STUDIES AND RESEARCHES CONCERNING THE MEASUREMENT UNCERTAINTY ESTIMATION OF SUB-GRAIN APPARENT AVERAGE SIZE OF POLYCRISTALIN QUENCHED STEELS

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Abstract. *The estimation of the apparent average size of sub-grain domains is important to underline the mechanical and electrical properties of metallic materials. Further, the size estimations of the sub-grain domains can be used to calculate the dislocation densities of the measured samples. Subsequently, the size and dislocation density provides a thoroughly understanding of the steels physical properties variation. In the paper are presented the values of the measurands and their measurement uncertainties estimated according with SR EN 98-3:2010 standard and error propagation low. There were considered two types of quenched steels: 316L stainless steel and 100Cr5 bearing steel.*

Keywords: *X-ray diffraction, apparent average size, measurement uncertainty, quenched steels*

1. INTRODUCTION

X-ray diffraction (XRD) is a well-known method used to investigation the mono and polycrystalline structures [1, 2]. Polycrystalline metallic materials have a long distance atomic order structure at the sub-grain level, which are, in fact, real crystallites. This is the main reason that X-ray diffraction technique is frequently used to study the distortion crystal lattice. The structural parameters measured by X-ray diffraction are the lattice parameters, apparent average size of the crystallite (sub-grain domain) and dislocation density. Due to the influence factors the results of the XRD measurements are affected by significant uncertainties [3].

2. UNCERTAINTY ASSESSING PROCEDURE

Determination or rather estimation of the average size of the "apparent" crystallite (sub-grain) size by X-ray diffraction investigation started since the second decade of the 20th century [4]. The first paper on this subject was published by SCHERRER in 1918 [5]. To obtain comprehensive information on the distribution of crystallite sizes of the sample it is mandatory to use advanced physic-mathematical model of the X-ray diffraction by distorted structures. But, for the present aims we used the well known formula given by Scherrer to estimate the apparent average crystallite size:

$$L = \frac{c \cdot \lambda}{\beta \cdot \cos(\theta)} \quad \textbf{(1)}$$

Where:
c - Constant, λ – X-ray wavelength, β – full-width at half maximum (FWHM) of the considered XRD peak.
According to the error propagation law, the standard deviation is calculated using the equation:

$$S_L^2 = \left(\frac{\partial L}{\partial c}\right)^2 \cdot S_c^2 + \left(\frac{\partial L}{\partial \lambda}\right)^2 \cdot S_\lambda^2 + \left(\frac{\partial L}{\partial \beta}\right)^2 S_\beta^2 + \left(\frac{\partial L}{\partial \theta}\right)^2 \quad \textbf{(2)}$$

Where: S_L is standard deviation of L

$$\beta = \frac{3.14 \cdot B}{180} \quad \textbf{(3)}$$

Where:
β - FWHM in radians
B – FWHM in degrees
Replacing the relations of the derivation of L in (2) one obtains:

$$S_L^2 = L^2 \frac{S_c^2}{c^2} + L^2 \frac{S_\lambda^2}{\lambda^2} + L^2 \frac{S_\beta^2}{\beta^2} + L^2 \cdot tg^2\theta \cdot S_\theta^2 \quad \textbf{(4)}$$

The S_L is the main contributor to the uncertainty budget of L. The estimation of L uncertainty measurement could be improved by repeating the measurement 5-10 times, but in the XRD practice it is restricted by time and cost reasons. Thus, the S_L is the most used indicator of the L uncertainty measurement.

3. RESULTS AND DISCUSSIONS

XRD measurement were performed with DRON 3 diffractometer equipped with Mo tube (λ_{Mo} = 0.71 A) in the „X-ray Diffraction Laboratory" of the „Physical Metallurgy and Metallic Materials` Science Department" of „Materials' Science and Engineering Faculty" from the „Politehnica" University of Bucharest, Romania. The phase nature identification is based on the positions of the diffraction lines e.g. on interplanar distance d_{hkl} calculated with Bragg formula:

$$2d_{hkl} \cdot \sin(\theta) = \lambda \quad \textbf{(5)}$$

Where:
2θ is the angular position of the hkl peak
The measurement uncertainty is estimated in accordance with SR EN 98-3:2010 which make use of the error propagation for two steels: stainless steel 316L and bearings RUL1.
The XRD diffractogram of RUL 1 is given in figure 1.

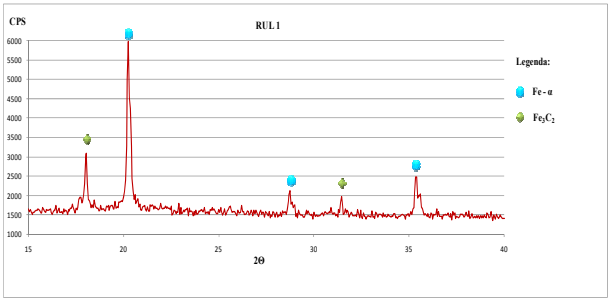

Figure 1 XRD diffractogram of Rul 1

The ICDD files No.6-696 and No 18-964 were the main files used to estimate the RUL 1 phase content. The phase content of RUL 1 is given in Table 1.

Table 1 Indexing data file

Angle	Practical result [nm]	Fe α	Fe_5C_2
		Theoretical data [nm]	Theoretical data [nm]
18.05	2.264		2.26
20.25	2.020	2.026	
28.75	1.430	1.433	
31.45	1.310		1.32
35.35	1.169	1.170	

Based on data given in Table 1 it is quite sure that the sample contains Feα (BCC) phase and Fe_5C_2 carbide.

Table 2 Elemental composition of RUL 1 specimen

C	Si	Mn	P	Cr	Mo	Co	Fe
0.97	0.28	0.38	0.001	1.36	0.04	0.014	96.6

The elemental composition of RUL 1 specimen (Table 2) and the data from the literature [6] underpin the exactness of the phase analysis given in Table 1. The criterion for choosing the XRD lines for apparent average size measurement is the peak intensity e.g. the peak maximum intensity is well above the background. Thus, following peaks have been used: Feα (110), Feα (200), Feα (211) for ferrite and (002), (802) for Fe_5C_2 carbide. The measurements results are shown in Table 3.

Table 3 Experimental data and apparent average sizes associated to Rul 1 specimen

No.	Angle	Half-Height Distance	hkl	L[nm]
1	18.05	0.38	002	31.08
2	20.25	0.6	110	19.75
3	28.75	0.35	200	34.41
4	31.45	0.2	802	60.59
5	35.35	0.45	211	27.21

The uncertainty assigned to L consist of standard uncertainty of A type (for β, θ) and uncertainty (standard) of type B (for C, λ, π). The uncertainties assigned to the quantities involved in (2) are given in Table 4.

Table 4 The uncertainties assigned to β, θ quantities

β	θ	C	λ	π
0.002	0.05	0.001	0.001	0.0016

The standard deviation of L and its relative standard deviation corresponding to the values given in Table 3 are shown in table 5.

Table 5 The standard deviation of L for different <hkl> directions corresponding to Table 3

S_{RL}	$\sqrt{S_{RL}}$	Standard error [Å]	Measurement error [%]
0.0040	0.063	1.96	6.29
0.0062	0.079	1.55	7.87
0.0063	0.079	2.73	7.94
0.0086	0.093	5.63	9.28
0.0091	0.095	2.59	9.52

Taking into account the uncertainties of the Ls one may conclude that the α L values for different <hkl> seem to be comparable in the range while for Fe_5C_2 the Ls values are significantly different. This aspect denotes that the carbide crystallite pattern is disk shaped. Further researches should be performed to establish the morphologies of α and Fe_5C_2 crystallites.

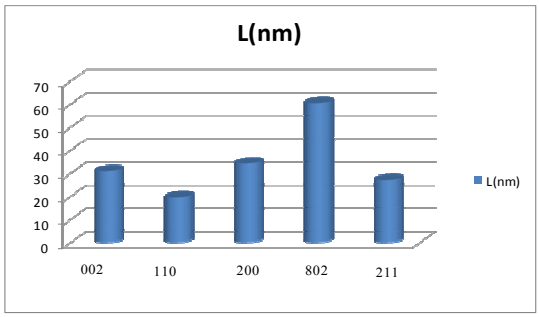

Figure 2 Distribution of mosaic blocks

The diffractogram of the 316L specimen was obtained with the DRON 3 -2 using Cu characteristic X-radiation (λ=1.540598 A) (Fig. 3)

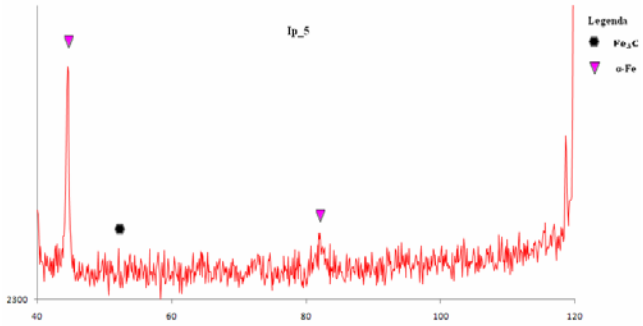

Figure 3 XRD diffractogram of the 316L specimen, code IP5

The ICDD files No.6-696 and No 18-964 were the main files used to estimate the RUL 1 phase content. The phase content of RUL 1 is given in Table 6.

Table 6 Preliminary results

Angle	Practical result [nm]	Fe α	Fe_3C
		Theoretical data [nm]	Theoretical data [nm]
44.5	2.036	2.01	
52.1	1.76		1.76
81.9	1.176	1.17	1.18

Based on data given in Table 6 it is most probable that the sample contains Feα(BCC) phase and Fe_5C_2 carbide.

Table 5 Data on apparent average estimation in the sample mosaic blocks Ip_5

No.	Angle	Half-Height Distance	Hkl	L[nm]
1	44.5	0.22	110	110
2	81.9	0.19	211	127

The elemental composition of Ip5 specimen (and the data from the literature [6] underpin the exactness of the phase analysis given in Table 5. The criterion for choosing the XRD lines for apparent average size measurement is the peak intensity e.g. the peak maximum intensity is well above the background. Thus, following peaks have been used: Feα(110), Feα(200). The measurement results are shown in Table 6.

Table 6 Method of calculating the standard deviation of the average apparent crystallite size for each diffraction line

S_{RL}	$\sqrt{S_{RL}}$	Standard error [Å]	Measurement error [%]
0.0062	0.08	9	8
0.0114	0.11	14	11

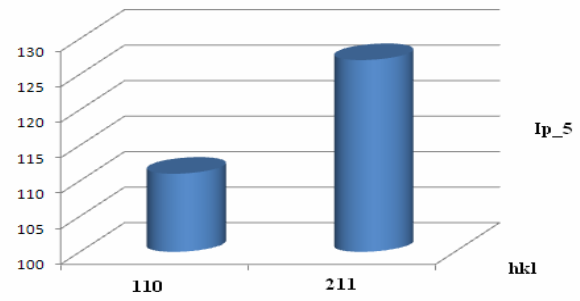

Figure 4 Distribution of mosaic blocks

4. CONCLUSION

The authors implemented an improved the procedure for apparent average crystallite size measurement in the „X-ray Diffraction Laboratory" of „Materials Science and Engineering Faculty". The main improvement consists in measurement uncertainty assessing in accordance with "SR Ghid ISO 98-3". The measurement uncertainty is required by "SR EN ISO/CEI 17025" which specifies „the quality of a numeric test result is quantified by the expanded uncertainty U(p%), where p is the level of confidence (p≥ 95%)."

The αL values for different <hkl> directions are quite the same and give reasons to consider that the morphological pattern of the sub-grain domains is of spheroid shape.

The L values for Fe_5C_2 indicate a disk like pattern shape.

The measurements of relative uncertainties for all L results are around 10%. It means that the measurement procedure has to get further improvement by:

- Theoretical model improving of XRD in imperfect crystallites
- Reference Materials (CRMs) usage for model validation

The development of nanotechnology implies crystallite size measurement with adequate exactness (precision and accuracy). In this context, the estimation of measurement uncertainty of the average crystallite size becomes a must. Thus, as the measurement uncertainty is smaller the quality of the result is higher. In this direction, the present procedure is a first step on the way of developing a measurement procedure for XRD crystallite size measurement intended to fulfill the requirement of the present technology in the field of metallurgy and in other advanced material technology fields.

5. REFERENCES

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