# SURFACE HARDNESS MEASUREMENT AND MICRO-STRUCTURAL CHARACTERISATION OF STEEL BY X-RAY DIFFRACTION PROFILE ANALYSIS

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**Abstract**    An X-ray diffraction line will broaden considerably when steels change into martensitic structure on quenching. The results presented in this paper show that X-ray diffraction technique can be employed for a rapid and nondestructive measurement of hardness of hardened steel. Measurement on various quenched and tempered steels showed that the breadth of its diffraction peak increased with increasing hardness. Some empirical equations are derived to correlate the width of the diffraction line with the hardness. The diffraction profile analysis can offer good indication about the material's characteristics and the microstructural evolution caused by heat treatment or by mechanical loading.

**Key Words**    X-ray Diffraction, Nondestructive Tests, Hardness, Steels

چکیده    زمانی که در اثر سریع سردی ساختار فولاد مارتنزیتی میشودوپهنای قله پراش ایکس بطرز قابل توجهی افزایش می یابد. نتایج ارائه شده در این مقاله نشان میدهد که تکنیک پراش پرتو ایکس می تواند بعنوان یک روش سریع و غیر مخرب جهت اندازه گیری سختی فولاد بکار رود. اندازه گیری روی چندین نمونه فولاد پس از عملیات حرارتی سریع سردی و بازگشت بمنظور کسب سختیهای متفاوت نشان داد که پهنای قله پراش با افزایش سختی متناسب است. جهت برقراری رابطه بین پهنای قله پراش و سختی، با استفاده از نتایج آزمایشات معادلاتی حاصل و ارائه گردید. ضمنا آنالیز منحنی پراش میتواند اطلاعات مفیدی درمورد تحولات ریزساختاری ناشی شده از عملیات حرارتی به ما ارائه دهد.

## INTRODUCTION

Hardness is one of the most important properties of materials. For metals, hardness usually implies a resistance to permanent or plastic deformation. To the design engineer, hardness often means an easily measured and specified quantity which provides information about the strength and heat treatment of the metal. There are different types of hardness measurements but only indentation hardness is of major engineering interest for metals. The widely used indentation hardness methods are Brinell, Rockwell and Vickers. Perfect correlation cannot be expected between the results of the hardness test and other properties of materials. Nevertheless, because both tensile strength and hardness are indicators of a metal's resistance to plastic deformation, they are roughly proportional. However, empirical relations have been developed between these two properties, at least in the case of certain steels [1]. The 0.2 percent offset yield strength can be determined with good precision from Vickers hardness measurement [2]. For most steels,

the tensile strength in megapascals is approximately 3.45 times the Brinell hardness number [3]. In France, IRSID along with twelve other laboratories from steels making plants, carried out an extended study on a large number of steels with different compositions and microstructures. The research of this group resulted in the compilation of French standard NF A03-173 in 1980 with the title of "hardness-tensile strength conversion table for steel" [4,5]. Steel and Spence's investigation confirms the fact that the extent of plastic deformation in steel can be estimated with hardness test [6]. Marburger and Koistinen [7] showed that the hardness of certain quenched and tempered steels is related to the breadth at half-maximum intensity. Kurita [8] also showed that X-ray diffraction technique using the Gaussian curve parameter allowed the measurement of hardness of hardened steels. It has been recognised, since Warren-Averbach's method appeared in the fifties, that the analysis of broadened X-ray diffraction profiles can be used to study the microstructures of cristalline materials [9].

## X-RAY DIFFRACTION METHOD

What differentiates a single crystal from polycrystalline aggregates is the presence of grain boundaries and grains. The effect of these grains and grain boundaries on X-ray diffraction is characterised by the line form or line broadening (due to grain size, grain strain and grain number contribution) and the texture effects (grain orientation) [10]. Therefore, the line form is related directly to all microstructural features such as heat treatment, work hardening, etc, in addition to the initial defects such as grain boundary or chemical composition inhomogeneity. Therefore, the interest is in the line form evolution in the case of heat treated steel. When a steel is hardened by quenching, the X-ray lines become very broad because of the martensite formation and the presence of very high dislocation density in the quenched mate-

rial. The corresponding hardness is also important. With annealing, the quenched steel releases its internal micro-stresses. Due to the thermal effect, the initial very high dislocation density decreases and subsequently the hardness decreases compared with the quenched steel. In order to explain the modification of the diffraction line after having undergone plastic deformation, the material is divided into small crystal domains called coherent diffraction domains. Each domain is formed from series of unit cell columns. These columns of the length L are perpendicular to diffraction planes. The avcerage of the lengths L of these columns is the *average coherent domain Size D*. Each domain, itself, can be deformed elastically of a length $\Delta L$, which makes the introduction of the distortion $\varepsilon_L = \Delta L/L$ for each column possible (Figure 1). Considering all columns with L length, from $\varepsilon_L$ one can define *root mean square strain* $<\varepsilon_L^2>^{1/2}$. Regarding the line form, it is accepted qualitatively that a diffraction line is broader when coherent domain is smaller and deformed elastically. Moreover, several analysis methods can be used to obtain a semi-quantitative or quantitative description of the microstructure evolution in the material such as stacked energy and dislocation density [11, 12]. The quantitative analysis involving Fourier analysis of the diffraction lines from the unknown and from the standard can be applied to several orders of diffraction such as hkl, 2h2k2l,... [9] or to a single peak [13, 14]. After smoothing, theoretical corrections and background subtraction of the peak, the computer can eliminate the instrumental broadening by the Stokes correction.
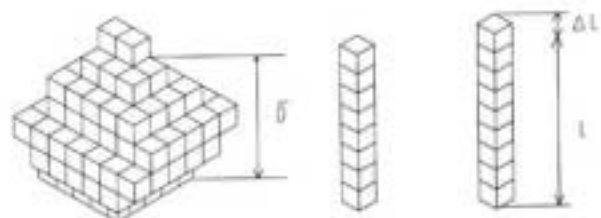


**Figure 1.** Schematical representation of a coherent diffraction domain.

## Fourier Analysis on Several Orders of Diffraction

The distribution of the diffraction intensity can be represented by the following Fourier integral.

$$I_{hk(n)} = k \, C_L \, \exp\,[-2\pi L(S-S_{hkl})] \, dL \qquad (1)$$

where

$S = (2\sin\theta)/\lambda$

$S_{hkl} = (2\sin\theta_{hkl})/\lambda$

$\theta_{hkl}$ = centre of gravity of the diffraction line hkl

$C_L = A_L + iB_L$

k = arbitrary constant

For a material without stacking fault, the peaks are symmetrical. Therefore, only the real part $A_L$ which can be considered as the product of two coefficients are concerned. One coefficient is related to the domain size $A_L^D$ and the other is related to the strain $A_L^S$:

$$A_L = A_L^D \cdot A_L^S \qquad (2)$$

with

$$A_L^D = 1 - L/D \qquad \text{(small L)} \qquad (3)$$

$$A_L^S = <\cos 2\pi L \cdot \varepsilon_L \cdot \varepsilon_{hkl}> \qquad (4)$$

Considering that strains $\varepsilon_L$ are distributed according a Gaussian distribution, the following relation can be obtained:

$$A_L^S = \exp\,(-2\pi^2 L^2 \cdot <\varepsilon_L^2> \cdot S^2_{hkl}) \qquad (5)$$

by using Equations 3 and 5, Equation 2 can be written as follows:

$$\text{Ln } A_L = \text{Ln } A_L^D - 2\pi^2 L^2 <\varepsilon_L^2> \cdot S^2_{hkl} \qquad (6)$$

Plotting $A_L$ as a function of $S^2_{hkl}$ will give series of lines for different values of L. The slope of these lines

which equals to $-2\pi^2 L^2 <\varepsilon_L^2>$ makes obtaining the $<\varepsilon_L^2>$ values possible and the origin gives $\text{Ln}A_L^D$ value. The $A_L^D$ values are plotted against the L values. The tangent to the origin of the curve $A_L^D = f(L)$ intersects the abscissa axis at a value equal to D. Due to anisotropy of mechanical properties of materials, it is necessary to use several diffraction order for Equation 6. However for cubic materials, only two orders are used. Experimental condition, particularly recording the diffraction peak can be the source of error. Special care should be taken. Figure 2 shows an example of the results for a steel.

## Single Diffraction Profile Method

The advantage of this method is to reduce the recording time by a factor of two. On the other hand, some supplementary hypotheses are necessary, particularly the hypothesis concerning the shape of the $<\varepsilon_L^2> = f(L)$ curve. The method first was proposed by Gangulee [13] and then by Mignot and Rondot [14]. The mean square strain is modelled into an hyperbolic form $<\varepsilon_L^2> = C1 + C2/L$ and the $A_L^D$ in a linear form $A_L^D = (1-L/D)$. The real part of the Fourier transform is thus fitted to a 3rd degree polynomial whose coefficients are used for the calculation of the searched parameters. Another parameter $\varepsilon_1$ can be defined which is the distortion factor and equal to $<\varepsilon_L^2> = \varepsilon_1/L$.

## Diffraction Line Width Method

The diffraction line breadth is a global indicator of the microstructural state of the material. In the case of heat treatment, the line breadth is in close relation with dislocation density and its distribution and also with work hardening capacity.
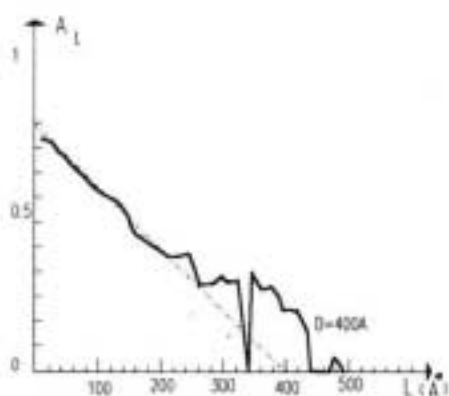
The diffraction line breadth is usually referred to as the breadth measured at half-maximum intensity. However some investigators prefer to determine the **integral breadth** of a diffraction line $I_w$. The integral breadth is given by the ratio between the integral
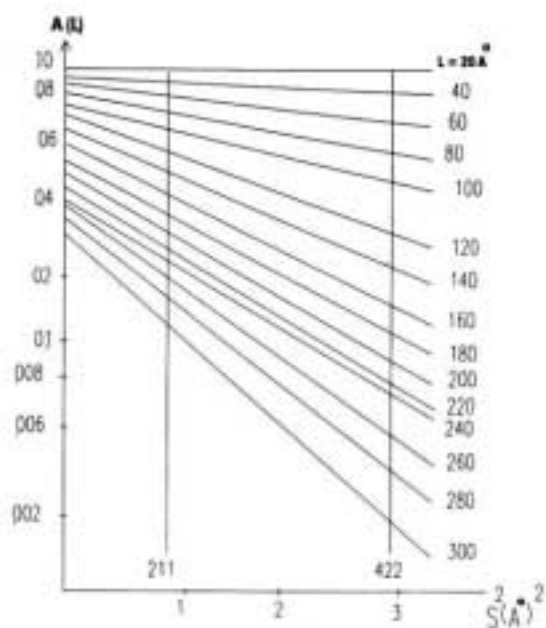
(a)
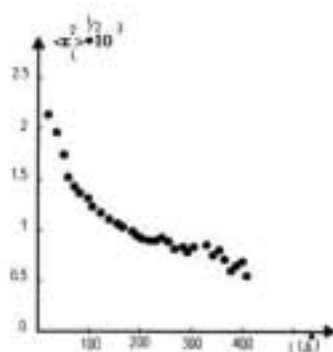


(b)



(c)

**Figure 2.** An example of the profile treatment by Fourier analysis.

intensity and the maximum intensity. The diffraction line breadth can be determined with an automated X-ray stress measurement apparatus (such as; SIEFERT, SIEMENS, JEOL, ...) using a microcomputer. The diffraction line can also be plotted on a chart recording (Figure 3) and the width be determined manually by smoothing the diffraction peak and the subtraction of background from the observed intensity. Figure 4 shows the method of peak positioning and the determination of the diffraction line width. Line position cannot be measured with sufficient precision on a chart recording. When using a computer, observed X-ray intensities (counts) must be corrected for both background and $LPA$ factors to obtain true diffracted X-ray intensities. One factor controlling the relative intensities of the diffraction line is the Lorentz-polarization ($LP$) factor. This factor can vary considerably over the width of a single line, when the line is broad and in the high-angle region. Measured intensities are to be divided by $LPA$ in order to make the lines almost symmetrical, before determining the line centre by the least square method. If the background is high, because of fluorescence by the specimen, bettrer accuracy is attainable by subtracting the background, assumed linear across the line, before applying the $LPA$ correction and finding the line centre [10]. The method of finding the centre of a diffraction line, broad or narrow, is to fit a parabola to n points on the top of the line and take the axis of the parabola as the centre. Measurement of line
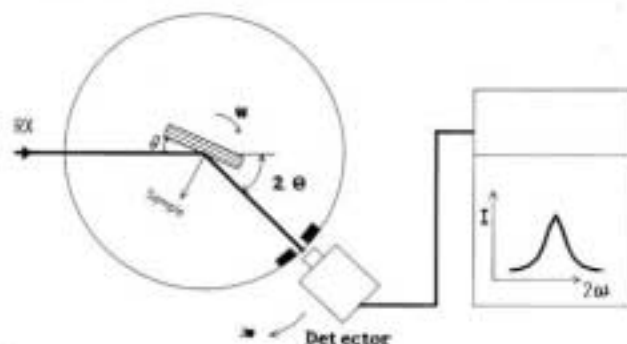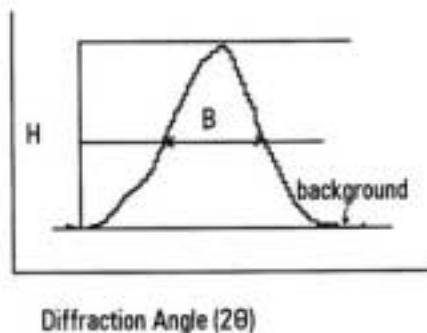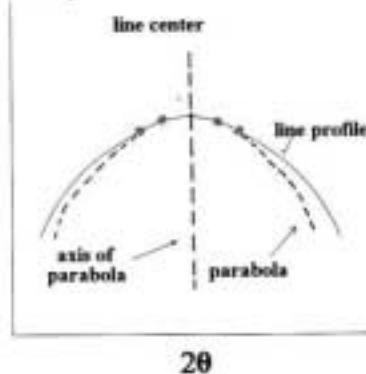


**Figure 3.** X-ray diffractometer.

X-Ray Intensity



Diffraction Angle (2θ)

(a)

Intensity



2θ

(b)

**Figure 4.** Method of peak positioning and measuring of diffreaction line width; (a) manual fitting; (b) automatic fitting.

This study was performed on two steels: AFNOR XC48 of composition (wt%) 0.49 C, 0.73 Mn, 0.18 Si remainder Fe, AFNOR 35NCD16 of composition (wt%) 0.36 C, 0.4 Mn, 0.36 Si, 3.85 Ni, 1.75 Cr, 0.4 Mo remainder Fe. Specimens with $15 \times 15$ mm² section and a thickness of 7 mm were cut and tempered at various temperatures after quenching from their austenitized temperature. After the heat treatment, they were ground and finally polished with emery papers. The diffraction profile was recorded with an automated X-ray stress measurement apparatus using a PDP 11/23 computer. The calculation of **B** and $I_m$ was completed at almost the same time that the X-ray intensity measurement was completed. In these experiments a diffractometer equipped with a Ω type goniometer (CGR/GS 2000), a position sensitive detector, PSD, and pulse height analyser (INEL) were employed. Chromium radiation ($\lambda_{K\alpha}$=0.22895 nm; 25 kV, 22 mA) was employed to examine the {211} peak (2θ= 156°). The irradiated spot on a specimen was 2 mm in diameter. Hardness was measured on a Vickers hardness tester with a load of 300g. The results of the hardness measurement are based on an average of five hardness values.

## RESULTS AND DISCUSSION

Line-width observations are nearly always made in back-reflection, whether by a photographic technique or by the diffractometer. It is then necessary to remember that the observation applies only to a thin surface layer of the specimen. Some automatically recorded profiles of the 211 lines are shown in Figure 5. The Figure also illustrates back-reflection pinhole patterns and microstructure of specimens corresponding to the profiles. The diffraction pattern in (c) exhibits the broad Debye line produced by quenching. This line becomes narrower for tempered specimens and mark-
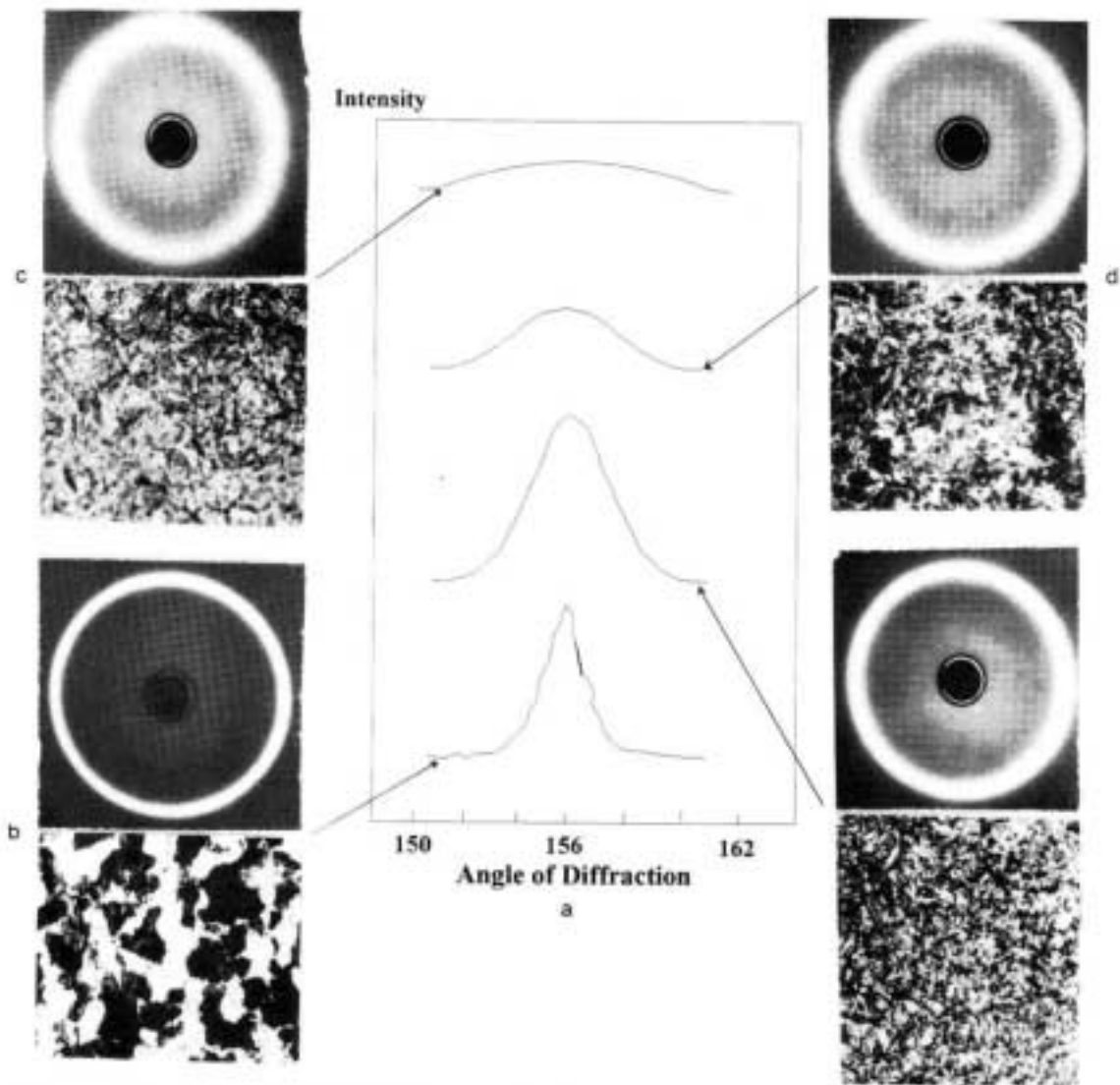
position makes the determination of the integrated intensity, peak level, and the width of the diffraction line possible. The above mentioned parameters $I_m$, D and $\varepsilon_i$ are associated with the distribution of the crystallographic defects when there is a microstructural change or a mechanical state modification. This current study mainly concentrates on one parameter, the diffraction line breadth, to follow the microstructure evolution in relation with the hardness values for quenched and annealed steels. However a few profiles are analysed for distortion measurement.

**Figure 5.** Line sharpening by tempering (a); back-reflection pinhole patterns and microstructure of (b) annealed specimen, (c) quenched specimen, (d) tempered at 200°C, (f) tempered at 600°C.

edly narrower for annealed specimens, as shown in (b). It is known that the width of the diffraction line depends upon the microscopic residual stress and dislocation morphology. It is also known that the heterogeneity of the substructure of low temperature martensite is rather high and there may exist microscopic residual stress peaks so that the width value is usually large.

Figure 6 shows a variation of the integral line breadth $I_w$ as a function of hardness $H_v$. Figure 7 presents the evolution of the line breadth B against hardness $H_v$. One can notice that the line breadth $I_w$ or

B increases linearly with the increasing hardness whatever the steel chemical composition.

From the results, the value of surface hardness is given by the following equations:

$$H_v = 43.331 + 107.042 \ B \qquad (7)$$

$$H_v = 32.386 + 0.986 \ I_w \qquad (8)$$

From the same results, the value of line breadth can be obtained by the following equations:
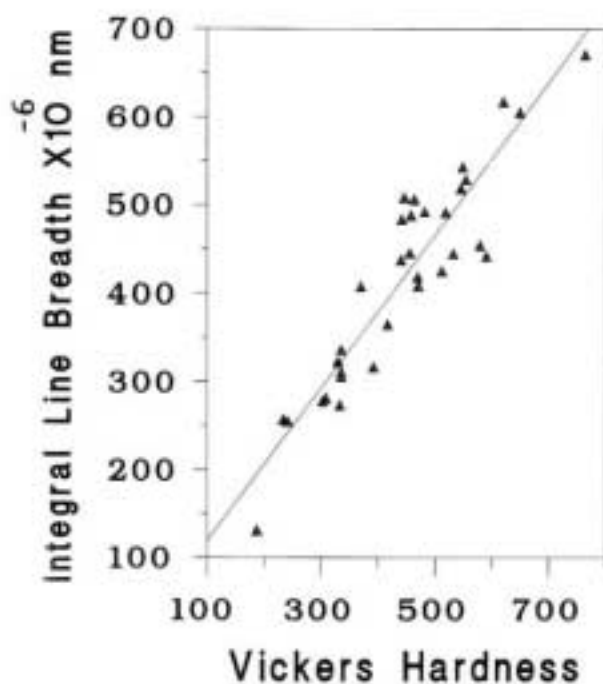
$$B = 0.034 + 0.008 \ H_v \qquad (9)$$

**Figure 6.** Variation of the integral breadth of the diffraction line with the Vickers hardness.
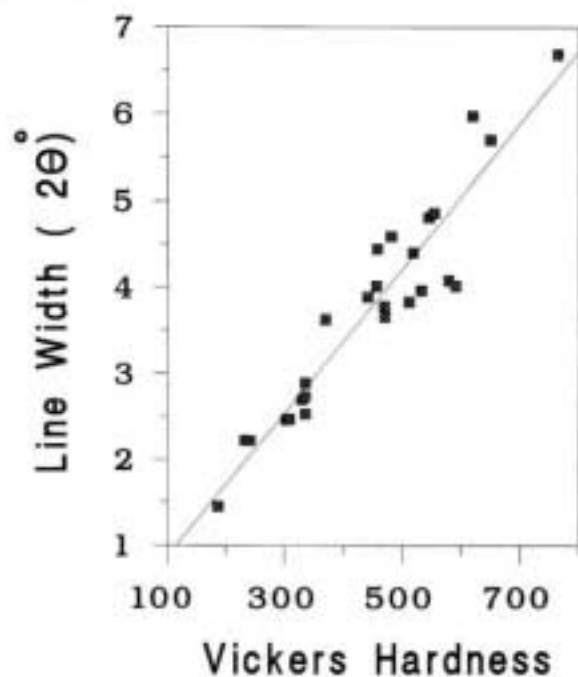


**Figure 7.** Variation of the width of the diffraction line with the Vickers hardness.

$$I_w = 32.262 + 0.867 \, H_v \qquad (10)$$

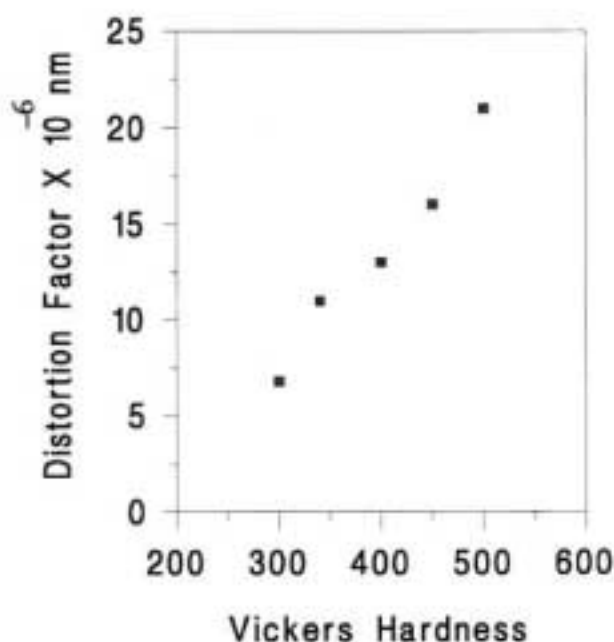In the above equations, B is in degree. $H_v$ is the



**Figure 8.** Distortion factor as a function of Vickers hardness.

Vickers hardness number and $I_w$ is $10^6$ times of the integral breadth in nm.

Figure 8 gives a linear relationship between distortion factor and the material hardness.

The saturation effect of carbon atoms and the existence of a high density of dislocation due to the quenching treatment in studied steels caused a high density of crystallographic defect in the specimens. Therefore the diffraction line is more broadened. Annealing treatment has two opposite aspects:

(a) the restoration of the microstructure and the carbon atom migration from the solid solution leads to a decrease in the defect density which reduces the line breadth;

(b) at the same time, the formation of carbide in the matrix creates the distortion.

Both of these two processes are thermally activated. According to a previous study [15], the X-ray diffraction technique is rather sensitive to changes in the defect density and their distribution, but not to the precipitation effect. Thus, it is the microstructural restoration and carbon atom migration that are studied

by diffraction line breadth.

Therefore, by using the diffraction line breadth, we can estimate, non-destructively, the hardness values, the distortion and the dislocation density for quenched and annealed steel specimens. In this study, small specimens was used, but for the field or a factory environment and for larger pieces portable equipment such as: RIGAKU, CETIM, PARS [16] and SET-X [17, 18]- can be employed.

## CONCLUSIONS

X-ray diffraction line broadens considerably when steels change into martensitic structure on quenching. The width of diffraction lines on various quenched and tempered steels increased linearly with increasing hardness according to some empirical equations. Hence, the X-ray diffraction technique may be employed for a rapid and nondestructive measurement of hardness of hardened steel. In fact, due to a very little penetration of X-ray in steel, in this way, one can measure superficial hardness. For instance, the penetration of Cr $K_\alpha$ radiation in steel is about 6 µm. Therefore, this technique may be useful for hardness measurement on thin films. By X-ray diffraction, one can also obtain some information on the material's characteristics or the microstructural evolution induced by heat treatments.

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