on new themes in aluminum science and technology are read in each meeting; 2/3 of the papers are from academic and 1/3 are from private enterprises.

2-2 The Light Metal Educational Foundation

The Light Metal Educational Foundation, Incorporated was established on 26th January 1955 with donations from Toyo Aluminum K.K.(and others) to commemorate the company's twenty-fifth anniversary and to contribute to research and education of light metals, mainly in academic circles.

The Light Metal Educational Foundation has contributed greatly for many years to the promotion of aluminum research activities in academic circles in Japan, particularly through large numbers of grants for 66 professors and subsidies for 24 associate professors and others (1996), as shown in Table 3, which lists the working expenses for the last five years and the budget for 1997.

2-3 Establishment of Alithium, Ltd. (Research Institute of Al-Li Alloys)

To endeavour to catch up with the US and European countries in research and production of Al-Li alloys, seven aluminum campanies (members of the Japan R&D Center for Metals (JRCM) established "Alithium Ltd." (Research Institute for Al-Li Alloys) on 24 March 1989. The need was identified by JRCM's Subcommittee on Functional Aluminum-based Materials (chairman, Prof. Y. Murakami). This company was financed by the Japan Key Technology Center. The program had a schedule of seven years and one month, ending in March 1996, with the objective to develop process and equipment for production of 4 ton ingots on a commercial scale, the total budget being about 2,000 million Yen.

The research programme made satisfactory progress and successfully attained its targets on time. The photograph shows a view of a 4 ton melting furnace, with which Al-Li alloys registered by the American Aluminum Association, such as 2090, 2091 and 8090, were satisfactorily produced as sound ingots, followed by rolling into plates and

Photo. View of a 4 tons melting furnace for Al-Li alloys

sheets. However, the further operation of these facilities is now stagnant because of the economic and some technological factors which have occurred world-wide, although much effort is being exerted to re-commence operation.
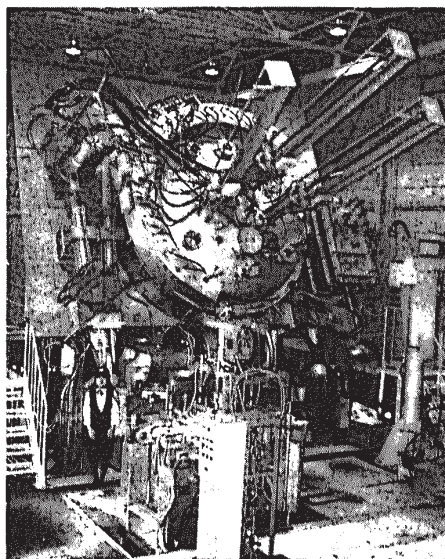
Table 3 The Light Metal Educational Foundation Working Expenses for the Last Five Years
and the Budget for 1997 (Million Yen)

Year / Items	1992	1993	1994	1995	1996	1997 (Budget)
(1) Educational Research Grants	24.4	24.3	18.0	17.0	17.5	18.5
(Number of Themes)	(80)	(79)	(69)	(65)	(66)	
(2) Subsidies for Research	6.0	5.6	3.15	3.60	3.60	3.9
(Number of Themes)	(30)	(28)	(21)	(24)	(24)	
(3) Grants for Advanced Joint	16.43	16.81	10.25	5.07	0.43	-
Research (Number of Themes)	(3)	(3)	(2)	(1)	(-)	
(4) Grants for Specific Research	14.08	15.82	5.0	5,89	4,5	6.0
Themes (Number of Themes)	(3)	(3)	(1)	(1)	(1)	
(5) Grants for University Students	5.94	5.10	4.80	3.90	1.8	3.0
(number of Persons)	(5)	(7)	(5)	(4)	(3)	
(6) Subsidiary Expenses For	2.6	1.65	1.90	1.30	-	1.6
Travelling Abroad (No.of Persons)	(13)	(9)	(11)	(8)		
(7) Publication,Seminars or	about	about	about	about	about	about
Symposia and Others	173.0	1400.0	788	1,137.0	1,162.0	1,150
Total	about	about	about	about	about	about
(Million Yen)	87.23	83.51	51.26	48.93	39.41	46.20

3-1   The two kinds of new aluminum alloys with higher strength of 100 to 150 kgf/mm^2 UTS

3-1-1 Higher strength nano-crystalline aluminum alloys with almost 150   kgf/mm^2 UTS [7]

Aluminum amorphous alloys have recently attracted great interest because of the fact that their tensile fracture strengths are about two or three times higher than the highest values of conventional Al-based crystalline alloys. Prof. Inoue's group, Tohoku University, [7] Y-H. Kim, A. Inoue and T. Masumoto, Materials Transactions, JIM,**32** (1991), 599, has recently developed amorphous $Al_{88}Y_2Ni_{10-x}M_x$ (M= Mn, Fe, or Co) alloys containing nanoscale particles by rapidly solidification, with composition ranges from 0 to 2% Mn and from 0 to 5% Fe or Co .

Dark-field electron micrographs of a rapidly solidified $Al_{88}Y_2Ni_9Fe_1$ alloy show fcc-Al particles with an average size of 5 nm (marked with arrows) dispersed homogeneously over the entire micrograph (Fig. 2). The volume fraction (V_f) of the fcc- Al particles can be varied by changing the rotation speed of the roller ; fcc-Al particles have nanoscale diameters of 2 - 30 nm and their size tends to increase with a decrease in cooling rate   (with an associated increase in V_f). Fig. 3   shows the relation between H_V, E or $\sigma_{f\,max}$ and V_f for rapidly solidified $Al_{88}Y_2Ni_{10-x}M_x$(x 5%, M=Mn, Fe or Co) alloys. The σ_f and ε_f are highest at $V_f = 5$ - 25%.   Combined characteristics of good ductility and ultrahigh σ_f above 1300 MPa are obtained in a wide V_f range from 5 to 35%. The σ_{fmax} reaches 1400 MPa for $Al_{88}Y_2Ni_5Fe_5$ and 1470 MPa for $Al_{88}Y_2Ni_8Mn_2$. This increase in σ_f is presumably due to the increase of the resistance to shear slip caused by an homogeneous dispersion of the nanoscale fcc- Al particles.

3-1-2 Higher strength AlZnMgCu alloys obtained by controlling the mesoscopic structures [8,9]

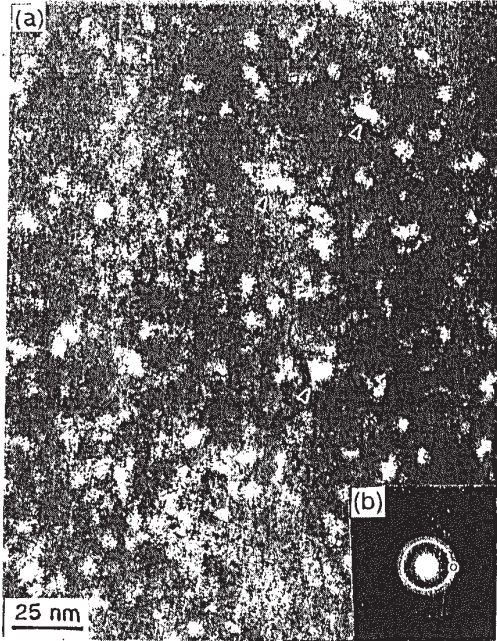A large improvement in the mechanical properties of AlZnMgCu alloys has been achieved by

Fig. 2   Dark-field electron micrograph (a), and electron diffraction pattern (b) of a rapidly solidified $Al_{88}Y_2$ Ni_9Fe_1 alloys. (Prof. Inoue's group)
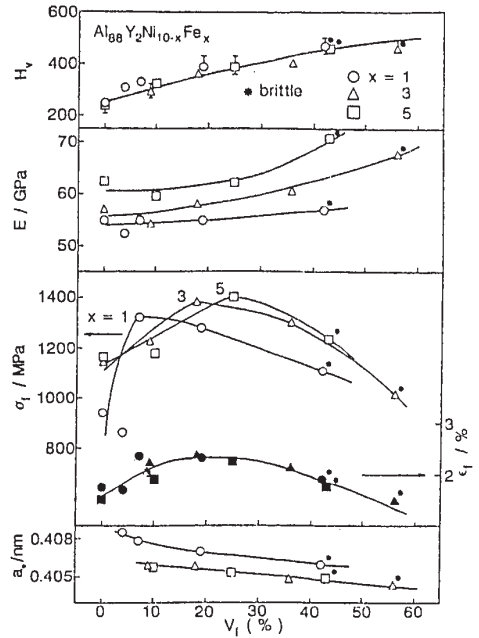
Fig. 3 Vickers hardness (H_v), Young's modulus (E), tensile fracture strength (σ_f), fracture elongation (ε_f) and lattice parameter (a_o) as a function for V_f for rapidly solidified Al_{88} Y_2 Ni_{10-x} Fe_x ($x \leq 5\%$)alloys. (Prof. Inoue's group)

cooperation between   Prof. K. Osamura, Kyoto University and Toyo Aluminum K.K.,[8] K. Osamura *et al.*, Metall.Transactions, A,**26** (1995), 1597; [9] J. Kusui *et al.*,Mat. Science Forum, **217-222** (1996), 1823, by controlling the mesoscopic morphology of metastable fine precipitates, such as GP zones and/or η'-$MgZn_2$ phase, as well as quasi-ternary intermetallic compounds, $Al_{11}Mn_3$($ZnCu_2$), which contribute to the fiber-reinforcement.

The chemical compositions of the alloys used in their study are summarized in Table 4. The base alloy was Al -9 - 9.5% Zn - 3% Mg - 1.5% Cu, and extra elements such as Mn, Ag and Zr were added systematically. The powder, prepared by air atomizing, was pressed into a rod-like shape by cold isostatic pressing.The rod was first pre-heated in an argon atmosphere at   773K for 3.6ks and then extruded to an extrusion ratio of 10. This standard fabrication condition is called Ar(10). When the rod was hot-pressed under vacuum at 773K and hot-extruded to a ratio of 20 , it was termed HP(20). The products were heat-treated to the T6 condition.

The mechanical properties of the alloys are shown in Table 5.   The base alloy has a tensile strength of 764 MPa, which is similar to reported data. Data from small-angle neutron scattering indicates the existence of fine precipitates of GP zones and/or η'-$MgZn_2$, of which the Guinier radius was assessed to be 2.3 nm. As is well known, age-hardening is caused by these precipitates which are coherent with the matrix. When 4% Mn was added, the UTS increased slightly, and, when the extrusion ratio was increased from 10 to 20 , the UTS increased appreciably. As shown in Fig. 4 [9], rod-like precipitates with lengths and widths of a few hundred nanometers are observed aligned with the extrusion direction. These precipitates were identified as $Al_{11}Mn_3(ZnCu_2)$ by TEM.

Table 4 Chemical Composition of the High-Strength Alloys Developed by Prof. Osamura's group

Alloy	Zn	Mg	Cu	Mn	Ag	Zr
9Zn-3Mg-1.5Cu (base)	9.62	3.01	1.48	—	—	—
Base-1.9Ag	8.91	3.34	1.42	—	1.8	—
Base-0.2Ag	9.20·	3.39	1.57	—	0.23	—
Base-0.08Ag	9.31	3.45	1.58	—	0.079	—
Base-0.04Ag	9.48	3.53	1.61	—	0.040	—
Base-4Mn	9.03	2.97	1.56·	3.67	—	—
Base-4Mn-0.02Ag	9.28	2.83	1.58	3.79	0.019	—
Base-4Mn-0.04Ag-0.02Zr	8.84	2.87	1.56	3.78	0.044	0.17
Base-4Mn-0.04Ag-0.5Zr	8.71	2.92	1.52	3.74	0.04	0.48

*Other impurities were Fe (<0.09) and Si (<0.09).

Table 5 Mechanical Properties of the High-Strength Alloys developed by Prof. Osamura's group

Alloy	σ_{UTS} (MPa)	σ_{YS} (MPa)	Elongation (Pct)	Fabrication Condition
9Zn-3Mg-1.5Cu (base)	764	739	7.6	Ar(10)
Base-1.9Ag	719	694	6.1	Ar(10)
Base-0.2Ag	780	757	3.6	Ar(10)
Base-0.08Ag	786	773	3.8	Ar(10)
Base-0.04Ag	796	783	3.8	Ar(10)
Base-4Mn	787	754	1.1	Ar(10)
Base-4Mn	879	845	1.3	Ar(20)
Base-4Mn-0.02Ag	886	820	1.6	Ar(10)
Base-4Mn-0.02Ag	915	891	1.3	HP(20)
Base-4Mn-0.04Ag-0.2Zr	908	861	0.9	Ar(10)
Base-4Mn-0.04Ag-0.5Zr	919	907	0.6	HP(20)

*The specimens were heat-treated under T6 condition of 393 K/86.4 ks.

diffraction, X-ray diffractometry and EDX analysis. The dimensions of the rod-like compounds are almost to those of SiC whiskers which are used normally as reinforcements for MMCs. In this research, these compounds were formed during the process of heat treatments and extrusion, and their aspect ratio were kept at relatively high values of 5 to 20, which could contribute effectively to fiber reinforcement. When a small amount of Ag was added to the base (Al - 9Zn - 3Mg- 1.5Cu) - 4% Mn alloy, the UTS was increased by about 100

MPa. Further increase in UTS was observed when 2% Zr was added to the (base - 4Mn - 0.04 Ag) alloy probably by grain refinement due to precipitation of metastable Al_3Zr particles. Table 5 shows the change of UTS when extra elements were added step by step to the AlZnMgCu alloys. The highest value of UTS of 943MPa was obtained in the (base - 4Mn -0.04 Ag - 0.5 Zr) alloy aged at 383K for 86.4 ks, although an extruded bar showed a UTS of 858MPa under the same temper.   This remarkable strengthening might be attributed to the multiple effect of precipitation hardening by various very fine precipitates and fiber reinforcement by rod-like compounds. The present higher strength alloys could be produced using conventional materials and inexpensive, mass-production processing.
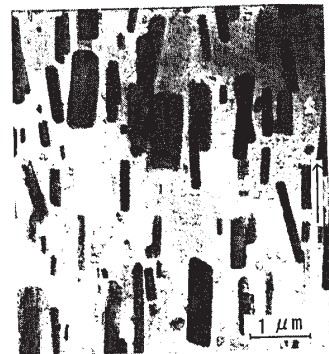

Fig. 4 TEM micrograph of the Base- 4 Mn alloy, arrow is the extrusion direction.

3- 2 Very high strain-rat superplastic aluminum matrix composits   [10, 11]

It is well known that aluminum alloy matrix composites reinforced with ceramic whiskers or particulates exhibit excellent specific mechanical properties at room temperature and also have good wear resistance and heat resistance. However, the composites have lower ductility than matrix alloys, resulting in poor workability. Recently, a new area of high strain rate superplastic forming has been opened to aluminum matrix composites. The original work by Nieh et al. [10] T.G.Nieh, Metall. Trans. A, **15** (1984); T.G.Nieh *et al.*, Scripta metall., **18** (1984),1405, revealed that a conventional powder metallurgy processed 20 Vol% SiC_w - 2124 aluminum composites behaved in superplastic manner at a high strain rate of 3.3   x 10 $^{-1}$ s^{-1}. To date, it is accepted that grain-boundary sliding plays an important role in superplastic flow for metallic materials as well as in composites. In particular, plastic deformation of the reinforcements is believed to be impossible at superplastic temperatures for metal matrix composites; therefore special mechanisms for the accommodation process are required under plastic flow. It was recently reported by Nieh et al. [10] that high strain-rate superplasticity might be a result of the presence of a liquid phase at the reinforcement / matrix interface or near the interface region. Furthermore, Prof Higashi's group has investigated the high strain-rate superplasticity in detail. They evaluated theoretically the local stress around the matrix - reinforcement interface to be nearly 8 MPa, and this value is very comparable with typical superplastic flow stresses of about 4 - 5 MPa obtained experimentally for the Al-Mg-Si alloy matrix composite reinforced with Si_3N_4 particles of 0.2μ m in diameter. Therefore, it is clear that a stress concentration is caused around matrix / reinforcement interfaces during superplastic flow for metal matrix composites.

The experimental results, [11], by Higashi's group are as follows. The total elongation at 773 - 848 K for the Si_3N_{3p} - (Al-Mg-Si) composite is shown in Fig. 5 (a) as a function of strain rate. A maximum elongation at each temperature was obtained at 2 s^{-1}. The variation in total elongation at 2 s^{-1} is shown inFig. 5(b) as a fuction of testing temperature between 773 and 848 K. Values of elongation increase with increasing temperature, reach a maximum at 833 K and then decrease rapidly to a small value of less than 100 % at 848 K. The optimum superplastic temperture for the maximum elongation is 833 K. The result of a DSC run is shown in Fig. 5 (c). It is worthwhile to note the appearance of a sharp endothermic peak. The onset temperature for partial melting is determined from the intercept of the two lines indicated in the figure. The melting point of the composite is 830 K, which is indicated by an arrow in Fig. 5 (b). It is important to note that the optimum superplastic temperature of 833 K is very close to the melting point of the composite. A photograph from *in-situ* TEM observation at 827 K is shown in Fig. 6 (a). It appears that local melting occurred at matrix-reinforcement interfaces and/or grain boundaries. A SAD pattern at the interfacial region, as shown in Fig. 6 (b), shows a diffuse ring, indicating   direct evidence of local melting in the interfacial region near the optimum superplastic temperature of 833 K. It is concluded for the Si_3N_{4p} - (Al-Mg-Si) composite that the optimum superplastic temperature of 833K, at which the maximum elongation was obtained, is almost equal to the melting point of 830K found by DSC and also 827K by *in-situ* TEM observation. Most importantly, the result from this investigation clearly suggests that the interface and grain boundaries melt at the optimum superplastic temperature. Also, the cavity nucleation rate increases rapidly with increasing stress. It is therefore suggested that a high strain concentration from interfacial sliding, which is accommodated only by diffutsonal or plastic flow, easily causes excessive cavity nucleation at the interface so that premature fracture occurs. In conclusion, Prof.Higashi's group proposes that superplastic flow in matrix composites is controlled by a grain-boundary sliding mechanism accommodated by relaxing the stress concentration via an isolated liquid phase at matrix - reinforcement interfaces.
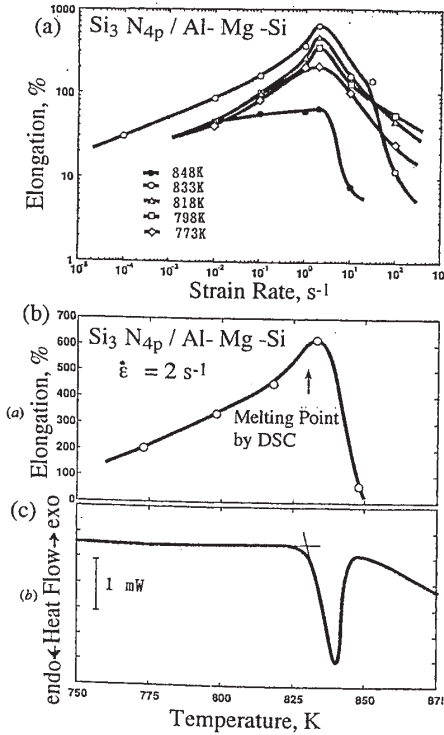
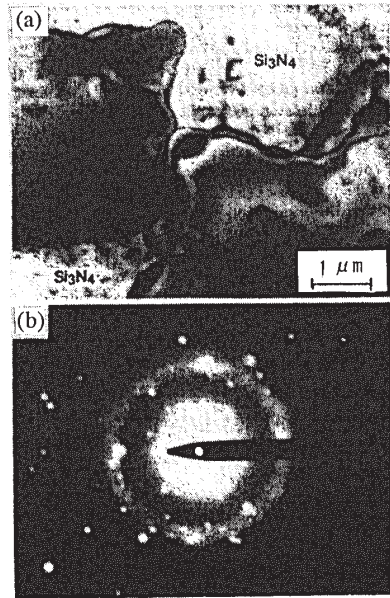Fig. 5 The variation in total elongation (a),(b) and DSC run (c).

Fig. 6 A TEM micrograph (a), and SEM pattern at the interfacial region by by the *in-situ* TEM observation at 827 K (b) for the Si_3N_{4p}-(Al- Mg-Si) composite.

3-3 Development of a Piston with an MMC - Reinforced Ring Groove by the Toyota Motor Co. [12]

The development of SiC whiskers or alumina-based short fiber, such as ICI's SAFFIL fibers, and their incorporation into aluminum alloys by squeeze casting has been extensively exploited in industrial applications, particularly in the automotive industry, where high temperature properties, thermal fatigue, and wear resistance are important criteria, together with cost performance.

The most successful example of an application is a diesel engine piston with an MMC - reinforced ring groove, developed by the Toyota Motor Co., as shown in Fig. 7. The squeeze casting method is used to allow permeation of the aluminum alloy melt into a reinforcement preform of SAFFIL - Al_3O_2 (crystallized) or kaowool (amorphous) fibers. The present production volume is 400,000 units per month, which is quite a large volume, and not found elsewhere in the world for MMC parts.

Fig. 7 Piston with MMc-renforced ring groove developed by Toyota Motor Co. [12]

3 - 4   Advanced Purification Technologies for Aluminum Recycling [13]

In Japan, secondary aluminum produced mainly by recycling of aluminum scrap is more than 1 million metric tons per year, although the total consumption of aluminum is more than 4 million metric tons per year, or 15% of the world demand. The volume aluminum scrap has increased rapidly since the 1980s and will further rise toward the year 2010. If this trend continues, Japan, which is at present the world largest importer of aluminum scrap, will become an exporter instead. To avoid further increase of scrap, it is essential either to increase the volume of secondary aluminum used in the making of wrought products or to prolong the service life of window sashes and automobiles, which are the main products of aluminum in Japan.

According to a survey of the aluminum scrap balance in 2010 , only 13.45 % scrap part for the current total wrought products of about 220 million metric tons has to be increased to 37.1 % scrap part of the wrought products of about 300 million metric tons in 2010, and thus greater advance of recycling technologies are urgently in demand. They include the development of methods for control of impurities and technologies for separation and sorting. Innovative technologies for refining metals and eliminating defects so as to achieve quality equal to that of primary metals are also required. As secondary aluminum production rises, on the other hand, there occur such problems as increases in dross and residual ash. Countermeasures for treating and for depositing or utilizing these materials are also needed.

Table 6   Theme Contents for Aluminum Recycling

Title of Theme	Outline of Technology to be developed inside Melt
(1) Technology to remove impurities from molten scrap by a fractional crystallization process	Molten low grade scrap is locally cooled with a cooler placed inside melt, causing initially crystallized aluminum to be removed. This process is repeated, e. g. to purify automobile scrap as far as a composition of 3.6%Si, 0.2%Fe and 1.5%Cu.
(2) Technology to remove zinc from scrap	Molten zinc-containing scrap is sprayed into a vacuum chamber to be separate by the difference in vapor pressure between aluminum and zinc for the purification to 0.1% or less zinc. This process is also effective for removing.
(3) More efficient non-metal inclusions removal technology	To make molten scrap clean enough to use as wrought materials, its flow is controlled through the combination of flux gas blowing and filtering to remove non-metallic inclusions.
(4) Residual dross and ash treatment and utilization	Deoxidation by high - temperature treatment of the now mostly discarded residual dross and ash, and development of effective use as refractory, civil construction and other materials.

To meet the above demand, R & D of the technologies needed to promote aluminum recycling was started in 1993 at JRCM, based on results of the identification of The Subcommittee on Functional Aluminum-Based Materials (chairman is Prof. Y. Murakami) with the emphasis on advanced purification technologies. This project is implemented by the New Energy and Industrial Technology Development Organization (NEDO) and has been entrusted   to   JRCM, where

aluminum companies and universities are carrying out researches with the main target focused on the element technologies relating to post-dissolution process of liquid-, gaseous, or semi-molten state purification, assuming that the supplied solid scrap is already separated or sorted out. Table 6 shows the theme contents of the programme. These technologies have been well prepared for success, and thus satisfactory results will be obtained within the time limit.

4 Concluding Remarks

In order to introduce the situation of research on aluminum in Japan, I have talked on some subjects for the early years of research on aluminum in this country and also to refer to some recent researches. I have also noted the aluminum research supporting organizations, and some others, peculiar to the likes of which not found in other countries. Japan has now grown to be one of the advanced countries in aluminum, but has many problems to be solved, such as energy economization and ecology, similar to other countries with aluminum industries. The future of aluminum involves the large field of structural as well as functional materials, and thus the development of aluminum materials has to be promoted strongly to meet the users' social and economic needs. New applications will lead to new barriers that will have to be overcome, but, due to the long-established successful co-operation between aluminum producers and users and academic circles in Japan, I have every confidence that surmounting these barriers will not be too difficult.

References

[1] I. Igarashi and G. Kitahara, J. Iron and Steel Inst Japan, 23 (1937), 35.
[2] I. Igarashi and G. Kitahara, J. Japan Inst Metals, 3 (1939), 66.
[3] I. Igarashi, *ibid.*, 6 (1942), 316.
[4] H. Nishimura, *ibid.*, 1 (1937), 1.
[5] G.G.Urazov and M.S. Mirgalovskaya, Metals Handbook, 8th edition, 8 (1973),386.
[6] A. Miyata and S. Setoh, Science Papers of Inst for Phys. and Chem. Research , Tokyo, 17 (1932), 189.
[7] Y-H. Kim, A. Inoue and T. Masumoto, JIM, 32 (1991), 599
[8] K.Osamura, O. Kubota, P. Promtit, H. Okuda, S. Ochiai, K. Fujii, J. Kusui, T. Yokote and K. Kubo, Metall. Trans., A, 26 (1995), 1597.
[9] J. Kusui, K. Fujii, K. Yokoe, T. Yokote, K. Osamura, O. Kubota, and H. Okuda, Mat. Sci. Forum, 217 - 222 (1996), 1823.
[10] T. G. Nieh, Metall. Trans. ,A, 15 (1984).
[11]   M. Mabuchi and K. Higashi, Phil. Mag. Lett., 70 (1994), 1.
[12]   T. Kaneko, Proc. 2nd Int. Conf. on Advanced Materials and Technology, New Composites '91, Hyogo, Kobe, Japan, 165.
[13]   Report for R & D of Recycling Technologies of Non-ferrous Materials, edited by Japan Research Center for Metals, March, 1997.