

An X-ray Study of Textural Stresses in Two-Phase Alloys

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Internal or residual stresses of the textural stress class are discussed, with special attention to that type resulting from a difference in thermal expansion of two phases present in an alloy. X-ray stress measurements were made on various brasses and steels, and the results were found to be in reasonable agreement with the quantitative theory of F. László.

1. Introduction

The term "textural stress" will be used in this study in the sense adopted by E. Orowan in a symposium of the Institute of Metals in 1948 [1].¹ His most inclusive term, internal stress, is defined to be a stress existing in a body upon which no external force is acting. (Some writers use the term "residual stress" in this wide sense.) This field is divided into two classes. One class consists of body stresses, which arise when an external factor has affected different macroscopic parts of a body differently, even though the material of the body may have been quite homogeneous. The other class consists of textural stresses, which result from inhomogeneities of the material, even though the activating external influence may have itself been homogeneous. Under ordinary circumstances, where the grain size is small compared to the macroscopic dimensions of the body, these classes are closely equivalent to the widely used terms, macrostress and microstress, respectively. The former implies a stress system constant over many contiguous grains in a body, balanced by opposing stresses in other grains at some distance; the latter implies a stress system that varies from grain to grain or within a grain, and is balanced over short ranges.

The most detailed and quantitative theoretical analysis of textural stresses arising from the presence of two phases in an alloy is to be found in a series of four articles between 1943 and 1945 by F. László [2]. The four articles were reviewed in a nonmathematical manner by F. R. N. Nabarro in 1948 [3].

László discussed textural stresses arising from various sources, such as anisotropy of the grains and differences in thermal or mechanical properties, and the effect of external stress on the textural stress. Much numerical detail was presented with regard to iron alloys. The greater part of the work dealt with an involved mathematical analysis of textural stresses caused by the difference of coefficients of thermal expansion of two phases present in various proportions, and with various grain shapes. The solution was only approximate, of course, because of the simplifications that were necessarily made. In particular, three types of idealized grain shapes were assumed: Lamellar, with alternating layers of the two constituents, and compound spheres and compound cylinders, with coverings and cores of the two different phases. These compound blocks

were assumed to have surfaces free of normal stress and to pack together to form the body of material, although strictly speaking only prismatic or polyhedral shapes could do so. The formulas for the approximate values of the principal stresses in these units presented in part I were used in this study. In László's article, part II, the ideas were extended to other special cases, such as capped cylinders, and the physical rigor was somewhat enhanced; but the results there were presented in the form of systems of simultaneous equations of high order, and the value of the much more difficult solutions of these equations was questionable in the light of the experimental margin of error anticipated in the study at hand.

László suggested four technological effects in which such textural stresses might be of importance. Yield, the formation of cracks, fatigue, and corrosion, the last three of which are most likely to be significantly influenced by such stresses. Although the mathematical erudition and the extensive calculations from theory displayed in these articles were very impressive, no attempt at verification by X-ray measurements was made. On the contrary, it was specifically stated by László, in part I, that these stresses could not be revealed by X-ray diffraction. In spite of this opinion, one might observe some stress with X-rays for the following reasons: X-ray diffraction is a highly selective process, one aspect of which is the sorting out of diffractions from crystals of the different phases at different Bragg angles. An averaging of the triaxial stress state over many grains of one phase within the material would result in a pseudohydrostatic stress. The relaxation of the component normal to a free surface, however, would result in a biaxial state of stress, the average value of which would be measurable by the two-exposure X-ray method described later. Even if the sensitivity of this method, which depends on the observation of very small shifts in the diffraction line, were not sufficiently high for application, thermally induced textural stresses might be indicated by line broadening. In fact, broadening from this source was observed in a qualitative degree with duplex aluminum-silicon alloys by J. P. Nielsen and W. R. Hibbard in 1950 [4].

If the experimental technique is skillful enough, however, actual net shifts of the line can be measured; such methods have been used by several investigators to study textural stresses due to differences in mechanical properties of two phases in plastically

¹ Figures in brackets indicate the literature references at the end of this paper.

deformed specimens. One of the earliest of these was D. V. Wilson [5], who in 1950 attempted measurements on the ferrite and cementite constituents in 1.26 percent carbon steel compressed 90 percent. Although his sensitivity of measurement of the cementite lines was very poor, he found a definite indication of opposing stresses in the two phases. In 1955 L. Reimer [6] published the results of work with iron and steel specimens deformed plastically by bending. He showed the influence of textural constraints between the ferrite matrix and imbedded cementite. There was some indication that the residual stresses were proportional to carbon content by volume. In the same year, V. Hauk [7] made a study of plastically extended specimens of some two-phase alloys, not only steel but also a copper-aluminum alloy. He observed that not only an orientation dependence, but also the difference in elastic limits of the matrix and the intermetallic phase, contributed to the resulting stress system.

In view of the success in detecting by X-ray diffraction textural stresses arising, as above, from differences in mechanical constants in plastically deformed two-phase alloy specimens, it appeared worthwhile to investigate the possibility of observing, in annealed specimens, thermally induced textural stresses arising from differences of coefficients of thermal expansion. It has been previously observed at the X-ray metallographic laboratory of the Bureau that textural stresses are not completely relaxed by sectioning procedures [8]. For this reason it was hoped that the usual method of X-ray stress analysis on a free surface would permit detection of the strain resulting from a biaxial stress condition in the surface, which was set up when the quasi-hydrostatic system of textural stresses in the two phases was sectioned. This is to say that, rather than the qualitative and somewhat ambiguous line broadening observed by some, the effect to be looked for was a small but definite line shift, which would indicate a nonzero net change in the lattice parameter of a given phase. The materials chosen were a two-phase brass and a 1.05 percent carbon steel.

2. Stresses in Brasses

Two types of brass were studied; one was a two-phase alpha beta brass, and the other was a single-phase alpha brass. By adding a sufficient amount of zinc to molten cartridge brass, a two-phase alloy was produced, containing 58 percent of copper and 42 percent of zinc. This alloy consisted of approximately equal volumes of the alpha and the beta phases.

After a procedure of heating and rolling, small pieces of the proper size for mounting were cut from this material, and a heat treatment to give an apparent optimum combination of small grain size and minimum of preferred orientation of grains was determined. This was found to be ½ hr at 900° F in air, followed by cooling in the furnace tube removed from the heating coils.

The single-phase alpha brass material, cartridge brass, contained 70 percent of copper and 30 percent

of zinc. The absence of the beta phase was confirmed by metallographic examination and X-ray diffraction. An optimum annealing condition for this material, 800° F for ½ hr, was similarly determined.

The technique employed for finishing the surface on the brass specimens consisted of many cycles of alternate mechanical polishing and electropolishing, ending with the latter. The progress of the procedure was followed by the growing sharpness of the diffraction spots on stationary X-ray patterns.

The diffraction patterns were made with X-rays from a cobalt target in the usual manner of the high-angle, or back-reflection, powder technique. To correct for changes of dimensions of the film and to provide a precise value for the specimen-to-film distance, a calibration ring, caused by diffraction from aluminum powder, was put on the film. The Bragg angle for alpha brass, using the (400) diffraction, was about 75°, and for beta brass, using the (310) diffraction, it was about 73°. Two patterns were made with each specimen, one with the X-ray beam normal to the surface of the specimen, and one with the beam making an angle of 30° with the normal. Only the second side emulsion was developed on each film, which resulted in sharper lines and greater contrast than is ordinarily obtained.

Measurements of the patterns were made with a modified traveling microscope calibrated to the order of 0.001 mm by the Shops Division of the National Bureau of Standards. The $K\alpha_1$ and $K\alpha_2$ lines were, in all cases, resolved, and in most cases readings were made on each. Two complete sets of data were taken independently by two different observers, with results in good agreement. Simple precision analysis of the data indicated that the probable error in the average of each setting was less than ¼₀₀ mm, which, in a typical case, would correspond to 0.00008 Å in d -spacing.

From the radii of the diffraction rings and the known Bragg angle of diffraction for the calibration powder, it was possible to calculate the cosine of the complement of the Bragg angle of diffraction from the specimen. From this and the known value of the X-ray wavelength, the lattice spacing was calculated by Bragg's Law. The two values of lattice spacing and the polycrystalline elastic constants enabled one then to calculate the stress by means of the well-known formula:

$$\sigma = \frac{E}{1+\nu} \cdot \frac{1}{\sin^2 \psi} \cdot \frac{d\psi - d_{\perp}}{d_{\perp}}$$

where E is Young's modulus, ν is Poisson's ratio, ψ is the angle between the surface normal and the diffraction plane normal, and the d 's are the lattice spacings [9].

In connection with this formula, some investigators have found that the ordinary mechanical values of Young's modulus and Poisson's ratio were satisfactory [10]. However, since the values for beta brass were not well established, it was decided to calculate these parameters for the alpha and beta phases from the fundamental single crystal elastic constants [11] by one of the polycrystalline aggre-

gate-averaging procedures. The three principal procedures were discussed in 1955 by H. H. Stadelmaier [12]. One formula, by A. Reuss, is based upon the simplifying assumption of equal stress in all grains; one by W. Voigt assumes equal strains in all grains. Of these two, the former has perhaps been used more, but G. B. Greenough [13] found that an average of the two results was preferable in his X-ray stress measurements. The averaging procedure of A. Huber and E. Schmid [14] makes no assumption except that all grains behave as if they have equal elastic moduli. This procedure, though in best agreement with observed moduli, is seldom used, perhaps because the answer does not proceed from a simple formula but from the numerical evaluation of the leading terms in an infinite series, representing the numerical solution of an integral. Nevertheless, it was this procedure that was used to obtain the values employed in these calculations. These values were, for alpha brass, $E=14.5 \times 10^6$ lb/in.² and $\nu=0.43$; and for beta brass, $E=9.95 \times 10^6$ lb/in.² and $\nu=0.29$.

The observed relative lattice changes, $\eta=(d_\psi-d_\perp)/d_\perp$, and the calculated values of the stresses, σ , are presented in table 1. A positive sign indicates

TABLE 1. Stresses in brass

Specimen	Alpha brass		Beta brass	
	η	σ	η	σ
Two-phase...	-2.58×10^{-4}	$\frac{\text{lb/in.}^2}{-5.27 \times 10^3}$	4.47×10^{-4}	$\frac{\text{lb/in.}^2}{6.53 \times 10^3}$
Single-phase...	-1.39	-2.83	-----	-----
Theoretical values of stress				
Two-phase...	-----	-4.33	-----	5.33

Precision in experimental σ about $\pm 3.2 \times 10^3$ lb/in.²

tension and a negative sign indicates compression. This table also shows the probable error in the stress measurement and the theoretical values of the stress calculated for the two-phase brass with the formulas derived by László.

The value shown for probable error was based upon two sources, an uncertainty in the average of the settings of the traveling microscope on the lines of the pattern of $\pm 1 \times 10^{-2}$ mm, and an estimated uncertainty of the temperature of $\pm 3^\circ$ C during each of the two exposures. These estimations were probably somewhat greater than was actually the case. They contributed the same order of magnitude of error, and, when they were combined, the total estimated probable error in stress was $\pm 3.2 \times 10^3$ lb/in.² This is in the neighborhood of the precision ordinarily claimed in X-ray stress measurements. Although the actual error was probably not quite this large, one could not expect great precision

because the Bragg angles of diffraction for both brasses were less than 76° .

The theoretical stress values were computed with the aid of the formulas in the first paper of László previously referred to. It is to be expected that the stress revealed by the measurement of lattice spacings in the randomly distributed crystals of the aggregate would be proportional to the cubic dilatation of the grains. If k is the modulus of compression, or bulk modulus, and p is the effective hydrostatic pressure, the cubic dilatation is

$$\Delta = -p/k.$$

But p is related to θ , the first stress invariant, by

$$p = -\theta/3,$$

where

$$\theta = \sigma_1 + \sigma_2 + \sigma_3,$$

where σ_1 , σ_2 , and σ_3 are the principal stresses. Therefore

$$\begin{aligned} \Delta &= -(-\theta/3)/k \\ &= (1/k) \cdot (\sigma_1 + \sigma_2 + \sigma_3)/3. \end{aligned}$$

That is to say, the dilatation is proportional to the average of the three principal stresses. Therefore, it was this average stress, as calculated from László's formulas and multiplied by a surface relaxation factor, that was compared with the observed value. The relaxation factor referred to is 1 minus Poisson's ratio, according to László, equaling, in most cases, approximately 0.7, which corrects for the increase in stress components in the surface when the normal component is reduced to zero.

The coefficients of linear thermal expansion used in the calculations were, for alpha brass, $22.4 \times 10^{-6}/^\circ\text{C}$, and for beta brass, $24.7 \times 10^{-6}/^\circ\text{C}$. These values were calculated from data published by P. D. Merica and L. W. Schad [15]. The temperature change was 455° C. It can be seen in the table that the results of the calculations of stress in the two-phase alloy were well within the experimental margin of error of the observed X-ray stress values, with the beta brass, having the larger coefficient, in tension, and the alpha brass, having the smaller coefficient, in compression.

The value of stress measured for alpha brass was somewhat closer to zero than the estimated probable error, but the authors felt that, since it was carefully checked by both remeasurement and reannealing, this stress was at least qualitatively real. Although no definite second phase with a specifically differing thermal expansion coefficient was present, some thermal stresses could theoretically be caused by small amounts of disordered material of different thermal properties present at or near grain boundaries. It is possible, also, that some small degree of surface preparation distortion might still have been present, thus contributing to the nonzero result.

3. Stresses in Steels

The material from which the steel specimens were prepared contained 1.05 percent carbon, with minor constituents totaling 0.74 percent. Three specimens were prepared with different heat treatments in such a manner that one showed a structure of fine lamellar pearlite, one had a spheroidized structure, and one was decarburized. The decarburized specimen, containing a minute amount of cementite to assure saturation of the ferrite with carbon, was included to provide a specimen of an essentially single-phase alloy having the same composition as the ferrite in the pearlitic and spheroidized steels. Such a specimen should contain no textural stresses ascribable to the presence of a second phase, and might provide a base line for the measurements on the pearlitic and spheroidized steels.

The decarburized specimen was prepared by cutting $\frac{3}{16}$ -in.-thick disks from the stock material and decarburizing them with wet hydrogen at 1,650° F. Several attempts were made before a homogenized specimen containing a trace of pearlite was obtained. The heat treatments to obtain the pearlitic and spheroidized conditions were conventional. The decarburized portions were removed and the whole specimens were given a prolonged electropolishing treatment in order to diminish the possibility of body stresses resulting from a carbon concentration gradient. The specimens then were mounted in Bakelite and polished by repeated cycles of light mechanical and electropolishing treatments until the sharpness of the X-ray diffraction indicated that the surfaces were essentially free of cold work.

The same two-exposure method of obtaining X-ray data for surface stress calculations was followed with each of the three types of steel as had been followed with the brass specimens. From the steels, diffraction lines were obtained only from the ferrite and, during the same exposure, from a calibrating powder spread thinly on the surface of the steel. Diffraction from the cementite is not observed in the high angle region from steel with this carbon content. Three pairs of patterns were made from each of the three specimens with three different radiations and slightly varying conditions. In one case, cobalt radiation was used with tungsten as the calibration powder, iron radiation was used with tungsten calibration also, and chromium radiation was used with silver calibration. Subject to the limitations of the speci-

men holder and Bragg angles, the angles of incidence for the inclined exposures of each pair were 45°, 30°, and 45°, respectively.

The mechanics of getting the patterns, processing the films, reading and averaging the data, etc., were in no way significantly different with the steel specimens than with the brasses. The results, along with the probable errors and the computed theoretical values, are presented in table 2.

When examining this table, one should first notice the variation in the observed probable errors in the stress; this error is nearly three times as large for iron radiation as it is for cobalt. The reason for this lies in the sizes of the Bragg angles given in the table. The stresses measured by cobalt and by chromium radiation at the two larger angles, however, do show the expected trend, which is the same as that in the theoretical values, in the order: pearlitic, spheroidized, and decarburized. The mass absorption coefficients, μ/ρ , of iron for the three radiations are also shown in the table. The cobalt radiation is the most penetrating, and is, therefore, the most reliable, because it reveals by diffraction the condition of the largest number of crystallites in the largest volume and is affected least by any residual surface preparation distortion that might be present. The average values of the stress shown in the table were weighted with respect both to the probable error in observation and the penetration of the radiation.

The theoretical values in this table were computed with the aid of formulas for the specific lamellar and spheroidal cases given in László's first paper. The elastic constants used were those cited by László: $E=30 \times 10^6$ lb/in.²; $\nu=0.30$, the coefficient of linear thermal expansion of ferrite equals $14.98 \times 10^{-6}/^\circ\text{C}$, and that of cementite $12.16 \times 10^{-6}/^\circ\text{C}$. The temperature drop in the calculations was taken to be 680° C. Again, the agreement between the theoretical values and the observed averages was reasonably good.

4. Conclusions and Discussion

In previous investigations involving X-ray stress measurement, it has generally been assumed that carefully annealed material was strain-free. László's theoretical work showed that inequalities of thermal expansion would cause textural (tessellated) stresses in two-phase alloys, but no direct measurement of

TABLE 2. *Stresses in steel*

Specimen			Pearlitic		Spheroidized		Decarburized		Probable error in stress
Radiation	θ	μ/ρ	η	σ	η	σ	η	σ	
Co	81	54	3.99×10^{-4}	14.12×10^3	1.00×10^{-4}	3.56×10^3	0.16×10^{-4}	0.57×10^3	1.92×10^3
Fe	73	72	1.30	5.57	1.59	6.84	2.45	10.52	5.34
Cr	78	115	4.20	14.38	3.18	10.89	2.72	9.29	2.02
Weighted average				12.74		6.05		4.58	2.53
Theoretical				9.2		6.2			

these stresses had been made. Contrary to the implication in one of László's papers, the results of this work demonstrate that textural stresses of this type can be measured by changes in the lattice constants if sufficient care is taken. The major experimental limitation is the problem of surface preparation, the routine of repeated cycles of light mechanical polishing and light electropolishing apparently being the best method available at present.

In the present investigation, good agreement was obtained between theoretical stress values and those obtained by X-ray diffraction in the case of alpha-beta brass, and at least qualitative agreement in the case of 1.05 percent carbon steels with various microstructures. The experimental precision was, in most cases, thought to be such that the probable error in stress measurement was a few thousand pounds per square inch.

In addition to the quantitative aspect of agreement with theory, these results reveal, by direct measurement, the impossibility of obtaining complete strain relief in a material that contains more than one phase. They will, perhaps, thus serve to emphasize the necessity of considering textural stresses as an ever-present factor in stress corrosion, surface distortion, fatigue, and similar problems. In this manner they may play some part in the very broad field of study relating the internal stress system in a metal specimen to the mechanical and physical properties and behavior of the body.

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