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Containerless Processing of Amorphous Ceramics

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## ABSTRACT

The absence of gravity allows containerless processing of materials which could not otherwise be processed. High melting point, hard materials such as borides, nitrides and refractory metals are usually brittle in their crystalline form. The absence of dislocations in amorphous materials frequently endows them with flexibility and toughness.

Systematic studies of the properties of many amorphous materials have not been carried out. The requirements for their production is that they can be processed in a controlled way without container interaction. Containerless processing in microgravity could permit the control necessary to produce amorphous forms of hard materials.

## INTRODUCTION

Amorphous forms of some ceramic materials are of scientific and technological interest because of their unique properties. Compared to their crystalline counterparts, amorphous materials are frequently tougher, more flexible and offer superior corrosion resistance. It is also possible to transform an amorphous phase to a crystalline one after processing it.

The formation of amorphous ceramics is potentially easier than formation of amorphous metals. This is because ceramics are polyatomic molecules, crystallization then requires the condensation of an appropriate set of atoms in lattice sites. In these circumstances, structure in the liquid phase and the processing route<sup>(1-3)</sup> can influence the ability to form a particular phase. The effect of convection can be to enhance mixing and therefore make crystallization possible in situations where diffusional limitations would prevent this. Impurities and condensed phases can have a strong effect on the ability to form amorphous materials.

## EXPERIMENTAL REQUIREMENTS

Some desirable experimental requirements are given below:

1. Processing of high melting point non-conductors above the liquidus.
2. Control of nucleation. The ability to process in the absence of heterogeneous nucleation sites is important.
3. Control of material purity.
4. Control of processing atmosphere.
5. Quiescent samples, free from convection.

Some of these conditions can be obtained by containerless processing on Earth. The very limited capability for containerless processing of non-conducting, low viscosity liquids presents a significant experimental limitation. While it is possible to process small samples for very short periods<sup>(4,5)</sup>, longer experiments with samples larger than a few hundred milligrams are not possible with presently available ground-based technology.

There is frequently a need to achieve all of the above experimental conditions simultaneously. Microgravity processing can allow this because the positioning forces are small and buoyancy-driven convection is eliminated.

## SCIENTIFIC RESEARCH OPPORTUNITIES

1. Elimination of Heterogeneous Nucleation: This would extend the range of materials from which glasses could be made. Work by Day et al<sup>(6)</sup> in a single-axis acoustic levitator in microgravity has demonstrated a factor of ca. 2.7 reduction in critical cooling rate (Rc) for a silica-based glass. The reduction in Rc was attributed to the absence of heterogeneous nucleation. Many glass systems of scientific or technological interest<sup>(7)</sup> cannot be formed in the presence of nucleation sites or with presently available hardware.

2. Preparation of Pure, Clean Materials: In most cases, the properties of materials have been measured by contained techniques. The purity of the sample has been assumed equal to that of the starting material. Containerless experiments by the authors<sup>(8)</sup> indicate that this assumption is not always correct. Containerless processing in a controlled atmosphere at high temperature provides a means to produce clean, pure samples.

Figure 1 shows a plot of pressure vs. inverse temperature for the vapor species over silicon and aluminum with an ambient oxygen pressure of  $10^{-6}$  atm. This illustrates that the liquid elements can be cleaned of oxide by evaporation. Further oxidation is suppressed by cleaning the atmosphere with a blanket of vapor from the specimen.

3. Quiescent Samples Free From Convection: This potentially allows accurate measurement of diffusion coefficients in liquids. Convection effects can mask diffusion effects by stirring of liquids. This makes it difficult to perform studies of the effects of diffusion on nucleation in multi-component liquids.

4. High Temperature Liquid Phase Processing Without Contamination: Impurities can arise from two sources; induced and inherent. Induced impurities occur due to dissolution or reaction with a container. Thermodynamic criteria show that the solubility of the container increases with temperature. While some kinetic inhibition of container dissolution can be expected, at temperatures above about 1500 K, impurity concentrations of tens of parts per million can be expected. Containerless processing completely eliminates this source of contamination.

Inherent impurities are those present in the starting material. The concentration of these can sometimes be decreased by appropriate processing techniques. For example, some impurities can be removed as vapor, volatile oxide or other species.

5. Superheating Above the Liquidus: Nucleation can be caused by sparingly soluble impurities. Materials such as carbides, nitrides or oxides of similar density to the bulk material can remain suspended in the liquid and act as nuclei. If the liquid is superheated, the impurities can be dissolved.

6. Impurity Nucleation Studies: It has been shown that nucleation of gas bubbles in liquid iron can be controlled by adjusting the ambient oxygen pressure<sup>(9)</sup>. Refractory metal oxides can be formed from impurities at low activity such as aluminum. Once the ambient oxygen pressure is sufficient to form the refractory oxide, this provides a nucleation site.

7. Complete Processing Without Contact: This provides the possibility of repeated heating, melting and solidification without the introduction of impurities from handling.

#### HARDWARE REQUIREMENTS AND CAPABILITIES

The basic hardware requirements are outlined below. Operation in microgravity is necessary to achieve the requirements of many experiments. The minimum requirement for instrumentation is noncontact temperature measurement.

- \* Position control of poorly conducting liquid samples.
- \* High temperature capability, 2000-2500 K.
- \* Controlled atmosphere operation.
- \* Instrumentation: NCTM, Emittance, Gas Analysis, Video Imaging.

Following is a brief discussion of some flight hardware. These are described more fully in other papers in these proceedings.<sup>(10, 11)</sup>

Processing furnaces: Figure 2 shows the High Temperature Acoustic Levitator (HAL). This and the Acoustic Levitation Furnace (ALF) operate on a similar principle to the SAAL but with temperature capability to 2000 K or above. They are highly developed versions of the space hardened SAAL and incorporate a

advanced features<sup>(12)</sup>. Both use six acoustic transducers laid out in opposed orthogonal pairs. This produces spherically symmetric energy wells which can be used to process a liquid sample in a controlled atmosphere. The HAL system has been used to process liquid samples in a program aboard the KC-135<sup>(13)</sup>.

Instrumentation: The Division of Amplitude Polarimetric Pyrometer (DAPP) combines an accurate radiometer and an ellipsometer to provide true noncontact temperature measurement<sup>(14,15)</sup>. This application of an ellipsometer enables the emittance of the surface to be measured. The DAPP eliminates one of the major sources of uncertainty in noncontact temperature measurement.

#### CONCLUSIONS

The ability to process materials in microgravity extends the range of materials which can be studied and processed. The addition of microgravity processing experiments to a ground-based research program offers the opportunity to quantify the effects of heterogeneous nucleation, impurities and convection on specific systems.

Appropriate hardware already exists to make high temperature studies of ceramic materials. It is important that this technology is put into use and that flight opportunities become available in the near future. This goal could be achieved by extending the KC-135 program and supplementing this with sounding rocket experiments.

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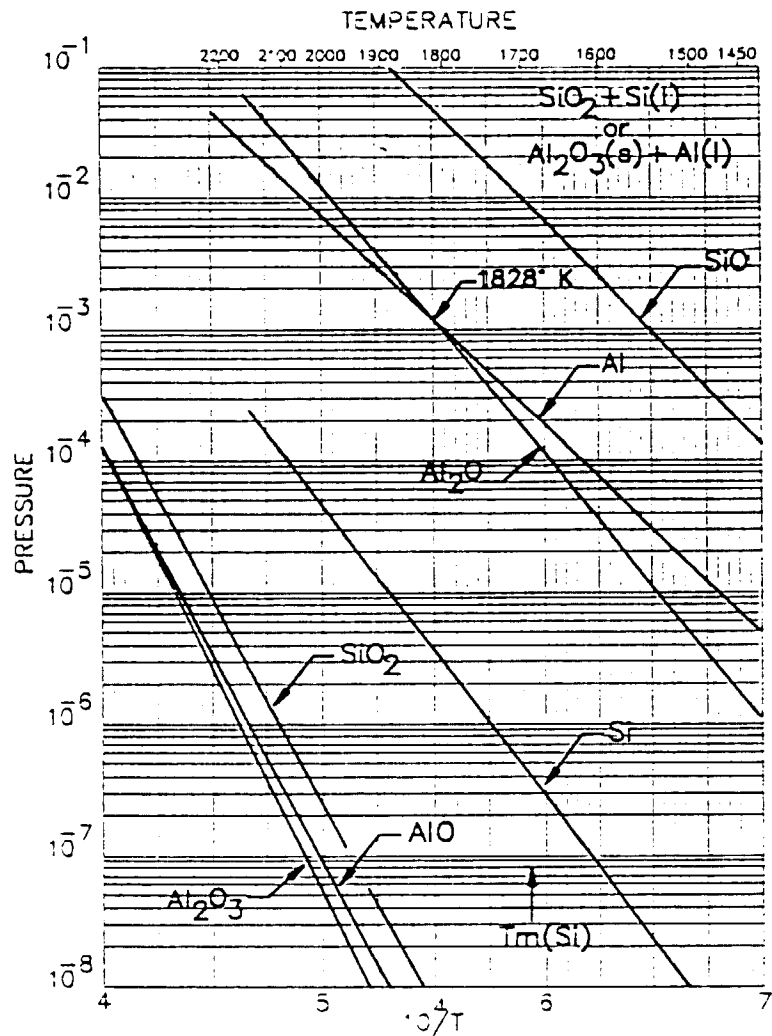


Figure 1

Plot of Vapor Pressure of Species over the Al-O and Si-O Systems as a Function of Inverse Temperature.  $p_{\text{O}_2} = 10^{-6}$  Atm.

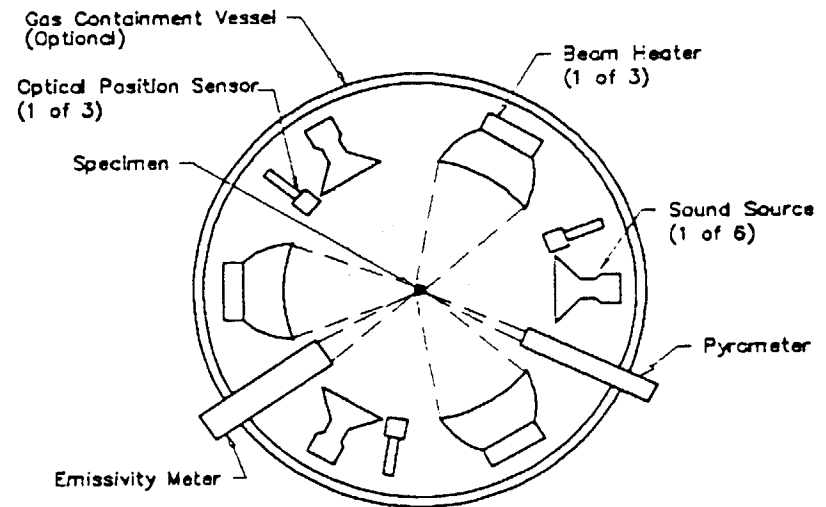


Figure 2

Schematic View of the Intersonics High Temperature Acoustic Levitator.