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**Research Article**

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# Non- Destructive Doppler Broadening Technique (NDDBT) to Study Defect Properties Wrought Aluminum Alloy (3004)

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**Abstract** Positron Annihilation Doppler Broadening Spectroscopy (PADBS) is non-destructive technique used in material science to detect mechanical working in the form of deformation effect. Plastic deformation basically produces dislocation defects by using hydraulic press at room temperature. To distinguish between valence and core electrons of wrought 3xxx Al-alloys, Doppler broadening line-shape (S and W parameters) have been measured. An exponential increase in the S-parameter accompanied with an exponential decrease in the W-parameter with increasing the degree of deformation was observed before reaching saturation for both of them. The positron parameters were determined (S-and-W parameters, FWHM, Trapping rate, trapping efficiency, trapping coefficient, dislocation density, and defect density). On the other hand some microstructural parameters (e.g mean crystallite size, micro strain, flow stress and stored dislocation energy) were determined by Using DBT which had a good correlation with positron annihilation lifetime technique (PALT) estimated parameters.

**Keywords** Non- Destructive Doppler Broadening Technique, NDDBT, Defect, Wrought Aluminum Alloy (3004)

## 1. Introduction

Aluminum and its alloys are widely used in aircraft automotive and in construction industries because of their desirable physical properties. There are many kinds of crystalline defects that can cause a significant improving in the properties of materials, including interstitial point defects and vacancy in addition to dislocation line defects [1]. Aluminum alloys grouped into wrought and cast alloys. Wrought alloys which mechanically worked to final shape consist of 4 digits based on major alloying elements, while cast alloys compositions are optimized for casting to final shaped. The non-heat treatable aluminum-manganese series (3*xxx*) as FCC materials cannot be strengthened by precipitation hardening; these alloys are primarily hardened by cold work (plastic deformation due to compression). Plastic deformation is a permanent shape change that is irreversible even after removing the applied load. It occurs in metals and alloys when they are subjected to loads exceed the energy required to bring dislocations movement. Selection of structural materials involves compromise among ductility and strength since high ductility materials normally have low strength, and vice versa [2]. Commonly, BCC metals offer higher strength while FCC metals offer higher ductility. A highly desirable feature for structural materials is to become stronger when deformed by cold work up to a certain degree of deformation. The energy expended in plastic deformation of a metal is mainly divided into two parts, one part is converted into heat depending on the type of the loading and on the degree of deformation and the other part is stored inside the defects in the form of a strain energy [3]; means the metal becomes a battery of energy. The stored energy during cold work was experimentally determined by many authors [4–6].

Positron annihilation spectroscopy (PAS) technique emerged as the most effective technique in point defect studies because of its high resolution and nondestructive nature. PAS takes advantage of the fact that positively

charged positrons tend to localize in open volume regions (*e.g.*, free volume, vacancies, dislocations, voids, etc.), where there are missing positively charged atomic nuclei [7]. Furthermore when a positron and an electron annihilate, the resulting gamma rays yield information about the annihilation site, making PAS a sensitive probe for vacancies. Studying the positron annihilation process characteristics can detect the state of electrons in metallic substances [8].
In order to observe the evolution of defects and to monitor the damage process directly, the positron annihilation Doppler Broadening technique (DBT) has been used. Upon trapping, a positron annihilates with an electron into two 511 KeV photons. The detection of the slight differences in energy of the 0.511 MeV annihilation quanta due to the energy and momentum of the electron in the positron-electron annihilation is the basis of Doppler broadening measurements. A distribution spectrum of the number of counts versus annihilation quanta energy in a given material is collected, resulting in a nearly Gaussian-looking peak at 0.511 MeV as shown in blow. In an annihilation spectrum these shifts lead to a broadening of the 511 keV photo peaks which is quantified by the S- and W-parameters [9]. S- And W- parameters reflect annihilations with valence and core electrons, respectively, therefore its necessary to monitor the generation and evolution of deformation induced defects and the chemical environment at the positron trapping sites. S-parameter is the ratio of the area under the central part of the peak to the total area and W-parameter being the ratio between the area under the wings of the peak and the total area. A material with a higher S value will have more defects than a material with a lower S value [10].

## 2. Experimental Procedures

Al-alloy 3004 with dimensions (12 x 12 x 3) $mm^3$ and chemical composition given in table (1) were cleaned, chemically etched, homogenized for 6 hours at 723 K in a non-vacuum furnace, and then annealed to room temperature. These prepared samples were deformed at room temperature up to 25% of their original thickness using a hydraulic press. The system which has been used in the present work to determine the Doppler broadening line-shape is consists of an Ortec HPGe detector with an energy resolution of 1.95 keV for 1.33 MeV line of $^{60}$Co, an Ortec 5 kV bias supply 659,Ortec amplifier 575 and trump 8 k MCA. Figure (1), shows a schematic diagram of the experimental arrangement. Doppler broadening is caused by the distribution of the velocity of the annihilating electrons in the directions of gamma ray emission. The signal coming from the detector enters the input of the preamplifier and the output from the preamplifier is fed to the amplifier. The input signal is a negative signal. The output signal from the amplifier is fed to a computerized MCA.
The positron source of 1μCi free-carrier $^{22}$NaCl, was evaporated from an aqueous solution of sodium chloride and deposited on a thin Kapton foil of 7.5 μm in thickness. Radioactive material can be deposited directly on the samples or separation foils can be used to allow using the source repeatedly. The source can also consist of a single radioactive metal foil. The source has to be very thin so that only small fractions of the positron annihilate in the source. A sandwich configuration has been used to guide the positron into the deformed 3004 Al samples. The source-samples configuration was then wrapped in a thin aluminum foil. Each sample spectrum was measured for 1800 seconds.

**Table 1:** represents chemical composition of3004 Al-alloy

| Element | Si | Fe | Cu | Mn | Mg | Zn | others | Al |
|---|---|---|---|---|---|---|---|---|
| Wt % | 0.3 | 0.7 | 0.25 | 1.0-1.5 | 0.8-1.3 | 0.25 | 0.15 | reminder |

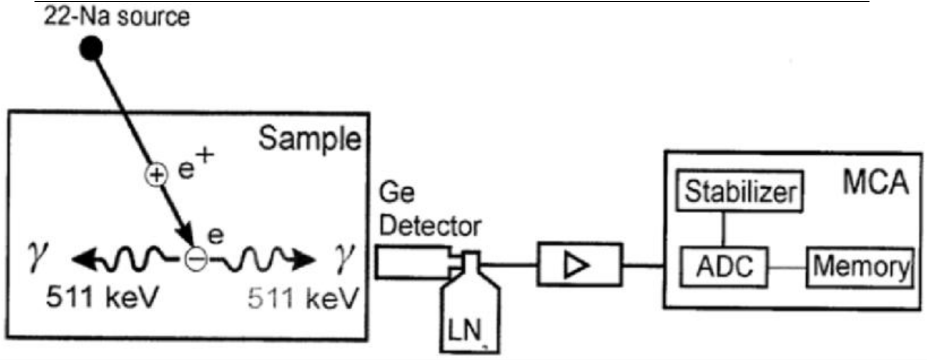


*Figure 1: Block diagram of HPGe-detector and electronics fo rDoppler broadening line shape measurements*

## 3 Results and Discussion

### 3.1. Estimation of the Positron Parameters by DBT

(S-and-W parameters, FWHM, Trapping rate, trapping efficiency, trapping coefficient dislocation density, defect density).

The Doppler broadening line shape parameters were measured for the investigated samples of the 3004 wrought Al alloy. Four spectra of the considered samples at 511 keV of annihilation at different deformations are shown in figure (2). This figure shows that the measured line shape profile of higher deformed samples reveal a little higher line shape counts than non-deformed sample. The Full Width at Half Maximum (FWHM) of the measured spectra is nearly the same for both samples with higher deformation, while it is a little more broadened for samples at lower deformation as represented in figure (3).

The SP software program was designed for the analysis of the annihilation line. The SP program is quite versatile. It finds the S- and W-parameters automatically subtracting the background [11]. It can fit the Gaussian curve to a spectrum.

A location of the maximum of the Gaussian curve defines the 511 keV positions, which is important for finding the integration ranges for S- and W-parameters.

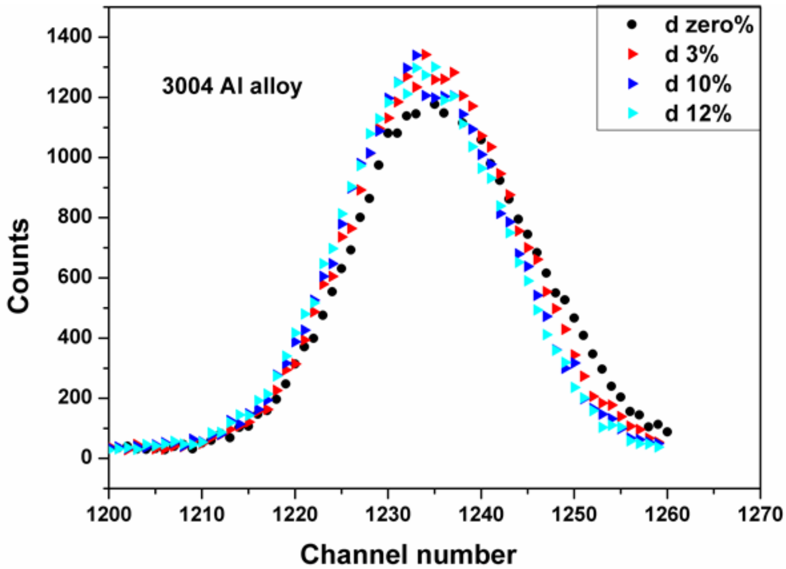


*Figure (2.a): Spectra of 3004 Al alloy at different degree of deformation*

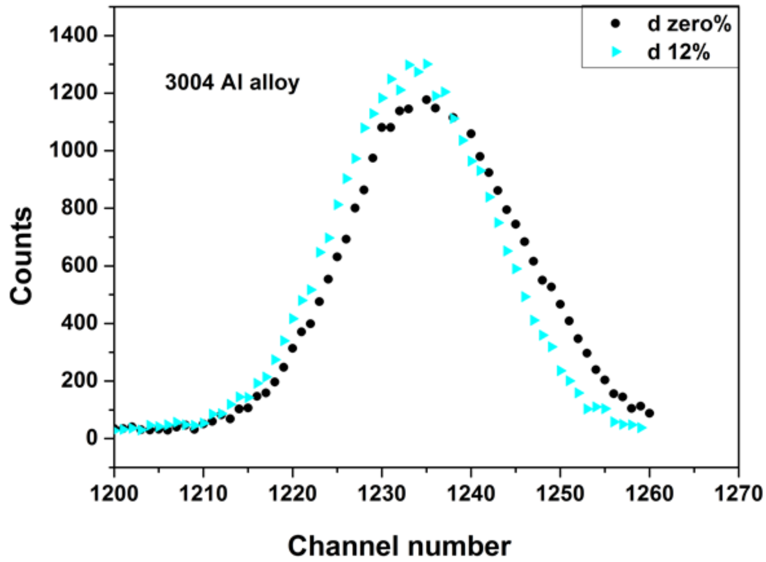


*Figure (2.b): Spectra of 3004 Al alloy at different degree of deformation*

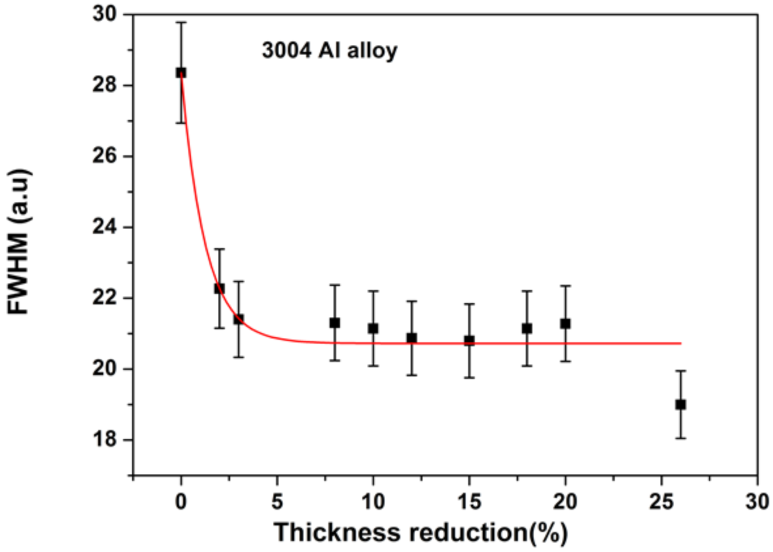


*Figure (3): FWHM versus thickness reduction at different degree of deformation of 3004 Al alloy. (Note that (a.u) means arbitrary unit)*

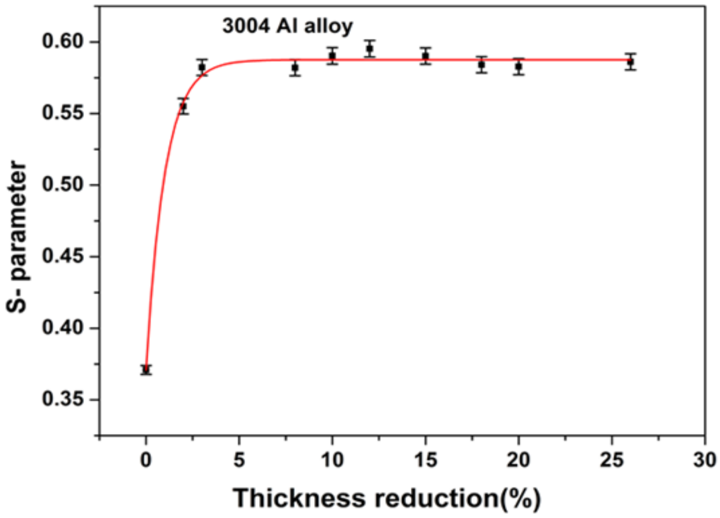


*Figure 4: S-parameter of3004Al alloy versus thickness reduction*

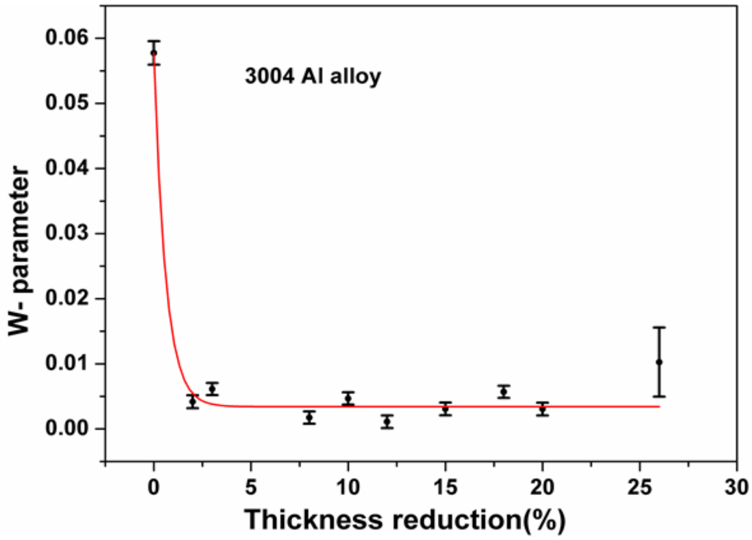


*Figure 5: W-parameter of 3004 Al alloy versus thickness reduction*

In this work, the change of the positron annihilation line-shape S- and W- parameters were determined. The relation between the experimentally measured S- and W- parameters (data points) and the degree of deformation is shown in figures (4, 5). The S-parameter value of the undeformed (annealed) sample is $S_f$(0.3709$\pm$0.0031) after which, an increase in the S-parameter, values were observed with increasing thickness reduction until 3% degree of deformation is reached. Above 3% thickness reduction, the obtained results of S-parameter are approximately kept constant in the region for saturation trapping of positron in defect states. S-parameter value of about $S_t$(0.5885$\pm$0.0057) was measured for saturated dislocation samples. On the other hand, an exponential increase in the S-parameter accompanied with an exponential decrease in the W-parameter with increasing the degree of deformation.

An abrupt increase in the S-parameter value (0.3709±0.0031) associated with an abrupt decrease in the W-parameter value (0.5885±0.0057) was obtained at zero deformation until saturation is produced at 5% deformation as shown in figure (7).Plots of (S versus W) can be used for defect identification [12]. If only a single type of vacancy is present, the relation between these parameters is linear [12]. A straight line connects the defect state (higher S-parameter values) and the bulk (lower S-parameter values) as shown in figure (6).

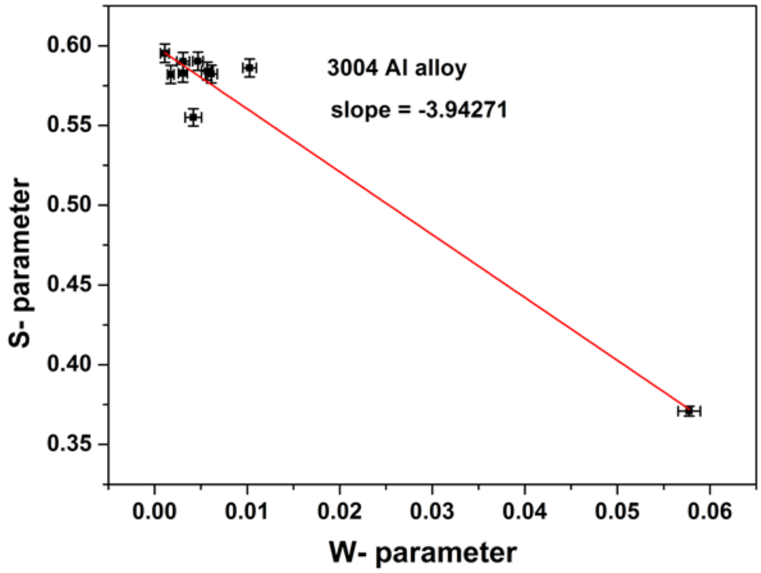


*Figure 6: S-parameter versus W-parameter of 3004 Al alloy*

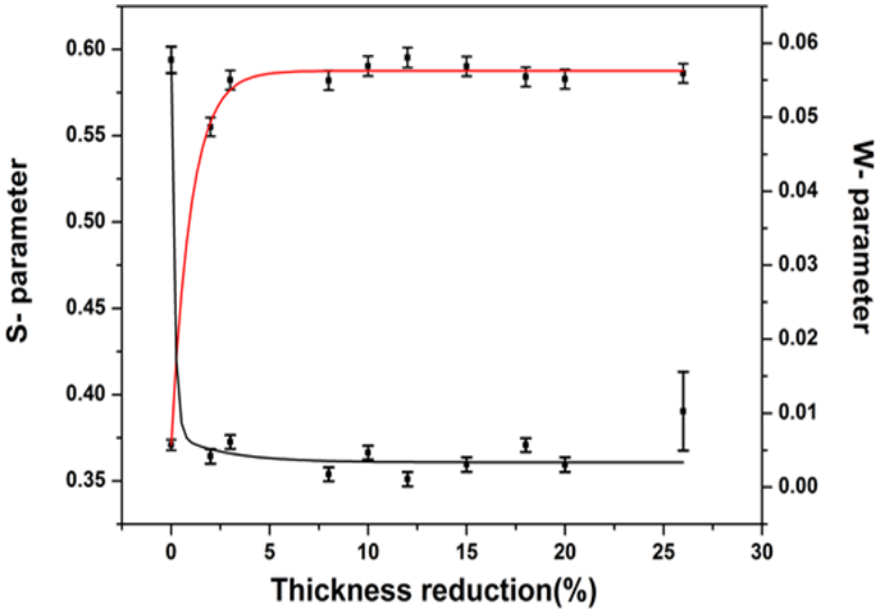


*Figure 7: S-and-W parameters variations versus thickness reduction*

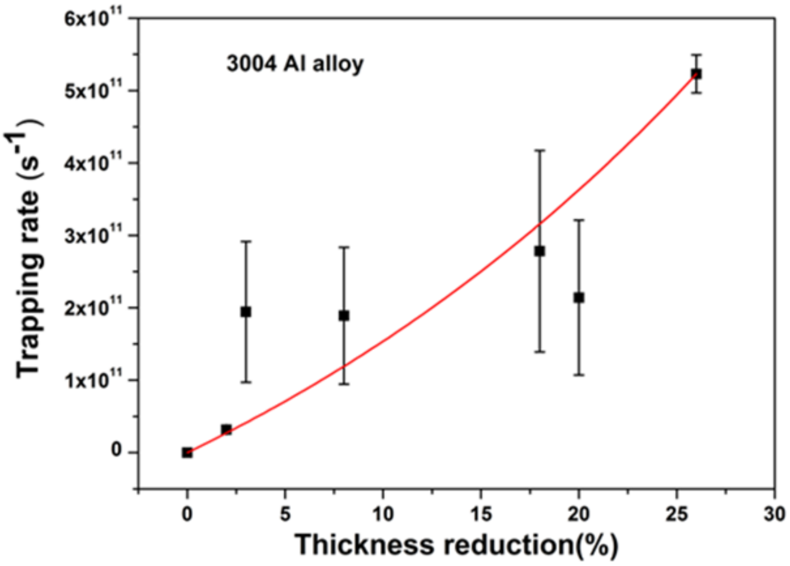


*Figure 8: Trapping rate variation versus thickness reduction of 3004 Al alloy*

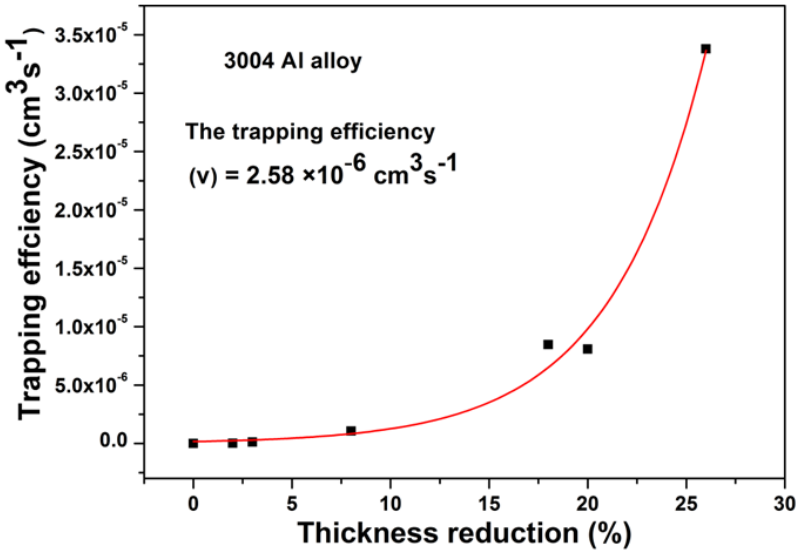


*Figure 9: Trapping efficiency versus thickness reduction of 3004 Al alloy by DBT*

The results of the Doppler broadening S-parameter were analyzed in terms of the simplest case of trapping model. The simplest case of the trapping model, including only one open-volume defect type can be used to determine the positron trapping rate ($k_d$). Using values of theS, $S_b$ (free) and $S_d$ (trapped) parameters measured from figure (4) the trapping rate can be calculated from the following equation [13]:

$$K_d = \frac{1}{\tau_b}\frac{(S-S_b)}{(S_d-S)} \quad (1)$$

Where ($\tau_b$=173 ps) is the bulk (free) lifetime of 3004Al alloy measured by PALT [14]. The positron trapping rate$k_d$ values of equation (1) was determined using equation (2):

$$k_d \ = \ 1.248 \ \times \ 10^{-3}[\log\,(1-R)]^2\,\frac{\nu}{b^3} \quad (2)$$

Where R is the fractional thickness reduction and b is Burger's vector of 3004 Al alloy that is estimated to be equal to 2.8598Å. Since the trapping efficiency (ν) is related to the trapping cross-section (σ) according to equation (3):

$$\nu = \sigma V \quad (3)$$

Where $V$ is the mean thermal velocity of the thermalized positron and from the Maxwell distribution one obtain that:

$$V=\sqrt{\frac{8K_bT}{\pi m}} \quad (4)$$

Where $k_b$, T and m are the Boltzmann constant, temperature and mass of the positron respectively. The positron trapping efficiency ($\nu$) value which gives the best fit of the experimental measured data point of figure (4) after substituting in the positron trapping rate equation (1) was determined by (DBT) to be $2.58 \times 10^{-6} cm^3 s^{-1}$. On the other hand, the trapping efficiency was determined by PALT in the previous work to be $2 \times 10^{-6} cm^3 s^{-1}$ [14].

The trapping rate $k_d$ is proportional to defect density ($\rho^{-}$); number of trapping sites per unit volume; as shown in the equation (5) [15]:

$$k_d = \nu\, \rho^{-} \quad (5)$$

The defect density is related to dislocation density $\rho(cm^{-2})$; length per unit volume; according to equation (6) [15].

$$\rho^{-}(cm^{-3}) = \frac{\rho(cm^{-2})}{b} \quad (6)$$

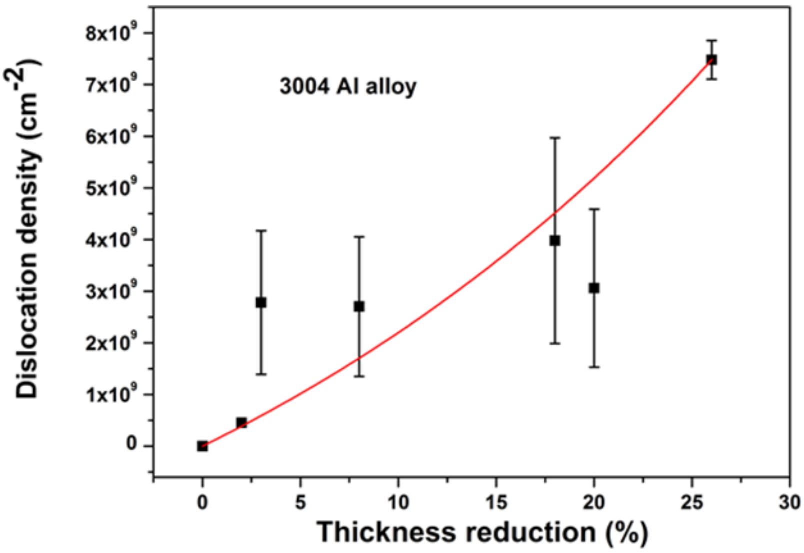


*Figure 10: Dislocation density of 3004 Al alloy versus thickness reduction*

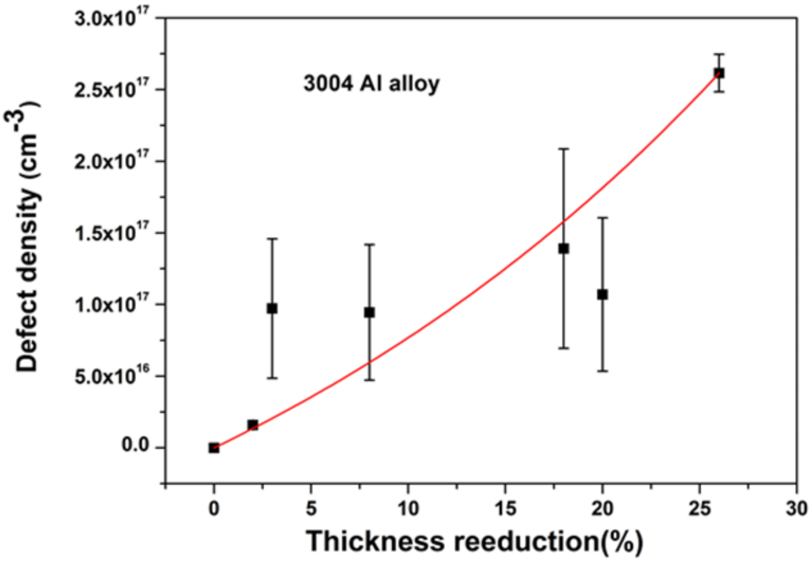


*Figure 11: Defect density of 3004 Al alloy versus thickness reduction*

The trapping coefficient ($\mu$) can be calculated by substituting equation (6) into equation (5) [15]:

$$k_d(s^{-1}) = \frac{\nu}{b}\rho(cm^{-2}) \quad (7)$$

A linear relationship for the dislocation density (ρ) as a function of the positron trapping rate ($k_d$) is depicted in figure (13).

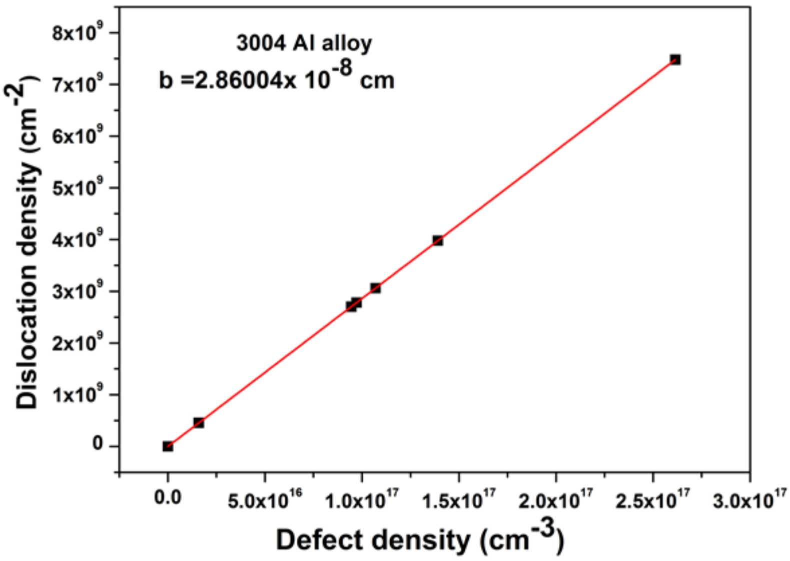


*Figure 12: Dislocation density versus dislocation density of 3004 Al alloy*

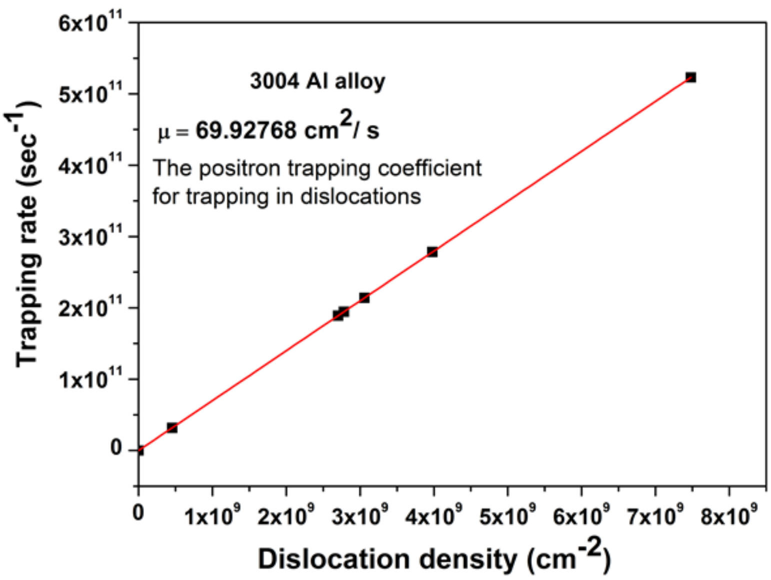


*Figure 13: Trapping rate as function of dislocation density of 3004 Al alloy*

The positron trapping coefficient which is the ratio between the positron trapping efficiency and the Burgers vector of the 3004Al-alloy can be determined by DBT from above figure as $\mu$ = 69.93($cm^2 s^{-1}$) but from the previous work by PALT was determined to be $\mu$ =68.58($cm^2 s^{-1}$) [14].

**3.2. Estimation of Some microstructural parameters by using DBT for 3004 Wrought Aluminum alloys** (e.g mean crystallite size and microstrain, flow stress and dislocation stored energy).

Positron Annihilation Spectroscopy (PAS) can also be used in determination of physical characteristics of alloys such as grain size. The positron annihilation parameters are found to be sensitive function of grain size [13] as shown figure (14).

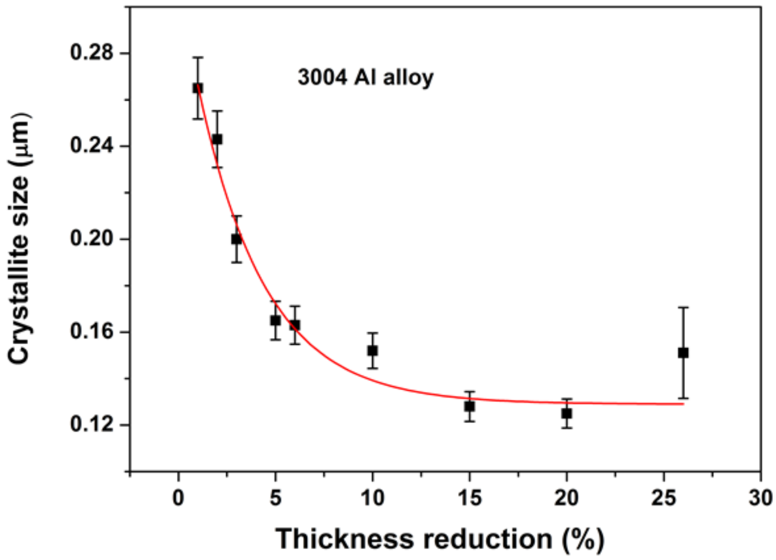


*Figure 14: The crystallite size versus thickness reduction of 3004 Al alloy by PALT*

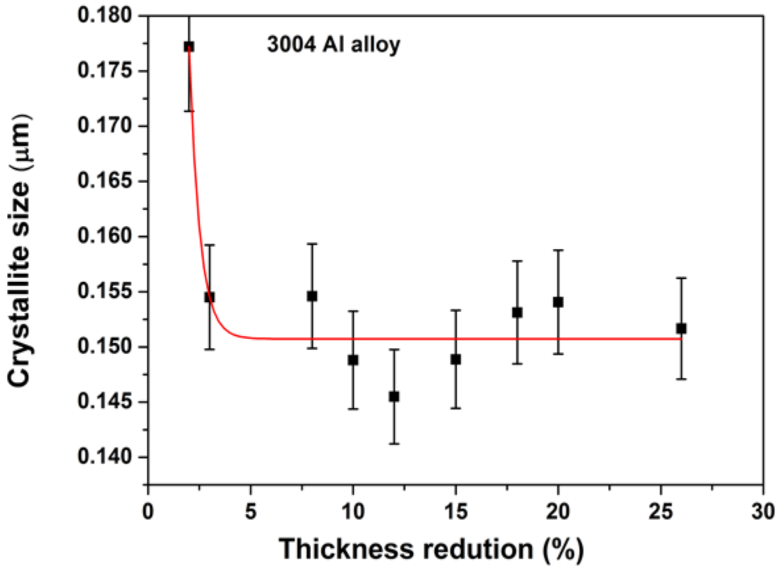


*Figure 15: The crystallite size versus thickness reduction of 3004Al alloy by DBT*

The grainsize or crystallite size (G) and the mean life time (τ) are related to the bulk (free) lifetime ($\tau_b$) and the defect (trapped) lifetime ($\tau_d$) as [13]:

$$\tau = \tau_b + [(\tau_d - \tau_b)\frac{L_+}{G}] \quad (8)$$

Using S, $S_b$ (free) and $S_d$ instead of $\tau$, $\tau_b$ (free) and $\tau_d$ respectively, one can evaluate the grain size using S-parameter values. Equation (8) is written in this case as [13]:

$$S = S_b + [(S_d - S_b)\frac{L_+}{G}] \quad (9)$$

Where $L_+$ is the positron diffusion length, $L_+$ is limited due to the finite lifetime of positrons in the defect-free bulk ($\tau_b$) as [13]:

$$L_+ = \sqrt{\tau_b D_+} \quad (10)$$

Where ($D_+$) is the positron diffusion coefficient.

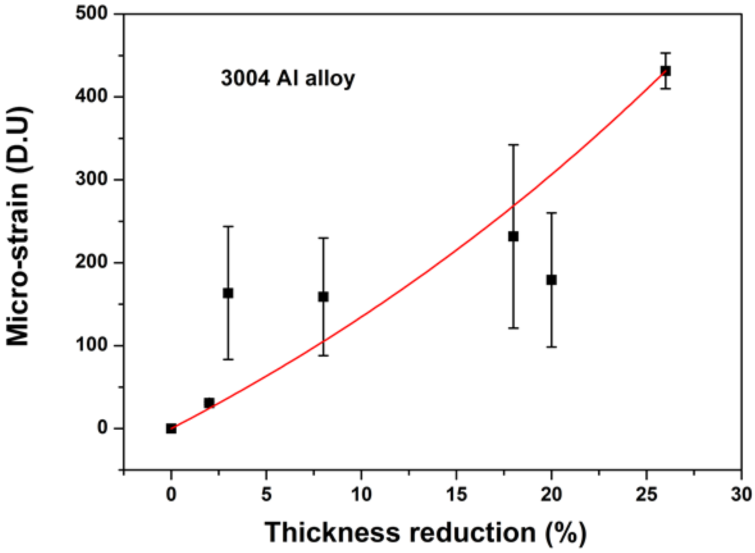


*Figure 16: The micro-strain versus thickness reduction of 3004Al alloy by DBT (Note that: D.U, means dimensionless unit)*

The residual micro-strain$< \varepsilon^2 >^{1/2}$is given by this relation [16]:

$$< \varepsilon^2 >^{1/2} = \frac{\rho\ G\ b}{3\sqrt{2\pi}} \quad (11)$$

Where$\rho$ is the dislocation density, $G$ is the mean crystallite size, b is the burger vector of FCC material 3004 Al alloy.

Generally, the flow stress in terms of the resolved shear stress ($\tau$) relates to the square root of dislocation density as follows [16]:

$$\tau = \tau_0 + \alpha\ Gb\sqrt{\rho} \quad (12)$$

Where $\tau_0$ is the friction stress, $\alpha = 0.5$ is a constant, G=26 GPa is the shear modulus of Al, and b is Burgers vector. In terms of tensile stress, by taking the friction stress equal to zero and $\sigma$ = M $\tau$, the flow stress ($\sigma$) isestimated for materials by the relationship [16]:

$$\sigma = M\alpha\ Gb\sqrt{\rho} \quad (13)$$

Where (M)is average Taylor factor. Then the above relation is given by [16]:

$$\tau = \alpha\ Gb\sqrt{\rho} \quad (14)$$

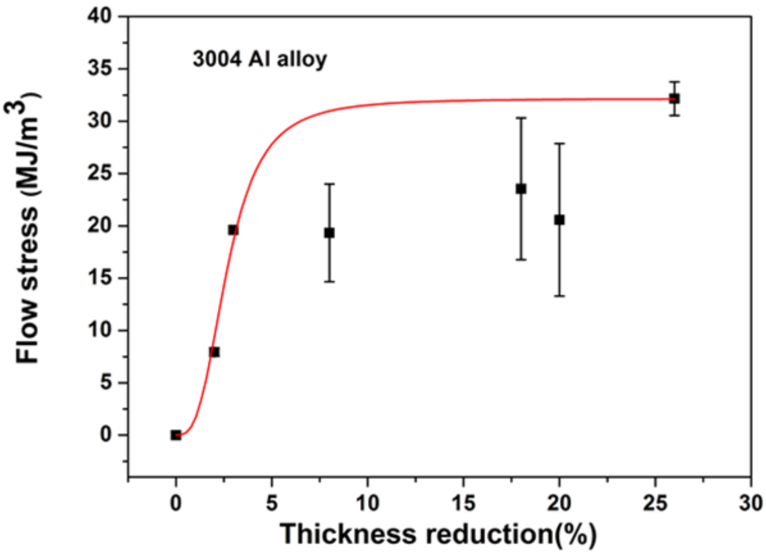


*Figure 17: The flow stress versus thickness reduction of 3004 Al alloy by DBT*

### 3.3 Estimation of the Stored Dislocation Energy

Stored energy is related to the amount of cold work (plastic deformation) acquired by the material [17]. A plastically deformed metal or alloy contains normally large stored dislocation energy and with annealing at higher temperatures and during recovery and recrystallization, it will typically return to a lower energy state by structural growth [14].The stored dislocation energy (E) due to the generation of crystalline defects can be determined on the basis of the dislocation theory. The dislocation density $\rho$ is related to the dislocation stored energy (E) as [14]:

$$E = \alpha\, G b^2 \rho \quad (15)$$

Where $\alpha$=0.5 is the dislocation interaction parameter, G is the bulk modulus of Al-alloy (G = 26 GPa) and b is the burger vector (b = 2.86 Å).

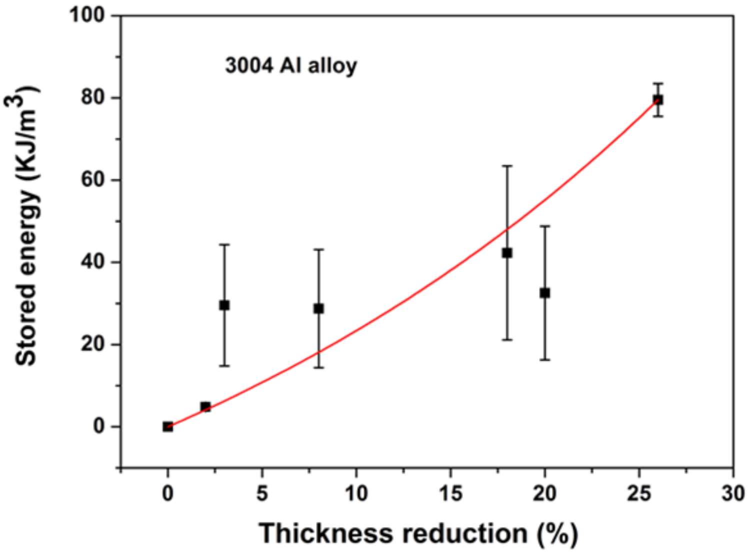


*Figure 18: The stored energy versus thickness reduction of 3004 Al alloy*

### 4. Conclusion

According to the results obtained from the measurements of the positron annihilation Doppler on 3004 wrought aluminum alloy from the above figures, the following conclusions can be made:

- Doppler broadening is a sensitive nondestructive nuclear technique to detect he degree of deformation of 3004 Al alloy.
- S- Parameter showed an exponential increase, while W- parameter decreased exponentially with increasing the degree of deformation before reaching saturation where, The S-parameter value of the undeformed (annealed) sample was$S_f$ (0.3709$\pm$0.0031) and $S_t$ (0.5885$\pm$0.0057) was measured for saturated dislocation samples.
- The Full Width at Half Maximum (FWHM) of the measured spectra is nearly the same for both samples with higher deformation, while it is a little more broadened for samples at lower deformation.
- The trapping rate of defects of 3004 Al alloy increases from $3.2\times10^{10}$ $s^{-1}$for nondeformed sample to$5.23\times10^{11}$at 26% deformation by DBT but in the previous work by PALT was found about $7.7\text{x}10^{10}$ $s^{-1}$ at saturation dislocation region of 6 % thickness reduction [14].
- The positron trapping efficiency ($\nu$) was determined by (DBT) to be $2.58\times10^{-6}cm^3s^{-1}$. On the other hand, the trapping efficiency was determined by PALT in the previous work to be $2\times10^{-6}cm^3s^{-1}$ [14].
- The range of the defect density (concentration of defects) changes from $10^{16}$ to $10^{17}$ $cm^{-3}$ but the range of the dislocation density changes from $10^8$ to $10^9$ $cm^{-3}$.
- The trapping coefficient ($\mu$) was calculated by (DBT) as$\mu$ = 69.93$(cm^2s^{-1})$ but from the previous work by PALT was determined to be $\mu$ =68.58$(cm^2s^{-1})$ [14].

- The grain size or crystallite size (G) was determined to be in the range from 0.178($\mu m$) to 0.152($\mu m$) by DBT and 0.2($\mu m$) to 0.151($\mu m$) by PALT in the previous work [14].
- The residual micro-strain changes from 30.71 to 431.39 by DBT.
- The flow stress changes from (7.93 to 32.15) MJ/$m^3$ by DBT.
- The stored dislocation energy varies from (4.84 to 79.528) KJ/$m^3$ by DBT.

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