

# Investigation of Precipitation in Aluminum-Copper Alloy Using Positron Annihilation Lifetime Technique

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*Positron annihilation lifetime studies (PAL) have been carried out on Al-Cu alloy, in order to get information about solute surroundings at the annihilation site. Exceedingly large numbers of positrons are found to annihilate at the matrix-Al<sub>2</sub>Cu precipitate interfaces. The defect structure of Al-Cu alloy with a variety of Cu concentrations is deduced from the PAL measurements at room temperature. The change of the average positron lifetime with different Cu concentrations (up to 25 wt. % Cu) has been discussed. PAL measurements revealed saturation trapping of positrons at large concentration of copper in the alloy, specifically in matrix-precipitate interface. Complementary techniques such as X-ray diffraction (XRD), scanning electron microscope (SEM), and Vickers hardness are used in the present work. The nucleation and growth of the Al<sub>2</sub>Cu particles lead to reduction in the positron lifetime and raising in the Vickers hardness. Hence, the hardness variation is qualitatively consistent with that of positron annihilation parameters. The information obtained from PAL studies provides direct observations of positron trapping at the misfit interfaces between the Al-rich matrix and the Al<sub>2</sub>Cu phase.*

## **1. Introduction:**

The properties of aluminum-based alloys are considered significant for a wide variety of uses [1]. Many of the wrought aluminum alloys are inexpensive and prepossessing because of their low density (2.71 g/cm<sup>3</sup>) compared with competitive metallic alloy systems. They also have good corrosion resistance and good workability. Because many of the fabricated aluminum alloys are based on the Al-Cu alloy, much attention was given to the phase transitions in Al-Cu alloy [2-5]. Information in a phase diagram permits the determination of boiling points, melting points, and sublimation points.

Application of positron-annihilation spectroscopy (PAS) to the characterization and study of defects in metals and alloys has grown rapidly in recent years [6, 7]. Owing to the ability of the positron to annihilate from a variety of defect-trapped states in metals and alloys, PAS can yield defect-specific information which has made a significant impact upon our knowledge regarding lattice defect properties in metals and alloys. PAL has also been applied to study the association of solute elements at vacancies during precipitation in binary alloys [8].

Since positrons provide information only about the environment in which they annihilate, they are considered as a selective probe of the atomic environment around defects. This selectivity is determined by the distribution of the positron in the defect. The list of the most important potential positron traps in an alloy containing the misfit regions is at the interface between the alloy matrix and a precipitate with a different crystal lattice [8]. In the present work, the  $\text{Al}_2\text{Cu}$  formation and its further growth is detected through the positron annihilations at the non-coherent precipitate-matrix interfaces.

On the other hand, the correlations between the results of the positron lifetime technique, which are obtained at various Cu concentrations in the Al matrix, and the results of other techniques such as SEM, X-ray and Vickers hardness are discussed.

## 2. Positron Annihilation in Materials:

The lifetime of a positron is directly related to the probability it will run into an electron. Positrons in materials have lifetimes that are governed by the electron density in the materials; the higher the electron density, the shorter the lifetime. The electron density determines the value of the bulk lifetime,  $\tau_B$ . Thus, positron spectroscopy is a sensitive probe of electron density in materials. If the defect density is lower than the bulk density, then the defects contain fewer electrons, so positrons in the defects have a longer lifetime. In addition, these defects have fewer nuclei, so they attract and trap positrons, which are repelled by the positive charge of nuclei. These characteristics have made positron spectroscopy a standard, well-established technique for defect detection and characterization in materials [9, 10].

Lifetime spectra may be resolved into two (or more) components with lifetimes  $\tau_i$  and corresponding intensities  $I_i$ . The component characterized by lifetime  $\tau_1$ , the shortest lifetime, and intensity  $I_1$  represents positrons annihilating in the matrix and dislocation loops. The component with  $\tau_2$ , the longer lifetime, and  $I_2$  characterizes positrons trapped and annihilating in three-

dimensional monovacancies and vacancy clusters. The size of the vacancy-like defects determines the value of  $\tau_2$  and whose concentration influences the intensity  $I_2$ . Theoretical calculations [11] have been done to establish a correlation between the positron lifetime in defects and the vacancy size of the clusters, which are assumed to be of spherical shape.

### **3. Experimental:**

Al – x wt.% Cu samples were prepared from 99.75 wt.% pure Al and 99.99 wt.% pure Cu. After homogenization in an electrical furnace at 850 °C, the ingot was cast into a graphite mould to produce rods of 25 mm length and 8 mm diameter.

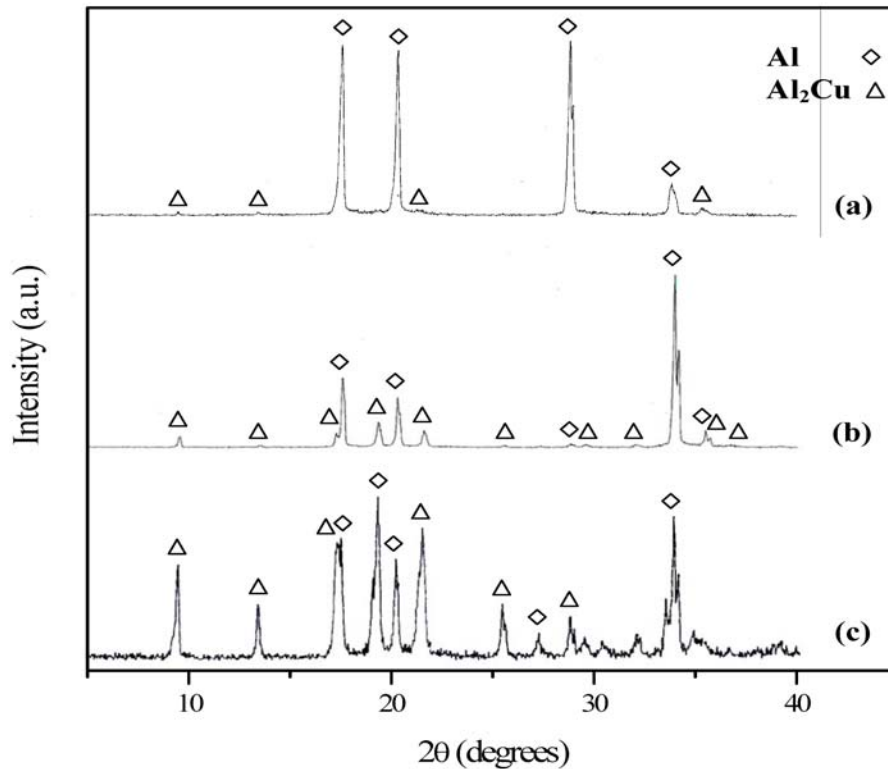
The positron source used in performing the positron annihilation measurements is a 20  $\mu$ Ci source of  $^{22}\text{NaCl}$ . The positron source is deposited on a kapton foil of a thickness of approximately 7 $\mu\text{m}$ , which is sandwiched between the two identical alloy samples. The fast-fast timing coincidence system is used in these measurements of the positron lifetime spectrometer using plastic scintillations, with a resolution (FWHM) of 266 ps. For more information about the positron lifetime spectrometer, reference [12] can be useful. All the spectra were about one million coincidences which are analyzed into the individual components by using the fitting software PATFIT [13]. The spectra are analyzed assuming one lifetime component.

The identity and sequence of  $\text{Al}_2\text{Cu}$  phase evolution with different Cu concentrations in Al-Cu alloys were studied by means of XRD and SEM techniques. XRD analysis were carried out using a Philips-PW 1710 vertical goniometer with a curved crystal monochromator using Mo-K $\alpha$  radiation ( $\lambda = 0.711 \text{ \AA}$ ) by scanning in the  $2\theta = 5 - 40^\circ$  range with  $0.5^\circ$  steps. For SEM examination, samples' surfaces were coated by a thin Au layer using fine coat JFC-100 E ion sputter Joel type for 10 min at 10 mA, the surface microstructure was studied by a Joel type JSM-1200 scanning electron microscope.

The hardness was measured using Vickers hardness instrument Leitz Welzlar Germany at a load of 5 Kg. Five to ten indentations have been taken for each data point.

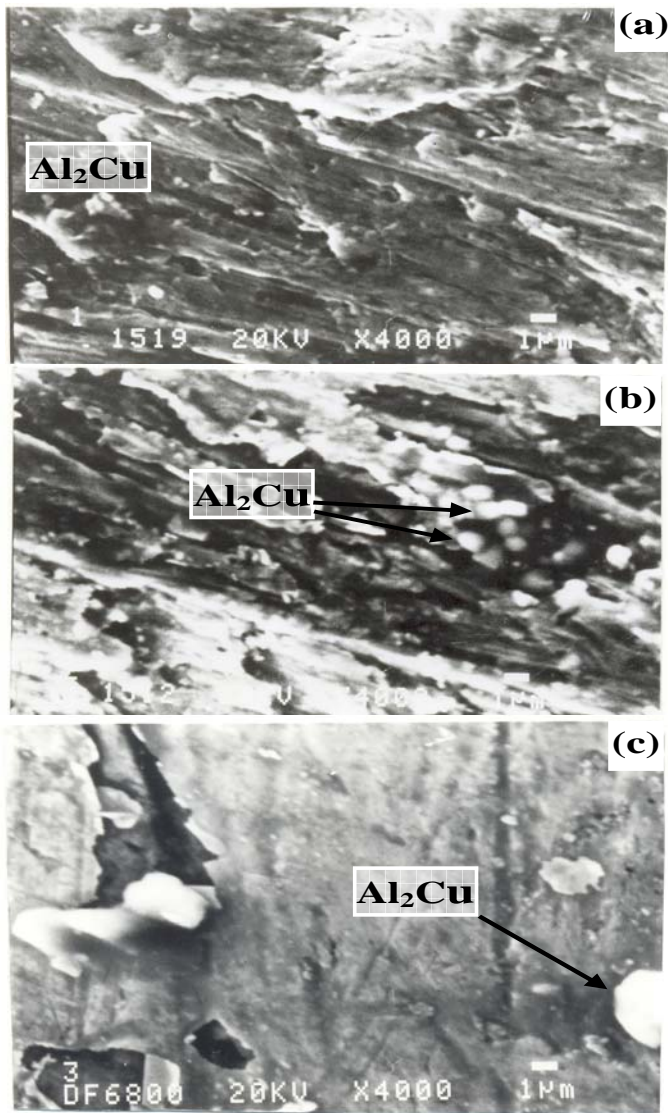
#### 4. Results and Discussion:

The XRD profiles of the as cast samples (before heat treatment) of composition Al-2.9 wt.% Cu, Al-12.3 wt.% Cu and Al-25 wt.% Cu are shown in Fig. (1) with indexed peaks. Corresponding XRD lines of Al and Al<sub>2</sub>Cu phases are detected. The profiles reveal the Al<sub>2</sub>Cu phase evolution in intensity between 2.9 wt. % Cu and 25 wt.% Cu. The Presence of these phases is confirmed by the subsequent SEM analysis.



**Figure (1):** X-ray diffraction patterns for as cast Al – x wt.% Cu: a) x = 2.9, b) x =12.3 and c) x =25.

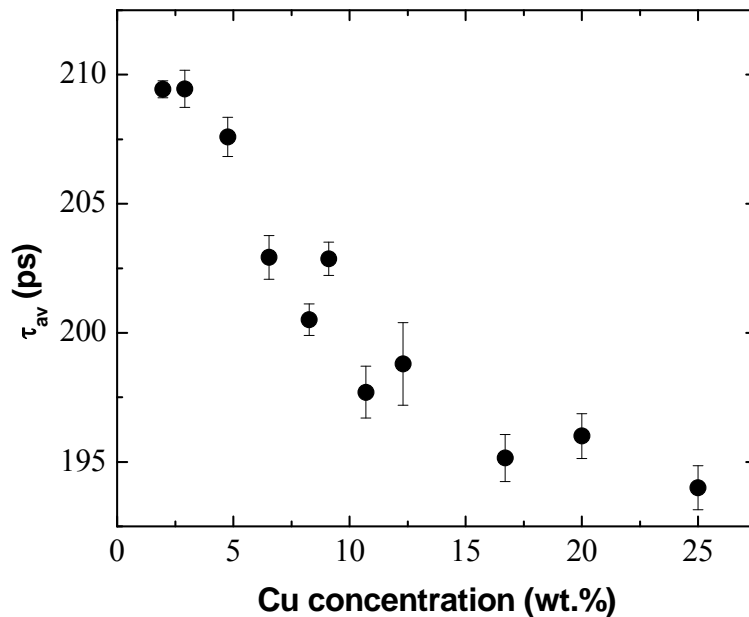
Scanning electron micrograph of Al-2.9 wt.% Cu, Al-12.3 wt. % Cu and Al-25 wt.% Cu are represented in Fig. (2). The Al<sub>2</sub>Cu phase is detected in the Al-matrix as very small particles in the range of 1 μm as shown in Fig. (2a). This particle size increases by increasing the wt.% Cu, the particle size reaches ~ 2 μm for the Al-25 wt.% Cu sample, as shown in Fig. (2c), which is still very small compared with the normal grain size of the Al matrix. Thus, Fig. (2) substantiates the results of XRD investigation in Fig. (1) that the Al<sub>2</sub>Cu phase is enlarged with the raise of the Cu content in the Al-Cu alloys.



**Figure (2):** Scanning electron micrograph of as cast Al – x wt.% Cu: a) x = 2.9, b) x =12.3 and c) x =25.

In order to detect the formation and further evolution of  $\text{Al}_2\text{Cu}$  precipitates, different Cu concentrations in the Al-Cu alloy were carried out in the range from 1.96 wt.% Cu to 25 wt.% Cu. Fig. (3) demonstrates the dependence of the average positron lifetime ( $\tau_{av}$ ) on the Cu concentration for Al-Cu alloy. The positron lifetime decreases with the increase of the Cu

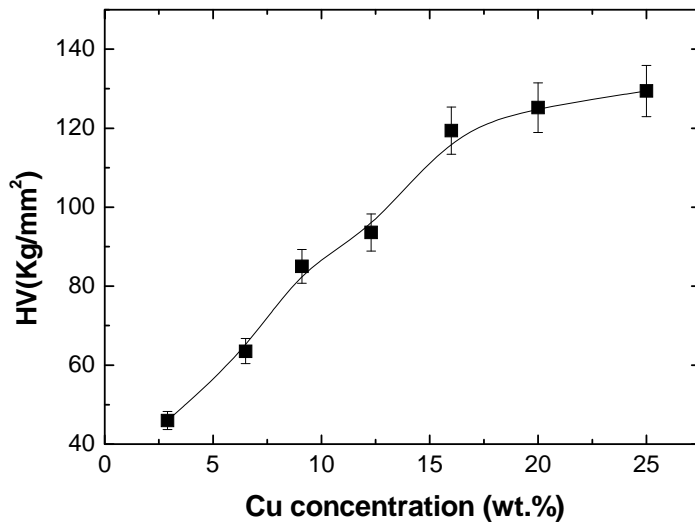
concentration [14, 15]. At low Cu concentration (1.96 wt.% Cu), the  $\tau_{av}$  value has about  $209 \pm 0.7$  Ps, which is in agreement with the values of 205 ps, 203 ps and 207 ps found by Krause *et al.* [16], Somoza *et al.* [17] and Cada *et al.* [18] respectively. The reduction of the positron lifetime was found up to 16.7 wt% Cu concentration. From 16.7 to 25 wt.% Cu concentration the value of  $\tau_{av}$  is almost constant and saturates at about 195 ps. This behavior may be understood as a formation of semi coherent  $\theta'$  ( $Al_2Cu$ ) particles and positron trapping at misfit dislocations of these particles [19]. This type of microstructure, with high number density of precipitates promotes intense positron trapping [8]. In this case, trapping occurs at misfit interfaces between the matrix and the precipitates. The decrease of  $\tau_{av}$  from about 209 ps at 2.91 wt.% Cu to about 195 ps at 16.7 wt.% Cu may be interpreted as an increase of the Cu content in the surroundings of the positron traps which are now the misfit dislocations of  $\theta'$  precipitates.



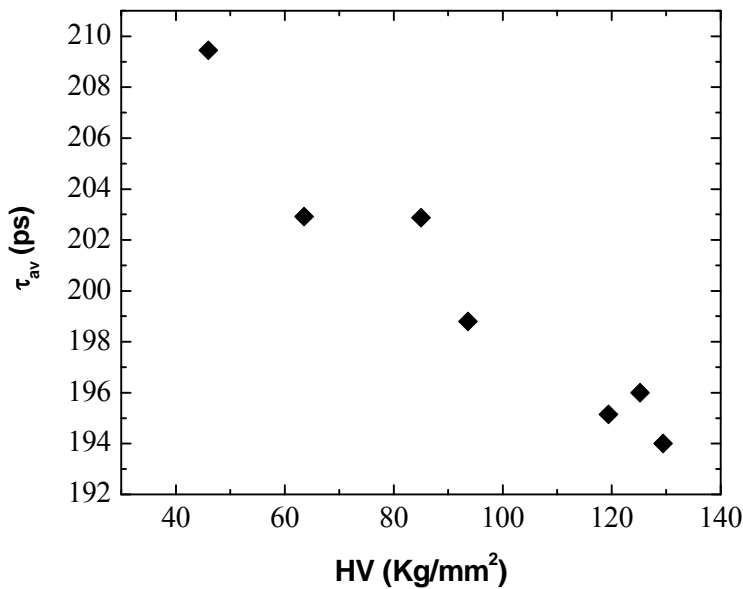
**Figure (3):** The average positron lifetime as a function of the Cu content for the as Cast Al-Cu alloy.

Vickers hardness measurements, shown in Fig. (4), have revealed an increase in the hardness with the Cu concentration [20] from 1.96 wt.% Cu up to 16.7 wt.% Cu, as a consequence of a well-dispersed distribution of semi-coherent metastable precipitates [8, 21]. In this first stage, the calculated rate of increasing of hardness was about 5.44 HV per 1 wt.% Cu added to the Al matrix. The observed hardness values from 16.7 wt.% Cu to 25 wt. % Cu exhibit little raise in the hardness curve compared with the previous part of the

curve. This trend can be considered as a sort of saturation in the hardness value. According to the above results, we may say, coarsening of  $\theta'$  precipitates results in the decrease of  $\tau_{av}$  [19]. This is highly consistent with the increase in Vickers hardness. Fig. (5) shows a correlation coefficient of 96.7 % between the  $\tau_{av}$  and the HV parameters.



**Figure (4):** The variation of Vickers hardness as a function of wt% Cu for as cast Al – x wt. % Cu, x=2.9 – 25. The line has been drawn as a guide for the eye.



**Figure (5):** The correlation between the average positron lifetime and the Vickers hardness.

## 5. Conclusions:

The present results emphasize the significance of PAL technique in investigating the atomic mismatch of the solute with the matrix, which leads to the formation of semi coherent  $\theta'$  ( $\text{Al}_2\text{Cu}$ ) precipitates in Al-Cu alloy. It is found that the misfit regions at the interface between the alloy matrix and the precipitates represent attractive sites for positrons. Lifetime of positrons inside these sites exhibits a decreasing trend from about 209 ps at 2.91 wt.% Cu to about 194 ps at 16.7 wt.% Cu showing saturation up to 25 wt. % Cu. This trend indicating more accumulation of the Cu atoms at the misfit dislocations of  $\theta'$  precipitates by increasing the Cu contents in the investigated alloy which is in agreement with XRD and SEM measurements. The increase of HV is consistent with the formation of clusters or solute-dislocation interactions. Accordingly, a correlation coefficient of 96.7 % between  $\tau_{\text{av}}$  and HV is obtained.

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## References

1. Andrey Z. Zhuk, Alexander E. Sheindlin, Boris V. Kleymenov, Eugene I. Shkolnikov and Marat Yu. Lopatin, *Journal of Power Sources*, **157**, 921 (2006).
2. R. Schülbe and U. Schmidt, *phys. stat. sol. (a)*, **103**, 29 (1987).
3. V. A. Phillips, *Acta Metall.* **21**, 219 (1973).
4. H. Arami and A. Simchi, *Mater. Sci. Eng. A.*, **464**, 225 (2007).
5. Hyukja Lee, Shankar Mall, Jeffrey H. Sanders, Shashi K. Sharma and Russell S. Magaziner, *Tribology International*, **40**, 1301 (2007).
6. Jerzy Dryzek and Ewa Dryzek, *Tribology International*, **40**, 1360 (2007).
7. N. de Diego, F. Plazaola, J.A. Jiménez, J. Serna and J. del Río, *Acta Materialia*, **53**, 163 (2005).
8. Dupasquier, G. Kögel and A. Somoza, *Acta Materialia*, **52**, 4707 (2004).
9. Y. Shirai, H. Araki, T. Mori, W. Nakamura and K. Sakaki, *Journal of Alloys and Compounds*, **330–332**, 125 (2002).
10. S. Dannefaer and D. Kerr, *Diamond and Related Materials*, **13**, 157 (2004).
11. P. Hautojärvi, J. Heiniö, M. Manninen, R. Nieminen, *Philosophical Magazine*, **35**, 973 (1977).
12. F. Becvar, J. Cizek and I. Prochazka, *Acta Phys. Pol. A*, **95**, 448 (1999).

13. P. Kirkegaard, M. Eldrup, O. E. Mogensen and N. J. Pederson, *Comput. Phys. Commun.*, **23**, 307 (1981).
14. Bharathi and C. S. Sunder, *Materials Science Forum.*, **105**, 905 (1992).
15. U. H. Gläser, G. Dlubek and R. Krause, *Materials Science Forum.* **105**, 1025 (1992).
16. R. Krause, G. Dlubek, G. Wendrock and O. Brümmer, in: Proc. of *European Meeting on Positron Studies of Defects*, Wernigerode PL7,1-22 (1987)
17. Somoza, A. Dupasquier, I. J. Polmear, P. Folegati and R. Ferragut, *Phys. Rev. B.*, **61**, 14464 (2000).
18. P. Cada, M. Cieslar, P. Vostry, F. Becvar, I. Novotny and I. Prochazka, *Acta Physica Polonica A.*, **88**, 111 (1995).
19. G. Dlubek, P. Lademann, H. Krause, S. Krause and R. Unger, *Scripta Materialia*, **39**, 893 (1998).
20. L. F. Mondolfo, "Aluminum alloys: Structure and Properties", part 2, Butterworths, (1976).
21. O. Novelo-Peralta, G. González, G.A. Lara-Rodríguez, Article in press, *Materials Characterization*, **59**, 773 (2008).