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CAPABILITY OF THE X-RAY DIFFRACTION METHOD TO EVALUATE THE MACROSCOPIC STRESSES IN PLASMA SPRAYED DEPOSITS

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Mechanical properties of thermally sprayed coatings differ in comparison with properties of bulk materials. Different methods are used for residual stress measurements. Presented work discusses capabilities and different aspects of X-ray diffraction residual stress measurements on the basis of preliminary experiments on metallic and ceramic coatings.

The last 20 years have seen a great attention being paid to thermally sprayed materials [1,2]. The technology can be used for production of special protective coatings on various components for a wide range of applications. Plasma spraying utilizes very high temperatures and high velocities of plasma, flowing from a special plasma torch, and therefore materials with high melting points can be deposited. Solid particles, melted and accelerated in the plasma jet, impact the substrate or previous layers of impacted particles, flatten and solidify with a very high rate of 10^5 - 10^6 $K.s^{-1}$. Consequently, a typical lamellar microstructure consisting of many flat splats develops with a relatively large number of defects, quite different from the microstructure of bulk materials. Characteristic for the sprayed structures are the pores, caused by various processes such as the volume changes during solidification, phase changes during cooling in the solid state, etc. The pores are randomly oriented and their shapes vary from large irregular voids through spherical pores to „planar" break-down of cohesion between two splats. In addition to pores. the structure is often directionally solidified in the course of the heat transfer to the substrate and therefore exhibits anisotropic properties. The thermally sprayed materials, either coatings or self-supporting parts, are under residual elastic stresses, originating from the deposition process. These stresses can negatively affect the deposit properties and become especially important in thick coatings (>1mm). Therefore, an increased attention should be given to their full description and understanding.

Origin of residual stresses

Residual stresses in sprayed coatings arise from two main sources 1) Intrinsic stresses arise from the rapid quenching of molten droplets upon impact on the substrate with restricted contraction. Such a large temperature drop (~ 2000 °C) would lead to a stress level that the material could not withstand; therefore, the resulting level is limited by the splat's intrinsic strength and adhesion to the underlying substrate. 2) Secondary stresses are from the cooling of the coating / substrate couple from the deposition to the ambient temperature, when the stresses develop due to differences in the thermal contraction. Clyne ang Gill [3] reported levels up to 1GPa for the intrinsic stresses and levels about 100 MPa for the secondary ones. In addition, the stress levels are affected by a number of other factors, such as: temperature gradients during and after deposition, stress relaxation processes (plastic deformation, cracking, intersplat sliding, etc.), diffusion processes and phase transformations, compliance of the substrate and coating, etc.

Stresses measurement

Many authors have measured the residual stresses in metals and alloys and several attempts have been made to measure these stresses in bulk engineering ceramics. However, very few authors are dealing with the residual stresses in the thermally sprayed deposits because of the above described complexity of the structure. The same reason is behind the fact that even data on mechanical properties of plasma sprayed deposits in general are scarce.

Simultaneous occurrence and complex interaction of the above mentioned phenomena influencing residual stress, together with limited knowledge of the constituent's properties (which vary a great deal with processing parameters), hinders advancement of reliable models to predict the stress levels and underlines the importance of experimental methods. There is a number of approaches to experimental stress determination in plasma sprayed deposits, each of which has certain advantages, drawbacks and limitations [3].

Three main groups of methods are used to investigate residual stresses. The widely used and most popular method is measuring lattice strains of a selected phase using an X-ray or neutron diffraction. Commonly used methods are the material removal ones [4,5]. They are based on measuring macroscopic changes of strain due to gradual removal of their layers or near a drilled hole. These changes are monitored during removal and evaluated to give stresses. Third method of stress monitoring is based on shape changes of material during or after deposition. This curvature method is successfully used for stress evaluation in plasma sprayed coatings in the last time [3, 6]. Some other methods were developed for measurement of stresses based on different techniques (Raman spectroscopy. mechanical methods....[7])

Principles of the diffraction methods are well known. The most popular is the "$\sin^2\psi$" method. This method is based on determination of changes in crystal plane spacing in different directions with respect to the specimen surface, which exhibit themselves as shifts in angular positions of the respective diffraction peaks. From experimentally obtained strain of lattice in different directions to the sample surface, the stress can be calculated with the use of appropriate elastic constants. The simplified form of the formula is

$$\varepsilon = 1/2\ S_2\ \sigma_\varphi \sin^2\psi + S_1\ (\sigma_{11} + \sigma_{22})$$

where ε is the strain in a particular direction,

ψ is the angle between the sample surface normal and the crystal plane normal.

S_2 and S_1 are crystallographic elastic constants which are in isotropic materials related to theYoung's modulus and Poisson's ratio,

σ_φ is the stress in the surface plane in the measurement direction

$\sigma_{11}+\sigma_{22}$ is the sum of principal stresses in the surface plane.

The layer removal method uses a biaxial strain gage attached to the uncoated substrate side, which measures the substrate deflection as the stressed coating layers are being removed by grinding. There are two major problems with this method. First difficulty is to control the material removal and furthermore the removal must not induce additional stresses or damage the measured sample. The second problem appears with a relatively complex analysis of the obtained gage data. Recently, the curvature method is being used for evaluation of stresses in thermally sprayed coatings [3, 6]. This involves measuring of curvature of substrate/deposit couple and gives a possibility to measure the stresses during the time of deposition.

This work is concentrated on stress measurement in plasma sprayed coatings using the X-ray diffraction - $\sin^2\psi$ method. Preliminary experiments were made and the obtained results pointed us to snares of evaluation from obtained values.

Instrumental effects. Artificial effects on diffraction peaks have to be separated from those caused by stresses only. One important phenomenon occurring during this kind of measurement is the loss of focusing when the angle $\psi \neq 0$, with subsequent decrease in peak height and increase in width. This effect could be minimized by using (hkl) reflections with high diffraction angle. In order to assess the influence of defocusing on the peak position, measurements on stress-free reference (starting powder) at exactly the same conditions as stress measurements on coatings should be performed as calibration. This procedure, as well as proper instrument alignment, helps to avoid artificial influences on measured stress.

Texture and anisotropy. There are three different phenomena usually involved under "texture"

1) Preferred orientation (crystallographic texture) - non-random distribution of crystal orientations with respect to the specimen. It manifests itself by intensity variation of a particular (hkl) reflection in different directions. This phenomenon frequently occurs in coatings, especially those produced by CVD and PVD techniques. In the case of plasma spraying, rapid cooling may result in preferred crystal growth in the heat flow direction. This phenomenon was measured and confirmed in plasma sprayed alumina [8].
2) Shape of crystal grains - columnar grain structure is often observed in plasma sprayed coatings, which slowly diminishes towards the coating surface, where the grains become more or less equiaxed. In addition, ultrafine structures may occur as a result of grain recalescence.
3) Lamellar structure of the coating, consisting of thin, elongated splats and voids of various shapes, size and orientations. This is a major factor contributing to the macroscopic anisotropy.

Triaxial stress. Usually it is assumed that the stresses are the same in all directions of the coating plane. This was confirmed for our specimens. For thick coatings of largely unequal x and y dimensions (parallel to the coating surface) and for more complex shapes (e.g., cylinders) this may not hold true, therefore measurement in both directions is recommended. The stress in z-direction (perpendicular to the coating plane) was also determined from our measurements, following the procedure described in [9]. The value was effectively zero. This was expected, in view of coating structure and low penetration depth of x-rays. Nevertheless, non-zero values of σ_z within the penetration depth can occur in multiphase materials and after certain surface modifications.

Surface effects. Because of the low penetration of x-rays (about tent of μm), the surface state of the specimen can influence the results. For example, if the surface roughness is comparable to the penetration depth, stress relaxation on the asperities can contribute to lower measured values than would be observed under a smooth surface. When comparing the roughness with the effective penetration depth, we concluded that the values determined by x-rays may be slightly underestimated with respect to stress deeper below the surface. On the other hand, surface modification techniques, like grinding and polishing, that decrease the roughness, can alter the stress state to a significant degree. The magnitude of this effect depends on various factors, e.g. the loading force, speed, direction of motion and roughness of the grinding media. This phenomenon is not so significant in ceramics coatings. Nevertheless, changes in stresses due to grinding were observed in stabilized ZrO_2 coatings [10].

Stress Gradient. The different penetration of x-rays at different incidence angles allows for assessment of stress gradient in the thickness direction. If there are significant stress gradients present in the surface layer, the usual linear fit into ε vs. $\sin^2\psi$ data is no longer applicable and at least approximative procedures [7] for gradient evaluation should be used. As noted above, this value is limited by the yield stress and the splat's adhesion; it can be also affected by the surface preparation.

Correlation to macroscopic deformation. In order to correlate the deformation measured within individual crystal grains to the macroscopic deformation of the coating, series of measurements were taken during three-point bending of coatings. The measured strain vs. applied strain dependence showed initial increase, but after a relatively small strain (in the elastic range of the substrate) exhibited random oscillations with no clear correlation to applied strain. After unloading, the measured strain remained approximately the same as at maximum load. Low strength of the coatings and possibly loose of some bonding places of the top layer of splats is thought to be responsible for this behavior. Diffraction methods measure strain in the unbroken crystallites inside the splats. This strain needs to be correlated to the macroscopic strain of the whole coating (which can be imaginated as a composite of splats and different types of voids), in order to facilitate comparison with mechanical stress and strength data, which is necessary for engineering applications.

Conclusions

The capabilities and limitations of x-ray diffraction method are discussed for the case of application to thermally sprayed deposits. The assumptions underlying the use of common "$\sin^2\psi$" method are discussed on the basis of preliminary measurements. The results show that most of them are applicable; however, the coatings' unique structure makes their use somewhat complicated and the simple routines used on bulk materials should not be mechanically reproduced.

Comparison of different methods for residual stress measurement need to be done to better understand the stress-strain relationships in plasma sprayed coatings.

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