ESTIMATION OF DISLOCATION DENSITY FOR AISI 304 STEEL AFTER PLASTIC DEFORMATION USING NON-DESTRUCTIVE METHODS*

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Sustainable development is very important in today's world. This development forces the application of innovative solutions in widely understood industrial production. Knowledge of mechanical damage and structural changes occurring after plastic deformation is of great importance from the point of view of industrial applications and material quality. The article compares the results obtained using two non-destructive methods: microhardness measurement and positron annihilation spectroscopy to studies of commercial chromium–nickel austenitic stainless AISI 304 steel. Both these methods revealed changes in the microstructure after plastic deformation. Moreover, these methods showed their suitability for estimation of dislocation density.

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1. Introduction

In the current era, the largest global demand for austenitic stainless steels is observed. This grade accounts for around 70% of total stainless steel production. The properties that make this steel so popular are the highest corrosion resistance, good plastic formability, ductility and weldability. In addition, they have a single-phase structure, which enables the most favorable conditions for the creation of a passive state and to maintain its durability.

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Austenitic steels contain up to 0.15% of carbon and a minimum of 16% chromium. Nickel is also an important alloy additive, which, in combination with other elements, ensures for these steels good corrosion-resistant, durable of austenitic structure and retain this structure in a very wide range of temperatures. The higher the alloying content, the greater the corrosion resistance even in aggressive environments and high temperatures.

Moreover, in order to increase their strength, these steels are subjected to cold plastic deformation after homogenizing annealing, which increases their yield strength several times. As a result of deformation strengthening, there is an increase in strength properties, such as yield point, strength or hardness. Plastic deformation induces changes in microstructure. In general, the density of dislocations increases, which results in the increase of the critical stress required to move them (strain strengthening).

Knowledge of mechanical damage and structural changes occurring after plastic deformation or other processes is of great importance from the point of view of industrial applications of materials due to their physical, mechanical, technological and functional properties. Most mechanical damage starts with the change of the microstructure. These, in turn, can be observed using standard material engineering methods such as scanning or transmission electron microscopy. It is also useful to study properties that change with microstructure, *e.g.*, microhardness. Therefore, other techniques are being sought that allow rapid quantification of the defects of the microstructure, especially non-destructive techniques product testing. The positron annihilation spectroscopy (PAS) can meet such requirements. Unlike other techniques, this method creates unique possibilities of testing defects of the crystal lattice resulting from plastic deformation [1] or irradiation [2].

The aim of this paper is to show how the dislocation density changes after plastic deformation depending on the degree of thickness reduction of stainless steel samples. In the article, there are compared the results obtained using two non-destructive methods. The first technique was microhardness measurements (HV), the second one was the Doppler broadening annihilation line (DB). Additionally, in order to point out which kind of defects are created after plastic deformation, the positron lifetime spectroscopy (LT) was used. Received results were analyzed and compared with each other.

2. Methods

The material under investigation was commercial austenitic stainless AISI 304 steel. The chemical composition (presented in Table I) was determined using an atomic emission spectrometer with spark excitation. Steel samples for the study were cut from a 10 mm diameter rod using the diamond saw with a low cutting speed. The samples in shape of discs, 4 mm

high and 10 mm in diameter, were annealed for 1 h in the flow on N_2 gas at temperature of 650°C, and then slowly cooled to room temperature in order to remove any deformation effect due to cutting and remove possible oxides formed on the surface during annealing.

TABLE I

The chemical composition of the studied samples [wt. %].

Cr	Ni	Mn	Si	Cu	Mo	V	P	S	C	Fe
18.37	8.12	1.13	0.29	0.35	0.36	0.09	0.028	0.026	0.012	bal.

After annealing, surface layers were etched in the hydrofluoric acid and 25% solution of nitride acid in distilled water. Those applied treatments allow us to obtain samples with a relatively low density of crystal lattice defects and clean surfaces. X-ray pattern was carried out using X-ray K_α anode lamp radiation Philips X-Pert diffractometer. In Fig. 1, there is visible only one austenite γ phase.

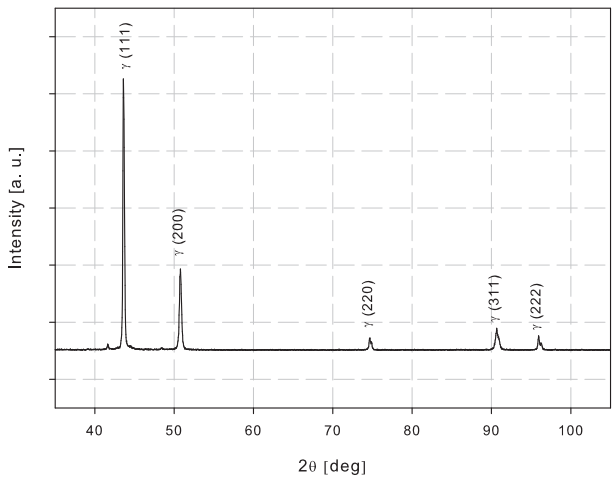


Fig. 1. X-ray diffraction pattern for the sample after annealing.

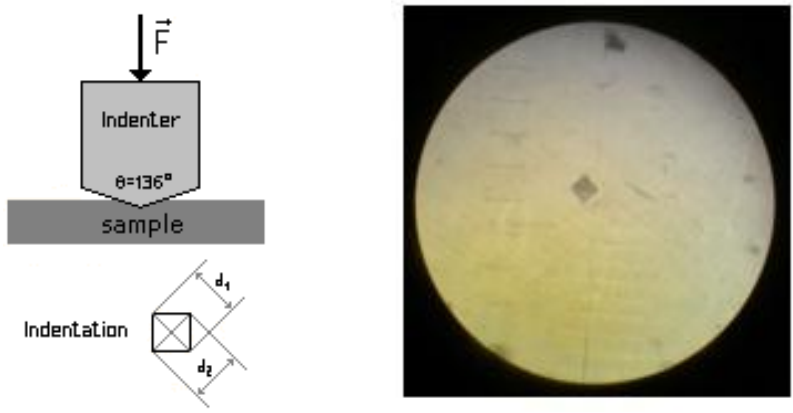
After that, samples in pairs were plastically deformed using a hydraulic press with pressure in the range from 12 to 18 MPa which gave a thickness reduction from 1 to 16% in relation to the initial value and using a larger press (1 TPa), resulting in an additional reduction of 50%. One of the pairs was not deformed, but preserved as a reference.

Measurements were carried out using two methods, which can be treated as non-destructive: The microhardness technique and positron annihilation spectroscopy. The latter comprises Doppler broadening of the annihilation line and lifetime positron spectroscopy [3].

Measurements of microhardness of AISI 304 steel were carried out using a Zeiss Neophot 21 microscope with Hanneman's indenter under load 100 G by means of the Vickers hardness test. The scheme of the first method is presented in Fig. 2. The hardness of the material in this method is determined as the ratio of the load F to the surface area S of the indentation. The indenter is a diamond pyramid with a square base and angle $\alpha = 136°$ between the opposite walls (Fig. 2 (a)). After pressing the indenter in the material, the length of the diagonals of the left imprint is measured. The value of HV for samples was calculated based on equation (1)

$$\mathrm{HV} = \frac{F}{S} = \frac{1.854F}{d^2}\,, \tag{1}$$

where F is a force applied to the diamond in kilograms-force and d represents lengths of diagonals in millimeters. For each sample, ten indentations were made in different places and the obtained results were averaged. An example of an indentation is visible in Fig. 2 (b).



(a) (b)

Fig. 2. Microhardness measurement using the Vickers method. A principle of microhardness measurement (a), an example of indentation (b).

For monitoring the Doppler broadening of the annihilation line, a coaxial high-purity germanium (HPGe) detector with an energy resolution equal to 1.38 keV (FWHM) interpolated at 511 keV was used. The ^{68}Ge/^{68}Ga isotope (activity 10 μCi) enveloped in a 7-μm-thick Kapton foil was used as a positron source. A typical measurement geometry was applied, *e.g.*,

the source was placed between two identical samples with a flat surface. The number of counts in the spectrum in the area of the annihilation line were 10^6. The obtained spectrum was analyzed using the SP-1 program by determining a so-called S-parameter. The S-parameter is defined as the ratio of the area A_S under the fixed central part of the annihilation line (for energy close to 511 keV) to the area under the whole annihilation line A. The parameter is sensitive to the annihilation of positrons with low momentum electrons which are present in open volume defects. An example of the annihilation line for the annealed sample and for the sample after plastic deformation is presented in Fig. 3. The increase of the S-parameter value caused by the sample deformation is clearly visible.

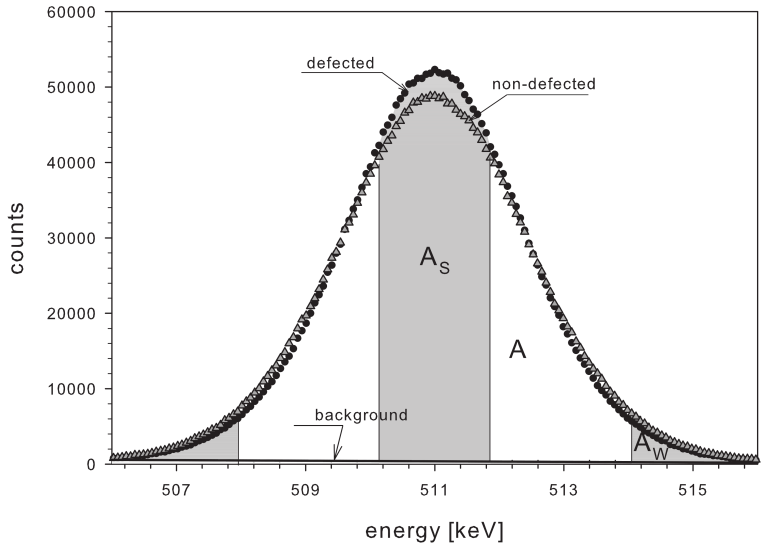


Fig. 3. The 511 keV lines for defected and non-defected samples of pure Fe.

The positron lifetime spectroscopy of more than 1.5×10^6 counts were measured for the initial annealed sample and deformed samples using the conventional fast–fast spectrometer with BaF_2 scintillators. The time resolution of the system was 280 ps (FWHM). The 115 μCi activity positron source containing this time ^{22}Na isotope was sandwiched between two stainless steel samples. All obtained spectra were analyzed using the LT code program [4].

3. Results and discussion

Figure 4 presents the results of microhardness measurement as a function of thickness reduction for AISI 304 steel. We can notice that the microhardness increases with increasing thickness reduction with an exponential ten-

dency. The growth of microhardness means that after plastic deformation the material becomes more tough for two reasons: First, the deformation causes the formation of the martensitic phase α' in the volume of austenite γ [5, 6], second, it causes an increase in density of dislocations in austenite γ. It means that plastic deformation generates a great amount of crystal lattice defects.

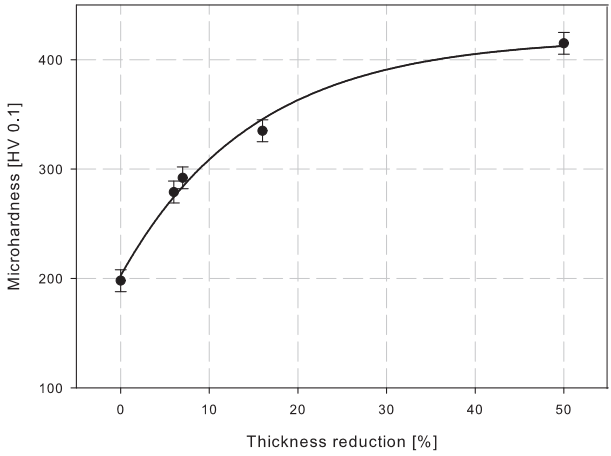


Fig. 4. The dependence of microhardness measurement with thickness reduction for AISI 304 steel.

The initial value of microhardness was 197 HV, which is consistent with the results given in [7]. Knowing the values of microhardness, we can calculate the material yield limit using an approximate relation (2) linking yield strength with the Vickers hardness (HV) [8]

$$\sigma_y = 3.55 \text{ HV} . \tag{2}$$

The yield strength is expressed in MPa, and the Vickers hardness in kg/mm^2.

This relation is important from a practical point of view, because the hardness measurements are easier to carry out than other tests determining the mechanical properties of the material. This is important, for example, in the case of steels used for pressure vessel construction in nuclear power plants. The monitoring of mechanical properties of these steels is of particular importance as they can change under the influence of radiation. In the case of austenitic steels, the correlation coefficient between the change in yield strength $\Delta\sigma_y$ and the change in microhardness ΔHV was determined as 3.03, and for ferritic steels as 3.06 [8]

$$\Delta\sigma_y = 3.03\,\Delta\text{HV} . \tag{3}$$

If the dislocations are caused by the plastic deformation and they are responsible for the strengthening of the material, the yield strength is related to the density of dislocation with the Taylor relationship [9]

$$\Delta\sigma_y = \alpha G b \sqrt{\rho}\,, \tag{4}$$

where α is constant and equal to 0.5, G is a shear modulus and for studied steel is 86 GPa, and b is a Burger's vector and for examined steel is equal to 2.58×10^{-10} m [10].

Table II contains estimated values of yield strength and dislocation density correlated with the thickness reduction.

TABLE II

Estimated values of yield strength and dislocation density for AISI 304 steel samples after plastic deformation.

Δd [%]	HV	ΔHV	$\Delta\sigma_y$ [MPa]	ρ [10^{14} m^{-2}]
6	279	81	245.43	4.89
7	292	94	284.82	6.59
16	335	137	415.11	14.0
50	415	217	657.51	35.1

It can be seen that the density of dislocations increases with the degree of deformation. Its value varies from 4.89×10^{14} to 3.51×10^{15} m^{-2}. The estimated dislocation densities can be compared with the results obtained by Shintani and Murata [7] based on the analysis of X-ray diffraction. In the conducted research, they showed that the maximum value reached 5.0×10^{14} m^{-2} at 60% elongation. Density values estimated in this paper are slightly higher, but this may be due to the fact that the use of formula (4) is a rough estimate. Moreover, in addition to dislocation density, the boundaries of small grains can contribute to hardness.

Figure 5 presents the results of Doppler broadening measurement for the deformed steel samples. The S-parameter exhibits an increase with deformation and saturation. This saturation takes place at a thickness reduction of approx. 15%. The solid line presents the curve given by Eq. (5) fitted to the experimental points:

$$S = S_{\mathrm{SAT}} + (S_{\mathrm{f}} - S_{\mathrm{SAT}}) \exp(-c\varepsilon)\,, \tag{5}$$

where ε is the relative thickness reduction expressed as a percentage, c is a constant, S_{SAT} and S_{f} are the values of the S-parameter, assuming the saturation of annihilation in defects and for the non-deformed (annealed) sample, respectively. The following values of fitted parameters were obtained: $c = -0.1863 \pm 0.0232$, $S_{\mathrm{f}} = 0.4798 \pm 0.0029$, $S_{\mathrm{SAT}} = 0.5092 \pm 0.0017$.

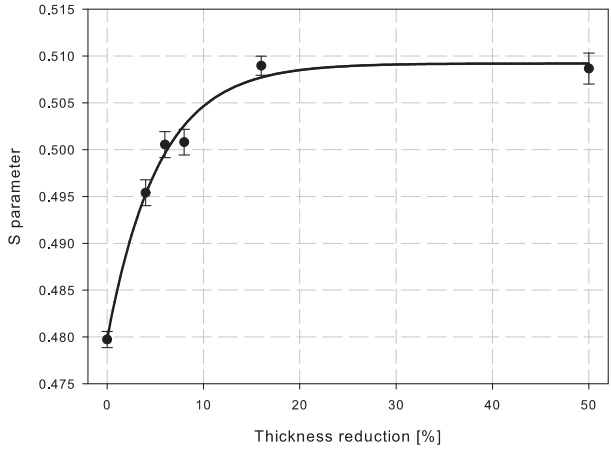


Fig. 5. The S-parameter as a function of thickness reduction for austenitic stainless AISI 304 steel. The solid line represents the best fit of the exponential function (5).

The increased value of the S-parameter indicates an increase in contribution of positron annihilation in defects. Plastic deformation generates a huge number of dislocations which are shallow traps for positrons. Positron, moving along the dislocation line, can be localized in deeper traps such as jogs or vacancies on dislocation lines. Jogs and kinks arise as a result of the intersection of moving dislocations. However, motion of dislocation with jogs generates point defects: vacancy and interstitial atoms. From the dependency of the S-parameter as a function of thickness reduction we can also estimate the dislocation density.

The results of the positron annihilation experiment for samples containing defects that trap positrons were analyzed in terms of a two-states trapping model [11]. This model assumes that positron exists in one of only two states in the material, the free or Bloch state and the defect trapped state. By using the value of the S-parameter and the obtained fitted values for S_{f} and S_{SAT}, the values μ (trapping rate) can be calculated as:

$$S = S_{\mathrm{f}} \left(\frac{1 + \mu S_{\mathrm{SAT}}}{1 + \mu S_{\mathrm{f}}} \right) , \tag{6}$$

where S is a value of the S-parameter obtained from the Doppler broadening of the annihilation line measurement, S_{f} and S_{SAT} come from the fitted function (5). The positron trapping efficiency ν is calculated by using the trapping rate according to the following equation [12]:

$$\mu = 1.248 \times 10^{-3} \left[\log \left(1 - \varepsilon \right) \right]^2 \nu / b^3 , \tag{7}$$

where ε is the fractional thickness reduction and b is Burger's vector. The trapping rate μ is proportional to the defect density ρ'; the number of trapping sites per unit volume; as shown in equation (8)

$$\mu = \nu\rho' \,. \tag{8}$$

The defect density is related to dislocation density ρ, length per unit volume [12], according to equation

$$\rho' = \frac{\rho}{b} \,, \tag{9}$$

where b is a Burger's vector.

The results of calculations are presented in Table III. It seems that the dislocation densities estimated using the positron annihilation method are more scattered than those obtained from the microhardness measurements but more reliable. What is more, results correspond quite well with the results that can be found in the literature. The dislocation density for steel obtained by Park *et al.* [13] after plastic deformation was of the order of 10^{13} m^{-2}. Nakashima *et al.* [14] determined the limit of dislocation densities at the level of 2×10^{15} m^{-2}.

TABLE III

Values of trapping efficiency and dislocation density for AISI 304 steel after plastic deformation.

ε [%]	ν [cm^{3}s^{-1}]	ρ' [cm^{-3}]	ρ [m^{-2}]
6	9.45×10^{-17}	5.25×10^{16}	1.35×10^{13}
7	7.22×10^{-17}	7.22×10^{16}	1.86×10^{13}
16	7.30×10^{-16}	4.17×10^{17}	1.07×10^{14}
50	1.83×10^{-17}	6.59×10^{18}	1.70×10^{15}

The S-parameter actually informs us that density of dislocations increases when deformation becomes stronger. In order to find out whether any other defects are formed, we will use the second positron annihilation method: measurements of the positron's lifetime. Table IV presents the results of positron lifetime for steel depending on the thickness reduction. This table additionally contains results for the samples reduced by 1, 2 and 3 percent. For annealed sample, this time was 110 ps.

Interpretation of the obtained results requires knowledge of the positron lifetime (theoretical or experimental). For a non-disturbed lattice of iron (or when there are atoms in interstitial positions), this time is between 106 and 110 ps [15]. The lifetime of positrons increases when open volume defects appear in the structure. For edge dislocations, the theoretically

TABLE IV

The lifetime of positron *versus* thickness reduction for AISI 304 steel after plastic deformation with additional results for steel after deformation between 1 and 3 percent.

ε [%]	τ [ps]	ε [%]	τ [ps]	ε [%]	τ [ps]
1	146	3	160	16	161
2	156	7	163	50	168

calculated lifetime is 117 ps [15]. The lifetime measured for deformed iron by Hidalgo *et al.* [16] was equal to 150 ps and this result was ascribed to annihilation in vacancies on dislocation lines. Park *et al.* [17] received 142 ps lifetime for screw dislocations and 165 ps for edge dislocations. It should be emphasized that dislocations in deformed material may also be of mixed type. Theoretical calculations carried out by Kuramoto *et al.* [15] give 174 ps for vacancy on the screw dislocation line and 140 ps for vacancy on the edge dislocation line. For a single vacancy, the lifetime of positrons is well-defined both experimentally and theoretically, and is 175 ps.

In connection with the above figures and considerations, we can conclude that the changes of positron lifetime in the deformed steel depend on the thickness reduction of the compressed sample. For the non-deformed material, it amounted to (110 ± 1) ps, which is consistent with literature [15]. However, after the 1% thickness reduction, this value increased to 146 ps, and at 3% strain the lifetime of positrons reaches a value close to saturation, *i.e. ca.* 160 ps, slightly changing at higher strains. The obtained lifetimes can be compared with the results published in Somieski and Krause-Rehberg [18] for iron and iron alloys under tension. In the case of AISI 304 steel, the values of the lifetime for a given degree of deformation are very similar, despite the use of different methods of sample deformation. In both cases, they are about 10 ps higher than for pure iron. This may indicate that if clusters of several vacancies arise, their concentration is much lower than in the case of iron. It should be taken into account that the crystal lattice of α-Fe is a body centered cubic (bcc), and the austenitic stainless AISI 304 steel is face centered cubic (fcc).

4. Conclusions

We have demonstrated that the microhardness method and positron annihilation spectroscopy provide information about the crystal lattice defects induced by plastic deformation. Following the two methods independently, it was found that the density of dislocations increases with an increasing de-

formation degree. Although the two approaches reveal the same tendency, some discrepancies between the dislocation density values can be observed, especially for small deformations. The discrepancies for small deformations can be explained by approximate character of Taylor's formula. It seems that more correct and reliable results are obtained from the S-parameter. The use additional method — positron lifetime spectroscopy — allowed determination what kind of defects have been introduced into the crystal lattice.

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