Needs for Process Control in Advanced Processing of Materials

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SUMMARY

Recent advances in the synthesis of new materials with complex microstructures, coupled with an improved understanding of process/microstructure/property relationships, has created a new challenge for NDE—the redirection of a technology originally conceived for flaw detection/characterization to the nondestructive measurement of process and microstructure variables during materials processing. This review shows how NDE techniques could play the sensor role in automated process control. The techniques, originally developed for detecting cracks, show merit in monitoring solidification. Other ultrasonic techniques show promise in characterizing temperature distributions and porosity. Problems include: the need for inverse modeling/calibration/high speed data acquisition and reconstruction; and display and hardware able to survive the harsh processing environment. Feedback systems based on artificial intelligence combine heuristic and mathematical predictions with developing sensor technologies to drive process development.

INTRODUCTION

The traditional role of nondestructive evaluation (NDE) has been detecting and sizing flaws in completed engineering structures. Numerous techniques have evolved in this area including x-ray radiography, ultrasonics, acoustic emission and eddy-currents. Theories are being developed to overcome flaw detection problems associated with heterogeneous phase distributions and the polycrystalline nature of engineering materials.

During this same period, new laboratory and commercial processes, e.g., rapid solidification processing (RSP), composite processing, directed high energy beam welding, superplastic forming, advanced ceramics, polymer blends processing, etc., have evolved. Concurrently, a deeper understanding of the process-microstructure-property relationship has emerged in both the new processes and established technologies such as continuous casting, rolling, and other forming processes. Full exploitation of emerging and existing technologies is hampered by the controlling complexity of the processes and the resulting microstructures. The trend is toward using automated techniques which use on-line measurements of process variables and evolving microstructures.

While some strides have been made in process modeling, the measurement methods and their associated analysis techniques (sensors in the nomenclature of process control) have definitely lagged behind. These sensors must not disturb processing and thus are true nondestructive evaluation (NDE) techniques. They often rely upon precisely the same phenomena that plagues NDE techniques use in detecting flaws. For example, the scattering of ultrasound by grains of metals, which precludes the small flaw detection in welds, provides a physical basis for potential in-process measurement of grain size (a key microstructure parameter) during materials processing.

ADVANCED PROCESSING OF MATERIALS

The economic gains for industries choosing this new approach are potentially great because of improved relative productivity and quality. A strong driving force exists to develop sensors for on-line microstructure characterization and process control. These developments will enhance the incentive to design computer-aided process control systems for advanced processing.

Today's "intelligent" processing systems can be broken into three components: a process model, sensors, and control. This report emphasizes sensors, but to fully understanding their role, it is necessary to briefly consider the two other components.

Process Models

Process models relate process variables (temperature, pressure, time, composition, etc.) to microstructure/properties. To illustrate the information...
they convey and the role they serve, consider a modern model used for alloy solidification (Figure 1).2

During conventional (slow) solidification processes, compositional equilibrium (partitioning) is maintained at the moving growth front. The interfacial morphology is a function of thermal gradient (Gt) in the melt at the interface and the interfacial velocity or growth rate (R). At high values of the ratio Gt/R (left side of Figure 1), the interface advances on a planar (or smooth) front and there is no microsegregation of solute atoms transverse to the solidification front. In contrast, at lower values of Gt/R the interface becomes morphologically unstable and leads to cellular and dendritic growth morphologies. The solute-enriched microsegregation builds up in the remaining liquid between the cells or dendritic arms.

During rapid solidification, high interfacial velocities, the regime of morphological instability, is capped off (Figure 1). Morphological stability is reestablished, and plane-front ensures. Potential perturbations in the growth front are now much finer in scale, and surface tension imposes a dominant stabilizing influence. The equilibrium-partitioning assumption is not realistic at the high growth rates in question. Various models are available for the dependence of partitioning on interfacial velocity. In the limit, when the growth front moves at a rate exceeding the solute atoms’ diffusive velocity in the liquid, no partitioning takes place. A completely homogeneous solid is produced. This velocity range is shown as a cross-hatched bar in Figure 1. In general, as the solute content in a given alloy system increases, the dome-shaped region in Figure 1 expands outward—planar solidification becomes more difficult.

"Intelligent" Control

The trend in processing is toward "intelligent" computer-aided control based on artificial intelligence (AI), expert systems, and control theory. Expert systems are applied when there is a human expert and the knowledge-base is primarily heuristic. Model-based systems are applied when rules are mathematical. In general, a coupling of heuristic and
model-based systems is anticipated. Artificial learning systems can formulate predictive process models.

As an example materials processing control system implementation, consider atomizing methods for rapidly solidified metal powders production. Powder microstructure and properties of consolidated products depend on cooling and solidification rates which are profoundly influenced by powder particle size. In addition, powder particle size distribution influences consolidation processes. Figure 2 shows a hypothetical process control chart for atomization where a predetermined ideal particle size distribution is used as the desired product. On the diagram's left side, a particle size sensor establishes actual size distribution for comparison with the desired distribution shown in the center box. On the right side, a feedback control loop utilizes a process model and control laws to make appropriate changes in process variables affecting particle size distribution. Thus, the system functions in a self-correcting mode to consistently produce an ideal powder distribution. Finally, automated monitoring and control systems may eventually permit continuous atomization processes to be developed.

Sensors

Current NDE research is leading to methodologies (measurement methods, imaging, and interpretation) to characterize microstructural features (such as grain size, composition variations, inclusion size and content, second phase distributions, state of cure, etc.) together with process variables such as internal temperature. Sensors are also under development for in-process metrology and for detecting defects such as porosity.

Sensor development is often complicated by an insufficient knowledge base. The relationships between sensing mechanism/measurement and microstructure have yet to fully emerge. Further handicaps are supplied by hostile environments, limited measurement time, and the need to avoid processing interference. Better models and algorithms of sensor-microstructure interaction, both for collected data analysis and data reduction into a useful form for particular processes, must be developed.5,6 It will also be necessary to apply emerging high-speed imaging, associated data acquisition, reconstruction, manipulation, and display techniques to represent the necessary information on-line in a readily interpretable form.

Presently, sensor-microstructure interactions have not generally reached the stage where models may be reliably used. Therefore, the use of "indirect sensors" (sensors which cannot be used directly in-process) must also be explored to (1) calibrate direct sensors and (2) aid the development of predictive models.

DIRECT SENSORS FOR MATERIALS PROCESSING

Without better sensor systems, there can be no real advancement in process control in the process themselves. Each material and process imposes its own particular set of constraints upon measurement methodology. However, important principles common to many applications exist, as illustrated by considering some examples from current research.

Continuous Metal Solidification

Numerous sensor needs exist for continuous casting. Figure 4 identifies

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Figure 3. A schematic illustration of a continuous metal caster.

Figure 4. Sensor needs for automated process control of continuous steel casting.
needs in continuous casting of steel. These include sensors that assure melt cleanliness, measure oxygen content and melt temperature, check location of liquidus and solidus isotherms during solidification and post solidification temperature distribution, etc.

Solidification in engineering alloys occurs over a range of temperatures. The potential differences are finite. In the schematic of Figure 3, it is confined in the space between the liquidus and solidus isotherms. This region of liquid plus solid co-existence is termed the “mushy” zone. Its shape, depth, and extent controls microsegregation of alloy elements on both the dendritic scale and a large scale segregation—approaching the dimensions of the ingot itself. The latter macrosegregation leads to significant surface-to-centerline composition variations that can not be eliminated by subsequent thermomechanical processing.

The continuous caster productivity is improved by increasing casting speed. However, as the speed increases, the “skin” thickness diminishes. The depths of the metal pool and the “mushy” zone increase. This can result in ruptures (breakouts) or increased macrosegregation and hot tearing in the ingot. A critical balance between casting speed and macrosegregation can be achieved with an “intelligent” control system. The solidification/segregation models relating heat and fluid flow to composition variations have matured enough so that, when combined with good sensory data on the size and shape of the “mushy” zone, they can provide the basis of an “intelligent” control system. Furthermore, a sensor capable of measuring solidified “skin” strength (its thickness and temperature) may enable safe casting process speed-up.

Ultrasonics is currently being explored for melt inclusion detection, “mush” zone location/characterization, solidified ingot temperature distribution, and internal discontinuities detection (hot tears and porosity). Both sound velocity and alloy density vary with temperature. For example, in 304 stainless steel, ultrasound velocity varies with temperature as shown in Figure 5.8. The difference in density and velocity of liquid and solid metals results in a finite reflection coefficient, 0.1 for many metals (Figure 6). Thus, for planar solidification, a medium strength ultrasonic echo, about 10% of the incident signal, is anticipated to be reflected. In conventionally solidified alloys, where temperature gradients are low, dendritic solidification is more common. The reflection region is distributed throughout the “mushy” zone, resulting in a much less coherent reflected signal of reduced temporal amplitude.

Reflected signal detection affords an opportunity to locate and perhaps characterize liquid-solid interfaces. The work of R. L. Parker has demonstrated the feasibility of locating a single interface in several metals and alloys including steel (Figure 7). Little, however, appears to have been reported on the characterization of the “mushy” zone by detailed analysis of the reflected echo. In addition to continuous casting, development of this sensor will lead to many other applications in solidification technology (e.g. determination of the location/shape of the “mushy” zone during growth of single crystal turbine blades).

A second application variation of ultrasonic velocity with temperature is actually mapping the internal temperature distribution of solid metals (Figure 8). If the relation between velocity and temperature is known for a particular grade of steel (it varies from grade to grade), then tomographic reconstruction schemes can be devised for recovering the spatial variation of velocity and hence temperature. Work in progress, for example, attempts to determine the average temperature within concentric rings (annual pixels) in cylinders by measuring flight value time for an appropriate set of

![Figure 5. The variation of longitudinal ultrasonic velocity with temperature for type 304 stainless steel.](image)

![Ultrasonic Reflection at Liquid-Solid Interface](image)

![Figure 6. Spatial distribution of ultrasonic reflection for planar and dendritic solidification.](image)

![Figure 7. Ultrasonic detection of liquid-solid interface during unidirectional solidification of steel.](image)

![Reference: Robert L. Parker. National Bureau of Standards](image)
ultrasonic rays. Initial results, obtained using noncontact laser generated ultrasound have shown encouraging success (Figure 9) although only a few pixels have been recovered per sample to date. In the future, the two evolving techniques—for determining “mushy” zone location/shape, and temperature distribution in the solidified shell of a casting—need to be combined to maximize useful information for solidification process control. In addition, velocity changes, due to temperature gradients in the solidified shell, must be accounted for in interface location measurements.

Welding and Surface Modification

Both welding and surface modification processes are much more rapid than continuous metal solidification, and so less time is available to gather information. Even so, measurement techniques are emerging to measure liquid-solid interface position/velocity and detect imperfections.

Utilizing techniques similar to those used by Parker, Lott et al while exploring ultrasonics’ use as a sensor for the closed-loop control of arc welding (Figure 10), controlled experiments were done to determine the feasibility of measuring the instantaneous weld pool depth. It was necessary to not only detect and characterize the interface echoes (from the liquid/solid, solid/air, and liquid/air interfaces), but to make two significant corrections to the time-of-flight values due to steep temperature gradients. The temperature gradients changed the time-of-flight because (a) the average velocity was shifted, and (b) the ray paths were bent (refracted). Using a heat flow model to calculate the thermal field, and relatively simple codes to compute correction factors to time-of-flight values, Lott et al were able to deduce a linear relation between weld pool depth and modified ultrasonic transit time that demonstrated feasibility (Figure 10).

In rapid surface solidification, surface melts, typically less than 1 mm in depth, solidify in a small fraction of a second. Under these conditions, the properties of the modified layer are critically controlled by solidification interface velocity and temperature gradient. For this process, acoustic emission has been explored. While it could not characterize either variable, it has shown some promise in detecting flaw growth during solidification. Using electron beam pulses 7 ms in duration, surface melting and resolidification of aluminum alloys has been achieved. Typically, acoustic emission signals are observed for 10-20 ms during cooling, by which time the melt has cooled to 200°C (Figure 11). The emission intensity in commercial aluminum alloys is markedly enhanced when hot tearing occurs (Figure 12). This work promises the opportunity to detect defective processing, enabling the area to be retreated immediately. Acoustic emission techniques are also being investigated for other (slower) solidification processes, such as fusion welding.

Powder Processing

While metal powders are presently used in the production of engine discs in advanced jet aircraft, their full exploitation is limited by lack of process control, including assurance of powder cleanliness. In high performance ceramics, the overriding problem is again the fact that components with desired microstructures/properties cannot be reliably and reproducibly manufactured. In metals, new processes such as centrifugal atomization permit production of more rapidly solidified powders with tighter size distribution. Direct production of unagglomerated ceramic powders are realized through precipitation from liquids, condensation from vapor reactions and crystallization of aerosols. Many sensor needs are recognized for control of powder production, rheological behavior of particulate assemblies, consolidation, and microstructure evolution.

Figure 8. Tomographic reconstruction of ultrasonic velocity/temperature distribution in a solidified cylindrical ingot.

Figure 9. Example of temperature reconstruction in a solidified 304 stainless steel ingot with an actual centerline temperature of 350°C.
For metals with coarser particles in the size range of 5 to 200 μm or more, direct light scattering or optical imaging techniques are available that can be adapted to particular processes. For very fine (less than 0.1 μm) and intermediate (0.1 to a few micrometers) ceramic particles, both dynamic and direct light scattering techniques may be investigated.

In both metals and ceramics, the consolidation process is particularly important to monitor. On the one hand, it is important to sufficiently sinter to eliminate porosity and attain full density. On the other hand, it is desirable to still retain small grain and second phase size distributions. Several research groups have been exploring the use of ultrasonics to characterize pore size distribution, though not-to-date, for in-process characterization purposes.15,16

Figure 10. Weld process monitoring and control using ultrasonic sensors. Arc-welding concept is shown on the left side; middle and right side show laboratory bead-on-plate weld configuration and correlation of weld-pool depth with time difference between echoes.12


Figure 11. Acoustic emission as a function of time for the heating and cooling phases of electron beam surface melting.

Figure 12. Cross-sections of electron beam surface melted regions; hot tearing occurs in an Al-Cu alloy (left), no tearing occurs in pure aluminum (right).
Tittman, et al\textsuperscript{16}, have initiated studies of ultrasonic characterization of micropores in IN100 powder alloy used for engine discs. Using ultrasonic backscattering techniques, they have examined materials with different metallographically determined pore densities and size distributions (Figure 13). The backscattering spectra indicate much lower scattering at all frequencies for their sample C, the material with the fewest pores (Figure 14). Furthermore, theories based on scattering and diffraction are successful in predicting the backscattering behavior, thus providing the potential to ultimately deduce size distribution from measured scattering spectra.

**INDIRECT SENSORS**

The development of analysis methods to deduce quantitative information about microstructure, such as pore size or precipitate distribution from the scattering of ultrasound, etc., could be simplified and calibrated by using what we term indirect sensors. These interrogate the bulk structure and give statistical estimates of microstructure constituent distribution. Examples include small angle neutron scattering (SANS) and synchrotron radiation. Both are finding increased applications in metals, ceramics, and polymers.

Neutrons are an excellent, nondestructive probe of microstructure. Since thermal neutron energies are low and interact only with the atomic nucleus, they are not significantly absorbed and penetrate deeply into most materials without disturbing them. Scattering is controlled by magnetic properties of the nucleus that vary non-systematically with atomic number so that microstructure constituents, rich in elements with similar atomic number to the matrix, may be readily observed (e.g., magnesium and aluminum or iron and manganese).

Hardman-Rhyne and co-workers\textsuperscript{15} have reviewed numerous recent applications of SANS to ceramics. One of interest to both metal and ceramic researchers is the sintering process characterization. Using the beam broadening regime, they studied the densification process associated with the sintering of MgAl\textsubscript{2}O\textsubscript{4} (spinel) as a function of temperature. Figure 15 shows the intensity of scattered neutrons as a function of position (scattering vector) on a neutron detector. Although the spinel powders used for the study had been heated at 1300°C for 12 hours, little sintering had actually occurred, and considerable scattering from porosity in the 1-10 \(\mu\)m size was observed. Sintering at higher temperatures (e.g., 1500°C) reduced the porosity and the neutron scattering. Even the material treated at 1500°C is not fully sintered. With its full width at half maximum, it is significantly larger than that of the fully dense blank.

Historically, the development of SANS theory has been limited to single particle diffraction. The theory is presently expanding to include neutron scattering dominated by multiple scattering. Further advancement of this theory, coupled with the availability of a cold source (such as the one planned for the National Bureau of Standards reactor in 1985), should permit quantitative analysis of pore size distribution and volume fraction during in-situ sintering experiments using large neutron wavelengths (1.5 \(\mu\)m).

This research highlights the opportunity for the calibration of direct nondestructive sensors such as those stemming from Tittman, et al\textsuperscript{16}.
work on porosity measurement. Related work offers the opportunity of calibrating other direct sensors (e.g., those characterizing precipitates, inclusions, microcracks, etc., whose size range is 1 nm-50 μm).

**References**


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**MEETINGS CALENDAR**

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**GENERAL MEETINGS**

(Editor's Note: Entries in the General Meetings Calendar are listed chronologically by either the actual date of the event or by the "abstracts due" date—whichever is most immediately relevant. Accepted entries will run for a maximum of six months for the three months prior to the "abstracts due" date, and for the three months preceding the actual event.)

March

Inclusions and Residuals in Steels: Effects on Fabrication and Service Behaviour

**March 4-5, 1985,** Ottawa, Ontario, Canada
Contact J. D. Boyd, Physical Metallurgy Research Laboratories, CANMET, 568 Booth St., Ottawa, Canada K1A 0G1; telephone (613) 993-7129.

Seventh Symposium on Applied Surface Analysis

**May 15-17, 1985,** University of Maryland, College Park, Maryland
Abstracts due **March 9, 1985**. Contact Dr. James S. Murray, Naval Research Laboratory, Washington, D.C. 20375; or Dr. John T. Grant, Research Institute, University of Dayton, Dayton, Ohio 45469.

The Markets for Neodymium-Iron-Boron Magnets—Conference and Exhibition

**March 10-12, 1985,** Monterey, California
Contact Dr. Hugh Olmstead, Gomah International Inc., P.O. Box 8, Gorham, Maine 04040; telephone (207) 892-2216.

Instrumented Impact Testing of Plastics and Composite Materials

**March 11-14, 1985,** Houston, Texas

ASTM Symposium on Toughened Composites

**March 13-15, 1985,** Houston, Texas

Ultrasonic Velocity Evaluation of Iron Castings

**March 14, 1985,** Chicago, Illinois

Fatigue in Mechanically Fastened Composite and Metallic Joints

**March 18-19, 1985,** Charleston, South Carolina
Contact ASTM Staff Manager Matthew Lief, ASTM Standards Development Division, 1916 Race Street, Philadelphia, Pennsylvania 19103; telephone (215) 299-5516.

Copper Metallurgy '85

**March 20-22, 1985,** Legnica, Poland
Contact Instytut Metali Nizzelaznych, 44-101 Gliwice, ul. Sowinskiiego 5, P.O. Box 133, Poland.

ASTM Symposium on Case Histories Involving Fatigue and Fracture Mechanics

**March 21-22, 1985,** Charleston, South Carolina
Contact Matthew E. Lief, ASTM Standards Development Division, 1916 Race Street, Philadelphia, Pennsylvania 19103; (215) 299-5516

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