Title of Publication *Edited by*
TMS (The Minerals, Metals & Materials Society), Year

TIME-RESOLVED ANALYSES OF MICROSTRUCTURE IN ADVANCED MATERIALS UNDER MAGNETIC FIELDS AT ELEVATED TEMPERATURES USING NEUTRONS

Gerard M. Ludtka[1], Frank Klose[2], Roger Kisner[3], Jaime Fernandez-Baca[1],
Gail Mackiewicz-Ludtka[1], John Wilgen[3], Roger Jaramillo[1], Louis Santodonato[2],
Xun-Li Wang[2], Camden R. Hubbard[1], Fei Tang[1]

[1]Materials Science & Technology
[2]Spallation Neutron Source
[3]Engineering Science and Technology
Oak Ridge National Laboratory
1 Bethel Valley Rd, Oak Ridge, Tennessee, 37831, USA

Keywords: Time-Resolved, Microstructure, Materials, Magnetic Fields, Magnetic Processing, Neutron Science, Neutron Scattering, Temperature, Phase Transformations, Ferrous Alloys, In-situ Analyses, Quantitative

Abstract

Fundamental science breakthroughs are being facilitated by high magnetic field studies in a broad spectrum of research disciplines. Furthermore, processing of materials under high magnetic fields is a novel technique with very high science and technological potential. However, currently the capability does not exist to do in-situ time-resolved quantitative analyses at high magnetic field strengths and elevated temperatures. Therefore, most measurements are performed ex situ and do not capture the microstructural evolution of the samples during high field exposure. To address this deficiency, we are developing high field magnet processing and analyses systems at the High Flux Isotope Reactor and the Spallation Neutron Source at the Oak Ridge National Laboratory which will link the analytical capabilities inherent in neutron science to the needs of magnetic processing research. Our goal is to apply advanced neutron scattering techniques to explore time-resolved characterizations of magnetically driven alloy phase transformations under transient conditions. This paper will provide an overview of the current status of this research endeavor with preliminary results obtained on ferrous alloys.

Introduction

High magnetic field processing is proving to be an innovative and revolutionary new focus area [1-12] that is creating the basis for an entirely new research opportunity for materials and materials process development. Based on fundamental thermodynamic principles, our experimental and modeling efforts over the last several years [6-14] have clearly demonstrated that phase stability (conventional phase diagrams) can be dramatically altered through the application of an ultrahigh magnetic field. The ramifications of this research include the existence of entirely new phases, magnetically induced isothermal phase transformation reversal, bulk nanocrystalline materials, texture enhancement, reduced residual stress, carbon nanotube formation, advanced superconducting materials, and novel microstructures with enhanced performance. More broadly, fundamental science breakthroughs are being facilitated by high magnetic field studies in a broad spectrum of research disciplines such as condensed matter physics, materials, biology, and medicine. Therefore, incorporating in-situ neutron scattering characterization methods with high magnetic field research has exceptional potential.

Our ex-situ research [6-14] clearly shows the breakthrough science that can be accomplished using ultrahigh magnetic fields. However, consistent throughout this research (and throughout the refereed literature) is the fact that all microstructural analyses and modeling validation were conducted ***after*** the magnetic processing experiments were concluded. Therefore the influence of magnetic fields on phase equilibria existing during experiments was characterized on specimens without a magnetic field superimposed. There exists the strong possibility that phase equilibria shifting could have occurred while removing the magnetic field, even at ambient temperatures. This potential discrepancy can be significant when assessing the validity of calculations (e.g., the ab-initio Local Spin Density modeling endeavor of reference 7) which are critical to accurately predicting magnetically enhanced phase diagrams. Accurate, validated predictions are essential to propose future experiments and materials development efforts. Therefore, for our research we recognized the critical need to establish a robust ultrahigh magnetic field environmental system for use at both the upgraded Oak Ridge National Laboratory High Flux Isotope Reactor (HFIR) and recently commissioned Spallation Neutron Source (SNS) for conducting advanced time-resolved characterization studies. Such new capabilities would facilitate developing the fundamental science necessary to achieve future major breakthroughs based on magnetic field effects on materials across many disciplines.

Design, fabrication and test of sample environment system

Initially, neutron scattering measurements will be performed at the HFIR using a 5 Tesla superconducting magnet. The challenge was to develop a sample insert that could provide sustained sample temperatures of 800-1000°C while limiting the heat load to the magnet cryostat to less than 10 watts. A sample insert was developed that provides the capability for inductively heating steel samples to 1000°C while limiting the heat load to the magnet cryostat. To minimize deleterious interactions of the neutron beam with the sample insert, an air-cooled (rather than water-cooled) induction coil is used to heat the sample. High temperature thermal insulation is provided by low-density alumina fiber insulation. To facilitate the process of changing out samples, the thermally insulated steel samples are enclosed in a quartz tube that can readily be withdrawn from the magnet. Each sample tube includes a type-S thermocouple, and an inert purge gas manifold. A photo showing a quartz sample tube containing an insulated sample positioned within the aluminum induction heating coil is shown in Figure 1.

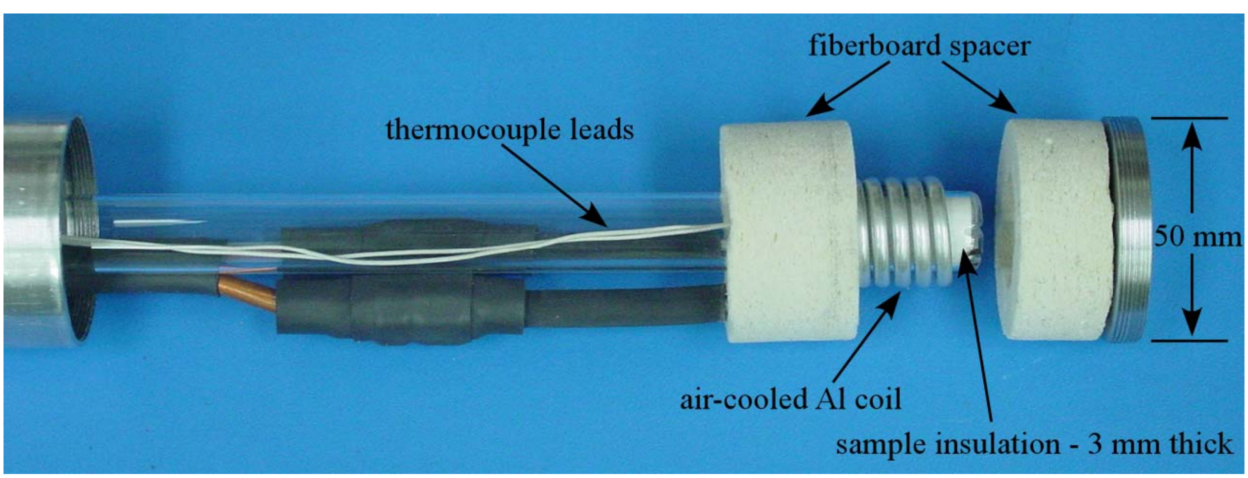

Figure 1. Photograph of specimen holder and key components.

After fabrication and assembly of the sample insert was completed, a 52100 steel specimen was given a heat treatment to confirm temperature control as well as heating and cooling requirements for sample temperatures in the 800-1000°C range. An S-type thermocouple was attached to the specimen and provided feedback control to the induction heating unit. The results of this test are shown in Figure 2. The figure plots the recorded temperature throughout the applied thermal cycle. The system was programmed to heat the 12 mm long by 8 mm diameter specimen to 1000°C in two minutes, hold for three minutes and cool to 750°C in two minutes followed by a six minute hold and air cool. The figure confirms the functionality of the insert and displays good temperature control.

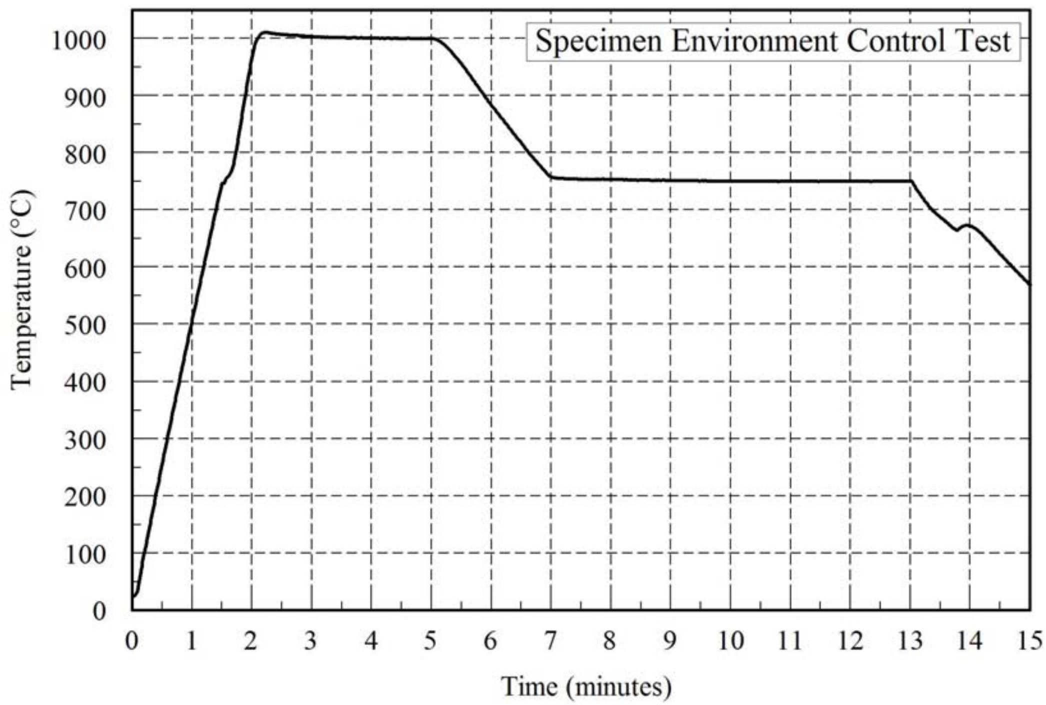

Figure 2. Temperature measurements displaying heating, cooling and temperature control for environment insert system.

Additionally, the sample insert was test-fitted into the 5 Tesla magnet to verify mechanical compatibility. A top view of the fully assembled system installed in the magnet cryostat is shown in the photo in Figure 3. Development of the high temperature sample insert was completed as planned and is currently available for deployment on the neutron beam line at HFIR. A data acquisition and induction heating control system will be employed on the experimental apparatus that will be used for neutron scattering under a high magnetic field at HFIR. The function of the system is to control heat-up, maintain steady-state heated conditions, and cool experimental samples.

Preliminary neutron scattering experiments

Preliminary neutron scattering experiments were performed at the HFIR using steel specimens. The experiments were conducted at room temperature, without high magnetic fields and used plated and non-plated specimens. The plating consisted of a 0.6 mm layer of copper. The goal of these experiments was to develop neutron scattering data analysis and characterization methods using the Wide Angle Neutron Diffraction (WAND) and Neutron Residual Stress Facility 2 (NRSF2) instruments. These two instruments compliment each other well as the NRSF2 provides high precision d-spacing measurements for determination of crystallographic lattice parameters and the WAND can obtain the full diffraction pattern and apply this information in a Rietveld analysis to determine phase weight fractions.

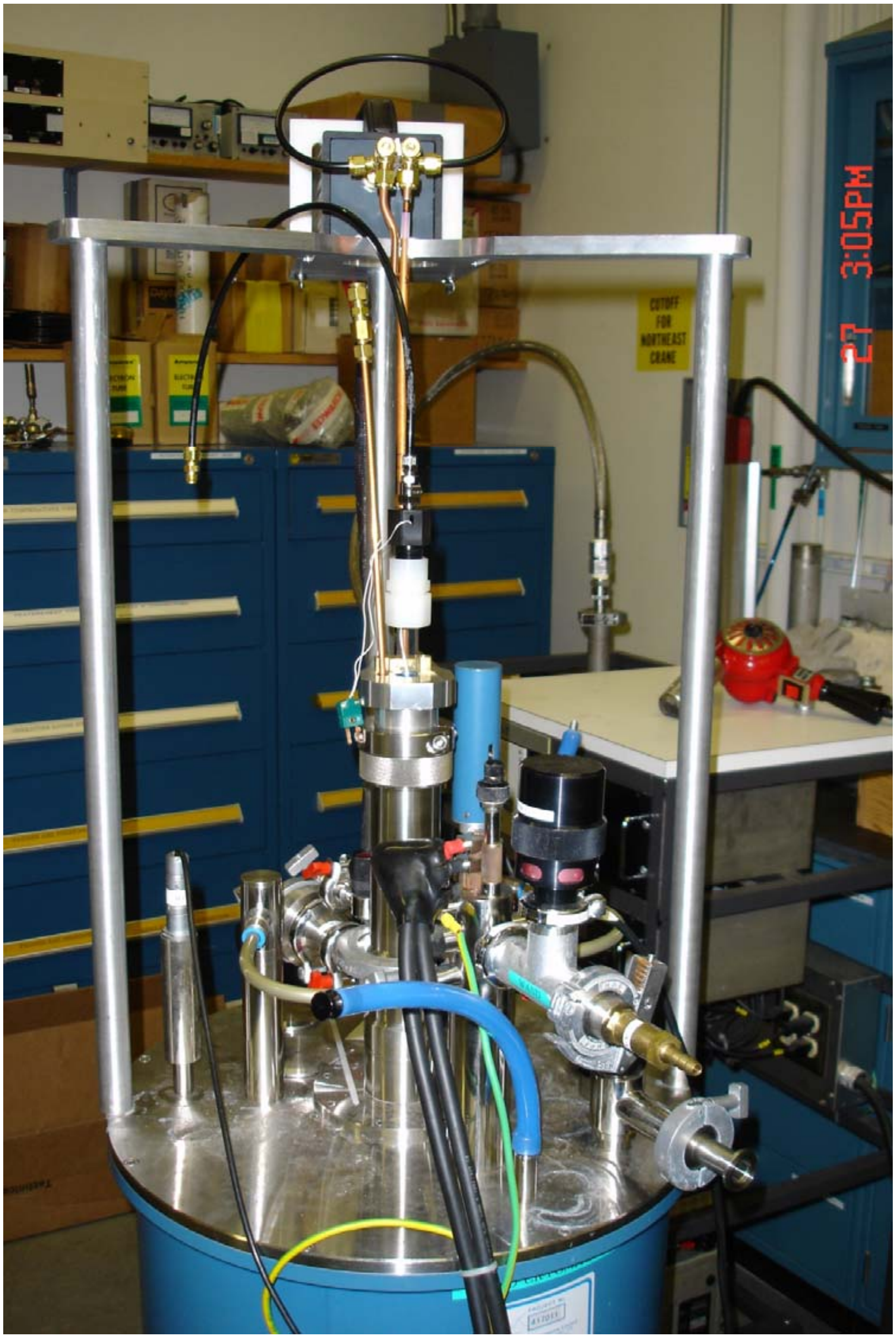

Figure 3. Heating/cooling insert shown installed on 5 Tesla superconducting magnet at HFIR.

Two steel alloys, a medium carbon 1045 steel and a high carbon, high chromium 52100 steel were probed and diffraction patterns were analyzed to determine crystal lattice parameters and phase fractions. For the sake of brevity, only results for non plated specimens and WAND data will be presented here. Table I shows lattice parameter measurements for ferrite (BCC) in both steels. The results are compared to X-ray diffraction measurements for 51XX series steel [15].

A diffraction pattern for an uncoated 52100 specimen is shown in Figure 4. A Rietveld analysis has been performed for estimating phase weight fractions. This particular analysis estimates 17% cementite (Fe_3C) and 83% ferrite (BCC). This result is reasonable although the cementite content is exaggerated compared to ThermoCalc predictions of ~15 wt.% cementite. These apparently high values for cementite fraction were consistent for 1045 results and suggest that some additional refinement may be required in the analysis. This accomplishment provided scoping data for developing and refining diffraction pattern analysis methodology and lays the foundation for later experiments.

Table 1. Ferrite lattice parameter measurements in angstroms.

Steel alloy	WAND ($\lambda = 0.1476\,\text{nm}$, $2\theta_0 = -0.24^\circ$)	X-ray measurements (Cavin et al)
1045	2.8659 ± 0.00002	2.8649
52100	2.8682 ± 0.00003	

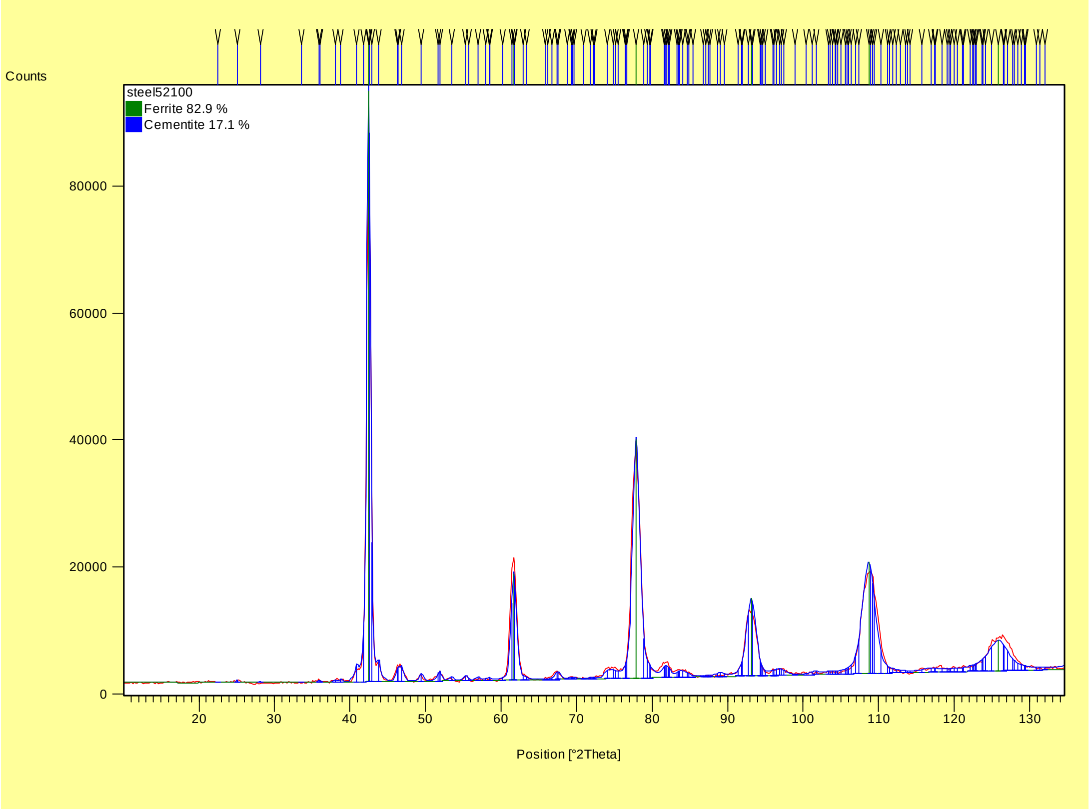

Figure 4. WAND diffraction pattern and applied Rietveld analysis for uncoated 52100 specimen.

Future Facility Development Activities

The next step in the project is to combine the elevated temperature capability with a 5 Tesla magnetic field and perform neutron scattering experiments. For the current HFIR operations schedule, these experiments should be conducted in the first half of 2007. High purity, levitation cast Fe-C alloys have been obtained and will be used in these experiments. The goal of these experiments is to obtain precise information regarding shifts in the Fe-C phase diagram due to the high magnetic field.

A major advancement of the program will be the addition of superconducting magnets with field strength much greater than 5 Tesla. One of the additions is a 16 Tesla superconducting magnet system at SNS (being built now and scheduled for commissioning in Spring 2009). A deliverable of our project is to provide an environment insert similar to the one described in this paper for the 16 Tesla magnet. The second addition is associated with the planned Zeemans high magnetic field facility at the SNS. For this program, specifications and designs are currently being written for a ~40 Tesla hybrid magnet. Such a magnet will merge the state-of-the-arts in magnet technology and neutron scattering. It is anticipated that an elevated temperature insert will be developed for this advanced system.

Acknowledgements

This research is sponsored by the Laboratory Directed Research and Development Program of Oak Ridge National Laboratory (ORNL), managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract number DE-AC05-00OR22725.

References

1. Choi, J-K., Ohtsuka, H., Xu, Y., Choo, W-Y. *Scripta Mater*., 43 (2000), 221.
2. Enomoto, M., Guo, H., Tazuke, Y., Abe, Y.R., Shimotomai, M., Metall. *Mater. Trans*., 32A (2001), 445.
3. Liu, W., Ou, D.R., Zhou, H.H., Tang, G.Y., Wu, F., *J Mater Res*., 16 (2001), 2280.
4. Joo, H.D., Kim, S.U., Shin, N.S., Koo, Y.M., *Mater. Lett*., 43 (2000), 225.
5. Ou, D.R., Zhou, H.H., Tang, G.Y., Zhu, , *J., Mater. Sci. Technol*., 19 (2003), 87.
6. Ludtka, G.M., Jaramillo, R.A., Kisner, R.A., Nicholson, D.M., Wilgen, J.B., Mackiewicz-Ludtka, G., Kalu, P.K., *Scripta Mater*., (51), 2004, 171-174.
7. D. M. C. Nicholson, R. A. Kisner, G. M. Ludtka, C. J. Sparks, L. Petit, Roger Jaramillo, G. Mackiewicz-Ludtka and J. B. Wilgen, Askar Sheikh-Ali, P. N. Kalu[2] “The Effect of High Magnetic Field Annealing on Enhanced Phase Stability in 85Fe-15Ni", Nicholson, et al, *Journal of Applied Physics*, (95) No. 11, June 2004, 6580-6582.
8. R. A. Jaramillo, S. S. Babu, G. M. Ludtka, R. A. Kisner, J. B. Wilgen, G. Mackiewicz-Ludtka, D. M. Nicholson, S. M. Kelly, M. Murugananth, and H. K. D. H. Bhadeshia, *Scripta Materialia*, (52), 2005, 461-466 .
9. Ludtka, G. M., Jaramillo, R. A., Kisner, R. A., Wilgen, J. B., Mackiewicz-Ludtka, g. M., Nicholson, D. N., Watkins, T. R., Kalu, P., England, R. D., “Exploring Ultrahigh Magnetic Field Processing of Materials for Developing Customized Microstructures and Enhanced Performance”, in Materials Processing in Magnetic Fields (Eds. Schneider-Muntau, H. J. and Wada, H.), World Scientific Publishing, New Jersey, pp 55-65 (2005).
10. Ludtka, G. M., Jaramillo, R. A., Kisner, R. A., Mackiewicz-Ludtka, G. and Wilgen, J. B., “Exploring Ultrahigh Magnetic Field Processing of Materials for Developing Customized Microstructures and Enhanced Performance”, *Oak Ridge National Laboratory Technical Report* ORNL/TM-2005/79, April 2005.
11. Jaramillo, R.A., Babu, S., Miller, M.K., Ludtka, G.M., Mackiewicz-Ludtka, G. Kisner, R.A., Wilgen, J.B. and Bhadeshia, H.K.D.H, “Investigation of Austenite Decompositions in high-carbon high-strength Fe-C-Si-Mn steel under 30 Tesla Magnetic Field", *Proceedings of the TMS Conference “Solid-Solid Phase Transformations in Inorganic Materials 2005”*, May 29-June 3, 2005, Phoenix, AZ.
12. Jaramillo, R.A., Ludtka, G.M., Kisner, R.A., Nicholson, D.M., Wilgen, J.B., Mackiewicz-Ludtka, G., Bembridge, N. and Kalu, P.N., “Investigation of Phase Transformation Kinetics and Microstructural Evolution in 1045 and 52100 Steel under Large Magnetic Fields", *Proceedings of the TMS Conference “Solid-Solid Phase Transformations in Inorganic Materials 2005”*, May 29-June 3, 2005, Phoenix, AZ.
13. J. B. Wilgen, R. A. Kisner, Gerard. M. Ludtka, Gail M. Ludtka, and R. A. Jaramillo, “Method for Non-contact Ultrasonic Treatment of Metals in a High Magnetic Field”, Department of Energy Invention Disclosure No. S-105,133, submitted April 6, 2005.
14. R. A. Kisner, J. Wilgen, G. M. Ludtka, R. A. Jaramillo, and Gail Mackiewicz-Ludtka, “Thermal and High Magnetic Field Treatment of Materials and Associated Apparatus” **,**U.S. Patent Application No. 11/109,376 filed 4/19/2005.
15. O.B. Cavin, D.A. Carpenter and C.R. Hubbard, ORNL unpublished data.