LA-UR-96- 2820

Title: **A New Approach to Texture Measurements: Orientation Distribution Function (ODF) Determination by *Rietveld* Refinement**

Author(s): **Robert Vondreele, MLNSC**
Allen Larson, MLNSC
Andrew Lawson, MST-5
Robert Sheldon, NMT-9
Stuart Wright, DOE/EXT

Submitted to: **DOE Office of Scientific and Technical Information (OSTI)**

Los Alamos
NATIONAL LABORATORY

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A New Approach to Texture Measurements: Orientation Distribution Function (ODF) Determination by Rietveld Refinement

Robert Vondreele*, Allen Larson, Andrew Lawson, Robert Sheldon, and Stuart Wright

Abstract

This is the final report of a three-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). This project sought to develop the experimental procedures and the mathematical treatment needed to produce an orientation distribution function (ODF) directly from full diffraction patterns from a sample in a limited number of orientations.

1. Background and Research Objectives

Many technologically important materials are subject to manufacturing processes that impart to the material a preferred orientation of the individual crystal grains within the material. The orientation of the grains can have a profound effect on the physical properties of a manufactured part affecting both its strength and possible failure modes. In the case of composite materials, which by proper combination of components can yield very high strength materials with light weight, the preferred orientation of both components with respect to each other as well as to the exterior dimensions of the part can strongly affect how well the component performs in service. In fact, the performance of a part made with composite materials can be enhanced by proper selection of a preferred orientation for the components. Moreover, in the case of new high T_c superconducting oxides, the preferred orientation has a profound effect on the electrical conduction in wires and other components made with these materials. For example, the crystal grains must be very highly oriented within a wire for it to carry appreciable current densities and much of the present development effort has been focused on achieving high preferred orientation in the finished wire. Present efforts in this direction are to develop a wire consisting of highly oriented oxide contained within a silver metal sheath. To achieve high superconducting currents, the oxide must be oriented with the

* Principal investigator, e-mail: vondreele@lanl.gov

long crystallographic axis (usually the c-axis of a tetragonal cell) normal to the wire. Evaluation of this texture by x-ray methods is difficult because of the extremely poor penetrating power of x-rays. The silver sheath is essentially opaque to x-rays so that sample preparation for conventional pole figure measurements involve removal of the sheath by chemical or mechanical means. This process can strongly alter the state of the oxide superconductor that is inside the wire thus invalidating the texture measurement. Moreover, the sample dimensions used in conventional x-ray analysis (typically 25-mm diameter disks) require a complex series of steps to properly orient an array of many wires so that the texture can be properly measured. Thus, methods of rapidly characterizing the preferred orientation of "raw" samples not subjected to elaborate preparation are essential to help guide the development of these materials into technologically useful components.

The preferred orientation of crystal grains within a manufactured part is described most fully by its orientation distribution function (ODF), which is a mapping of the probability of each of the possible grain orientations with respect to the exterior dimensions. Traditionally, an ODF is determined from pole figures for a relatively small number of reflections. These pole figures are measured with x-rays or neutrons using short detector scans over the center of an individual diffraction peak for a large number of different sample orientations. This is efficient if the selected diffraction peaks are reasonably strong (relative to background) and well separated, such as in pure fcc and bcc metals. It is also appropriate for constant wavelength sources where collection of individual diffraction peak intensities is a reasonably efficient use of the source. However, the traditional method is not very efficient for neutron diffraction at a spallation source such as LANSCE (Los Alamos Neutron Science Center) where the entire diffraction pattern is accessible for each sample setting. Moreover, a different approach is necessary for complicated diffraction patterns, such as from composite materials, intermetallic compounds, high T_c ceramics, polyphasic minerals and polymers where there is expected to be heavy overlap of adjacent diffraction peaks. In addition, the large number of settings normally collected for an individual pole figure may not be necessary, since the entire pattern is obtained at each setting. Thus, a new method of ODF analysis needs to be developed to handle the more complex diffraction patterns obtained from modern technological materials as well as take advantage of the particular characteristics of spallation neutron sources.

2. Importance to LANL's Science and Technology Base and National R&D Needs

The development of a simple experimental procedure and mathematical treatment needed to produce an orientation distribution function directly from full diffraction patterns will

advance the development of sophisticated materials such as high T_c superconducting wires and high strength composite materials. This project supports the Los Alamos core competency in nuclear and advanced materials.

3. Scientific Approach and Results to Date

Our experimental approach is to obtain diffraction data on a number of "standard" texture samples and more complex test specimens of, for example, granites, composite materials and high Tc ceramic components using the High Intensity Powder Diffractometer (HIPD) at LANSCE. During the first two years of the project, we completed measurements on several test specimens including 18 different settings of a hot-pressed alumina plate (NIST SRM 1976 intensity/texture standard), a 1/4" rolled stainless steel plate, a 1/4" stainless/carbon steel/stainless "sandwich" rolled plate, and several cross-rolled titanium plates, all of which comprise some 800 individual data sets. During the final year we have collected additional data sets with which to test the new spherical harmonic Rietveld refinement code. This includes a reexamination of the steel sandwich, new data from a rolled aluminum plate, a nanocrystalline aluminum rod, a shocked titanium plate, some tungsten rods to be used in the new LANSCE production target, niobium/copper alloy rod and silver/copper alloy rod, and some uranium/niobium alloy plates. This suite of data (from approximately 25 samples and comprising roughly 2400 individual patterns) is the test set for the code developed for ODF determination.

Our data analysis approach initially included the tools to extract full sets of intensities for all phases in a multicomponent sample for possible ODF determination from pole figures. This was successfully done using the "le Bail" method for extracting intensities from powder patterns. The method works quite well for multicomponent mixtures and has found considerable use in the solution of crystal structures from powder data. However, we have not pursued the use of these intensities for ODF determination since that approach does not lead to a rapid ODF determination method. To achieve this goal, we have proposed the simultaneous development of direct modeling of the ODF in the Rietveld refinement software. A major achievement has been the implementation of the generalized spherical harmonic description of texture into the Rietveld code GSAS operating on personal computers. This model was developed to be applicable to any crystal symmetry and to the most common sample symmetries (fiber, rolling, shear and centric symmetries) in as many as nine phases in a polycrystalline sample. Currently the maximum order of the harmonics is L=34 and up to 600 harmonic coefficients can, in principle, be refined for each phase. This analysis has been tested against most of the data collected and pole figures calculated from the refined coefficients

generally are satisfactory. As an example the (100) pole figure (Figure 1) of upforged tantalum metal was computed from the spherical harmonics coefficients determined in a Rietveld refinement of 46 powder diffraction patterns collected from a single cylindrical sample held in a variety of orientations in the neutron beam. This pole figure is virtually identical to that obtained by more conventional means with both neutron and x-ray diffraction.

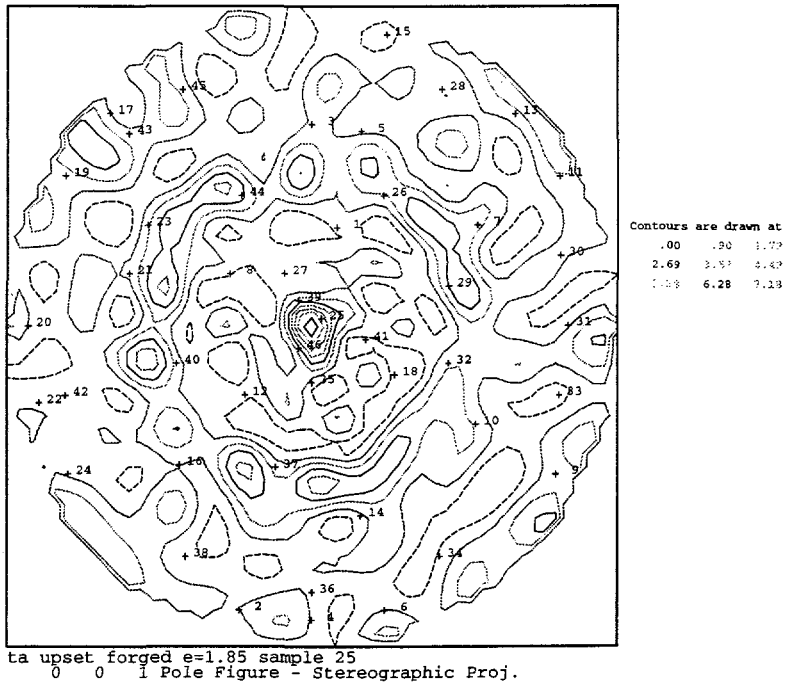

Figure 1. The (100) pole figure computed from 205 spherical harmonic coefficients determined via Rietveld refinement with 46 neutron TOF neutron powder diffraction patterns.

One related problem is the modeling of the strains that are associated with texture. Since strains affect the powder pattern by shifting the peak positions, a model to accommodate this effect is needed to prevent systematic errors in the intensity measurements associated with ODF determination. To deal with the effect of strain we have implemented a simple model that allows the strain to be described as having an isotropic and an anisotropic component. One unexplored facet of this simultaneous determination of strain and texture is the coupling of these two effects within a complete theoretical framework for the response of a material to a load.

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