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Effect of Nb and Ag on the Microstructure and Mechanical Properties of Rare Earth Modified AA2219 Aluminium Alloy

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Abstract

AA2219 (Al–6.5%Cu) alloy is a widely used age hardenable alloy in aerospace and is used in cryogenic tanks, fuselage and shells for space vehicles and defence applications

One of the problems associated with the development of AA2219 alloy is dendrite formation during casting of alloy, long homogenization cycle is needed to get a dendrite free structure. Also, during thermo mechanical working of this alloy (such as forging and hot rolling), the temperature should be made high as cracks will occur during low temperature. But if the temperature is made high, grains will be bigger and strength and ductility of the alloy decreased. The grain size should be reduced without losing the mechanical properties of the alloy. This research concentrates on decreasing the grain size by adding rare earth element scandium to AA2219 alloy with Niobium (Nb) to find the effect of Al_3 $(Sc_{1-x}\,X_x)$ $L1_2$ type particle which can give further grain refinement. Silver (Ag) is added to one billet which is known to accelerate the precipitation of Ω phase, (composition similar to that of $CuAl_2$) expecting the mechanical properties to increase further. In this investigation, six billets of different composition are made with Sc, Nb and Ag with AA2219 alloy and their tensile properties are studied and correlate with the microstructure in as-cast condition.

Keywords: Al-Cu alloys, AA2219, Vacuum Induction Melting, Thermomechanical treatment, Mechanical Properties

I. INTRODUCTION

Al-6.3 Cu aluminium alloy (AA 2219) is extensively used in aerospace structures. It has good strength, ductility, weld ability and excellent cryogenic and high temperature properties. It is being used extensively for liquid engine tankages and structures in the form of sheets, plates and forgings. In aerospace structures, it is necessary to design the components with optimal weight at the same time making them failsafe and damage tolerant.[1]

While casting of the AA2219 alloy main difficulty occurs is the formation of dendritic segregation [2-3] also during thermo mechanical working of the alloy the temperature should be made low to avoid grain cracking and if the temperature is high it will cause recrystallization. So, low temperature for rolling is preferred at the same time, grain size should be reduced for improving the mechanical properties of the alloy and to meet certain applications in aerospace industries. For this purpose, the rare earth elements are used. Previous studies have confirmed that Sc has good grain refinement effects in Aluminium and Al alloys [4-5]. With rare earth Sc and transition metal Nb, Ag, further improvement in microstructure and mechanical properties can be obtained [6].

The Al_2Cu **Ω** phase in AA2219 alloy with Ag is known to accelerate the precipitation of the **Ω** phase[7].In another study, the results show that the alloy with Ag is strengthened by a homogeneous distribution of coexistent θ and Ω precipitates on the matrix (001) and (111) planes8.In this study, 0.4% Ag is also added in one billet to study the effect of above said phase in AA2219 alloy containing Al-4.5%Cu.

Scandium can form $Al_3\,(Sc_{1-x}\,X_x)$ with transition metals(where X is transition metals),which has the same L12 structure. It can partially substitute the Sc in Al_3Sc structure and therefore more Sc atom will form Al_3Sc or Al_3 $(Sc_{1-x}\,X_x)$,[9,10] .Thus,partial substitution of Sc by other elements may increase the ductility of Al_3Sc (as observed in other intermetallics) and also decrease the cost of the alloy [11] .Xiang Qingchun et al [12] in their work shows refined grain structure and increase in tensile strength by adding Sc and Zr in as-cast Al-Cu alloy. P.Nagaraju et al [13] also shows the grain structure refinement when they added Sc and Zr in AA2219 alloy.

Aluminium combination with niobium shows grain refinement due to insitu Al_3Nb particle [14]. In the research study of Feng Wang et al in 2014 they added 0.5% Nb in Al and found that there is significant grain refinement [15]. The present Investigation is focussing on significant grain refinement by Al_3 $(Sc_{1-x}\,X_x)$ phase with the addition of Nb. Y. Harada a, D.C. Dunand and other researchers reported the effect of Sc with other transition metals Zr, Er, Ce etc [16-19].

It has been established by our research that the tensile properties of AA2219 alloy in as rolled at different rolling temperature revealed that, one with higher rolling temperature has poor tensile values and bigger grain size. One with lower rolling temperature produces cracks on the surface of alloy. In order to avoid cracks the higher temperature is taken sacrificing grain size and tensile values. The current Investigation focussed on reducing the grain size of the AA2219 alloy with higher rolling temperature and at the same time improving its mechanical properties by adding rare earth element Sc, Nb, Ag. The tensile properties are evaluated in as rolled condition and correlated with microstructure.

II. EXPERIMENTAL

II.1 *Melting and casting*

For present Investigation material used is Nb (0.5 to 1.0wt%), Ag(0.4wt%) and rare earth element Scandium (0.5wt%) as well as DC cast billets of AA2219 having nominal composition Al-6.3Cu-0.3Mn-0.12Zr. Direct Chill cast (DC) AA2219 alloy was used as starting material for processing of Al-Cu alloy with above elements. The chemical composition of Six billets of Alloy is shown in Table 1. The developmental process for casting of six billets was done in Vacuum Induction Melting (VIM) using dynamic argon atmosphere.

The charging was done in such a way that AA2219 blocks were kept on the bottom as well as on the sides of the graphite crucibles for all melts. Other alloying elements either in the form of master alloys or pure metal were put in the middle. After melting of the alloy, casting of the alloy was done in mild steel mould under argon atmosphere. After sufficient time, ingot was taken out from the VIM. Six melts of modified Al-Cu alloy AA2219 were taken using this technique.

For that, the AA2219, pure Niobium, pure silver and master alloy Al-2%Sc were used for melting of Al-Cu alloys. (designated henceforth as 2219+Sc, 2219+Nb,2219+Sc+Nb, 2219+Sc+Nb+Ag alloy)

II.2 Thermomechanical working and Characterization

II.2.1 Forging and Section rolling

The homogenization temperature of modified alloy is as chosen from the DSC experiment. The homogenized billets of modified AA2219 Al-Cu alloy as shown were scalped and forged in forging press to 45 mm diameter rod after preheating the billet at 450°C in furnace for 2 hrs with intermittent annealing at same temperature for 30 minutes. In addition to this, DC cast billets of AA2219 were also forged with above mentioned parameters. These hot forged rods of DC cast AA2219 were section rolled at 250°C, 300°C, 350°C, and 400°C.The rolling temperature for six billets of modified AA2219 alloy were so selected as 350ºC from the experiment done at different rolling temperature on DC cast AA2219 alloy alone. The reduction direction was changed in increments of 90^{o} by rotating the material one quarter turn in each pass. The hot forged rods of Al-Cu (AA2219) alloy of six billets with Sc and transitional metal Nb and Ag were section rolled to 12 mm diameter rods after soaking at 350ºC(623K) for 30 minutes with intermittent annealing.

II.2.2 *X-ray Fluorescence (XRF)*

The XRF method is widely used to measure the elemental composition of materials. Since this method is fast and non-destructive, it is the method of choice for field applications and industrial production for control of materials. All elements were determined using this technique.

II.2.3 Differential Scanning Calorimetry (DSC)

Differential Scanning Calorimetry measures the amount of energy (heat) absorbed or rejected by a sample as it is heated, cooled or held at constant temperature. DSC also performs precise temperature measurement. In order to establish the homogenization and precipitation hardening temperatures, differential scanning Calorimetry was done.

II.2.4 Microstructural analysis in as cast condition

To study microstructure of metals metallurgical microscope is used. Metal piece of size 10x10 mm were cut from the as- cast and intermediate and fine grinding was carried out on one face of specimen using emery papers of progressively finer grade. Four grades of abrasives were used: 220, 280, 400,600 and 800. Alumina of 1µm size was used for fine polishing. Final polishing of the specimen was carried out using 0.5µm diamond paste. The scratch free mirror like surfaces was etched with keller’s reagent (2ml HF, 3ml HCl, 5ml HNO_3, 180ml H_2O).

II.2.5 Tensile Testing

The INSTRON 4206 Universal Testing Machine (UTM) is used to evaluate the mechanical properties of the material at room temperature. For this purpose, standard dog bone specimens as per ASTM E8 were utilized. The evaluation of the stress-strain curve is briefly described below. The tensile testing was done on as rolled samples.

III. RESULT AND DISCUSSION

III.1 Chemical composition Analysis

DC cast billets of AA2219 having nominal composition Al-6.3Cu-0.3Mn-0.12Zr was used for this study. The cut pieces from these billets were re-melted to process Al-Cu alloys with Sc, Nb and Ag addition. The chemical analysis of modified cast billets of Al-5Cu alloy with different composition weighing 5-6 kgs of dia. 110mm were carried out after taking samples from Top and Bottom of the billets (B1 to B6). XRF was carried out using borings and coupons respectively taken from the as-cast and machined billet. The achieved chemical compositions of the billets are shown in Table 1.

From the Table-1, it can be noticed that in all the alloys Zirconium percentage is towards the lower limit value. This may be because Zr was not used as Al-Zr master alloy rather Zr is coming from AA2219 scrap, which is not the standard practice for addition of Zr. Generally, Zr is added as master alloy of Al-Zr for processing of Al-alloys. It can be seen from the table all other alloying elements are within the targeted range and homogeneously distributed throughout the billet. The maximum variation in the chemical composition of the samples taken from Top and Bottom was within 5%. Aluminium-Scandium master alloy will be used as raw material for scandium.

TABLE 1: Chemical composition of modified AA2219Alloy

Materials	Cu	Mn	Zr	Sc	Nb	Ag	Al
Targeted range(wt%)	**4.6-5.6**	**.15 - .25**	**.05 - .15**	**.3- .5**	**.3- .9**	**.25 - .35**	**Balance**
Billet 1 (B1)	5	0.21	0.06	-	-	-	Balance
Billet 2 (B2)	5.0	0.20	0.06	0.4	-	-	Balance
Billet 3 (B3)	4.9	0.21	0.07	0.4	0.4	-	Balance
Billet 4 (B4)	4.2	0.22	0.07	0.4	0.8	-	Balance
Billet 5 (B5)	4.8	0.22	0.06	-	0.8	-	Balance
Billet 6(B6)	4.5	0.22	0.07	0.4	0.5	0.32	Balance

TABLE 2: DSC peaks of Alloy

Alloys composition Billets (1 to 6)	B1	B2	B3	B4	B5	B6
	Alloy + pure Al	Alloy+0.5Sc	Alloy+0.5Sc+ 0.5Nb	Alloy+0.5Sc+ 1.0Nb	Alloy+1.0Nb+ pure Al	Alloy+0.5Sc+0.5Nb+ 0.4Ag
First peak	543	574	574	543	543	543
Second peak	-	-	-	574	-	574

III.2 Differential Scanning Calorimetry (DSC)

DSC was done on as-cast billets with a view to establish homogenization cycle. The peaks corresponding to the endothermic reaction taking place in each billet are tabulated in Table 2. It can be seen that melting peaks were found during heating from room temperature to 620^o C. The endothermic peak of temperature of 574^o C is found in billet2 and billet3, which means peak temperature increases from 543^o C to 574^o C with 0.5 Sc additions in billet2, and 0.5 Sc, 0.5 Nb additions in billet 3. In billet3, 0.5 Nb addition with Sc doesn't show any variation in the peak compared with billet2. The peak temperature of 574 ^{o} C is found in billet 4 and 6 in combination with peak 543^o C.It was noted that reappearance of peak 543 ^{o} C is found with the addition of 2219 0.5Sc 1.0Nb in billets 4 and 2219 1.0Nb billet 5 and in billet6 with 2219

0.5Sc, 0.5Nb 0.4Ag additions. At temperatures well below the endothermic melting peak was adopted for homogenization on all the ingots in order to avoid eutectic melting.

III.3 XRD

XRD done on section rolled samples of modified AA2219 alloy (shown in Fig 1). XRD pattern of billets in section rolled condition shows Al_3Sc, $CuAl_2$ (Ω phase) and Al_3Nb precipitate, because of the thermo mechanical treatments of billets. ($AlAg_2$) γ' phases (shown in Fig 1) are also found in billet-6 [20]. These precipitates might have formed during solidification of billet and air cooling of the rods after final pass of section rolling.

Plane [111] which show increase in intensity from billet-1 to billet-2 with addition of 0.5wt% Sc with Al-5Cu which shows maximum intensity of all the billets. Then intensity decrease in billet 3 and again shows increase in intensity with 1.0 Nb addition in billet4.Intensity of peak [111] is minimum in billet 6 which contains 2219+0.5 Sc+ 0.5Nb +0.4 Ag with Al-5Cu alloy.

The 2nd peak [200] has maximum height in billet 1, and then this peak shows decrease in height with Sc and Nb addition in rest of billets.3rd peak also shows similar character of 2nd peak. Billet 2 and 4 peaks variation is similar to annealed condition of Al alloy.

III.4 Microstructure

The microstructure of as-cast structure of each billet (Fig.2) was taken. Billet 1 shows small dendrites and characteristic of the as-cast billets [2], whereas billets with Sc are free of dendrites. Some billets have minor segregation. It can also be noticed that addition of Sc alone has resulted in major grain refinement as compared to addition of only niobium or niobium and scandium combination.

It was well known from the previous studies about the grain refinement action of Sc with Al and Al alloys because of the effect of Al_3 Sc structure [4,5]. In billet 4 in addition to Sc the 1.0 wt.% Nb addition shows small increase in grain size comparing with Billet2 and Billet 3. When adding 1.0 niobium and pure Al on billet 5 shows that Nb alone doesn't show any grain refinement. Adding of 0.4silver with 0.5 Sc and Nb in billet 6 also shows a large grain size compared to Billet 2 and 3. This shows that scandium is better grain refiner as compared to Niobium.

III.5 Mechanical Properties

The tensile properties under different rolling temperature of AA2219 alloy alone were experimentally investigated first. It was found that it has a tensile strength around 300 Mpa and yield strength of 260 Mpa experimentally investigated at rolling temperature of (625K)350^{o}C also found an elongation of 12.4%. (these results are taken from the previous research work; It is observed that from the experiment at low rolling temperature cracks occur in the surface of the alloy but the mechanical properties are high. While increasing the rolling temperature the cracks are absent on the alloy but mechanical properties are low at (625K) 350 ^{o} C as compared to 250 and 300^o C rolling temperature. In the current investigation rare earth element Sc is added with transition metal Nb and Ag to improve the microstructure and mechanical property at high rolling temperature, which is at 350^o C since this temperature does not produce cracks on the alloy.

The modified Al-5Cu alloys at 350 ^{o} C rolling condition shows decrease in tensile value when compared to AA2219 at same rolling temperature, because of the addition of Al-Sc master alloy addition to theAA2219 alloy which contains 1Kg pure Al. So, for comparison, the AA2219 is added with 1kg pure Aluminium in billet 1.

The mechanical properties such as tensile values of modified AA2219 alloy in section rolled at 350 ºC are shown in Fig 3. The tensile strength, yield strength %elongation and % RA were calculated. The addition of 0.5 wt% Sc with AA2219 alloy (billet2) shows UTS of 223 Mpa and YS of 201 Mpa and elongation of 16.92% which shows an increase in strength and ductility compared to billet1 (AA2219 and pure Al).

Billet-4 is the one which gives maximum UTS of 235 Mpa, YS of 213 Mpa and Elongation of 13.08% which has 2219+ 0.5 Sc + 1.0 Nb. It is because of a greater number of Al_3Nb and Al_3Sc precipitate formed during thermomechanical working. The ductility is maximum in billet-2 because of the grain refinement effect of Sc, as it can correlate it with microstructure as shown in Fig.2 and minimum in billet-5 (AA2219+1.0Nb + pure Al) which has bigger grain size Fig.2.

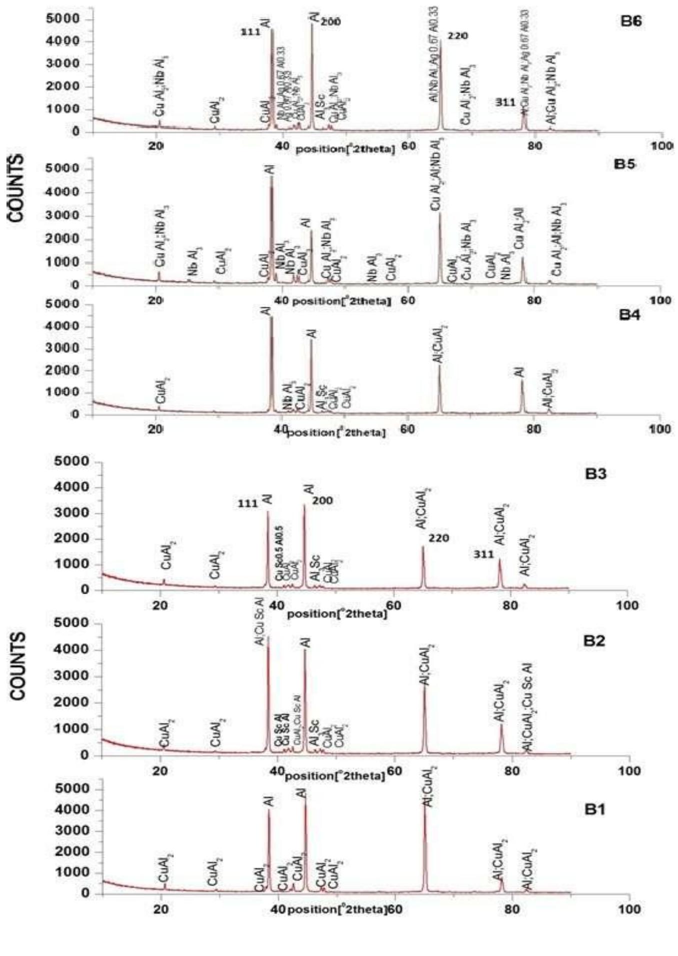

Fig .1- XRD graph of six billets of AA2219 (Al-5Cu) alloy in as rolled condition. (a): billets 1 to 3, and (b) billets 4 to 6

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It is noticed that niobium addition shows a small decrease in ductility of the alloy. The 2219+0.5Sc+0.5Nb +0.4Ag addition in billet 6 shows a UTS of 21 Mpa and YS of 209 Mpa which is very much similar to billet4. By observing the stress strain graph (Fig.4) it was found that billet-4 which contains AA2219 with 0.5 Sc and 1.0 Nb has higher strain hardening and elastic region when compared with others. It shows that rare earth Sc and transition metal Nb in AA2219 increases the strain hardening of the alloy. The billet 6 containing Ag in addition to Sc and Nb is the one which shows high strain hardening and elastic region after billet-4

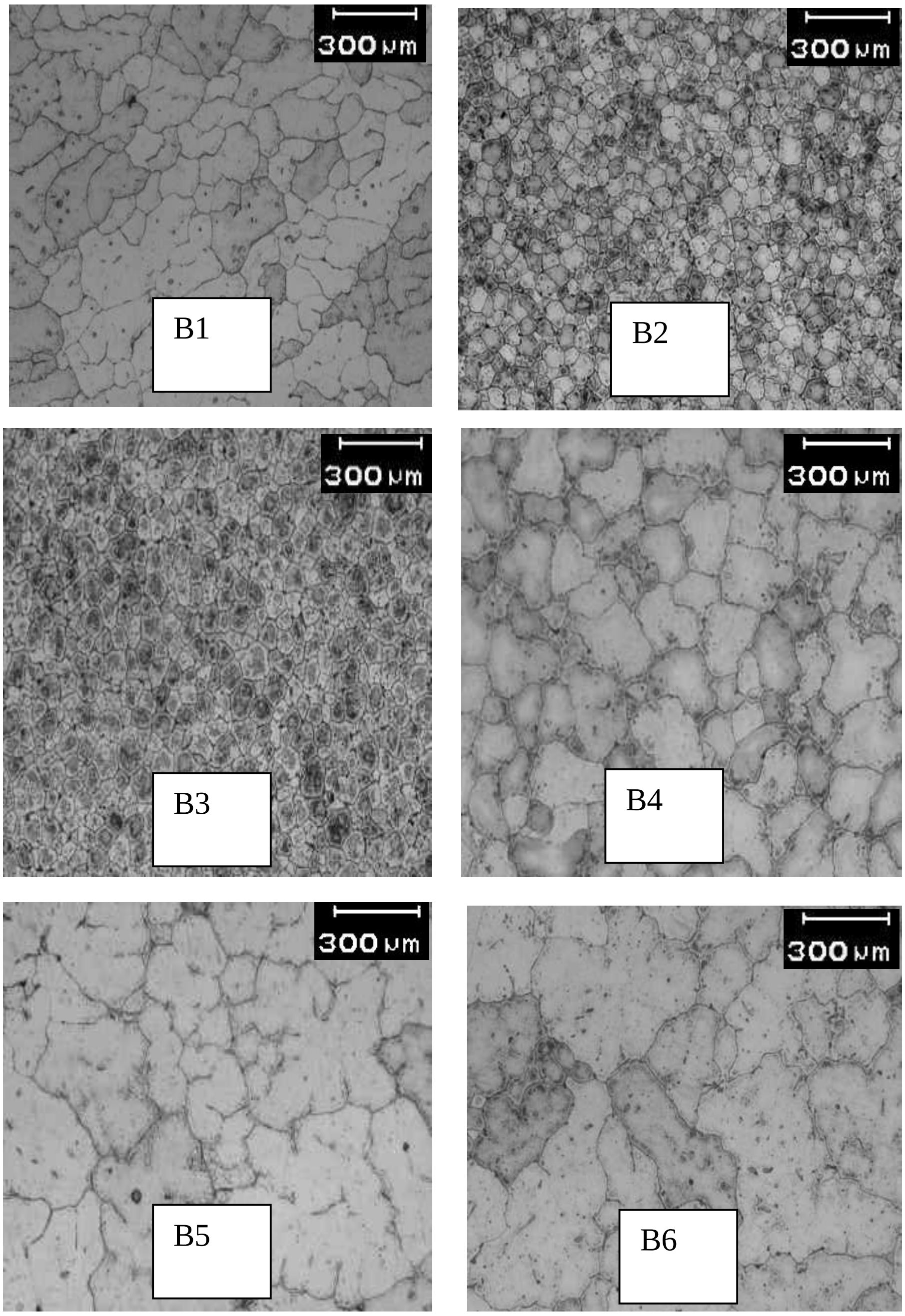

Fig .2- Optical microstructure of 6 billets of modified AA2219 alloy

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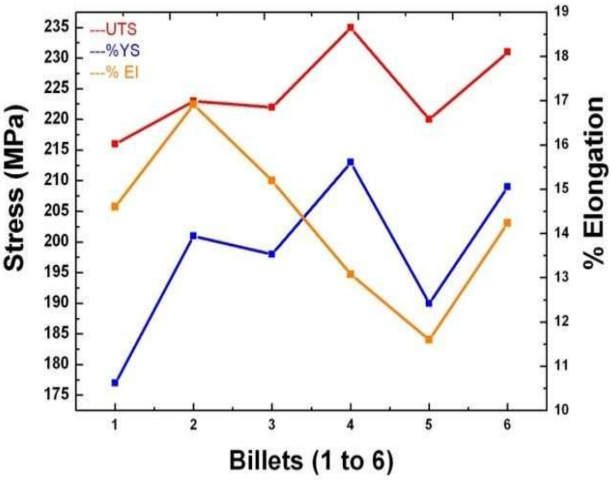

Fig. 3- Comparison of Mechanical properties after tensile test of 6 billets of modified AA2219

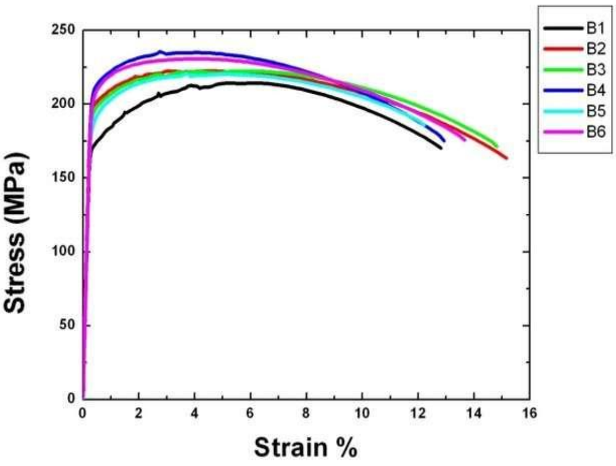

Fig .4 - stress strain curve of six billets of AA2219

IV. CONCLUSIONS

In the present work, vacuum induction melting (VIM) of modified Al-Cu alloy AA2219 of approximately 5 Kgs was carried out under argon atmosphere and ingots were cast in mild steel mould coated with graphite. These ingots were subjected to thermo-mechanical processing. Micro structural is evaluated as- cast condition and mechanical properties were evaluated on section rolled rods.

From the results obtained following conclusions are drawn:

- The XRD in as rolled condition show Al_3Sc, $CuAl_2$ and Al_3Nb, $AlAg_2$ precipitate as expected.
- The microstructure of the alloys is taken under as-cast condition and observed grain refinement in billet-2 which contains 0.5 Sc and billet-3 which contains 0.5 Sc and 0.5 Nb. It does not show significant grain size reduction in billet 4 ,5 and 6 compared to billet-2 and 3. This may be due to poor dissolution of Nb in Al alloy and the higher density of Nb with respect to Al.
- Mechanical properties in section rolled conditions were evaluated. The mechanical properties under rolled condition shows increase in mechanical properties for billet 4(2219+.5Sc+1Nb) compared to billet-1(2219 +1 kg Al). There is an increase of 12 % in yield strength and 10% increase in tensile strength in billet-4 compared to billet-1. This shows that addition of niobium and Scandium to AA2219 increases its mechanical properties. It can see that the combination Sc and Nb with AA2219 increases the tensile strength. In billet-6 (2219+.5Sc+.5Nb+.4Ag) the addition of silver also increases the mechanical property similar to billet-4 compared to billet 1.
- The formation of Al_3 $(Sc_{1-x}$ $X_x)$ phase with Nb did not result in much grain refinement as expected.There is considerable increase in tensile values. It is observed that the further adding of niobium in billet 4(2219+.5Sc+1Nb) doesn't show significant grain refinement as expected and it is same in billet 5 which has no Sc.
- The addition of Ag in the alloy in addition to Sc and Nb is the one which shows high strain hardening and elastic region.

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