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THE USE OF "SELF HEATING" CERAMICS AS CRUCIBLES FOR MICROWAVE MELTING METALS AND NUCLEAR WASTE GLASS

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ABSTRACT

Silicon carbide (SiC) crucibles were used to melt aluminum and copper in conventional and tuned microwave cavities at a microwave frequency of 2450 MHz. SiC crucibles were also used to vitrify and homogenize mixtures of nuclear waste and glass frit.

BACKGROUND

A number of materials can be heated by applying energy to them in the form of high frequency electromagnetic waves(1). The mechanism of heating depends on the nature of the material, i.e., its electronic, molecular and crystalline structure and its microstructure and impurity content.

"Dielectric " heating is caused by polarization of the charges in a non conducting material and the inability of this polarization to follow the extremely rapid reversals of the applied high frequency electromagnetic field. There can also be **heating due to** relaxation and resonance processes under the influence of the **alternating magnetic field**. In addition **conduction type heating** occurs due to charge particles forming conducting paths under the influence of the applied field.

The first application of microwave heating to ceramics was due to M. L. Levinson(2,3) who patented a "Microwave Kiln" in 1969 and "Methods of Firing Ceramic Articles Utilizing Microwave Energy" in 1971.

A furnace with more uniform heating and requiring less microwave power was patented(4) in1981 by S. Maeda, Y. Minowa and H. Komura. The unit employs ZrO_2 and ZnO as heating materials.

A microwave muffle furnace employing SiC components was described by E. D. Neas and M. J.Collins(5). The heating elements for the furnaces are housed in microwave transparent refractory insulators and the furnaces fit into conventional size microwave ovens.

Materials that have high absorption factors for microwaves are described as **"lossy"** and heat rapidly; even in conventional microwave units with nominal powers, e.g.,600 watts. Some examples of lossy ceramics are SiC(1), ZrO_2, ZnO, UO_2, U_3O_8, and PuO_2 (cf. paper 6-SXI-91).

The crucible material for the present studies was SiC. The first microwave application of SiC was its use as a high efficiency "dummy load" to absorb radar and prevent its escape during tuning of radar transmitters for military application; where secrecy of location was important to the security of the device.

MELTING NUCLEAR WASTE GLASS

SiC crucibles and a muffle furnace(5) constructed of SiC components, were both used to vitrify and homogenize mixtures of simulated nuclear waste and glass frit. The SiC for the crucibles and the muffle furnace was housed in microwave transparent fused quartz foam insulation.

The microwave systems used for melting the nuclear waste glasses were 850 watt multimode commercial units with furnace temperature controls to (+/-) 5 °C. The model MDS-205 microwave unit and muffle furnace was manufactured by the CEM Corporation, 3100 Smith Farm Road, Matthews, N.C. 28105. The model RMS-150 microwave unit was manufactured by Floyd Inc., 5440 Hwy. 55 East, Lake Wylie, S.C. 29710.

The silicon carbide crucibles were procured from the Norton Company Advanced Ceramics, #1 New Bond street, Worcester, MA 01615. The crucible identification was "Cryston" type CN-137 which is a silicon nitride bonded composition. The dimensions were .044 m (1.75 in.) O.D X .038 m (1.50 in.) I.D. by .089 m (3.50 in.) length. The length was reduced to .051 m (2.00 in.) for these experiments.

To localize the heating and preserve the SiC crucible for further melts, the material to be melted was placed in a Pt, MgO, Al_2O_3 or BN crucible which was in turn placed in the SiC crucible.

Metals, in powder, strip or other forms , do not arc when enclosed in SiC crucibles or Si C furnaces in the microwave cavity because the "lossy" SiC absorbs nearly 100% of the microwaves, heats rapidly and in turn heats the metal.

The SiC muffle furnace reached a temperatures of 1100 ^{o} C in about 15 minutes and the SiC crucibles in about 10 minutes due to their smaller mass. Mixtures of 3 to 15 grams of simulated nuclear waste and glass frit were melted and homogenized in platinum and Al_2O_3 crucibles in less than 10 minutes. These nuclear waste glasses are discussed by C.M. Jantzen in the next paper of this session (paper 2-JXVII-91) "Characterization of Radioactive Waste Melter Feed Vitrified by Microwave Energy". One of the experimental arrangements used is shown in Figure 1.

The molten liquid glass was observed to be extremely mobile during microwave melting; displaying a wave like and sometimes bubbling motion. This natural agitation is probably due to the high thermal and field intensity gradients inherent in microwave heating and considerably enhances homogenization and diffusion in the molten liquid.

MELTING METALS

Metals can be heated, melted or reacted with other materials in a microwave cavity by placing the metal in a SiC crucible or other ceramic crucible which has a high absorption factor for microwaves. The SiC crucible rapidly heats by absorbing nearly 100 percent of the microwaves then transfers the heat to the metal. **The caveat of metal melting is oxidation, hence all melting must be performed in an inert atmosphere or under vacuum.**

Copper was melted using a 2 kilowatt, Model, MCR -1300 tunable resonant cavity, Figure 2, manufactured by Wavemat, Inc., 44191 Plymouth Oaks Boulevard, Suite 100, Plymouth, MI 48170.

The MCR-1300 "tuned" single mode cavity provided a higher microwave field intensity through direct coupling of the microwave energy with the SiC crucible and the metal sample. In these experiments, The cavity dimensions were continuously adjusted , i.e., "tuned", to maximize microwave absorption as it changed with increasing temperature. The temperature was recorded via an optical fiber thermometer system. The copper was completely melted (m.p. = 1085° C) in less than 20 minutes using a net power of 575 watts.

The microwave cavity was designed so that an inert gas could continuously flow over the sample. Argon and nitrogen gases were employed in the present experiments; however nitrogen caused a black oxide to form over the melt. An open port is available in the top of the cavity to observe or video the appearance of the sample as it heats. After melting, the copper was observed to bubble profusely throughout the crucible until the microwave power was turned off.

As suggested in the case of the glass; the bubbling is believed to be due to the field intensity and associated thermal gradients inherent in the microwave heating process. The stirring action provided by bubbling should enhance diffusion and homogenization and eliminate the need for mechanical stirrers required for carrying out some sluggish chemical reactions,e.g., molten salt reductions of metal oxides (cf. paper 6-SXI-91)

To preserve the SiC crucible for further melts the copper cylinder, .016 m (0.625 in.) diameter X .038 m (1.5 in.) length,was inserted in an MgO crucible, .032 m (1.25 in.) OD X .025 m (1.00 in.) ID X .064 m (2.5 in.) length. The MgO crucible was in turn inserted in the SiC crucible whose dimensions were .064 m (1.8 in.) OD X .038 m (1.5 in.) ID X .064 m (2.5 in.) length. The MgO crucible was relatively transparent to the microwave radiation.

Metals may also be melted with the Floyd model RMS-150 and the CEM model MDS-205 multimode units discussed above as long as an inert gas or vacuum environment is provided .

For these units and the Wavemat Inc., MCR 1300; the mass of the "lossy" material, i.e. the SiC crucible, or the argon pressure must be adjusted to prevent breakdown of the argon into a plasma by the microwaves.

CONCLUSIONS

SiC crucibles heated by microwaves were used as secondary containers to melt and homogenize mixtures of nuclear waste and glass frit and to melt metals.

The microwave heating system is more flexible and efficient since the crucible's mass and geometry can be designed to provide the required power and be located closer to the sample than in conventional furnaces.

During microwave melting the molten liquid is agitated by bubbling and wave like motion; hence reaction rates and homogenization are enhanced.

For heating radioactive materials in shielded remote facilities; microwave heating equipment is simpler to remote(6) and requires less maintenance than conventional furnaces.

REFERENCES

1. R. C. Metaxas, R.J. Meredith, "Industrial Microwave Heating", Peter Peregrinus Ltd., London, UK., 1983.
2. M. L. Levinson, "A Microwave Kiln", U.S. Patent # 3,469,053, 1969.
3. M. L. Levinson, "Methods of Firing Ceramic Articles Utilizing Microwave Energy", U.S. Patent # 3,585,258, 1971.
4. S. Maeda, Y. Minowa, H. Komura, "Microwave Heating Oven" , U.S. Patent # 4,307,277, 1981.
5. E. D. Neas, M. J. Collins, Chapter 2, "Microwave Heating", pp. 28,30, "Introduction to Microwave Sample Preparation", H.M. Kingston, L.B. Jassie, ACS Professional Reference Book, American Chemical Society, Washington, D. C., 1988.
6. E. F. Sturcken, T. S. Floyd, D. P. Manchester, Chapter 9, "Remote Operation of Microwave Systems", pp. 188-202, "Introduction to Microwave Sample Preparation", H.M. Kingston, L.B.Jassie, ACS Professional Reference Book, American Chemical Society, Washington, D. C., 1988.

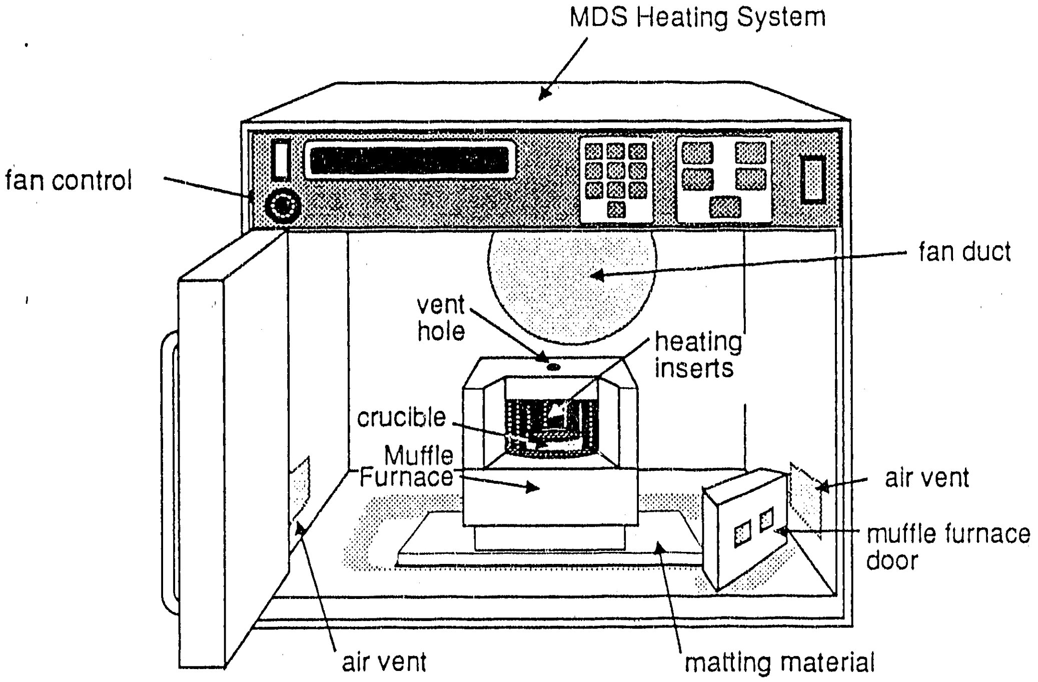

Fig. 1 Furnace Arrangement For Melting Glass. The furnace and MDS-81 microwave unit shown are manufactured by the CEM Corporation(5). The author has also used an RMS-150 microwave unit, manufactured by Floyd Inc., 5440 Hwy. 55 East, Lake Wylie, S.C. 29710.

In the Floyd furnace , an Al_2O_3 Crucible, containing the mixture of simulated nuclear waste and glass frit, was contained in an SiC crucible which was mounted in a block of fused quartz foam insulation. The insulation was split in two with cylindrical holes in the top and bottom so that the SiC crucible fit snugly into the holes. Heating was more rapid and required less power with the crucible arrangement since the heat source was more insulated and closer to the sample. The crucible arrangement is more flexible since one can design the fused quartz insulation and the number, geometry and mass of the SiC crucibles to fit the needs of the experiment.

The Floyd, Inc. RMS-150 microwave system will be more suitable for the DWPF and other WSRC operations since it is already remoted for operation in environments that are either corrosive or contain a high level of nuclear radiation that is harmful to personnel and deteriorate the physical, mechanical and electronic properties of the instrument.

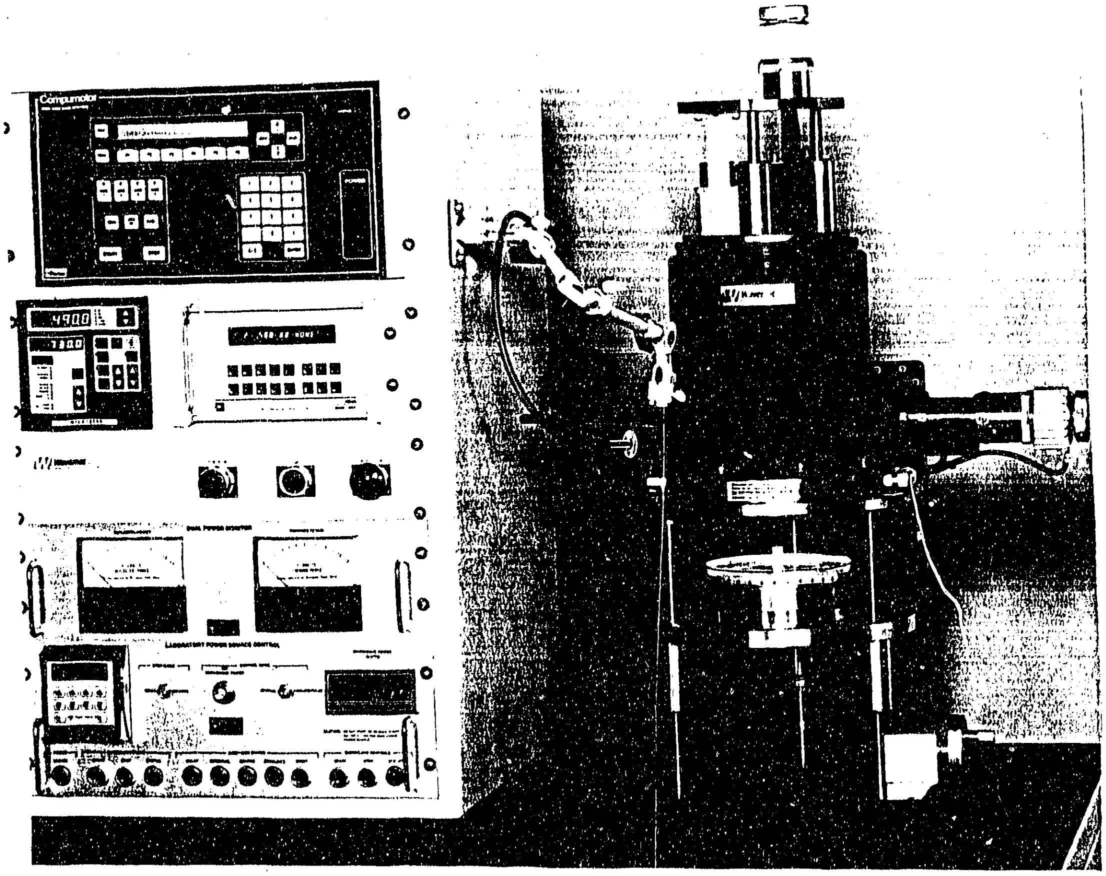

Fig. 2. Tunable Microwave Cavity. Copper was melted in the microwave cavity shown above; manufactured by Wavemat, Inc., 44191 Plymouth Oaks Boulevard, Suite 100, Plymouth, MI 48170.

As the microwave absorption changed with temperature increase; the absorption of microwaves by the SiC crucible containing the copper was continuously maximized by changing the dimensions of the cavity with the vernier controlled sliding short at the top of the cavity and the adjustable microwave "launch" probe in the wall cavity on the right side. The inlet and outlet for gas are located on the top and bottom of the cavity. The cavity is water cooled. The temperature is sensed through the front glass port by the optical fiber thermometer system attached to the right side of the instrument panel.

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