

Role of X-ray Diffraction in Evaluating Component Integrity

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ABSTRACT

X-ray diffraction (XRD) is the most powerful tool for non destructive study of the fine structure of matter. A substance always produces a characteristic diffraction pattern, which is the basis for chemical analysis as that substance actually exists and not in terms of its chemical constituents. Both qualitative and quantitative phase analysis are possible, as explained with reference to determination of retained austenite. XRD is also used for measuring the residual stresses (macro as well as micro) based on the shifting and broadening of diffraction peaks respectively. The elastic strains are converted into stresses with the help of X-ray elastic constants. A method for evaluating XEC is described. New developments based on these principles are also outlined.

INTRODUCTION

The diffraction of X-rays by matter is the basis of a unique scientific tool. Today, X-ray diffraction is used not only for structure determination of crystal but for such diverse applications as chemical analysis, stress measurement, study of phase equilibria, particle size, micro strain measurement and determination of orientations in a polycrystalline aggregate. It is proposed to describe the most useful details of some of these applications for evaluating component integrity. These techniques can be used for failure analysis, remaining life assessment and ways for life extension can be suggested based on the interpretation of data obtained.

RESIDUAL STRESS (RS) BY X-RAYS

Residual stress measurement in components can reveal the stress patterns produced by, and provide analysis of, the processing parameters causing the stresses. Residual (internal) stress occurs when portions of specimen tend to assume dif-

ferent volumes and the process is counteracted by cohesive forces. RS arises due to elastic response of a material to an inhomogeneous distribution of non elastic strains. The primary origin lies with virtually all forms of manufacturing and fabrication processes, in-service modifications, accidental overloading and even assembly operations. Residual stress as well as service loading stresses are prime contributors to overall structural performance. The unrecognized presence of an undesirable RS field can make the difference between reliable product performance and product failure especially when service loads are cyclic or when environment is conducive to stress corrosion cracking. Conversely, introduction of a desirable stress field can turn a product failure predicament into a rewarding condition of product reliability. The analysis and prediction of RS by theoretical approach is a numerical exercise on idealized systems and is not practical. The engineering understanding is normally obtained through experimental methods.

The X-ray diffraction provides the best researched method for determining RS. It can separate the macro and micro components without knowledge of the unstressed lattice parameter. The two types of RS are based on the scale of their existence or influence within a material. The macro RS exist over a few grains and is measured as the displacement of a diffraction line which is proportional to the lattice strains of the irradiated grains of polycrystalline material. The micro RS exists in one grain only and causes a general broadening of the diffraction lines. The diffractometer method is faster and more precise than the photographic method and can be used for accurate determination of RS even in case of broad peaks produced by quenched and tempered steels.

XRD methods offer a number of advantages compared to other methods for the measurement of near surface stresses. XRD methods are based on linear elasticity, in which the RS is calculated from the strain measured in the crystal lattice and are not usually significantly affected by material properties such as hardness, degree of cold work or preferred orientation. XRD methods are capable of high spatial and depth resolutions and can be applied to a wide variety of sample geometries of most polycrystalline materials and are non destructive on the surface. Any surface measurement is an exponentially weighted average of stress at surface and in layers immediately beneath it⁽¹⁾. The majority of failures of industrial components initiate from the surface where the stress and the ambient conditions are more stringent than in the interior. It is possible to measure absolute value of stress in individual phases, localised stresses without the stress free sample. XRD can be a more powerful diagnostic tool because of the additional information it provides on the metallurgical condition of the component. The data obtained using ψ^2 technique can be directly related to grain size, preferred orientation, surface cold working or hardness and process changes among other factors.

The accuracy of the stress value depends upon the measurement of the peak position 2θ i.e. the shift in diffraction peak due to stress as the sample is tilted at different angles ($\pm\psi$) to the incident X-ray beam, fig. 1. Thus, in the diffractometer method, a series of measurements can be made with the planes of the atoms oriented at different angles with respect to the sample surface, and hence the surface stress. Multiple exposure technique⁽²⁾ gives statistically superior accuracy and also indicates the presence of texture, stress gradient and shear stress. A straight line relationship is observed in the plot between peak shift and $\sin^2 \psi$ and the slope is given by

$$m = \frac{(1 + \nu)}{E} \cdot \sigma_{\phi}$$

where E and ν are Young's modulus and Poisson's ratio for the particular crystallographic plane used. Consequently, the stress in any chosen direction ϕ on the surface of the specimen is

$$\sigma_{\phi} = \frac{E}{(1 + \nu)} \cdot \frac{\cot\theta}{2} \cdot \frac{\Delta 2\theta}{\sin^2 \Psi} \cdot \frac{\pi}{180} \quad (\theta \text{ is Bragg angle})$$

Since the angle ϕ does not occur in the equation, the direction of the stress can be made to coincide with the longitudinal, transverse, circumferential or any other direction. The magnitude and direction of the principal stress can be determined by making four measurements at the normal spacing d_0 and three inclined spacings d at azimuth angles of ϕ and $\phi \pm 60^\circ$. Due to equilibrium conditions, biaxial stress at free (unloaded) surface and triaxial stress inside material is possible. In case of triaxial stress analysis such as depth gradient, information can be obtained from diffraction data at different tilt angles. For best accuracy, at least two, preferably three rotation angles (ϕ) and 14 inclination angles (ψ) up to 60° are required to be taken up for accuracy.

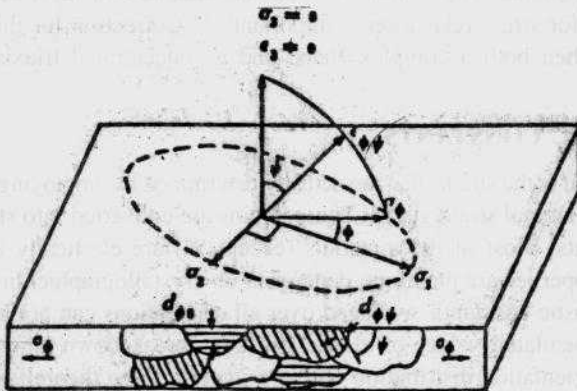


Fig. 1 : Plane Stress at a free Surface

Errors associated with Stress Determination by XRD

XRD method assumes an infinite number of randomly oriented grains. Non linear curve of peak shift vs. $\sin^2\psi$ mean the following :

- * Undulating curve could mean large grain size, preferred orientation or rough surface. If intensity at each ψ is uniform, then the surface is rough. Translate sample by 1/2" and repeat measurement in the same direction. If the integrated intensity vs. $\sin^2\psi$ pattern is the same, then texture is present. If the pattern is not similar then large grain size is the culprit for the undulations. Improvement can be obtained for large grain size by sample oscillation to cover larger areas. In a textured or plastically deformed material, use of high multiplicity planes obtainable with a shorter wavelength X-rays may have a higher probability of being oriented in a random manner and give better results.
- * The presence of splitting in positive and negative ψ profiles in curve of peak shift vs. $\sin^2\psi$ indicates that in addition to the strains in the plane of the surface, the shear strains are finite within the irradiated volume. The sample can be rotated by 180° , if the -ve and +ve ψ angles switch position, then the presence of shear stress is confirmed. In case there is no switch, the specimen is not aligned properly or there is a focussing circle error.
- * The curvature in the strain vs $\sin^2\psi$ plot shows the presence of different stress at different depths. Non destructive surface RS can not be corrected for errors caused by penetration of the X-ray beam into a varying stress field. Therefore, surface results must be interpreted with caution. The layer removal method can be used for stress distribution. Removal of material containing internal stress, is in general, associated with a redistribution of all stress components. The method of removal, thickness of layer removed and correction for stress relaxation is important^(3, 4). Correction for this effect is difficult when both a complex shape and a concentrated triaxial RS state occur.

THE ELASTIC CONSTANTS

By diffraction, it is the strains that are actually determined by employing interplanar spacing as an internal strain gauge. These strains are converted into stresses with elastic constants. Most of the materials (except W) are elastically anisotropic. The elastic properties are therefore, dependent on crystallographic direction. The mechanical elastic constants averaged over all orientations can not be used. All theoretically caculated values of elastic constants break down when there is a non-random orientation distribution within a material. The theoretical values of X-ray Elastic constants (XEC) may not agree with experimental ones because of

dependence of XEC on i) composition and second phase components, ii) grain size⁽⁵⁾, iii) microstructure⁽⁵⁾, iv) deformation and v) heat treatment. The magnitude of these effects depend on the hkl reflection employed. XRD is a powerful tool to determine the effective elastic constants of the material, even when it is inhomogeneous.

The experimental determination of XEC can be done on a rectangular beam specimen with strain gauges and generating the calibration curves on dead weight loading in a 4-point bend fixture, fig. 2⁽⁶⁾. From a knowledge of applied load, sample dimensions and moment arm of the 4-point bend fixture, the actual stress in the outer fibre of the sample is calculated. On the diffractometer, the fixture is clamped to minimize displacement errors. The lattice spacings of the selected set of planes is measured 5/6 times at ψ angles of 0 and 45°, at the same stress levels as used in calibration by loading. The peak shift is calculated between $\psi = 0$ and 45° for each measurement and plotted against the applied stress. The slope of the line m is determined by least square method. The elastic constant is

$$\frac{E}{(1 + \nu)} = m. \sin^2 \psi. d_0$$

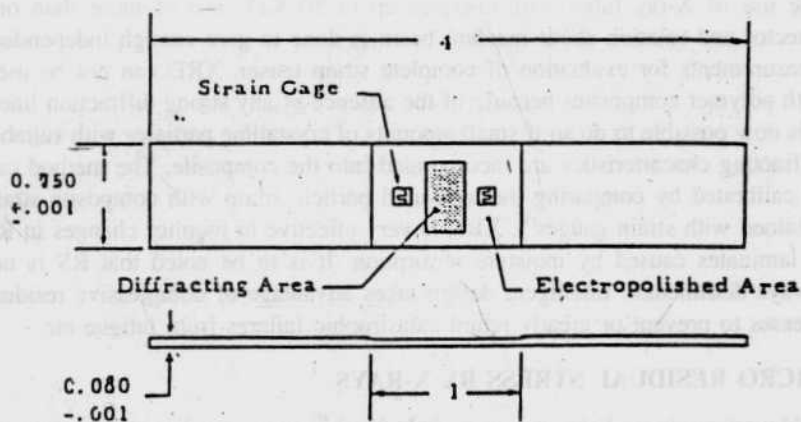


Fig. 2 : Schematic Diagram of Four Point Bending Sample for Determination of XEC

NEW DEVELOPMENTS IN STRESS MEASUREMENT

The lab models of XRD have restriction of sample size, are time consuming and have limited capacity for stress determination on actual industrial components. Light weight and compact, portable X-ray stress analysers have a mechanical

coupling between the miniature X-ray tube and the counter, the specimen being self supportive and independent. The use of position sensitive detectors reduce the data gathering and analysis times. The peak shift determination between two measured intensity curves is done efficiently and accurately by cross correlation function. This method is statistical which determines the relative peak positions. The results are unaffected by peak splitting, irregular background variations, scatter and even by slight adjusting errors of the goniometer. This is especially useful when for a given stress state, shifts of diffraction profile are smaller for larger Young's modulus e.g., ceramics requiring precise line shifts. The XRD method can now be used on even larger, irregular shaped components in different environments. It is used for process and quality control apart from failure analysis. Special hardware facilities for displacement and rotation of samples are also added. It can be used to study the stress profiles below the surface by slope etching method (single polishing operation to remove a wedge of material), making automatic stress depth analysis possible.

Stress at operational temperatures can be measured and phase changes examined in-situ with position sensitive detectors and a high temperature environment chamber. In-situ X-ray measurement of RS relaxation at elevated temperature during heating, holding and cooling stages of a tempering treatment can be done.

The use of X-ray tubes with energies up to 50 KeV, use of more than one detector and rotation about incident beam is done to give enough independent measurements for evaluation of complete strain tensor. XRD can not be used with polymer composites because of the absence of any strong diffraction lines. It is now possible to do so if small amounts of crystalline particles with suitable diffracting characteristics are incorporated into the composite. The method can be calibrated by comparing the measured particle strain with composite strain obtained with strain gauges⁽⁷⁾. XRD is very effective to monitor changes in RS in laminates caused by moisture absorption. It is to be noted that RS is not always detrimental. Intelligent design takes advantage of compressive residual stresses to prevent or greatly retard catastrophic failures from fatigue etc.

MICRO RESIDUAL STRESS BY X-RAYS

Cold work produces dislocation arrays which subdivide original crystallites (grains) into much smaller coherently diffracting domains. The same dislocations also produce tensile and compressive strains within the domains. The size and the micro strain are the main factors responsible for peak broadening. Micro RS arises during large scale plastic deformation due to inhomogenous nature of material because of i) grain boundary effects, ii) anisotropy of grains, iii) crystallographic orientation of grains and iv) constituents having different mechanical properties. The lattice planes usually become distorted such that spacing of

any particular (hkl) set varies from one grain to the other or from one part of grain to another. The sharp diffraction line is replaced by the sum of sharp lines, each displaced from the other.

When only relative numerical results are required, simple analytical procedures for deducing the breadth of pure diffraction profiles can be used. The broadening B by particle size alone ($< 0.1 \mu\text{m}$) was determined at half its maximum intensity and given by $0.9 / t \cos \theta$, where t = diameter of crystal particle, (Scherrer). Some investigators take integral breadth for B as the integrated intensity divided by the maximum intensity. This relation is used to find particle size of loose powders (strain free) and not size of individual grains in a solid aggregate. The broadening b (above instrumental broadening) produced by non uniform micro strain is obtained by $\sqrt{2}\theta = -2Vd \tan\theta/d$. However, the separation of size and strain broadening by the method of integral breadths depends upon explicit assumptions as to the shape of the broadening profile due to each effect. Fourier transform method does not depend on the acceptance of specific models of size and strain broadening. Fourier distribution (Stoke's method) and iterative folding Ergun⁽⁸⁾ of the measured intensities of diffraction lines yield information about the mean crystallite size of the coherent crystallite domains, distribution of crystallite sizes, nature and extent of lattice imperfections (elastic distortion) and/or deformation faulting. Warren and Averbach method⁽⁹⁾, requires a standard sample as a reference with composition similar to the unknown that shows no evidence of distortion due to crystal size or micro stress. The line profiles of two peaks of the standard (within $\pm 50^\circ$ of the two peaks of the unknown sample) are measured. The broadening of the line profiles of the standard are used for correction due to instrumental broadening. The interplanar distance are measured with at least two independent diffraction orders for the unknown sample. The observed line profiles, corrected for Lorentz polarization, background, $K\alpha_2$ contribution and instrumental broadening are expressed as Fourier series. The analysis of the Fourier coefficients of the standard and the unknown discloses both particle size and strain, without any prior assumption as to the existence of either. The real part of the Fourier coefficient of the produced lines is then graphed as a function of the coherence length. For the real part of the coefficients, a separation yields the contribution of the particle size and micro stress, since the size coefficient is independent of the order of the hkl plane and micro strain is a function of the order.

OTHER RELATED APPLICATIONS OF X-RAYS

Paterson⁽¹⁰⁾ showed that the fourier coefficients of the line profile could also disclose the presence of stacking faults caused by cold work. As the cold work increases, the broadening increases, until in the back reflection region, the $K\alpha$ doublet overlaps and becomes unresolved and can be taken as evidence of cold

work. The X-ray diffraction also shows the effects of recovery, recrystallization and grain growth. During recovery, both macro and micro RS reduce in magnitude, but the strength and hardness remains high. Much of this stress relief occurs due to polygonization, which can occur in individual grains of an aggregate just as in a single crystal. During recrystallization, new grains form, residual stress is practically eliminated, and strength and hardness decrease abruptly. Since non uniform strain due to residual micro stress is the major cause of line broadening, we find that broad diffraction lines characteristic of cold worked metal sharpen during recovery. The lines attain maximum sharpness when recrystallization takes place. During grain growth, the lines become increasingly spotty as the grain size increases. Conversely, when an annealed metal with sharp diffraction lines is progressively deformed, the lines progressively broaden and hardness increases. The hardness can be accurately measured from the breadth of its diffraction lines. This relation can be established experimentally for each metal/alloy and serves as a rapid method to determine RS.

XRD can be used to characterize wear and mechanism of boronized steels⁽¹¹⁾. The changes in surface RS and half value breadth during wear process and relation between changes in X-ray penetration and structure are investigated. X-rays are successfully applied to detect wear damage accumulation⁽¹¹⁾. X-ray fractography is developed for investigating plastic zone of fatigue cracks. Study of material strain and work hardening through displacement and broadening of X-ray diffraction peaks provides information on fatigue process. RS measurement at different depths is used to determine plastic zone of fatigue crack⁽¹²⁾.

RETAINED AUSTENITE BY X-RAYS

Retained austenite (RA) with a near random crystallographic orientation is found in the microstructures of heat treated low alloy, high strength steels with medium (0.40 wt. %) or higher carbon contents. The presence of RA may not be evident in the microstructure. The transformation of RA to martensite during service can affect the performance of steel. Quantitative microscopic examination is quite satisfactory when austenite content is fairly high, but becomes unreliable below about 15% austenite in many steels. The X-ray method is quite accurate in the austenite range of greatest practical interest. The direct comparison method for quantitative phase analysis by XRD does not require a sample of the pure phase because the required reference line comes from another phase in the mixture. It is usually impossible to obtain a reference sample of pure austenite, or of known austenite content, of the same chemical composition as the austenite in the unknown. The volume percent of retained austenite (1% and above) in steel is determined by comparing the experimental integrated X-ray diffraction intensity of ferrite (body centred cubic) and austenite (face centred cubic) phases with theoretical intensities⁽¹³⁾. This method is applicable to steels with near random

crystallographic orientations of ferrite and austenite phases. Chromium radiation is chosen to obtain the best resolution of X-ray diffraction peaks for other crystalline phases in steel such as carbides. No distinction is made between ferrite and martensite phases because the theoretical diffraction intensities are nearly the same. Mo radiation is used to produce numerous X-rays diffraction peaks. Substantial alloy contents in steel cause some change in peak intensities. In case of steels with total alloy content more than 15 wt.%, care is needed for application of this method. It is possible to calculate the theoretical correction factors for changes in volumes of unit cells for austenite and ferrite resulting due to variations in chemical composition.

The sample cutting and surface preparation should be done with minimum heat effects and plastic deformation to avoid partial decomposition of both the martensite and austenite. The X-ray diffraction peaks from other crystalline phases such as carbides must be separated from ferrite and austenite peaks. Three austenite peaks (111), (200) and (220) and three ferrite peaks (110), (200) and (211) can be obtained with Cr radiation. Four peak ratios (integrated intensities) of the resolved ferrite to austenite peaks are adequate to determine the RA content V_γ of near randomly oriented samples.

In a sample, integrated intensity I of all diffraction peaks for each phase is proportional to the volume fraction V of that phase.

$$I_\alpha^{hkl} = K \cdot R_\alpha^{hkl} \cdot V_\alpha / 2\mu$$

K is a constant independent of the nature of the sample. The parameter R is the structure factor and is proportional to theoretical integrated intensity. It depends on hkl , θ , crystal structure, composition of the phase and can be calculated from the first principles. μ is the linear absorption coefficient. For any pair of austenite and ferrite peaks,

$$I_\alpha^{hkl} / I_\gamma^{hkl} = [(R_\alpha^{hkl} / R_\gamma^{hkl}) (V_\alpha / V_\gamma)]$$

$$V_\alpha + V_\gamma = 1 \quad (\text{No carbide})$$

The absolute value of RA content can be estimated from a measurement of number of ferrite and austenite intensity ratios and calculation of R -value ratios using these equations. If carbides are present, the volume fraction of carbide should be determined from chemical extraction or quantitative metallographic or by diffraction methods. If we measure integrated intensity I_c of particular cementite line and calculate R_c , then V_γ / V_c can be found from

$$V_\alpha + V_\gamma + V_c = 1$$

With filtered radiation, the minimum detectable limit is about 2 volume % austenite, and with crystal monochromated radiation probably about 0.2 %⁽²⁾. There

is no other independent test method to determine an accepted reference value for retained austenite.

CONCLUSIONS

X-ray diffraction is shown to be the most fundamental and best researched technique to furnish absolute values with high precision of macro residual stress, X-ray elastic constants, micro strain, particle size and retained austenite in a non destructive way. Portable residual stress analysers are now available for conducting on site and in-situ jobs for evaluating residual stress and retained austenite. X-ray diffractometer is a costly equipment and can be justifiably employed if the application so demands.

REFERENCES

1. V. M. Hauk, E. Macherauch, "Advances in X-ray Analysis", 27, 81 (1984).
2. B. D. Cullity, "Elements of X-ray Diffraction, 2nd Ed. Addison-Wesley, USA (1978), p. 414.
3. Z. A. Kang, J. B. Li, Z. G. Wang, J. of Testing & Eval., 22, 217 (1994).
4. I. C. Noyan, Met Trans., 14A, 249 (1983).
5. G. Faneinger, J. Soc. Mater. Sci, Japan, 19, 42 (1970).
6. P. S. Prevey, "Advances in X-ray Analsis", 20, 345 (1977).
7. C. Balasingh, V. Singh, Bull. Mater. Sci., 20, 325 (1997).
8. S. Ergun, J. Appl. Crystallgr., 1, 19 (1968).
9. B. E. Warren, B. L. Averbach, J. Appl. Phys., 21, 595 & 599 (1950) and 23, 497 (1952).
10. M.S. Paterson, J. Appl. Phys., 23, 805 (1952).
11. N. Mori, T. Endoh, M. Kawakami, J. Soc. Mater. Sci. Japan, 34, 1093 (1989).
12. R. N. Pangborn, S. Y. Zamrik, "Non destructive Characterization of Materials IV", Ed. C. O. Ruud et. al., Plenum Press, NY (1991) p. 259.
13. ASTM Designation E 975-95.