A study on the effect of semi solid heat treatment of a cast Al-Cu alloy on its microstructure, mechanical property, physical property and ageing response

Rakoti Shyam Kumara[1], Jhilik Senb[2] and Ashish Bhattacharyyaa[3]
[1,2,3]*Department of Metallurgical and Materials Engineering, National Institute of Technology, Durgapur-713209,West Bengal, India.*

Abstract: The present work presents a novel investigation on ageing behaviour of a Al-10Cu alloy, in which Cu% is much above the solid solubility limit of α-solid solution at the eutectic temperature of 5480C. The heat treatment process consisted of isothermal holding at the semi solid temperature of 5600C for 2 hours followed by water quenching and subsequent ageing at 1500C. During semi solid heat treatment (SSHT) the lamellar eutectic fraction dissolved resulting in the formation of highly supersaturated α phase on quenching. The as quenched alloy was aged at 1500C for 8 hours to a peak hardness of 95 VHN. The resistivity changes on SSHT and ageing was recorded and the mechanical properties of the peak aged samples were also recorded. The changes taking place during this novel heat treatment was followed up by TEM examination.
Key word: Semi solid heat treatment, cast structure modification, copper distribution in α solid solution, improved conductivity.

I. INTRODUCTION

Aluminium alloys are widely used in aerospace, automobile and other engineering industries. Microstructure of as cast alloys influence their mechanical and physical properties. The microstructural features include grain size of the α-phase; morphology and constitution of the eutectic phase and distribution of the pro-eutectic phase. Microstructural modification of non-ferrous alloys by semi solid process was developed in the mid 1970's1. A number of investigations were subsequently carried out on the modification of the structure and properties of nonferrous alloys through several semi solid processing routes. Limodin et.al2 reported the overall and local microstructural changes occuring during partial remelting of an Al--15.8 wt.% Cu alloy.This study was based on in situ investigation by X-ray tomography. In 2008 Atkinson and Liu3 studied the microstructural coarsening of semi-solid aluminum alloys. Their work mainly concentrated on two potential SSM processes like RAP and CS of alloy 201 and 2014. In 2009 Terzi et.al4 investigated in situ X-ray micro-tomography characterization of the entrapped liquid formed during partial remelting of a cold-rolled Al-8 wt.% Cu alloy. In 2010 Atkinson and Liu5 again studied the coarsening rate of microstructure in semi-solid aluminium alloys 201 and 2014 under the same SSM processes like RAP and CS. Recently, Sahu and Behera6 investigated the effect of SIMA processing on grain size distribution, hardness and tribological behavior of Al-10Cu alloy. The present investigation aimed to study the effect of semi-solid isothermal heat treatment (SSHT) on the microstructure, mechanical property, physical property and ageing response of an Al-10Cu alloy through metallography, mechanical testing, resistivity measurements, EDS analysis, scanning and transmission electron microscopy.

II. EXPERIMENTAL

2.1 Preparation of Al-Cu alloy

In the present work the experimental alloy has been prepared in a vacuum induction furnace and the liquid mental poured in a metal mould in the form of circular rods and rectangular plates. The composition of Al-Cu alloy was analyzed by using Optical Emission Spectrometer and the result is presented in the Table. 1.

2.2 Heat Treatment

Small samples with dimensions of 10 mm x 10 mm x 10 mm were heated in a muffle furnace in normal atmosphere. The temperature was controlled with an accuracy of ±20C using a programmable controller.

2.3 Volume Percentage of Liquid in Alloy in Semisolid State

The normal liquid volume percentage during heat treatment was determined by applying the lever rule in the Al-Cu equilibrium binary phase diagram [ASM 1992]. The value of the temperature and volume fraction of liquid during semisolid heat treatment for the alloy has been given in the Table. 2.

2.4 Metallographic Sample Preparation

Samples having dimensions of 10mm x 10mm x 10mm were prepared for metallographic study by grinding and polishing as per standard procedure normally adopted. Etchant used was Keller's reagent (2.5% HNO_3, 1.5%HCl, 1% HF and 95%H_2O). Swab etching technique was applied.

2.5 Optical Microscopy

Optical microscopy and garin size analysis was carried out using a Leica image analyzer (Model: DM2500M, Leica Mircosystems, Wetzlar, Germany) interfaced with image analysis software.

2.6 Scanning Electron Microscopy

The microstructural investigation of the alloys was carried out using a scanning electron microscope (Nova Nano SEM 430, Netherland) coupled with energy dispersive X-ray (EDX) micro analyzer (Model: ISI300, Oxford Instruments Ltd., UK) for compositional analysis. The polished and etched specimens were observed using secondary electron (SE) imaging mode.

2.7 Transmission Electron Microscopy

To determine the progress of microstructure modification through semisolid heat treatment, a detailed microstructure characterization was carried out using a high resolution transmission electron microscope (JEOL-JEM 2200 FS, JAPAN) with inbuilt EDS facility. The sample used for TEM observations were prepared using ion-beam milling.

2.8 Hardness Tests

Macro-hardness measurements were carried out using a Vickers diamond indenter (Model: LM 700, LECO, Japan) operated at a load of 10 kg allowing a dwell time of 15 s for indentation.

2.9 Tensile Tests

Tensile tests were carried out using a universal testing machine (Instron Model: 8516, U.K.) The tests were carried out at room temperature with a cross head speed of 1.0 mm/min.

2.10 Resistivity Measurement

Resistivity changes with microstructural condition (as cast, as solution treated and as solution treated & aged) was recorded. Instrument - 2182A Nano-voltmeter, 6221 DC and AC current source, Keithley - Current Voltage source, USA.

III. RESULTS AND DISCUSSION

The EDS analysis of the as cast structure [Fig. 2.2(a-c)] shows that the solidification of the cast alloy deviated from the equilibrium condition. However an island of a non-equilibrium phase could be detected inside the α-phase. In this phase the copper content was in the hyper eutectic range (38.3%Cu). Obviously the heavier solute was not uniformly distributed before casting, resulting in survival of a copper rich hyper eutectic β-phase. Even the copper content (4.83%Cu) of the α-phase is much above the equilibrium concentration of copper. It is interesting that iron, present as impurity, also segregated into the melted liquid. The dendritic structure is refined during semi solid processing, spheroidized grains are formed. Eutectic liquid is entrapped inside the spheroidized grains, as well as between the grains. On SEM-EDS analysis the situation becomes clearer [Fig. 2.3(a-c) and 2.4(a-d)]. It is apparent that the eutectic lamellar structure is not completely removed during semi solid heat treatment. But the lamellae of the intermetallic copper aluminide phase are broken. On EDS analysis (spot 1, Fig. 2.4a) the composition appears to be near eutectic. The inter lamellar spacing is so small that the copper aluminide phase could not be analyzed separately. In spot 2, Figure. 2.4b the copper content in the α phase is >6.0% which is a significant deviation from the equilibrium diagram. In spot 3 and 4 of Figure. 2.4(c-d), the copper aluminide phase is quite massive and stand proud over the matrix. The copper content is nearly 48%, which suggests that this solid intermetallic phase separated out due to micro-segregation of cupper.

Ageing treatment at 1500C for 8 hours resulted in increased hardening. But EDS analysis [Fig. 2.6(a-c)] results do not provide any clue to the precipitation process. The SEM photographs also do not reveal any precipitate. Usually the previous reports on ageing of Al-Cu alloys suggest that the precipitates formed on ageing treatment are resolved only in TEM. The next attempt was therefore made for TEM examination of thin foils of the relevant samples.

On TEM examination, a fractured lamellae surrounded by α- phase visible in Fig. 3.1(a-c). In the surrounding the α- phase, the Cu content varied from 3.83% to 11.10 [Fig. 3.2, spot 4 and 5]. The lamellae of course shows 100% Al constitution. It therefore seems that dissolution of the eutectic copper aluminide lamellae in the liquid formed during SSHT raised the Cu content in the melt, but during the short heat treatment period copper content was not homogenized. On quenching, the α-matrix became supersaturated with respect to Cu. But the TEM photographs [Figure 3.3(a-f)] suggest that fine copper aluminide precipitated out, as the matrix could not hold the entire copper in solid solution. The copper concentration was higher near the α-lamellae. The solution treated sample shows clear sub-grains with dark precipitates. Dislocation tangles are visible at places [Figure 3.3(a-f)]. Presumably dislocation-solute atom-vacancy interaction occurred as seen in Fig. 3.4. The copper content varied from 11.86% inside the sub-grain to 21.55% at the sub-grain boundary [Figure 3.5, 3.6].

The mechanical properties of the as cast alloy has been compared with those of the semi-solid heat treated alloy. The data are presented in Table 3.1 and3.Table 3.2. The tables also illustrate the mechanical properties of solid state solution treated and aged Al-10Cu alloy. It transpires from these data that the yield strength and tensile strength of the alloy did not improve significantly by semi-solid heat treatment although the microstructure is transformed from a dendritic to equi-axed rounded grains. However, the mechanical properties improved after ageing. The resistivity data on the samples are given in Table 3.3. The resistivity of the as cast sample decreased significantly after semi solid heat treatment and quenching. The enhanced copper content in the α-solid solution [Figure 3.6] probably improved conductivity and correspondingly resistivity dropped. It dropped further on ageing. It is likely that a part of the dissolved copper precipitated as incoherent copper aluminide . This reduced lattice strain of the supersaturated α- solid solution and hence the resistivity dropped. The resistivity of the solid state solution treated Al-10Cu alloy is higher in the aged condition. In this case, the initial saturation of α- matrix was smaller [Figure 3.7(a-c)]. During the ageing mainly coherent precipitates formed. The increased lattice strain which resulted in an increase in resistivity.

IV. CONCLUSIONS

The present investigation aimed to find out the effect of SSHT on the micro structure, mechanical property, physical property and ageing response of an Al-10 Cu alloy. The following conclusions may be drawn on the basis of optical microscopy, SEM-EDS study, TEM examination, tensile and resistivity test:

After SSHT, the dendritic structure was replaced by rounded α- grains with eutectic mass at the grain boundary. Isolated precipitation of copper aluminide phase was also observed within the α-phase.

High rate of cooling (metal mould casting) leads to higher copper content in the α-phase than that of the equilibrium value in Al-Cu alloy system.

Water quenching after semi solid solution treatment further increases the copper content in the α-phase and correspondingly the copper in the eutectic theta-phase is less than the equilibrium value. This leads to formation of a gradient in copper concentration in the aged sample.

TEM examination indicates that SSHT raised the copper content in the melt, but during the short heat treatment period copper content was not homogenized. On quenching, the α-matrix became supersaturated with respect to Cu. The TEM photographs also suggest that fine copper aluminide precipitated out, as the matrix could not hold the entire copper in solid solution. The solution treated sample shows clear sub-grains with dark precipitates. Dislocation tangles are visible at places . Dislocation- solute atom-vacancy interaction might have occurred. Wide variation in copper content was observed from the center to boundary of the sub-grains.

The mechanical testing data suggest that the yield strength and tensile strength of the alloy did not improve significantly by semi solid heat treatment, although the microstructure is transformed from a dendritic pattern to equi-axed rounded grains.

The resistivity of the Al-Cu cast sample decreased significantly after semi solid heat treatment and quenching. A part of the dissolved copper precipitated as incoherent copper aluminide during ageing. This reduced lattice strain of the supersaturated α- solid solution and hence the resistivity dropped. The resistivity of the solid state solution treated Al-10Cu alloy is higher in the aged condition. In this case, the initial saturation of α-matrix was smaller. During the ageing mainly coherent precipitates formed. The increased lattice strain which resulted in an increase in resistivity.

V. REFERENCES:

[1] Flemings M C, Rick R G and Young K P, Mat Sci and Eng, 25 (1976) 103.
[2] Limodin N, Salvo L, Suery M, and DiMichiel M, Acta Materialia, 55 (2007) 3177.
[3] Atkinson H V, and Liu D, Mat Sci and Eng A, 496 (2008) 439.
[4] 4.Terzi S, Salvo L, Suery M, and Boller E, Scripta Materialia, 60 (2009) 671.
[5] Atkinson H V, Liu D, Trans Nonferrous Met Soc China 20 (2010) 1672.
[6] Sahu Ashutosh,and Behera Ajit, Materials Today: Proceedings 2 (2015) 1175.

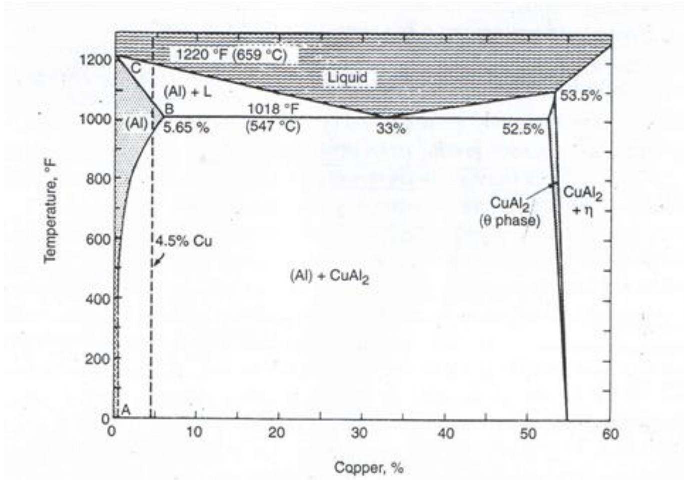


Figure 1. The relevant part of the aluminum-copper phase diagram.

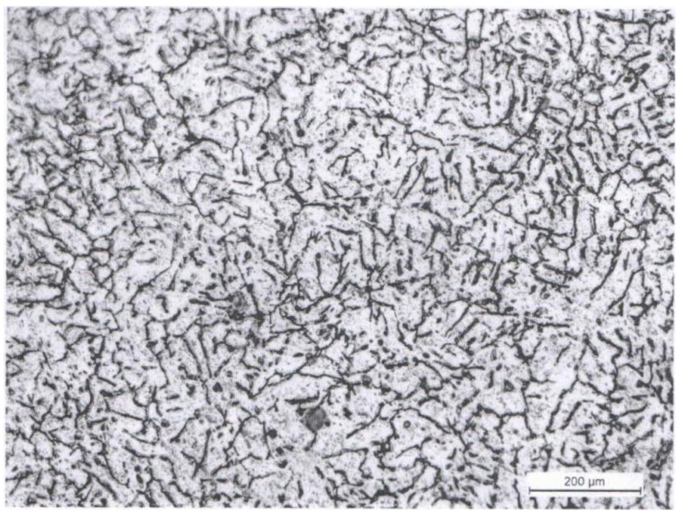


Figure 2.1a Optical miscrograph of the as cast alloy

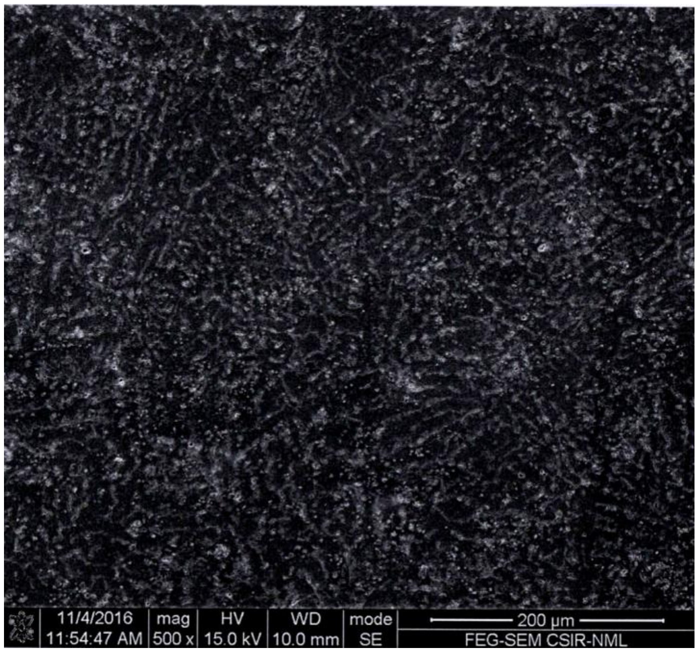


Figure 2.1b. SEM micrograph of the as cast alloy.

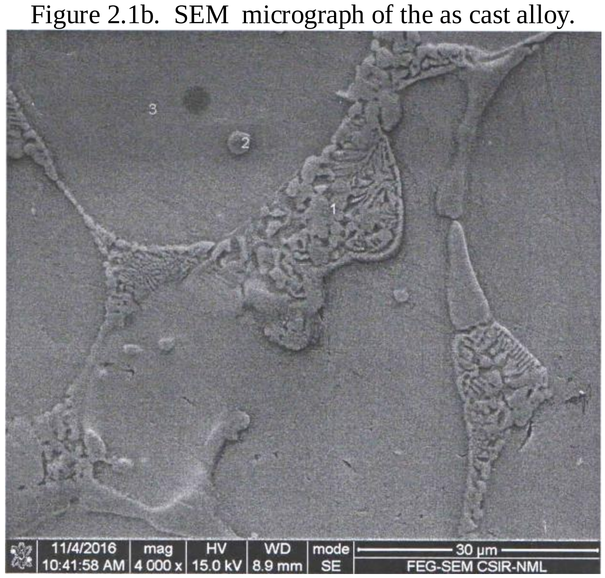
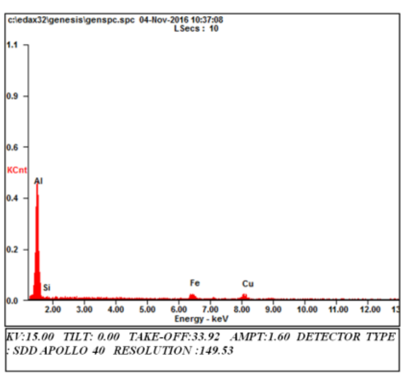


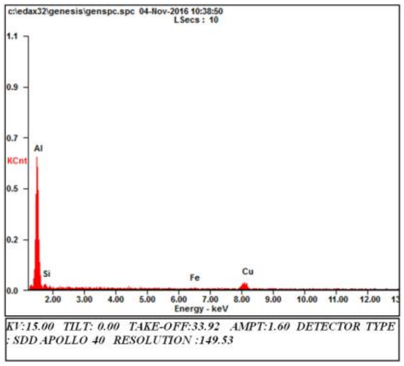
Figure 2.1c. The corresponding SEM micro-graph for EDS study.



Element	Wt %	At %
AlK	55.70	73.88
SiK	00.21	00.27
FeK	13.20	08.46
CuK	30.88	17.39

EDAX ZAF QUANTIFICATION STANDARDLESS SEC TABLE : DEFAULT

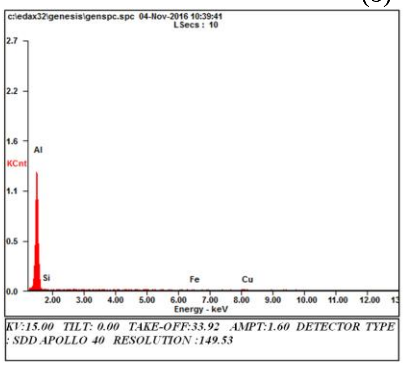
(a)



Element	Wt %	At %
AlK	58.77	76.36
SiK	01.08	01.35
FeK	01.84	01.16
CuK	38.30	21.13

EDAX ZAF QUANTIFICATION STANDARDLESS SEC TABLE : DEFAULT

(b)



Element	Wt %	At %
AlK	94.76	97.68
SiK	00.00	00.00
FeK	00.40	00.20
CuK	04.83	02.12

EDAX ZAF QUANTIFICATION STANDARDLESS SEC TABLE : DEFAULT

(c)

Figure 2.2. EDS study at different locations (1-3) as indicated in Fig.1c.

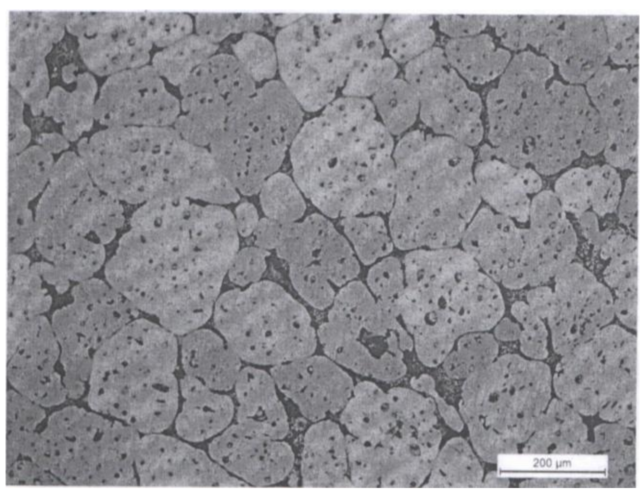


Figure 2.3a. Optical micrograph of the alloy after solution treatment (in semi solid range) at 5600C for 2 hrs.

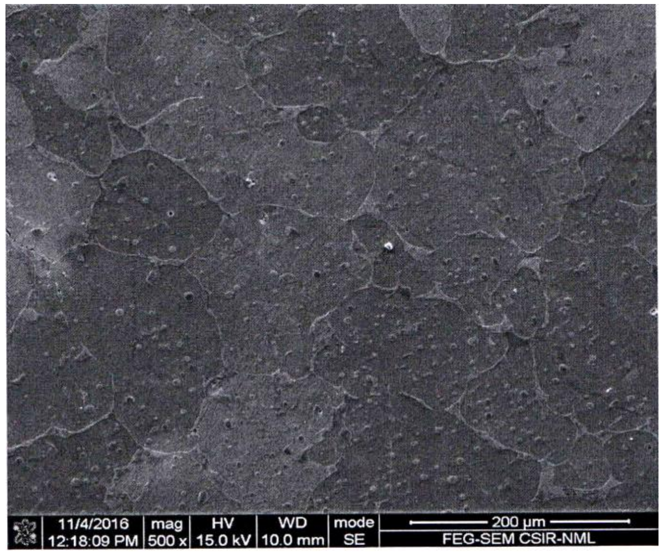


Figure 2.3b. SEM micrograph of the alloy after solution treatment (in semi solid range) at 5600C for 2 hrs.

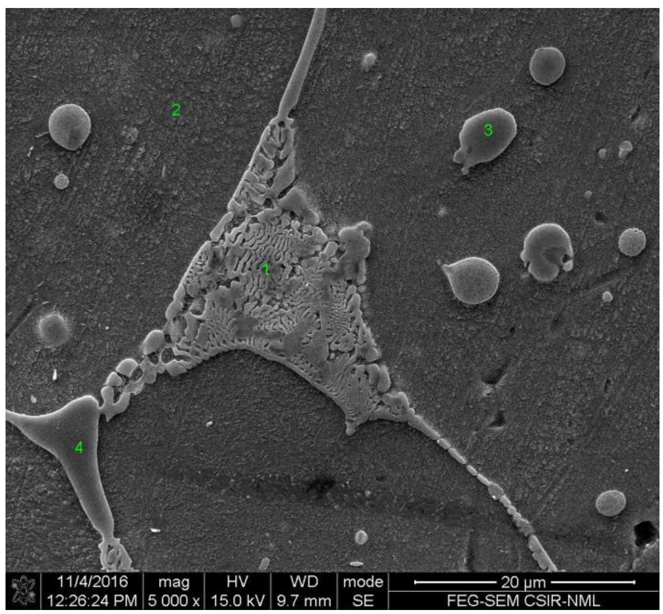
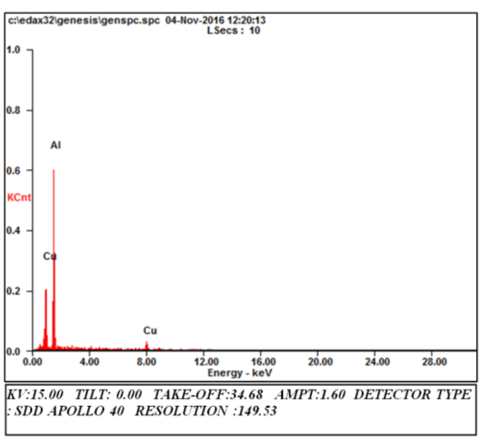
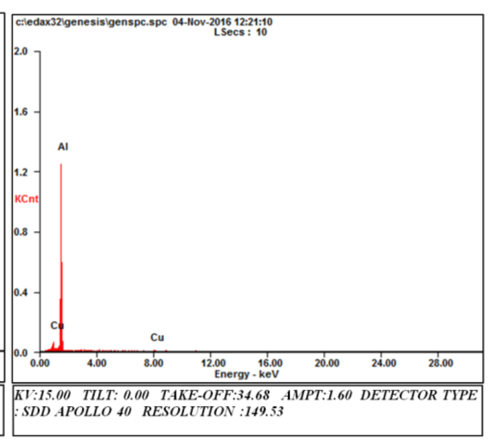


Figure 2.3c. The corresponding SEM micro-graph for EDS study



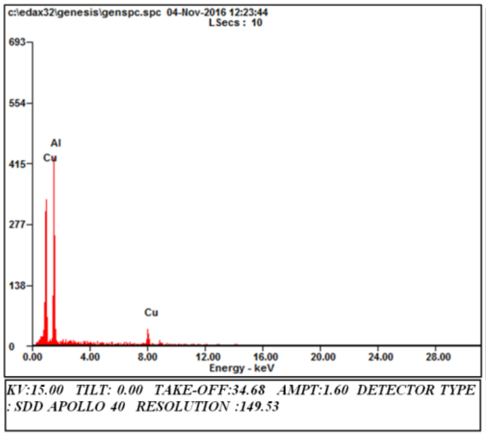


Element	Wt %	At %
AlK	65.86	81.96
CuK	34.14	18.04

(a)

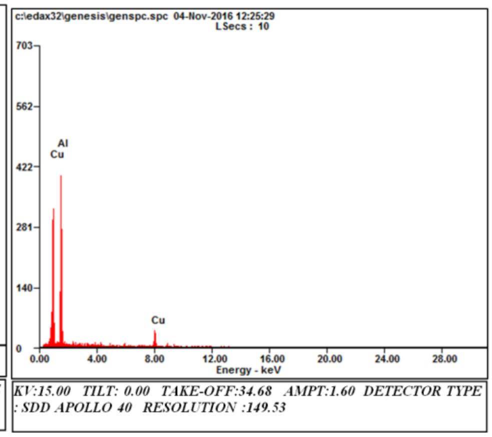
Element	Wt %	At %
AlK	93.66	97.20
CuK	06.34	02.80

(b)



Element	Wt %	At %
AlK	52.30	72.08
CuK	47.70	27.92

(c)



Element	Wt %	At %
AlK	50.92	70.95
CuK	49.08	29.05

(d)

Figure 2.4. EDS study at different locations (1-4) as indicated in Fig. 2.3c.

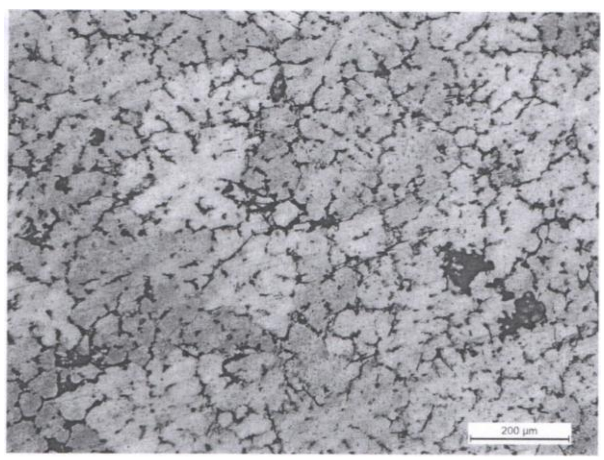


Figure 2.5a. Optical micrograph of the solution treated (in semi solid range at 5600C for 2 hrs). sample after aging at 1500C for 8 hrs.

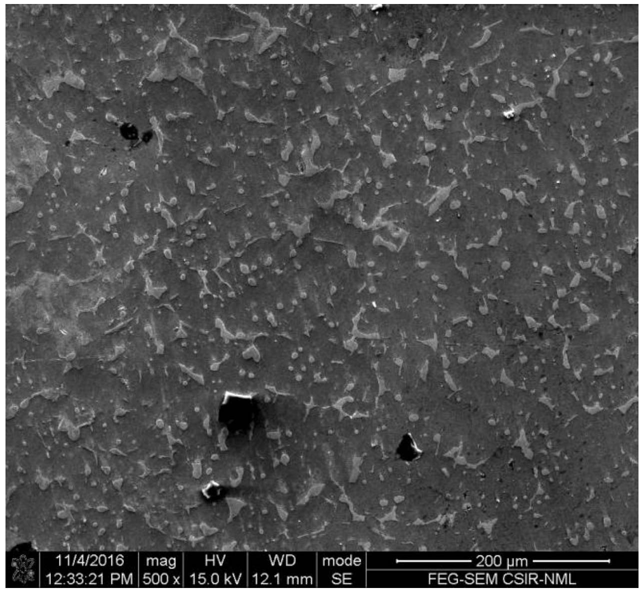


Figure 2.5b. SEM micrograph of the solution treated (in semi solid range at 5600C for 2 hrs). sample after aging at 1500C for 8 hrs.

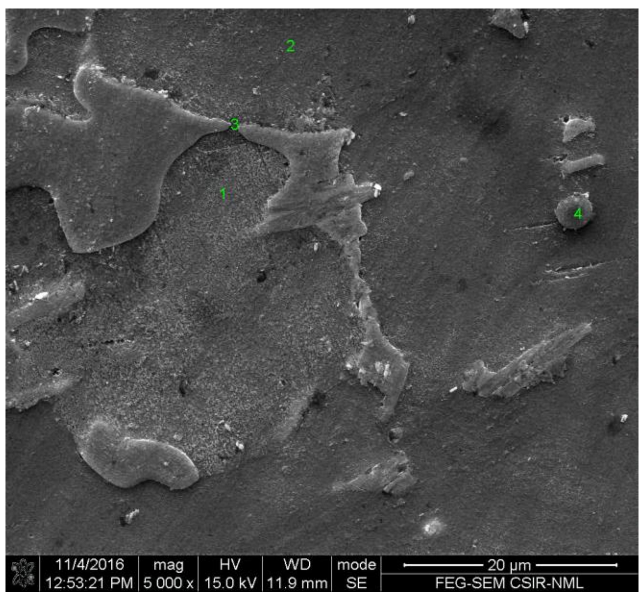
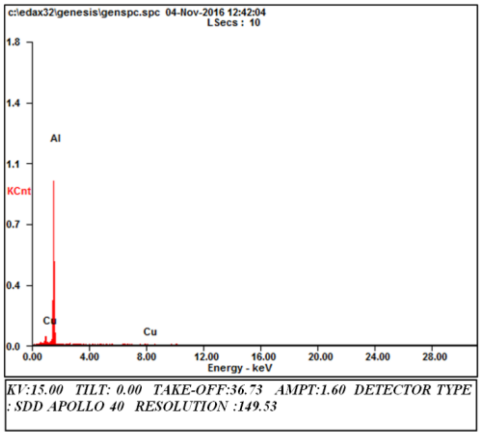
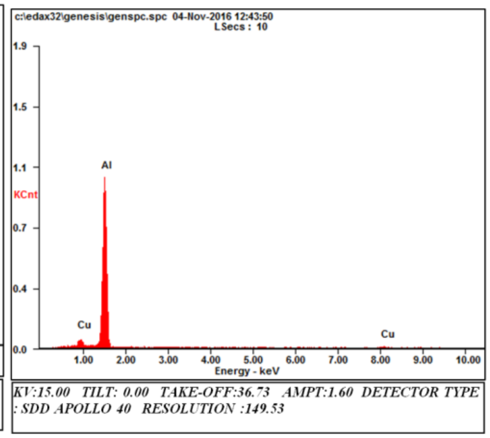


Figure 2.5c. The corresponding SEM micro-graph for EDS study.



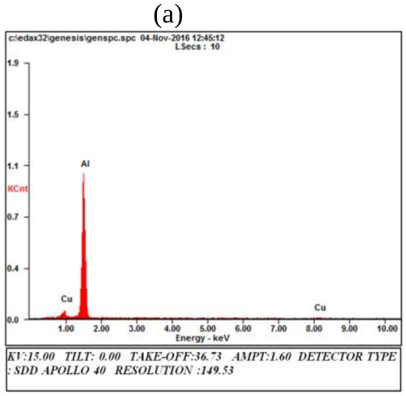
Element	Wt %	At %
AlK	94.32	97.51
CuK	05.68	02.49

(a)



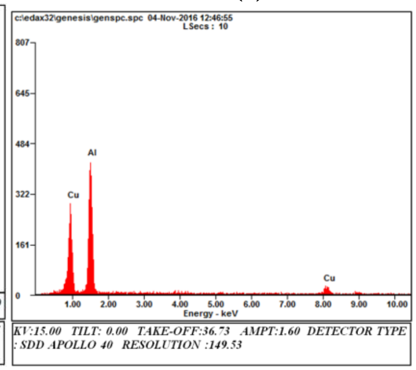
Element	Wt %	At %
AlK	95.77	98.16
CuK	04.23	01.84

(b)



Element	Wt %	At %
AlK	94.69	97.67
CuK	05.31	02.33

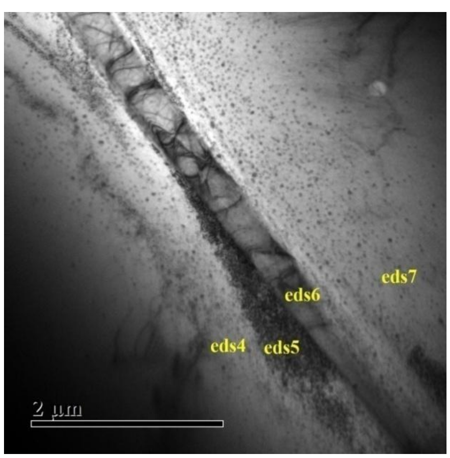
(c)



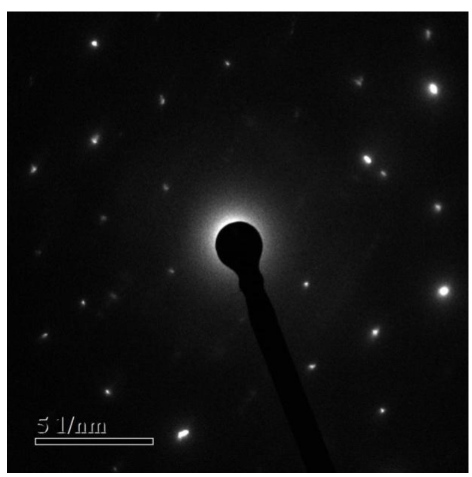
Element	Wt %	At %
AlK	58.24	76.66
CuK	41.76	23.34

(d)

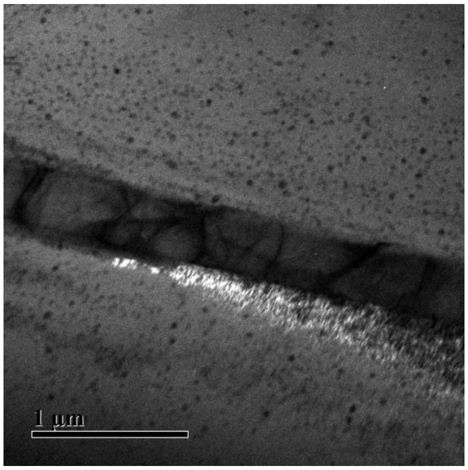
Figure 2.6. EDS study at different locations (1-4) as indicated in Figure 2.5c.



(a)



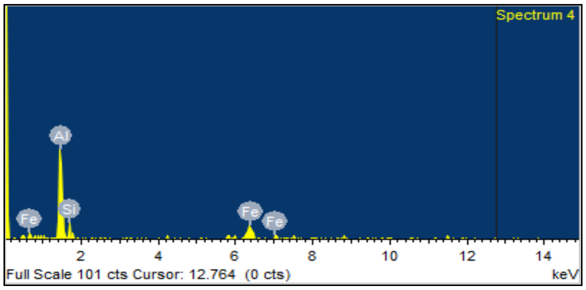
(b)



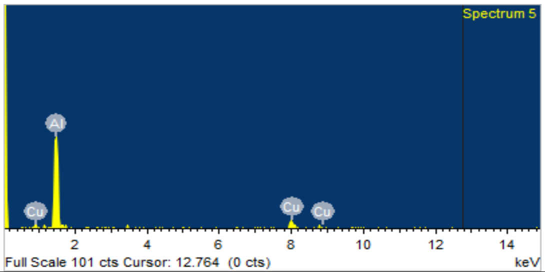
(c)

Figure 3.1. TEM study of as-cast Al-10Cu alloy (a) BF image of dendrite interior (b) Corresponding SADP (c) Same image (a) at higher magnification

Element	Weight%	Atomic%
O K	0.00	0.00
Al K	74.86	86.26
Si K	0.00	0.00
Fe K	21.31	11.87
Cu K	3.83	1.87
Totals	100.00	

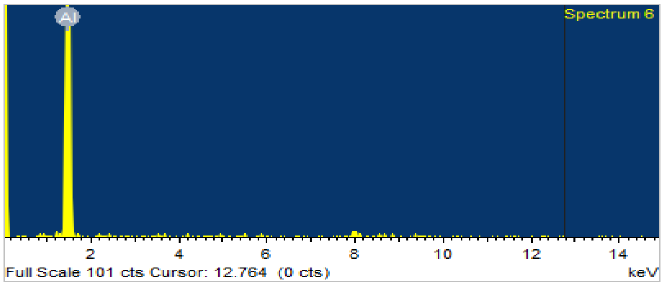


Element	Weight%	Atomic%
Al K	88.90	94.96
Cu K	11.10	5.04
Totals	100.00	



Element	Weight%	Atomic%
Al K	100.00	100.00

Totals	100.00	



Element	Weight%	Atomic%
Al K	93.11	96.95
Cu K	6.89	3.05
Totals	100.00	

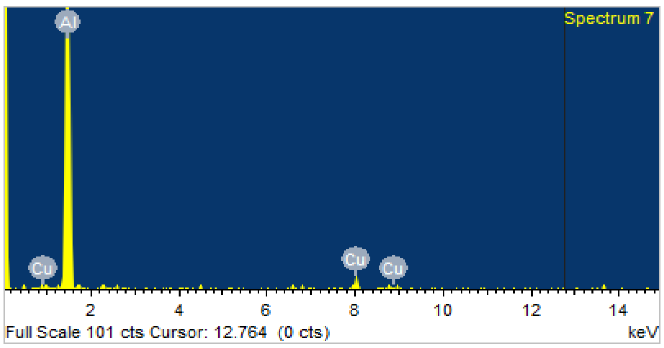
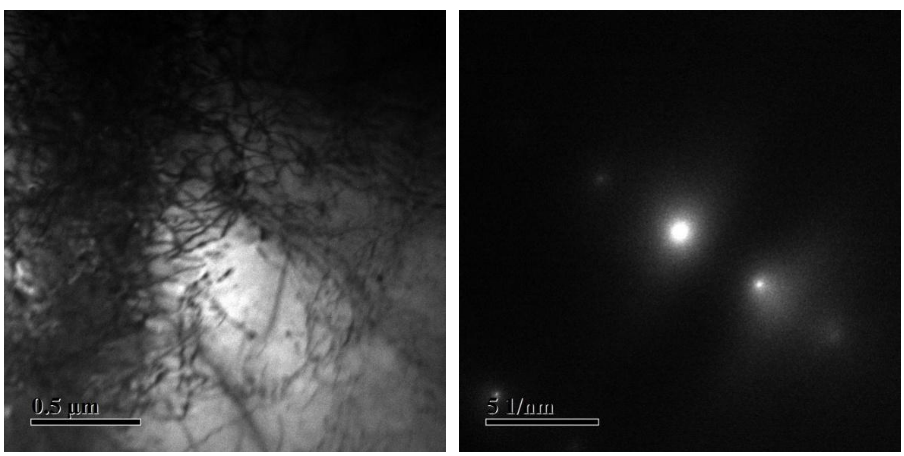
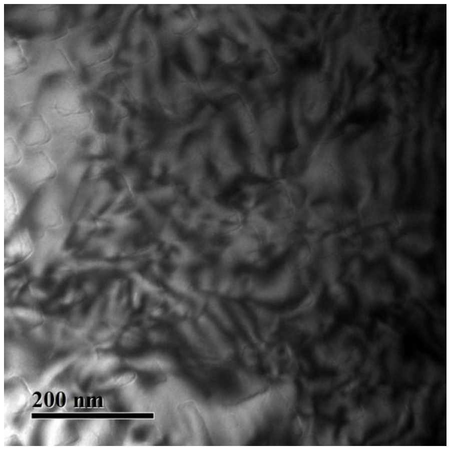
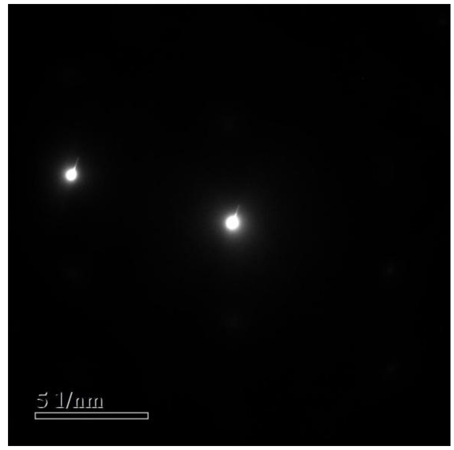


Figure 3.2.EDS study of cast Al-10Cu alloy Spectrum 4-7 correspond to Figure. 3.1(a and c)

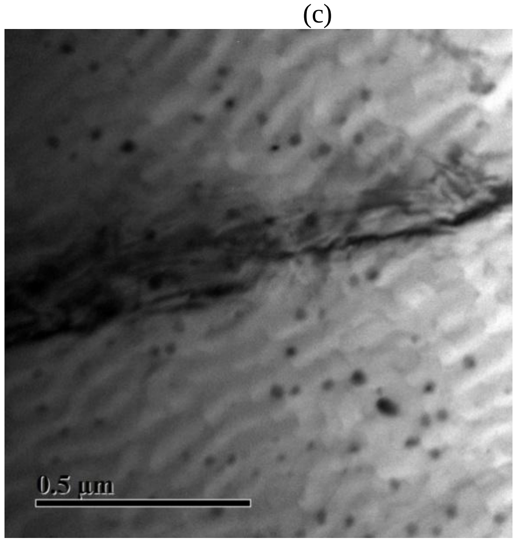


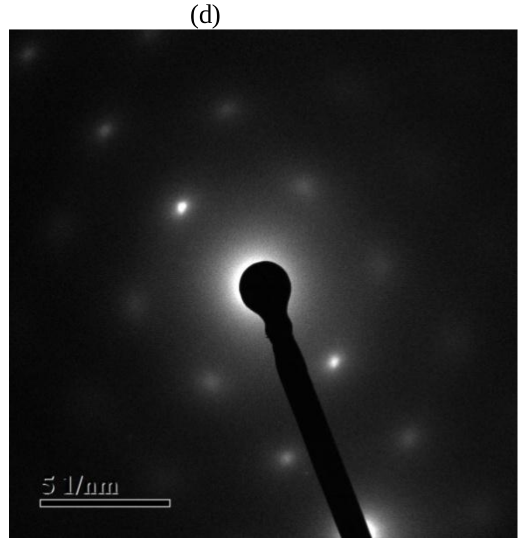
(a) (b)





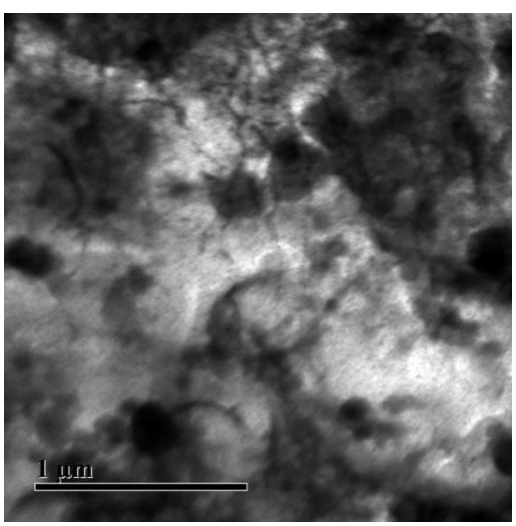
(c) (d)



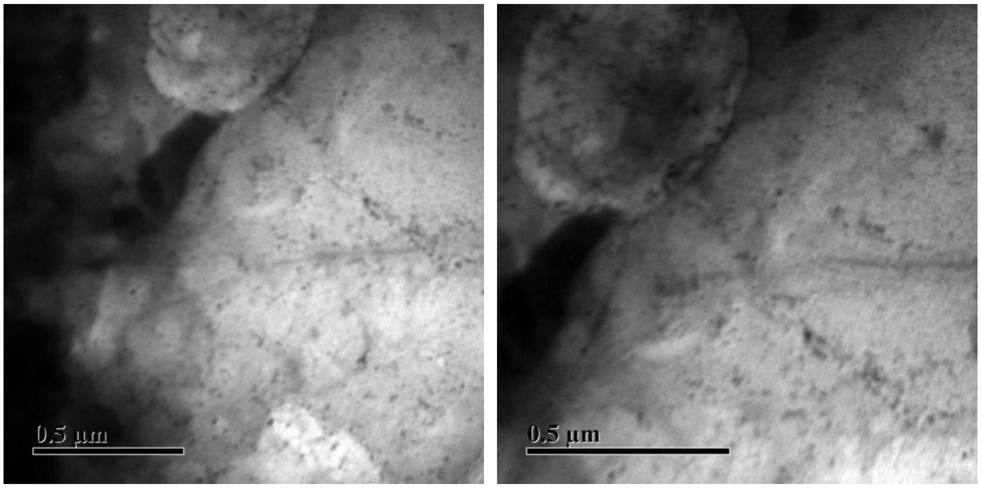


(e) (f)

Figure 3.3. TEM study of Al-10Cu alloy solution treated (in semi solid temperature range) at 5600C for 2 hours. (a) BF image showing that dislocations are generated in the grains due to high quenching stress (b) Corresponding SADP (c) BF image showing dislocation tangles (d) Corresponding SADP (e) Distinct sub-grains also formed. There is evidence of precipitation of inside the grains. Quenching cracks also appear. (f) Corresponding SADP.

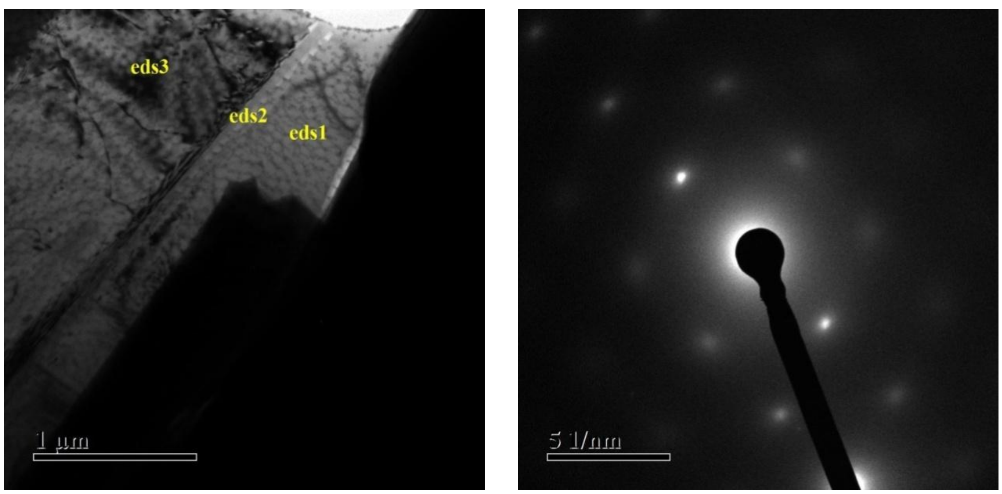


(a) (b)



(c) (d)

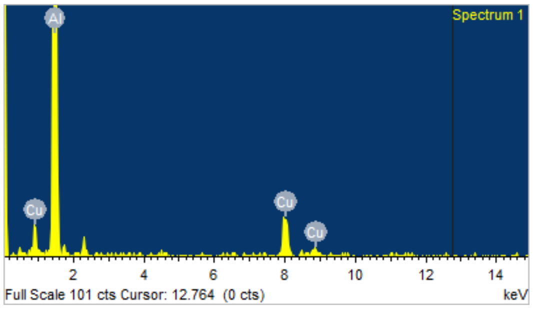
Figure 3.4. TEM study of (semi solid solution treated + aged at 1500C for 8 hours) Al-10Cu alloy
(a) BF image indicating a highly dislocated spot
(b) Corresponding SADP
(c) and (d) BF images show during ageing new grains are formed. Newly created precipitates are also visible in the grains.



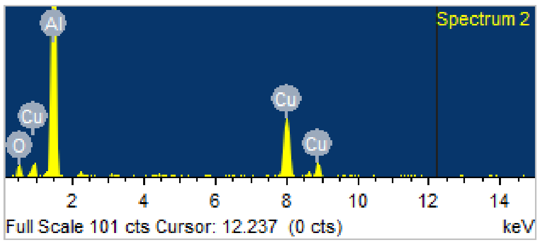
(a) (b)

Figure 3.5. Further TEM investigation of semi solid solution treated + aged sample
(a) BF image at a particular location inside the grain
(b) Corresponding SADP.

Element	Weight%	Atomic%
Al K	88.14	94.60
Cu K	11.86	5.40
Totals	100.00	



Element	Weight%	Atomic%
O	2.33	4.40
Cu	21.55	10.26
Al	76.12	85.34
Totals	100.00	100.00



Element	Weight%	Atomic%
O K	0.00	0.00
Al K	80.45	90.65
Cu K	19.55	9.35
Totals	100.00	

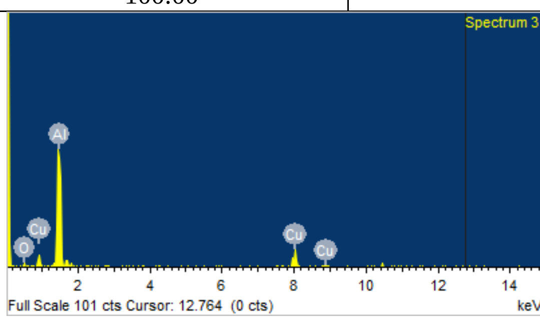


Figure 3.6. EDS study of the BF image (Figure 3.5a)

Sprectrum 1-3 indicates even after aging for 8 hours at 1500C distribution of copper is non- uniform. It suggest that the precipitation is not uniform throughout the grain during ageing.

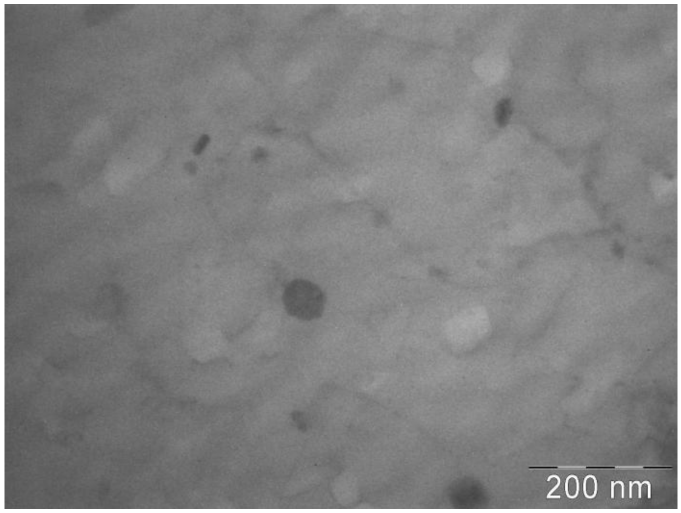


Figure 3.7a. BF image of the solid state solution treated (at 5300 C for 2hrs.) + aged at 1500 C for 8hrs.

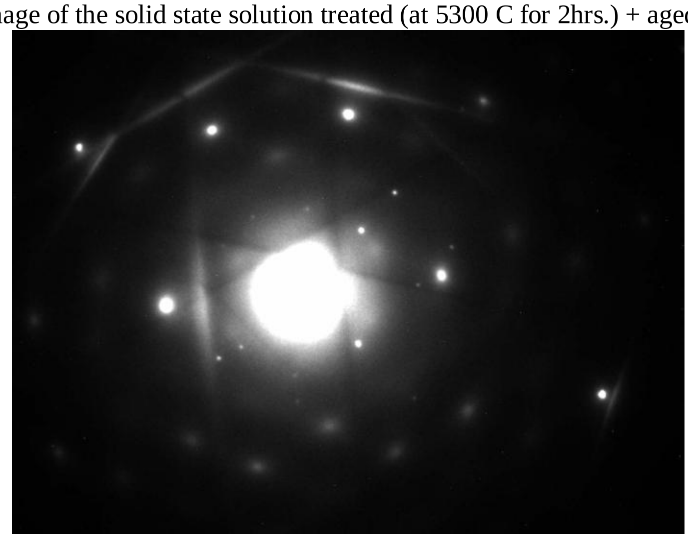


Figure 3.7b. The corresponding SADP.

Element	Weight %	Atomic %	Xpt
AlK	95.1	97.7	0.021
MnK	0.8	0.4	0.004
FeK	1.4	0.7	0.003
CuK	2.7	1.2	0.002
Total	100.0	100.0	

kV: 200.00 X Tilt: 20.00 Y Tilt: 0.00 Azimuth: 45.00
Thickness: 100.00 Elevation: 35.00 AmpT: 25.6
Det Type:SUTW, Sapphire Res: 133.03 Lsec: 100

Figure 3.7c. The EDS analysis of a location indicating average chemical composition of the solid state heat treated matrix (Fig. 3.7a).

.

Table 1: Chemical composition of the Al-Cu alloy used in the present study.

Al (wt%)	Fe (wt%)	Si (wt%)	Mg (wt%)	Mn (wt%)	Cu (wt%)
Balance	0.41	0.35	0.002	0.02	10.05

Table 2: Volume fraction of liquid in the semi solid state.

Temperature (oC)	Al-10Cu
560	20 (approximately)

Table: 3.1 Hardness of the alloy at different condition.

Sample Condition	Hardness (HB)
As-cast	87
Semi solid solution treated at 5600C for 4 hrs.	65
ST + aging at 1500C for 6 hrs.	87
ST + aging at 1500C for 8 hrs.	95
ST + aging at 1500C for 10 hrs	82
Solid state solution treated at 5300C for 2 hrs + aging at 1500C for 8 hrs	91

Table: 3.2 Tensile property of the alloy at different condition.

Sample Condition	Yield Strength (MPa)	UTS (MPa)	% Elongation
As-cast	158	228	3.52
Semi solid solution treated at 5600C for 2 hrs.	78	119	2.37
Then aged at 1500C for 8 hrs.	179	276	3.92
Solid state solution treated at 5300C for 2 hrs + aging at 1500C for 8 hrs	118	168	2.65

Table: 3.3Resistivity of the alloy at different condition.

Sample condition	Resistivity (x104Ω- m)
As-cast	1.23
Solution treated at 5600C for 2 hrs.	0.77
ST (5600C for 2 hrs) + aging at 1500C for 8 hrs.	0.68
ST (5300C for 2 hrs) + aging at 1500C for 8 hrs.	4.33