

LATTICE DEFORMATION MEASUREMENTS VIA “ON SITE X-RAY DIFFRACTION”

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The present paper is about the estimation of lattice deformation from data collected from manufactures directly on site. The aim here is to give evidence that the concept of the Mean Equivalent Lattice (MEL), when applied to “on site X-Ray Diffraction” is the basis for a reliable qualification of the material rheology to external solicitations. Such method allows for the identification of lattice deformations without resorting to the computation of the residual stress with using the elasticity constants (i.e. tensile, shear and rigidity constants E , μ , ν); these elasticity constants descend from the classical theory of solid mechanics, where the continuum mechanics and the material isotropic model are the fundamentals. Any MEL deformation is instead related to the variation of the d-spacing among lattice planes which are connected to the anisotropic atomic arrangement. So the macroscopic scale is constituted by a number of MELs and related boundaries. The recent on site X-ray diffraction technology may offer effective and easy solutions, with a significant impact on reliability of results, simplification, economy and time consuming.

Key words

Lattice deformation measurements, elastic and plastic deformations, x-ray diffraction, residual stress, mean equivalent lattice, quality control process.

I. INTRODUCTION

The material functional properties and their rheology to stress are in general determined according to macroscopic parameters. Nanometric description, macroscopic functional and rheology properties of materials still offer a wide context of discussion and research; this context is open especially in those sectors, where the familiarity is sensitive to scales higher than nanometers. In these sectors, the adoption of the classic theory of elasticity reaches a well consolidated fulfillment. The classic theory of elasticity has the assumption of continuity of the space around the investigated element from where the deformation vector is observed. Nevertheless, when considering microscopic atomic aspects, said fulfillment vanishes because of the lattice space anisotropy, which has intrinsic discontinuities. In fact, as a consequence of the atomic arrangement in the lattice space, stresses cannot be distributed homogeneously in all the crystallographic directions.

The standard EN15305 rules the determination of residual stress; when using XRD, the suggestion is given to resort to the empirical evaluation of the elasticity constants (*XEC*) as a minor risk of mistake than using values that are provided for other methods than X-ray diffraction analyses. The question rises to identify concepts and methods which are consistent with the qualification of material without resorting to the calculation of residual stresses and limiting the evaluation to the lattice deformations.

Among several diffraction methods, “on site” XRD is today the only available for data collection without contact on site, thus limiting any artificial manipulation of the specimen. The method and related technology (Berti *et al.*, 2008; Berti and De Marco, 2010) are capable to detect surface thickness, with a sufficient penetration to determine the *d*-spacing and related variations having a reasonable physical meaning. This *d*-spacing set defines the Mean Equivalent Lattice (MEL) as the concept to qualify the material rheology to either external solicitations or self-equilibrating internal rearrangement. The MEL concept points out the *d*-spacing set as the physical observable through X-ray diffraction pattern without resorting to elasticity constants. This concept results in a significant impact on reliability of results, simplification, economy and time consuming.

II. LATTICE DEFORMATIONS AND MEAN EQUIVALENT LATTICE

The mathematical theory of X-ray powder diffraction is the appropriate development basis to define the micro/nano structural asset in terms of physical observables. The tolerances $\Delta d/2$ of Figure 1(a) and Figure 1(b) are considered as a reasonable value to identify the atomic positions in a cubic lattice, considered as an example, either in its ideal regularity (i.e. elastic oscillations) or including a d' shift term (i.e with an additional effect) which is external to the tolerance limits and consequently dynamically plastic in fact. Among the possible descriptions of real lattices, the superposition of “*ideal*” [Figure 1(c) left] and “*paracrystalline*” [Figure 1(c) center] lattices is the effective one (Hosemann and Bagchi, 1962); when considering random deviations from the average position of nodes, the real lattice is described by “*Mean Equivalent Lattice*” (MEL) (Berti, 1995), i.e. the volume where the real lattice is equivalent on average to the ideal one. This equivalence applies within the $\Delta d/2$ tolerances which are intrinsically anisotropic as the d -spacing from where these tolerances are originated. The sequence of Figure 1 reports the planar projection of a three dimensional cubic arrangement taken as an example. In this projection we miss the three axial elliptical shape of the tolerance volume in the direction $[111]$; so the projected tolerances are reported as circles in Figure 1 (a) and Figure 1 (b).

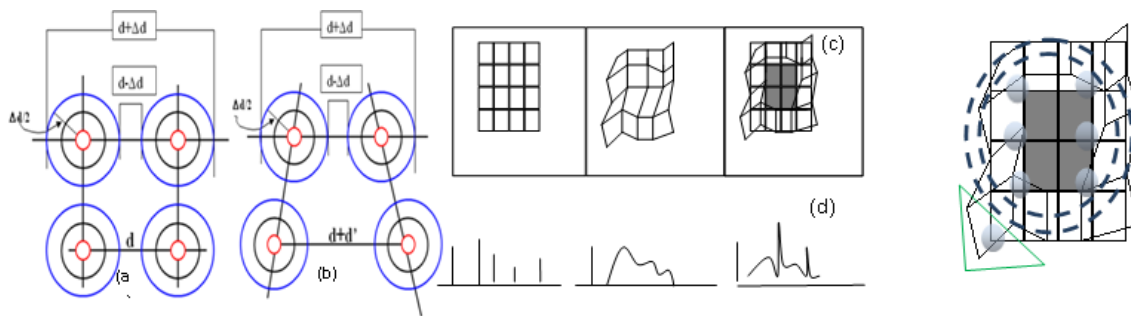


Figure 1[(a), (b), (c), (d), (e)]. Lattice frameworks and position of nodes with related tolerances. In (a) is reported the case of the ideal framework, in (b) the case of distorted framework, in (c) the sequence from left to right shows the ideal crystalline, the ideal paracrystalline lattices and the schematic arrangement of a typical real lattice named here the Mean Equivalent Lattice (MEL) where the ordering range may be considered as a quasi-long within the experimental errors (i.e. wide microstructural size). In (d) there are schematic examples of diffraction signals expected from the above mentioned lattices. In (e) the light blue balls indicate the uncertainty on the lattice node positioning and the dashed lines around MEL indicate that the microstructura core is surrounded by short ordering range where the complication on microstructural size or amorphous arrangements introduces dynamic deformation of lattice (i.e. the intrinsic mechanisms of creep and related energy activation).

The Bragg equation shall consider these tolerances on d , and λ which will results in uncertainty on the identification of the diffraction line position θ . This uncertainty is accidental in nature and shall be considered as additional term to not removable systematic instrument contributions (i.e.

calibration) (EN13925-3, Berti, 2001). When the accidental uncertainty on λ is negligible one obtains the known cotangent relation (Parrish, 1963).

On continuing with the superposition approach, we can consider the ideal atomic arrangement as the rigid body approximation which obeys the Bragg equation in its geometrical formulation i.e. the coherent domains = crystallites EN1330-11. When considering the approximation with random elastic oscillations governing the position tolerances (or uncertainty), the ideal paracrystalline lattice overlaps the ideal atomic one as in [Figure 1(c) right]. The schematic trend of signals related to (a), (b) and (c) of Figure 1 are reported in Figure 1 (d). These overlapping effects result in MEL boundaries; i.e.:

- a. zones where the d-spacing coherence vanishes the dashed rings in Figure 1(e);
- b. zones where the oscillation are no longer confined within the expected tolerances thus inducing plastic deformation which is dynamic in fact (e.g. the triangles in Figure 1(e)).

The addition of the ideal and rigid lattice with the paracrystalline and vibrating lattice implies the ensemble vibration of atoms influence on other MELs at either short or long range. As a consequence any small real lattice volume is confined in the limited space described in terms of MEL and smaller than the crystallite volume. Deformations and boundaries are then naturally introduced [shadow of Figure 1(c) left]. When these boundaries are vibrating with oscillations wider than $\Delta d/2$ they becomes plastic deformations in facts. As a consequence, the term “defect free” is inapplicable to real material lattices.

For all aforementioned a reference value of d is required where the Δd is known to be considered as the benchmark or “the Reference Value Material (RVM)” from where ranking the deformation from unknown specimens (PrEN 13925-4).

A certain criticism arises when considering the elasticity and plasticity in conjunction to lattice deformations. This conjunction is frequently used to determine the residual stresses to qualify the material usability resulting in macroscopic effects of phenomena originated at the micro/nano-scale. The concepts of tensile, shear and rigidity constants (E , μ , ν) are a macroscopic description descending from the classical theory of solid mechanics, where the continuum mechanics and the material isotropic model are the fundamentals. Now it seems reasonable to consider the isotropic model as the cumulative effect from a number of randomly aggregated MELs.

From a different point of view, Hobart defines the specific stress of Peierls (σ_p) (Caglioti, 1974) in terms of the distance between two crystallographic planes normal to the dislocation line (c), the width of the dislocation (w) and a scalar quantity (R). This scalar quantity can be iteratively determined as a function of k , the transversal elastic constant between contiguous planes and α , the longitudinal elastic constant. In the Hobart description α and k can be calculated via relations taken from the continuum mechanics (E , ν) and μ respectively, i.e. the Young's and shear modulus, along with the Poisson's ratio. Other parameters required for the k and α calculations are the cell parameters a , b , c .

On continuing on the same approach of MELs a conceptual inversion shall be introduced in the calculation of the macroscopic quantities (E , ν) and μ in terms of said cumulative effects of the microscopic ones; then possibly α and k should be calculated from the whole diffraction pattern parameters.

III. EXPERIMENT

Lattice deformation may be used as a power method for quality control process of metallic rings via "on site" x-ray diffraction. "On site X-ray Diffraction" is intended to perform measurements directly from the specimens under investigation at their site.

Differently from the traditional in lab XRD, the instruments adapt themselves as much as possible to the manufactures under investigation. This new inspection concept allows for the following: 1) it extends the applicability of X-ray diffraction to specimens, independently of their size, weight and shape, 2) it merges the consolidated knowledge on X-ray diffraction with the new technological aspects (e.g. robotics, communication technology) (Berti, 2007).

Figure 2 shows the instrument for the experimental measures. Technical specifications are reported in Table I. XRD analyses were carried out on industrial metal rings for aeronautic engine supports, manufactured in "Inconel 718", a nickel based alloy hardened by precipitation. Three distinct rings (called ONE,TWO,THREE) were produced by using three intentionally distinct processes.

TABLE I. Equipment and performances of DifRob[®]

Size (cm)	220x100x100
Weight (Kg)	250
Power of Rx Tube (nominal) (W)	2200
Power of Rx Tube (effective) (W)	450
Spot size and shape	1 mm wide diameter and circular
Cooling	Liquid
Detector	Proportional Counter
Interface	RS232
Movement devices	Wheels



Figure2. DifRob[®], the prototype for on site XRD measurements developed by XRD-Tools

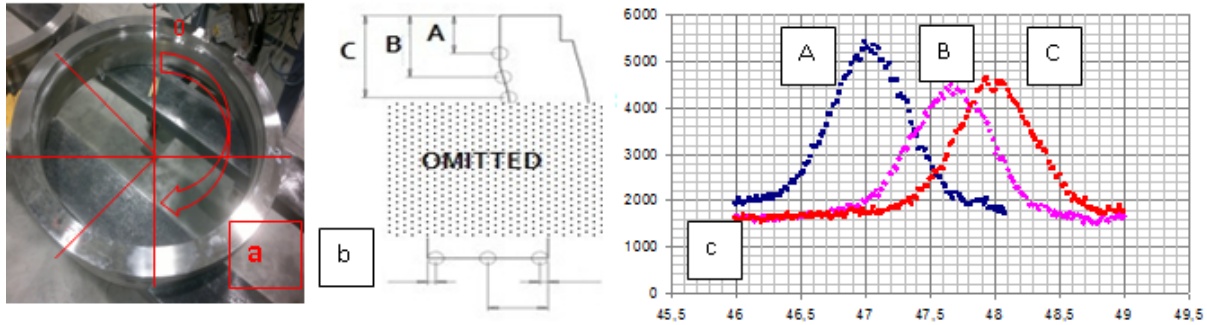


Figure 3[(a), (b), (c)]. The ring ONE of the three analyzed INCONEL718 rings (a) with radiuses at angles 0, 90, 180, 240, 270, 300 degrees. The other two rings were analyzed on 0°, 120°, 240°. ONE was taken as reference by chance and observed with more detail than the others. The cross section of said rings (b) where are locate the points A, B, C. Their relative distance is about 10 mm. They are the points where the data have been collected. An example of the variation of the diffraction line intensity distribution (c) in counts/sec. vs. θ in machine units.

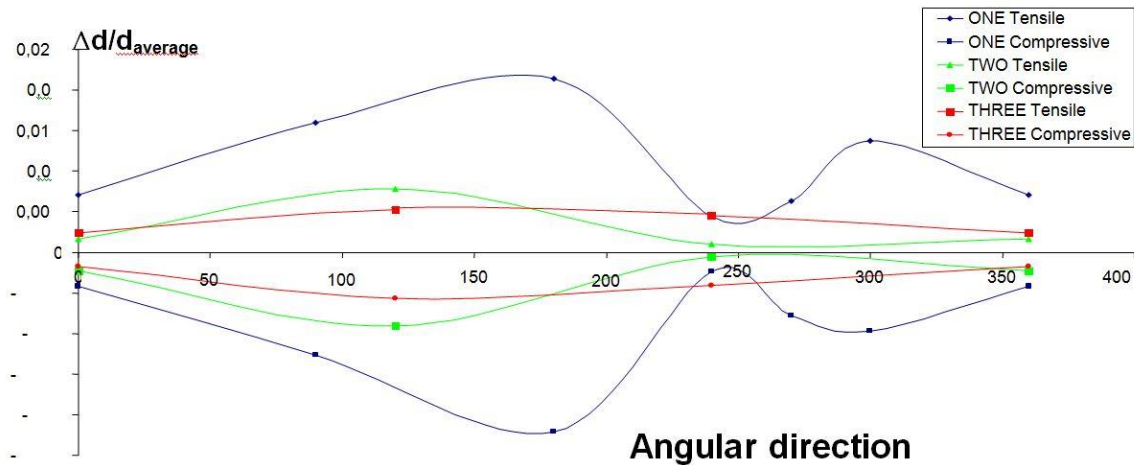


Figure 4. $\Delta d/d_{average}$ vs degrees of angular direction. For each specimen and each direction, the average value of d is calculated among A, B and C distances. Then, the maximum and the minimum values of d - spacing, recorded at any angular directions are considered. The quantities $d_{max} - d_{average} / d_{average}$ and $d_{min} - d_{average} / d_{average}$ are calculated, thus generating a compressive (negative) and a tensile (positive) curve for each ring. On observes here that ring ONE has been analysed with more details than other by chance on an higher number of angula directions than the two others. The THREE ring shows higher homogeneity of the relative deformation than TWO over the three observed points (0°,120°,240°). Although the observed point from ONE are a bit different in the angular directin from the two others. The internal unhomogeneity is evident and higher than from the two others. This observation implies that the qualification of the three rings can be ranked according to the homogeneity of the relative deformation. This relative deformation homogeneity is the effect of the homogeneous disttribution of the residual stress. This residua stress homogeneity id higher for THREE than for the other two rings.

The purpose of the study was for giving indications on the state of internal deformations of these components, in relation to the different manufacturing processes. Figure 3(a) shows one of these rings and the angular generation lines where the data have been collected. Figure 3(b) shows the section of said ring and the positions A, B, C, where data were collected. Figure 4(c) shows an example on the diffraction pattern from data collected in A, B, C respectively at the angular

direction 300 degrees of labeled “ONE” ring. Three analyzed rings had been produced with intentionally different methods and treatments:

- Ring one: Coarse grained structure (ASTM grain size $n=4$), no expansion effect experienced water quenched after solubilization.
- Ring two: Coarse grained structure (ASTM grain size $n=4$), expansion effect experienced, water quenched after solubilization.
- Ring three: Fine grained structure (ASTM grain size $n=7$), expansion effect experienced, air quenched after solubilization.

These production process was intentionally undelivered to the operator before starting the measurement experiment. So the ring ONE was selected as the reference by chance by the operator. On this specimen six special directions have been selected [Figure 3(a)] at 0° , 90° , 180° , 240° , 270° , 300° . On each angular direction, data have been collected on the ring side surface, at distances denominated “A”, “B” and “C” [Figure 3(b)], far from the side surface. The distance between the extreme points “A” and “C” is about 20 mm and “B” is more or less located at the intermediate position.

The direction selected for the ring TWO and THREE were selected at intermediate angular values 0° , 120° , 240° . For each of the three patterns, the elaboration of the observed data leads to the determination of the d -spacing, in a defined angular direction. These values [Figure 4] has been reported as $\Delta d/d_{\text{average}}$ vs. the angular directions. The diffraction lines at the positions 46,5 and 48,5 degree (θ) have been selected and pertaining to the lattice of the Nickel Chromium Inconel 718 alloy composition who investigate by the $\text{Cr } K\alpha = 2,291 \text{ \AA}$ radiation. This diffraction line is compatible with the plane $[111]$.

The variation of d -spacing obtained from the solution of the Bragg Equation has been estimated according to the following quantities:

$$d_{\text{max}} - d_{\text{average}} / d_{\text{average}} \quad (1)$$

$$d_{\text{min}} - d_{\text{average}} / d_{\text{average}} \quad (2)$$

d_{average} is calculated for any of the selected angular directions, as the average over the d -spacings calculated at the three selected points A, B and C.

Among the three d-spacing values, the minimum (d_{\min}) and the maximum (d_{\max}) are considered. Then the quantities (1) and (2) are calculated and plotted against the angular direction (Figure 4). The results are the curves obtained in Figure 4 for each ring.

IV. DISCUSSION

The paper focuses on the ability of the diffraction method to identify appropriate valued of the lattice deformation when using technologies for in service inspection or at least “on site”, i.e. at the site of the manufacturer or in the production line. The identification of said lattice deformation is quite important for its relation to the residual stress and related complications mentioned by EN15305.

The lattice induced deformations of materials determine a macroscopic calculable effect sometime used in the scale of Mega Pascal (MPa) to indicate the induced residual stresses. Figure 4 shows the effects of said induced residual stress over directional alignments of the analysed Inconel 718 rings. Said residual stress is defined as “Self-equilibrating internal stresses existing in a free body which has no external forces or constraints acting on its boundary” (e.g. EN 15305). Such definition implies that the atomic arrangement of boundaries is equivalent to a rigid body; the conceptual introduction of MELs implies that the atomic arrangement surrounding each MEL is subject to additional mean vibration (the d' of Figure 1(e) which is out of the tolerances $\Delta d/2$; it is a dynamic deformation which is plastic in fact. This plastic dynamic deformation is anisotropic; it nucleates the material creep intrinsic mechanism and the related propagation. See as an example the green triangles in Fig. 1 (e)

Fact is that many cases exist where the crystallite size plays a relevant role to modify the intensity distribution of the X-ray diffraction lines. So, when using X-ray diffraction the concept of plastic deformation shall include effects related to both no longer active external loads (i.e. residual stress) and vibrations wider than $\Delta d/2$. These two effects are no longer compatible with the concept of rigid body crystallites but respects the concept of MEL.

V. CONCLUSION

The experiment carried out here shows the effect recorded on the pattern from using original material with a different “grained structure”, which are probably connected with different MELs and the identical ideal crystallite sizes of Inconel. This implies that when considering the deformation effect revealed by the X-ray diffraction and caused by different original grained structure, this deformation results in different parameters of the MELs. So, the MELs are sufficiently representative of the lattice quality without providing efforts to determine the residual stress in the MPa scale.

From the microscopic point of view, the variations of the d -spacing have a qualifying meaning of the MEL feature which is connected to the residual (plastic) deformations and deviating from the crystallographic expected configuration.

The proposed technology based on DifRob® (Berti, 2007) results in a very effective one for collecting data directly from the industrial components on the manufacturer site. A new DifRob type diffractometer for measurement carried on site has been recently developed which improves the performances of the previous one of Figure 2. The improvement in the instrument mobility is also achieved through the reduced dimension of the whole diffractometer and equipments thus resulting in a more effective usability in the frame of the activities discussed in the present paper.

The new technology development for “on site” X-ray diffraction and the related conceptual consistence to real properties of material lattice will have a beneficial impact when the qualification of materials is faced with precise determination of residual stress. The ambiguities can be removed when rising from improper or erroneously use of the elasticity constants, which were not intended for being used with X-ray diffraction measures. The paper shows that it suffices the determination of relative deformation for the purpose of material qualification and production process qualification.

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Berti, G. (2007). “Diffractometer and Method for diffraction analysis,” US patent 7,260,178.

- Berti, G. (2001), "A method for routine comparison of XRPD measurements," *Powder Diffr.*, **16**, 1-6.
- Berti, G. (1995). "Detection and modelling of micro-crystallinity by means of X-ray Powder Diffractometry," *Adv. X-Ray Anal.*, **38**, 415- 412.
- Berti, G. and De Marco, F. (2010). "Fractional nanophotonics: On site NDT-XRD for Cultural Heritage and Environment," *Proceedings Convegno Nazionale sulle tecnologie fotoniche*, Pisa – Italy, 25-27.
- Berti, G., De Marco, F. and Nicoletta, A. (2008). "On site X-ray diffraction from distance for cultural heritage," in *In situ monitoring of monumental surfaces- Proceedings of the International Workshop SM08 27-29 October 2008*, edited by Edifir, Florence, pp. 409-414.
- Caglioti, G. (1974), *Introduzione alla fisica dei materiali* (Zanichelli, Bologna, Italy).
- Hosemann, R. and Bagchi, S. N. (1962). *Direct Analysis of Diffraction by matter* (North-Holland Pub. Co., Amsterdam).
- Parrish, W. and Mack, M. (1963). *Data for X-Ray Analysis* (Eindhoven, N.V. Philips' Gloeilampenfabrieken).
- prEN13925-4: "Non-destructive testing- X-ray diffraction from polycrystalline and amorphous materials- Part4: Reference Materials" – This label is still related to the WG10 of CEN/TC138 - Doc. 18 rev 5 still under study.
- UNI-EN 15305 (2008). "Non-destructive Testing – Test Method for Residual Stress analysis by X-ray Diffraction," CEN - AFNOR Paris. [<http://store.uni.com/magento-1.4.0.1/>] [accessed 12-Aug-2013].
- UNI-EN 13925-3 (2005). "Non-destructive testing- X-ray diffraction from polycrystalline and amorphous materials- Part 3: Instruments," – CEN - AFNOR Paris. [<http://store.uni.com/magento-1.4.0.1/>] accessed 12-Aug-2013.