Application of the Positron Lifetime Spectroscopy to the Study of Microstructural Defects in Steel

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Positron annihilation lifetime technique has been applied to study the microstructural changes in steel after heat treatment (tempering process). Positron lifetime spectra for the tempered samples in sandwich configuration with the positron source were accumulated at room temperature using a conventional fast-fast coincidence timing system. The measured lifetime spectra have been analyzed. A correlation between the macroscopic mechanical properties and the positron annihilation parameters has been established. Meanwhile, with the aid of image analysis method, the American Standards for Tools and Materials (ASTM) grain size in the tempered samples was determined. The two state trapping model has been applied for interpretation of the experimental results.

1. Introduction:

Investigation of the damaged state of a material is very important for industrial applications. Most mechanical damages start with a change in the microstructure of the material [1,2]. The physical basis for the use of positrons in defect studies is the fact that positrons injected into a material may get trapped at defects that represent regions where the atomic density is lower than the average density in the bulk i.e vacancies, vacancy clusters and dislocations. Since positrons and electrons are antiparticles an injected positron will annihilate with an electron of the material. As a result, gamma-rays will be emitted. These γ- quanta carry information about the state of the positron before annihilation and by proper measurement of the emitted γ- quanta it is therefore possible to obtain useful information about those defects that have trapped the positrons. Positron annihilation lifetime (PAL) spectroscopy has been extensively used for the investigation of vacancy- type defects in solids [3-6]. Thermalised positrons can be trapped in defects of open volume like dislocations, vacancies or microvoids [7]. The trapping rate is characteristic of the different types of defects. The most sensitive parameter in the positron lifetime spectroscopy is the positron mean lifetime that can be considered as an integral value of the defect density. Compared to other methods of defect characterization, positron annihilation lifetime spectroscopy is nondestructive and does not require special sample preparation. The sensitivity of positron annihilation spectroscopy (PAS) to vacancies and vacancy clusters has previously been applied to provide valuable information about the microstructure of damaged steels by mechanical stresses {tensile strain [8], creeping [9], Fatigue [10]}. In addition , by means of PAS the microstructure of heat- treated steel by high-powered laser beam [11,12] and by heat treated in various manners [13] was studied.

The aim of this work is to investigate the influence of heat treatment (tempering process) on the mechanical properties and microstructure examination of the tempered steel. Also, an attempt is done to establish a correlation between the macroscopic mechanical properties and the positron annihilation parameters.

2. Experimental:

The chemical composition of steel is given in Table (1). The thermal treatment of the steel samples was as follows: a) heating at 970 ^{o}C in an electric furnace for 30 minutes, b) quenching in water at room temperature (hardening process), c) reheating at 300, 400, 500, 600, and 700 ^{o}C for 30 minutes followed by air cooling (tempering process). Samples for positron annihilation are shaped as discs of 20 mm diameter and 2 mm thick.

Table (1): Basic chemical composition of steel in wt %

Fe	C	Si	Mn	P	S	V
98.753	0.3	0.12	0.79	0.024	0.004	0.009

The positron source was prepared by depositing about 20μCi of aqueous ^{22}NaCl on a thin kapton foil (7 μm thick). ^{22}NaCl spots were dried and covered with another similar foil glued together by epoxy glue and evacuated for a long time (more than 24 h) to be sure that no air between the two foils. The positron annihilation lifetime measurements were achieved by using a standard fast-fast coincidence timing technique. In this technique, two detectors are placed close to the source-sample sandwich to detect the emission (signaled by 1.28 MeV γ- quantum) and the annihilation (signaled by a 0.511 MeV γ- quantum) of the positrons, thus making it possible to obtain their lifetime, i.e. the time difference between the two signals. To calculate the time resolution of the system, the positron annihilation lifetime spectrum of the kapton sample has been measured. The kapton seems to be the only polymer with no positron yield for the long-lived component. The time resolution was calculated using RESOLUTION program [14] and is found to be 240 ps (full width at half maximum, FWHM). The sample/positron source/sample sandwich was put in a glass tube in order to perform the PAL measurements in vacuum at room temperature (about 25 ^oC). The accumulation time (three hours) provided excellent counting statistics namely 10^6 counts in the peak channel. In the present work, the lifetime spectra were analyzed to finite term lifetimes in terms of two lifetimes components τ_1 with concentration I_1 and τ_2 with concentration I_2 using the PATFIT program [14] with source correction. The shortest lifetime τ_1 is attributed to annihilation in the prefect lattice while the longer lifetime τ_2 is then due to positrons trapped in defects. Also, the hardness measurements are carried out at room temperature with the microhardness tester (ZEISS, Model mph 160). For microscopic examination, scanning electron microscope (JEOL, Model JSM- T200) at the central research lab. in El-Minia university was used.

3. Results and discussions:

The lifetime spectra collected for the steel specimens have been analyzed in terms of two lifetime components τ_1 and τ_2 with relative intensities I_1 and I_2. The variation in lifetime values is relevant to defect type while changes in the intensities reflect defect concentration. The positron annihilation lifetime in bulk (τ_b) and in defect (τ_d) can be determined from [15];

$$\lambda_b = 1 / \tau_b = I_1 \tau_1^{-1} + I_2 \tau_2^{-1} \quad \text{and} \quad \lambda_d = 1/ \tau_d = \tau_2^{-1}, \qquad (1)$$

where λ_b and λ_d are the decay rate of positron from bulk and defect, respectively. Meanwhile, the positron mean lifetime, τ_m can be calculated using the formula [15];

$$\tau_m = (\tau_1 I_1 + \tau_2 I_2). \qquad (2)$$

The relationship between the positron annihilation parameters (τ_2, I_2 & τ_m) and the tempering temperature is shown in Fig. (1). As seen in Fig. (1) the longest lifetime τ_2 decreases as the tempering temperature increases with an average value of 410 ps which is equivalent to a cluster of roughly 10 vacancies [3]. The noticeable increase in the positron mean lifetime τ_m is due to positron trapping at dislocations and small vacancy clusters as well.

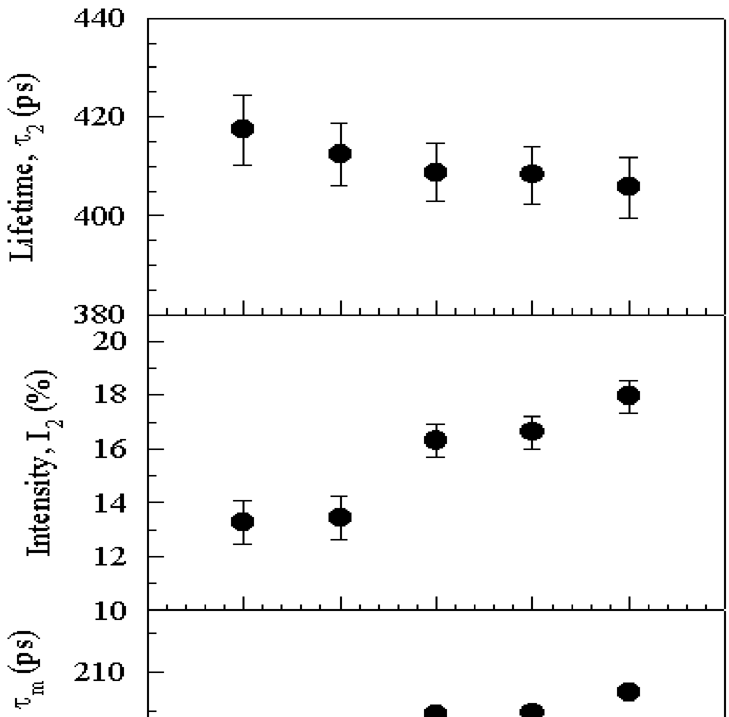


Fig. (1): Positron annihilation parameters τ_2, I_2, and τ_m as a function of tempering temperature.

A quantitative analysis of annihilation characteristics in terms of positron trapping rates and defects concentration is made using the so-called two state trapping model [7,16]. The model is based on the assumption that there are one or more types of (homogeneously distributed) defects that trap the positrons. According to this model, the following relations can be obtained, which relate the measured I_1, τ_1, I_2, and τ_2 with the total trapping rate $K = \sigma c_v$, where σ is the specific trapping rate and c_v is the defect concentration;

$$I_1 = \frac{\tau_b - \tau_d}{\tau_d (1 + \tau_b K) - \tau_b}, \tag{3}$$

$$I_2 = \frac{\tau_b \tau_d K}{\tau_d (1 + \tau_b K) - \tau_b}, \tag{4}$$

$$\tau_1 = \frac{1}{\tau_b^{-1} + K}, \tag{5}$$

Solving the equations (1-5) then;

$$K = \frac{\tau_m - \tau_b}{\tau_b(\tau_d - \tau_m)}, \tag{6}$$

which can be deduced from the experimental values I_1, τ_1, I_2, τ_2, and τ_m. The valance electron density in both bulk and defect specimens, n_b and n_d, respectively can be determined by the known formula [17];

$$n = [\lambda\,(n) - 2] / 134. \tag{7}$$

These values as well as the total trapping rate of the positrons are listed in table (2).

Table (2): Positron annihilation rates and densities of valance electrons in the bulk and in defects as well as the trapping rate.

T (°C)	λ_b (ns^{-1})	λ_d (ns^{-1})	n_b x 10^{-2} a.u.	n_d x 10^{-3} a.u.	K
300	5.38	2.39	2.52	2.91	0.45
400	5.50	2.42	2.61	3.13	0.53
500	5.38	2.45	2.52	3.36	0.57
600	5.39	2.45	2.53	3.36	0.61
700	5.40	2.47	2.54	3.51	0.64

From table (2) it can be seen that as the tempering temperature increases the density of valence electrons in the defect increases and the total trapping rate of the positrons increase. This indicates changes in the microstructure which is expected to be seen in microhardness variations and microscopic examinations. The experimental results have been interpreted by the two state- trapping model to deduce the vacancy formation energy in the tempered steel. Since [18]

$$K = A\,e^{-\Delta E / kT}, \tag{5}$$

where A is a constant characterizing the metals and k is the Boltzman constant, the formation energy ΔE for vacancies can be extracted. Figure (2) shows the trapping rate (ln K) as a function of 1/kT in the temperature range 300-700°C. The value of the formation energy is found to be 0.0416 eV. The small amount of vanadium (0.009 wt %) present in the steel under investigation makes it different from plain carbon steels. Tempering of micro-alloyed steel containing vanadium would result in softening and removing the internal stresses produced by quenching. Therefore, the small value of ΔE is due to the procedure of the heat treatment (tempering process) which reduced the value of the formation energy Δ E remarkable.

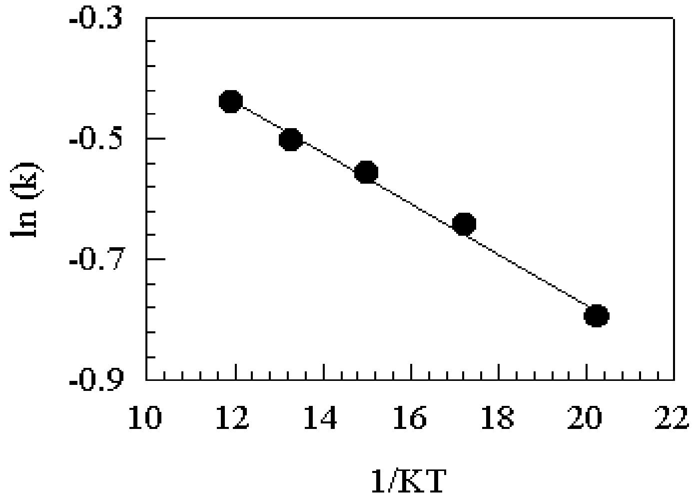


Fig. (2): The trapping rate (ln K) as a function of 1/kT

To establish a correlation between the macroscopic mechanical properties and the positron annihilation parameters, the tempered steel hardness was measured. Figure (3) shows the effect of the tempering temperature on the tempered steel hardness and the microstructure examination. It is clear from this figure that the hardness is rapidly decreasing with increasing the tempering temperature except at 600 ^{o} C.

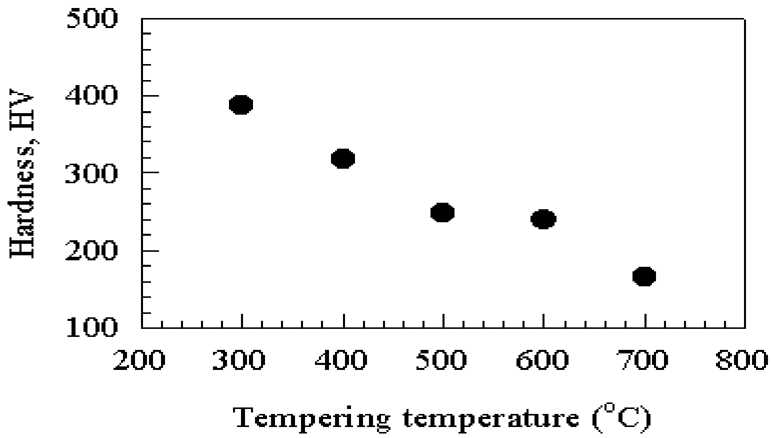
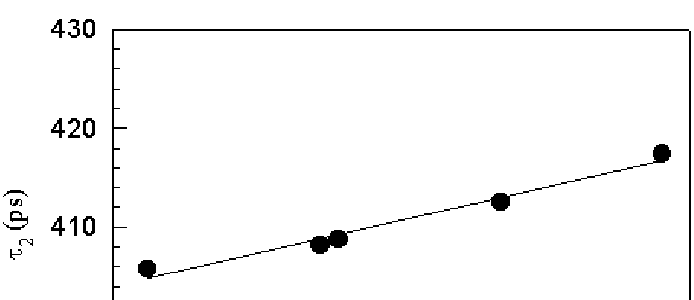


Fig. (3): Effect of tempering temperature on the hardness of the steel.

In the temperature range 300-500 ^{o}C, the martensite decomposes into ferrite and the precipitation of fine particles of carbide occurs. The fine granular

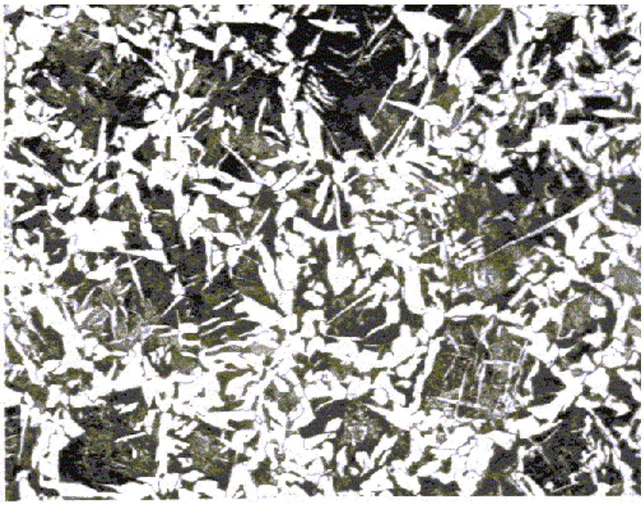


structure formed is known as secondary troostite. This results in some toughening at the expenses of hardness. At higher temperatures 500-700 ^{o}C, the carbide particles coalesce thus producing fewer and larger particles, which provide fewer obstacles to dislocations. The result is a decrease in strength and hardness while further increasing the toughness. This granular structure of carbide particles in a ferrite matrix is known as sorbite and is associated with maximum toughness [19]. The relation between the positron annihilation parameters (τ_2 & I_2) and the hardness is shown in Fig. (4). This figure shows a linear dependence of the positron annihilation parameters and the hardness i.e. the positive correlation is affecting the hardness variations.

Fig. (4): The dependence of the positron annihilation parameters (τ_2 & I_2) on the hardness.

Also, with the aid of image analysis method, the phases in the tempered samples i.e. microstructure examinations, and ASTM (American Standards for Tools and

Materials) grain size were determined from Figs. (5&6). The obtained data of grain size were found to increase with increasing tempering temperature, which leads to the increase in size defects and therefore increase the mean lifetime. These results are consistent with the data shown in Fig.(1). The microstructures in general show that the hardness values obtained are relevant to the microstructure itself being the result of tempering conditions.



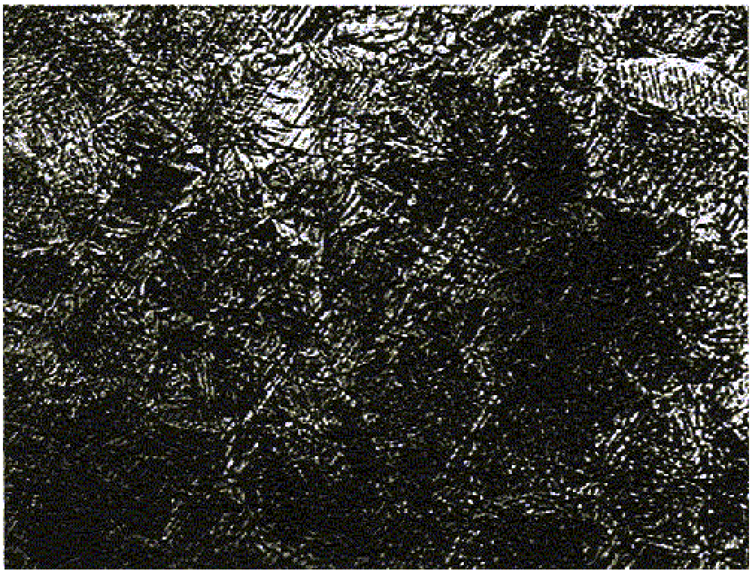
Condition : As received ***Magnification : 100***

Micro-structure: Structure shows fine grains of pearlite (dark) and ferrite (white).

Condition : As quenched ***Magnification : 100***

Micro-structure: Martensite and some retained austenite.

Fig . (5): Image analysis of microstructure showing grain size and phase percentage for the as received and quenched samples.



Condition : Tempering at 500 ^{o}C ***Magnification : 100***
Micro-structure: Structure shows coarse needles of tempered martensite

Fig (6): Image analysis of microstructure showing grain size and phase percentage for the Tempering sample.

4. Conclusion:

Positron annihilation spectroscopy has been a powerful tool to detect vacancies type. The average value of the longer lifetime component ~ 410 ps must be associated with voids that contain (on an average) ~ 10 vacancies or more, as mentioned above. The results show that the tempering of the steel leads to decrease in hardness values, which is in accord with the observed microstructure. Tempering at 300 ^{o}C was sufficient to relieve internal stresses but not enough to change the microstructure. Increasing the tempering temperature is the effective way to control the mechanical properties of such steels and improve their toughness. A correlation between the macroscopic mechanical properties and the positron annihilation parameters has been achieved (Fig 4). The presented results prove that providing information about the microstructure of technical materials is also useful.

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