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# MICROSTRUCTURE/PROPERTIES OF HIGH TEMPERATURE SPRAY DEPOSITED AI-Cu-Mg-X ALLOYS

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## Abstract

In airframe construction where temperature resistance and damage tolerance are important design considerations, the 2XXX series Al alloys are well established for most structural applications. Alloying and processing development have been investigated in this alloy system to improve the combinations of mechanical properties required for high speed aircraft. Based on the improved temperature capabilities of high Cu/Mg ratio alloys, the present work describes the mechanical property behavior of spray deposited alloy compositions in the Al-Cu-Mg-X alloy system, specifically N203 and N201. Cylindrical billets of each nominal composition were spray deposited and wrought processed into rectangular extruded bar. The age hardening behavior of the extrusions was evaluated by selected tensile and fracture toughness testing. Improved properties over conventional ingot metallurgy Al alloys were obtained due to the fine grain sizes and reduced second phase constituents, attributable to the spray deposition fabrication. Microstructural analysis was performed with a focus on developing relationships governing the mechanical property behavior. Further modifications in the aging practices to alter the volume distribution of strengthening phases were evaluated and compared to baseline mechanical property results.

#### Introduction

Precipitation hardened Al alloys for high temperature airframe applications have been exclusively limited to either 2618 or 2219 alloy compositions, strengthened primarily by S' or  $\theta$ ', respectively. Transition element additions, including Mn, Fe, Ni, Cr, V, and Zr have been added to 2618 and 2219 to provide both a modest component of dispersion strengthening and maintain preferred grain structures. Development efforts on several PM 2XXX Al alloys by Wald and Chellman have successfully exploited this approach [1, 2]. Secondary alloying element additions have been examined which promote the formation of more energetically favored and thermally stable precipitates [3 - 5].

The alloy design strategy that has proven to be most effective for concurrently improving both the ambient and elevated temperature strength of Al-Cu-Mg alloys originates from the research of Polmear and Couper [6]. For modest levels of Ag additions (0.3 to 0.5 wt. pct.), the  $\Omega$  phase has been shown to dominate the precipitation strengthening of high Cu/Mg solute ratio alloy

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compositions [6].  $\Omega$  forms on {111} matrix planes as a uniform dispersion of large, very thin hexagonal shaped plates [7], and is a considerably more stable and thermally resistant phase compared to  $\theta'$  and particularly S'. Microstructural observations support the coexistence of  $\Omega$  with other strengthening phases, which improves alloy flexibility in terms of alloying and heat treatment responses [8].

The inherently slow cooling rates in direct chill (DC) cast ingots represent a limitation on the fine scale of microstructural features in most Al alloy products. This is particularly true in highly alloyed compositions, where large constituent phases are present with ingot metallurgy (IM) practices. Spray deposition offers an alternate processing method and is applied in the present work. The spray deposition process involves atomization of an alloy melt into a spray of fine liquid droplets, then impingement on a cooled substrate [9]. Cooling rates for spray deposition  $(10^3-10^4 \text{ °C/sec})$  are much higher than typical of DC castings, although slower than rates associated with rapid solidification methods [10]. The difficulties of oxide film contamination and economics of multiple step processing related to rapid solidification methods are avoided in spray deposited Al alloys, and the higher solidification rates through spray deposition promote constituent phase refinement and grain size reduction. This is important in high Cu/Mg solute alloys, as Al<sub>2</sub>Cu and Al<sub>2</sub>CuMg phases are more amenable to dissolution during either homogenization or solution heat treatment. Reduction in grain size leads to a decrease in the length of dislocation pile-ups, and thereby offers improved ductility and fracture toughness. Since the microstructure of spray deposited alloys typically consists of fine, equiaxed grains, improved isotropy of mechanical properties is likely to be an additional attribute of combining new high temperature Al alloying principles with spray casting [11,12]. Research to exploit the extended composition range of spray deposited Al-Cu-Mg-X alloys has demonstrated encouraging results in terms of strength and thermal stability up to temperatures of 150°C [13].

The objective of this work is to characterize the mechanical property behavior and microstructure of spray deposited A1 alloy N203, with respect to suitability for primary airframe structural applications. Preliminary microstructural analysis and systematic mechanical testing have been performed to establish material property characteristics. Additionally, mechanical property data generated for spray deposited A1 alloy N201 are presented for comparison.

### **Experimental**

Extrusions of spray deposited Al alloy N203 (Al-5.0Cu-0.5Mg-0.5Mn-0.4Zr-0.4Ag), were procured from Alcan-CoSpray, Banbury, UK. Spray deposited billets of approximately 80 kg were fabricated in the Alcan prototype demonstration unit as per previous research [14]. The extrusions measured 1.9 cm in thickness x 6.4 cm in width x L. Cylindrical billet charges were extruded at 420°C at an extrusion ratio of 24.6:1. After extrusion, the N203 material was solution heat treated by Lockheed at 520°C for 3.0 hours, cold water quenched (CWQ), and isothermally aged to two different -T6X tempers, specifically 190°C/5 hours and 177°C/12 hours. Flat extruded bar of the N201 composition was provided by the Swiss Federal Institute of Technology in Zurich, Switzerland, and heat treated by Lockheed to the -T6 condition using the same 195°C/7 hours practice as Polmear and Couper [6].

Room temperature tensile tests were conducted on N20X Al type samples in longitudinal (L), long-transverse (LT), and short-transverse (ST) orientations relative to the extrusion direction for the -T6X conditions. Elevated temperature tensile data was generated over a temperature range of 107°C to 200°C. Thermal stability of N201 was evaluated through elevated temperature tensile tests on samples which were isothermally exposed for 100 hours at 149°C and 200°C.

Compact tension (CT) fracture toughness specimens were machined from the extruded bars and tested for the aged samples in the L-T orientation at room temperature. ASTM E399 procedures were used to analyze the CT toughness data.

Mechanical property test results for spray deposited N201 and N203 alloy extrusions are presented in the next section. Various published findings are also cited for comparison, namely, results of Polmear and Couper [6], and Beffort, Uggowitzer, and Speidel [13]. Table 1 lists the chemical compositions of all alloys mentioned in the mechanical property results.

A microstructural analysis is presented on the N203 extruded bar, both as-received, and after heat treatment to the -T61 and -T62 conditions. Microstructural analyses of spray deposited high temperature 2XXX A1 alloy materials have been previously discussed by Chellman and Bayha [11,12]. Scanning electron microanalysis was conducted on a JEOL JSM-35 with EDS capability at 25kV. Electron transparent specimens for transmission electron microscopy (TEM) were prepared by cutting 3.0 millimeter discs, electro-polishing, and finally ion thinning. Thin foil samples were examined on a Phillips EM400T with EDS attachment, operated at 120kV.

Alloy	Cu	Mg	Mn	Ag	Zr	Ti	Other
Spray N201	5.93	0.51		0.46			1.07Fe, 1.01Ni
Spray N202 [11]	6.23	0.36	1.84	0.40	0.31	0.19	0.22V
Spray N203	4.89	0.50	0.48	0.42	0.40	0.20	0.22V
Beffort [13]	6.30	0.40	1.80	0.40	0.50	0.25	0.25V
Polmear, Alloy C [6]	6.0	0.45		0.50			1.0Fe, 1.0 Ni

Table I. Chemical Compositions of Spray Deposited N20X Al Type Alloys, and Similar Alloys Used for Comparisons of Mechanical Properties.

## Results and Discussion

#### Mechanical Properties

Tensile isotropy in the L and LT orientations for the N203 and N201 extrusions are exhibited in Figure 1. The N203 alloy extrusions show superior strength and ductility properties in comparison to previous work on Al N202 [11]. Strength differences in L and LT are slightly larger by approximately 9 pct., while elongation values are identical. The lower overall strength of the N201 extrusions is consistent with results of Polmear and Couper (Alloy C) [6]. Evidence for recrystallization and extensive formation of Cu-containing intermetallic phases are largely attributable to the reduced N201 alloy strength. Improved isotropy of tensile properties noted in spray deposited N20X Al type alloys is a consequence of the uniform and fine grain structures.

The airframe durability and damage tolerance potential of spray deposited N20X alloys is shown in Figure 2, where compact tension (CT) fracture toughness results are plotted versus tensile yield stress for heat treatment tempers previously described in the aging study. Recent results on strength and toughness combinations for spray formed Al-Cu-Mg-Mn alloy extrusions and pancake forgings are also shown for purposes of comparison [13,15]. The major alloying variants for the three spray deposited Al alloy products are based on Cu and Mn contents, with Mg, Ag, Ti, V, and Zr maintained nearly equivalent. Spray cast N203 extrusion results show higher toughness than all other N20X Al composition variants, with a high attendant yield stress of approximately 480 MPa at room temperature. A consistent toughness and strength trend is noted for the spray deposited Al alloys as a function of the total Cu and Mn content. Both volume fraction and size of primary constituent and dispersoid phases seem to exert a major role in the toughness and strength property combinations. From microstructural observations, the presence of Al20Cu2Mn3 particles may dominate the fracture mechanisms of these high solute Al alloys. With a gradual reduction in the Cu and Mn levels, an increase in the interparticle spacing between Al<sub>20</sub>Cu<sub>2</sub>Mn<sub>3</sub> particles contributes to a continuous improvement in fracture toughness values. The contribution of these particles to fracture toughness properties in 2XXX series Al alloys have been reported in recent studies [1, 15]. These observations and property trends indicate that it is possible to tailor the Cu and Mn compositions in this alloy system to meet fracture toughness requirements.

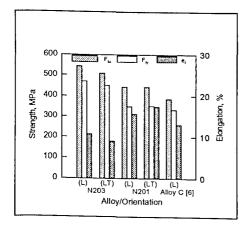


Figure 1. Tensile Isotropy of Spray Deposited Al N20X Extrusions.

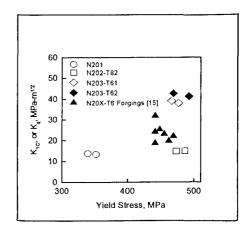
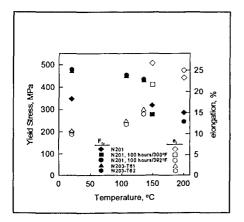


Figure 2. Fracture Toughness vs. Tensile Yield Stress for Spray Deposited Al Alloys.

The elevated temperature tensile response of N203 and N201 extrusions are shown in Figure 3, as a function of testing temperature. The N203 data signifies only a slight (4 pct.) degradation in tensile yield stress when tested at 107°C. Yield stress loss at 135°C was approximately 9 pct. N201-T6 samples also show expected strength reductions as the testing temperature is increased. Elevated temperature testing of N201-T6 after 100 hours isothermal exposure shows an influence due to continued aging of the alloy. There is no evidence in Figure 3 of a tensile ductility minimum with increasing test temperature, such as occurs in rapidly solidified aluminum alloys.

The higher elevated temperature properties of the N203-T6 materials can be attributed to the incorporation of Ti- and Zr-based dispersoids. Although the  $Al_{20}Cu_2Mn_3$  dispersoid particles exert an important influence in fracture toughness properties, their role is not strongly indicated by the current results on elevated temperature tensile properties. The contribution of these finely dispersed particles to creep resistance above 105°C for spray deposited Al alloys represents an area of current research interest [13].



20 μm

Figure 3. Elevated Temperature Tensile Properties of Spray Deposited N20X Alloy Extrusions.

Figure 4. Scanning Electron Micrograph of Spray Deposited N203 Microstructure.

#### Microstructural Observations

Spray deposition processing of aluminum alloys contributes directly to the formation of fine and more equiaxed grain structures with sizes between about 1.5 and 4.5 microns. As seen in Figure 4, at the high solute levels of these alloy compositions, based either on Cu/Mg or Fe/Ni additions, the sizes of primary constituent phases are significantly refined. The efficient use of alloying additions by spray deposition is accomplished through the formation of strengthening phases and dispersoid particles for grain structure control. Evidence of coarse, incoherent

particles originating from excess solute during spray casting is typically limited, but occasionally is observed along grain boundaries, with sizes ranging from 0.5-15.0 microns (Figure 5). These constituent particles were more prevalent in the N202 alloy [11] compared to N203 based on an examination of the energy dispersive spectra (EDS). The high Fe and Ni additions in the N202 spray formed alloy resulted in the formation of Al<sub>9</sub>FeNi particles, with an average size of about 15.0 microns [11]. The intermetallic dispersoid particles are considerably smaller in size than Al<sub>9</sub>FeNi particles reported in 2618 Al fabricated by conventional ingot metallurgy practices [10].

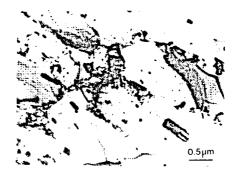


Figure 5. Transmission Electron Micrograph of As-Extruded Spray Deposited Al N203 Microstructure.

Previous microstructural analysis [11] on spray deposited Al N202 revealed that solution heat treatment (SHT) times longer than the conventional 1.5 hours at 520°C promoted nucleation and growth of  $\Omega$  phase strengthening precipitates to the exclusion of S' and  $\theta'$  phases. The prolonged solutionizing time apparently "annealed out" some of the matrix dislocations. Dislocations in the matrix are preferred nucleation sites for the heterogeneous formation of S' and  $\theta'$  phases. Selected area diffraction patterns indicated reflections for these phases, but it was noted that  $\Omega$  phase reflections are becoming stronger for longer solution heat treatment times. Experiments are currently underway to maximize the extent of  $\Omega$  phase formation by using novel heat treatment sequences. In this study, 3 hour solution heat treatment times are combined with various aging time/temperature combinations.

Figure 6 shows the effect of heat treatment on the N203 microstructure. The micrograph in Figure 6a is from a -T61 sample, and shows a high density of  $\Omega$  phase, and some constituent particles. In Figure 6b (-T62), the diameter of the  $\Omega$  platelets is approximately 60 pct. smaller and the number density is higher than for the -T62 microstructure. Both micrographs in Figure 6 are in the <112> orientation.

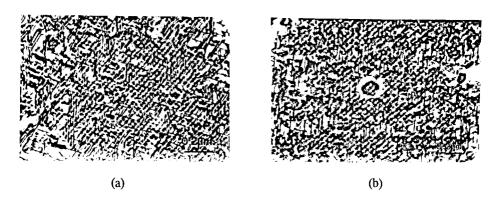


Figure 6. Transmission Electron Micrographs of Spray Deposited Al N203 Microstructure, <112> Orientation: a) -T61; and b) -T62.

## **Conclusions**

1. Spray deposition processing of Al N20X alloys produces a refined and homogeneous microstructure compared with conventional ingot metallurgy methods. Constituent particles and grain structures are significantly reduced in size, with average grain dimensions of approximately 1.5 to 4.5 microns. The fine and uniform distribution of dispersoids and precipitates contributes to good overall balance of properties.

2. Spray deposited N20X alloys have isotropic tensile properties, which are a consequence of the uniform and fine microstructure.

3. Spray deposited Al N203-T6X extrusions have potential for airframe structural applications, due to excellent combinations of tensile strength and fracture toughness.

4. Extrusions of Al N203 and N201 display attractive elevated temperature strength and thermal stability for high temperature airframe structures. No indication of a ductility minimum with increased service temperature is observed.

5. Heat treatment variations can be imposed on spray deposited Al N203 to create a primarily  $\Omega$  phase strengthened microstructure.

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#### **References**

1. G.G. Wald and D.J. Chellman, "Supersonic Cruise Vehicle Technology Assessment of an Over/Under Engine Concept - Advanced Aluminum Alloy Evaluation", NASA Contract Report No. 165676, 1981.

2. D.J. Chellman, "Development of Powder Metallurgy Al Alloys for High Temperature Aircraft Structural Applications-Phase II", NASA Contract Report No. 165965, 1982.

3. J. H. Taylor, B. A. Parker, and I. J. Polmear, Metal Science, 1976, Vol. 12, pp. 478-482.

4. A. Garg and J.M. Howe, Acta Metall., Vol. 40, 1992, pp. 2451-2462.

5. L.B. Blackburn and E.A. Starke, Jr., <u>Aluminum-Lithium Alloys</u>, eds. T.H. Sanders, Jr. and E.A. Starke, Jr., MCE Publications Ltd., Birmingham, UK, 1989, pp. 751-766.

6. I.J. Polmear and M.J. Couper, Metallurgical Transactions, Vol. 19A, 1988, pp. 1027-1035.

7. K. M. Knowles and W. M. Stobbs, Acta Crystallographica, Vol. B44, 1988, pp. 207-216.

8. R.J. Chester and I.J. Polmear, <u>The Metallurgy of Light Alloys</u>, Institution of Metallurgists, London, 1983, pp. 75-81.

9. A. G. Leatham, et. al., <u>Proceedings, First International Conference on Spray Forming</u>, September 1990, pp. 1-12.

10. N. J. Grant, <u>Proceedings, Symposium on High-Strength Powder Metallurgy Aluminum</u> <u>Alloys</u>, The Metallurgical Society of AIME, 1982, pp. 3-18.

11. D. J. Chellman, T. D. Bayha, Qiong Li and F. E. Wawner, <u>Proceedings, 2nd International</u> <u>Conference on Spray Forming</u>, Swansea, UK, September 1993, pp 427 - 434.

12. T.D. Bayha and D.J. Chellman, "Elevated Temperature Fracture Behavior of Spray Deposited 2618 Extrusions", to be published in <u>Proceedings</u>, PM<sup>2</sup> TEC '94, Toronto, Canada, May 1994.

13. O. Beffort, P. J. Uggowitzer, and M. O. Speidel, <u>Proceedings, Third International</u> <u>Conference on Aluminum Alloys</u>, Vol. 1, 1992, pp. 46-51.

14. T.C. Willis, Metals and Materials, August 1988, pp. 485.

15. J. White, et. al., to be published in <u>Proceedings, 14th SAMPE '93 International Conference</u> and Exhibition, European Chapter, Birmingham, UK, 19-21 October 1993.