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Alternative Crucibles for U-Mo Microwave Melting

March 2017

BW Kirby

U.S. DEPARTMENT OF ENERGY

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Pacific Northwest National Laboratory
Richland, Washington 99352

Summary

Microwave melting will be used in the casting process to produce a low-enriched uranium-molybdenum (U-Mo) alloy at the Uranium Processing Facility at the Y-12 Complex. The crucibles used currently contain silicon carbide (SiC) in a mullite ($3Al_2O_3$-$2SiO_2$) matrix with an erbia coating in contact with the melt. These crucibles are causing issues with silicon impurities in the U-Mo that are likely transported via the gas phase. Pacific Northwest National Laboratory has investigated alternative crucible materials that are susceptible to microwave radiation and are chemically compatible with molten U-Mo at 1400-1500°C. Recommended crucibles for further testing are: 1) high-purity alumina (Al_2O_3); 2) yttria-stabilized zirconia (ZrO_2); 3) a composite of alumina and yttria-stabilized zirconia; 4) aluminum nitride (AlN). Only AlN does not require an erbia coating. The recommended secondary susceptor, for heating at low temperature, is SiC in a “picket fence” arrangement. Zirconia and possibly the alumina/zirconia composite would not require a secondary susceptor.

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1.0 Introduction

Microwave melting will be used in the casting process to produce low-enriched uranium-molybdenum (U-Mo) alloy at the Uranium Processing Facility at the Y-12 Complex. The crucibles used currently contain silicon carbide (SiC) in a mullite ($3Al_2O_3$-$2SiO_2$) matrix with an erbia coating in contact with the melt. These crucibles are causing issues related to silicon impurities in the U-Mo that are likely transported via the gas phase. Pacific Northwest National Laboratory has investigated alternative crucible materials that are susceptible to microwave radiation and are chemically compatible with molten U-Mo at 1400–1500°C.

2.0 Microwave Heating

Microwaves are defined as electromagnetic radiation in the frequency range of 300 MHz to 300 GHz, which corresponds to wavelengths from 1 m to 1 mm. Microwave energy interacts with ceramic materials to produce heat primarily by charge transport mechanisms and dipole reorientations. Interactions vary with temperature (because electrical and ionic conductivity change), and also with the specific frequency of incident radiation (because frequency must be near the timescales of the relevant physical processes in order to interact).[1]

The complex permittivity of a material (ε) is given by:

$$\varepsilon = \varepsilon_0(\varepsilon' - i\varepsilon'')$$

where ε_0 is the permittivity of free space (8.86×10^{-12} F/m), ε’ is the relative dielectric constant, and ε’’ is the relative dielectric loss factor.

A common metric for quantifying dielectric loss is given by the dissipation factor, tanδ (“tan delta”)[1]:

$$tan\delta = \frac{\varepsilon''}{\varepsilon'} = \frac{\sigma}{2\pi f \varepsilon_0 \varepsilon'}$$

where σ is the total effective conductivity in the sample (S/m) and f is the frequency (Hz). This report uses tanδ to compare the microwave susceptibility of various potential materials.

Power, P (W/m^3) dissipated in the material is given by[1]:

$$P = \sigma|E|^2 = 2\pi f \varepsilon_0 \varepsilon' tan\delta |E|^2$$

where $|E|$ (V/m) is the magnitude of the internal field. So dissipated power is proportional to the frequency, the dielectric strength, the internal field strength, and tanδ. Higher frequency typically leads to more dissipated power, even though tanδ tends to decrease with increasing frequency. The most common wavelength used in commercial microwaves is 2.45 GHz. Higher frequency systems are generally more costly. It is believed that the Y-12 system operates at a fixed frequency in the range near 2.45 GHz.

3.0 Alternative Crucible Materials

3.1 Current Material

Current crucibles in use at Y-12 for U-Mo melting are a composite of mullite ($3Al_2O_3$-$2SiO_2$) with SiC. An erbium oxide coating on the crucible is what actually contacts the melt. SiC is an excellent absorber of microwave energy, including at room temperature. The SiC heats the mullite matrix until it reaches a temperature at which the alumina itself also begins to absorb microwave energy. At that point, the whole crucible body is producing heat, ultimately melting the U-Mo alloy and reaching a temperature of 1400–1500°C.

When the U-Mo alloy is cast after this melting process, undesirable silicon impurities have been found to be present. The current understanding is that silicon species are transported in the gas phase (argon atmosphere) from the crucible body to the melt. The goal of this study is to recommend alternative crucible materials that do not contain silicon.

3.2 Comparison of Alternative Materials

A review of refractory ceramics and their dielectric properties was performed and the results are presented in Figure 1. Data were not available to compare all materials at the full range of temperatures and microwave frequencies. The frequencies of measurement are noted for each material. Tanδ tends to decrease with increasing frequency, so for the materials listed at frequencies higher than 2.45 GHz, these data should be considered a lower bound on tanδ.

Low-loss materials are considered to be those with a tanδ less than approximately 0.01. This region of Figure 1 is labeled as "Transparent." Some small amount of heating can still occur, but processing times will be longer. The region labeled "Reflective" is for a tanδ greater than approximately 2, when materials do not allow penetration of microwaves beyond a small skin depth. Metals, including molten metals, are typically microwave reflectors. Lossy materials are those that absorb a significant portion of microwave energy, with $0.01 < \tan\delta < 2$. This region is labeled "Absorbing" in Figure 1. In the discussion of these materials, a distinction is made between the crucible material (such as mullite, currently), and the secondary susceptor (such as SiC, currently). At high enough temperature, the crucible material also becomes a susceptor. For the sake of clarity, a secondary susceptor is a material that begins heating at or near room temperature and leads to the heating of the main crucible material. This process is generally referred to as hybrid microwave heating when the goal is to sinter or heat treat the primary body.

Room temperature data for the secondary susceptors graphite and magnetite (Fe_3O_4) are shown in Figure 1. Both of these materials experience rapid temperature rise in a microwave field, much like SiC. High-temperature values of tanδ were not found. An additional value for the magnetic tanδ for magnetite is shown. Magnetite interacts with the magnetic field as well as the electric field, producing heat from both processes. The critical temperature where ferromagnetism breaks down in magnetite is 850°C, above which only the electric field interaction would produce heat.

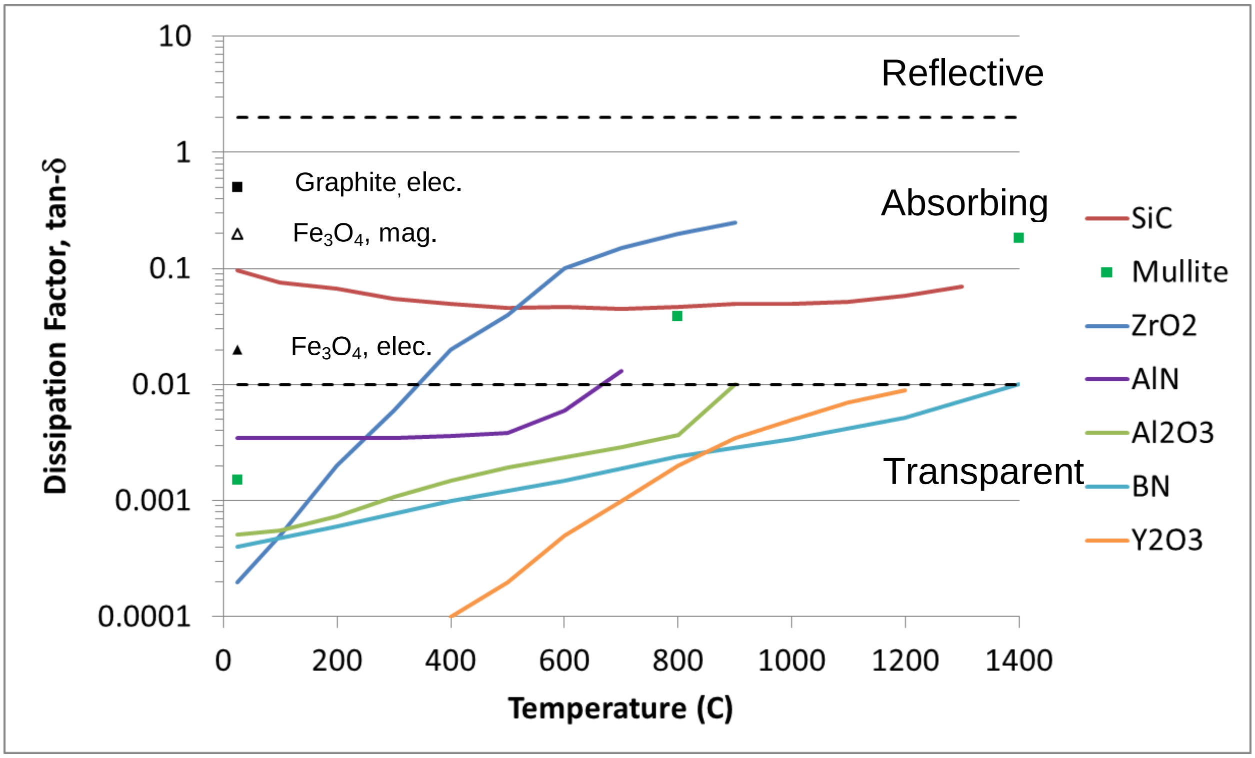

Figure 1. Tanδ for various candidate crucible and secondary susceptor materials: SiC, 3.02 GHz[2]; mullite, $3Al_2O_3$-$2SiO_2$, 3GHz[3]; Zr_2O_3 (cubic, unknown dopants), 2.45 GHz[4]; AlN, 8.5 GHz[5]; Al_2O_3, 3.7 GHz[5]; BN, 4.9 GHz[6]; Y_2O_3, 5 GHz[7]; graphite, 2.45 GHz[3]; Fe_3O_4, 2.45 GHz[3].

These values should be considered representative. Particle size, phase, dopants, density, and overall geometry will affect microwave absorption characteristics. A brief discussion of each material follows. Thermal conductivity data are also given. Thermal conductivity affects the ability of a material to distribute heat internally. Non-uniform microwave fields combined with low thermal conductivity can lead to hot spots and thermal runaway as hotter regions become better and better absorbers of microwave energy.

3.2.1 Oxides

Uranium oxide is extraordinarily stable; very few oxides have a more negative free energy of formation, as seen on a standard Ellingham diagram. Most oxides would be at risk of reduction to their metallic state by the U-Mo melt. Oxides that are more stable than UO_2 include yttria, erbia, and magnesium oxide (up to 1400°C). The current alumina crucible must be coated with erbia to prevent reaction with the melt.

3.2.1.1 Mullite ($3Al_2O_3$-$2SiO_2$)

Mullite is the main crucible material used currently for microwave melting. It is combined with a SiC secondary susceptor in the form of a composite crucible with SiC powder. Mullite must be protected from the melt by an erbia coating. Only a few data points for tanδ at high temperature were available, but clearly mullite absorbs significant microwave energy at high temperature. Mullite is a possible source of the observed silicon impurity in the U-Mo melt. The thermal conductivity of mullite is 6 W/m·K.

3.2.1.2 Alumina

Alumina reaches the threshold of tanδ = 0.01 at around 900°C. Alumina must be protected from the melt by an erbia coating. Alumina is the most common ceramic, and experience with this material may make it an attractive candidate; however, it must be paired with a secondary susceptor. Brosnan et al.[8] were able to couple to an alumina sample at 2.45 GHz with a SiC powder bed used as a secondary susceptor. A sample temperature of 900°C was reached in 10 minutes with further heating at 45–60°C/min. Details of secondary susceptor materials are discussed below. The thermal conductivity of alumina is ~32 W/m·K.

3.2.1.3 Zirconia

Cubic stabilized zirconia (typically with yttria dopant) has high oxygen conductivity and therefore interacts strongly with microwave radiation. It reaches the threshold of tanδ = 0.01 at around 350°C. Yttria-stabilized zirconia may be a candidate material for direct heating, i.e., without a secondary susceptor. It must be protected from the melt by an erbia coating.

Baeraky[9] found that pure ZrO_2 and MgO-doped ZrO_2 do not couple strongly to microwaves, but Y_2O_3-doped ZrO_2 does. Janney[10] describes a series of experiments firing 3% and 8% yttria-doped zircona. Rapidly rising tanδ, combined with low thermal conductivity, led to runaway heating and localized hot spots and sample cracking in a non-uniform 2.45 GHz furnace. Such non-uniformity is typical of consumer microwave ovens in which interference patterns in the cavity lead to widely varying field strengths in regions that are only a few centimeters apart. Experiments in a 28 GHz high-quality and highly uniform furnace gave rapid, uniform heating without sample cracking. The researchers developed a "picket fence" arrangement of SiC secondary susceptor rods surrounding the zirconia body. Figure 2 depicts what this arrangement might look like in the U-Mo furnace. At temperatures up to 600°C, the SiC rods absorbed microwaves and heated the zirconia. Above 600°C, the zirconia absorbed more strongly than the SiC, causing the SiC to cool because most of the microwave energy was being absorbed by the zirconia. Heating rates had to be limited to avoid cracking to 2°C/min below 500°C and 3.5°C/min above 500°C. Zirconia has one of the lowest thermal conductivities of any ceramic, 2 W/m·K. A highly uniform microwave furnace may mitigate the formation of hot spots and allow higher heating rates than those discussed above.

3.2.1.4 Yttria

Pure yttria is one of the few oxides more stable than uranium oxide, so it could be used without an erbia coating. Overall microwave susceptibility is low, however, and a secondary susceptor would be required to reach ~1200°C before significant absorption by yttria would occur. The thermal conductivity of yttria is 8–12 W/m·K.

Mathew[11] successfully sintered nanoscale yttria powder in a microwave furnace using a SiC powder bed as a secondary susceptor. The heating rate was 40°C/min up to 1540°C using two 1.1 kW magnetrons at 2.45 GHz. An additional experiment used the same microwave heating with the SiC powder bed and additional heating from resistive molybdenum heating elements. That additional heating reduced the sintering temperature to 1450°C.

3.2.1.5 Other Oxides

MgO is more stable than uranium oxide up to 1400°C, but low microwave susceptibility was disqualifying. No microwave susceptibility data were available for erbia (Er_2O_3). Other exotic lanthanide and actinide oxides were not considered due to cost.

3.2.2 Non-Oxides

3.2.2.1 Aluminum Nitride

Aluminum nitride shows promising microwave susceptibility, reaching the threshold of $\tan\delta = 0.01$ at around 700°C. Higher temperature data were not available, but $\tan\delta$ would be expected to increase further at higher temperature. Aluminum nitride is stable in inert atmospheres at temperatures over 2000°C. Bulk oxidation occurs in air at 1380°C. It is much more stable than both molybdenum nitride and uranium nitride, so it could be used without an erbia coating. Aluminum nitride has an exceptionally high thermal conductivity, 100–200 W/m·K.

3.2.2.2 Boron Nitride

Boron nitride is stable in inert atmospheres up to 2000°C, and in air up to 850°C. Grade XP refers to pure boron nitride without additives or binders, and is the required grade for extreme temperature use. It is chemically stable in the presence of Mo and U. Microwave susceptibility is low, however, which may disqualify boron nitride as an option here. The thermal conductivity of boron nitride is 71 W/m·K.

3.2.2.3 Other Non-Oxides

Boron carbide, B_4C, was considered, but is not stable at high enough temperatures.

3.2.3 Secondary Susceptors

3.2.3.1 Silicon Carbide

SiC is used at the secondary susceptor in the current crucible, presumably as particles dispersed in the mullite matrix of the crucible. SiC is the most common secondary susceptor, either in powder or solid form. SiC is a possible source of the silicon contamination seen in the U-Mo melt, but the contamination may also be coming from the mullite itself, so SiC should not be completely ruled out as a secondary susceptor. Microwave absorption is excellent across a wide temperature range.

SiC can be incorporated with another ceramic as a composite, as is done now. One possibility would be full encapsulation of SiC in a pure alumina (Al_2O_3) crucible.

SiC can also be used in a "picket fence" arrangement, where solid rods of SiC are placed around the crucible[10] (see Figure 2). Radiative heat transfer from the SiC rods works to raise the temperature of the crucible until it begins absorbing microwave energy.

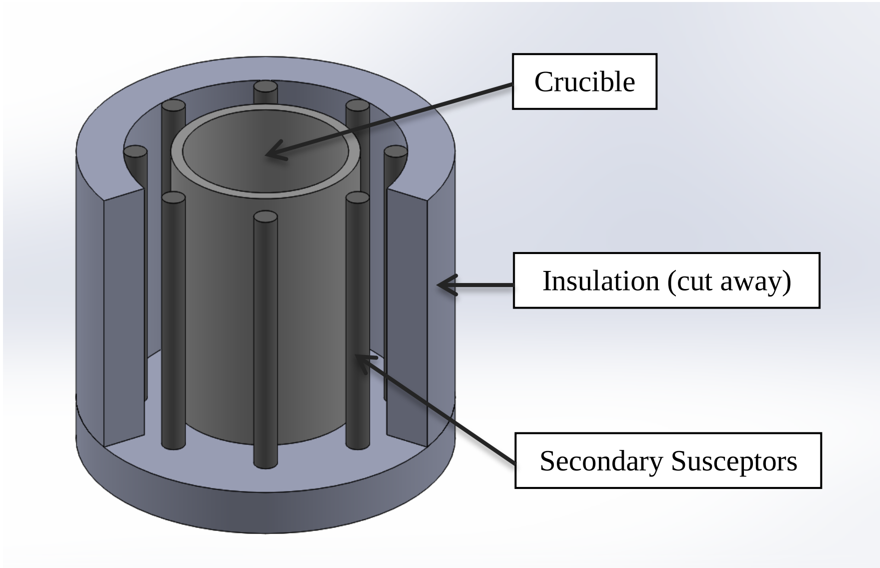

Figure 2. Picket fence arrangement of secondary susceptor rods around a crucible for hybrid microwave heating.

3.2.3.2 Molybdenum Disilicide ($MoSi_2$)

Specific dielectric loss data were not available, but $MoSi_2$ has been used as a secondary susceptor in the picket fence arrangement,[12], much like for SiC. $MoSi_2$ is a common high-temperature resistive heating element. It may be a preferable option for limiting Si volatility.

3.2.3.3 Graphite

Graphite is another common microwave susceptor, absorbing even more strongly than SiC at room temperature. Graphite crucibles are used in the induction melting process for U-Mo. Graphite can be used in composite form or in the "picket fence" arrangement discussed above.

3.2.3.4 Zirconia (ZrO_2)

Zirconia is a strong enough microwave absorber at room temperature that has been used as a secondary susceptor. Yttria-stabilized zirconia is the most noted material for high microwave susceptibility. Zirconia could be used in a composite with alumina to increase susceptibility.

Samuels[13] used zirconia fibers packed into an alumina container as a secondary susceptor, though this required preheating in a conventional furnace. Later experiments in a more powerful microwave furnace with a "novel susceptor design" did not require preheating.[13]. Lee et al.[14] used a porous zirconia cylinder in a 600 W, 2.45 GHz microwave and achieved steady-state temperatures (only due to the secondary susceptor) of 1112–1519°C depending on geometry.

3.2.3.5 Magnetite (Fe_3O_4)

Magnetite strongly absorbs microwave energy at room temperature, and interacts with both the electric and magnetic fields. Magnetic susceptibility shuts down above the Curie temperature of 585°C, while electrical susceptibility continues. Composites of Fe_3O_4 with ceramics may be challenging due to thermal expansion mismatch. Thermal expansion coefficient of Fe_3O_4 is 23 $10^{-6}K^{-1}$ to 1000°C. Additionally, the low partial pressure of oxygen in the melt furnace may reduce magnetite to metallic iron.

3.3 Recommendations

The material(s) selected for microwave melting of U-Mo must be adequate absorbers of microwave energy and be chemically compatible with molten U-Mo. The current mullite/SiC crucible causes silicon impurities in the melt. Carbon contamination from induction melting crucibles has also been a concern. Alternative crucible materials and secondary susceptors that are likely to satisfy these requirements are recommended below. These recommendations are a starting point and should be reviewed by subject matter experts in uranium processing and microwave systems.

3.3.1 Crucible Materials

Table 1. Recommended candidate crucible materials. Good absorbance occurs at tanδ > 0.01.

Crucible Material	Coating Required?	Secondary Susceptor Required?	Thermal Conductivity (W/m·K)	tanδ 25°C	tanδ 500°C	tanδ 900°C
Al_2O_3	Yes	Yes	32	.0005	.002	.01
ZrO_2	Yes	No	2	.0002	.04	0.25
AlN	No	Yes	100–200	.004	.004	>0.01

Option 1: Alumina

To limit the possibility of any impurities, high-purity alumina would be an excellent choice. An alumina crucible would require an erbia coating and must be paired with a secondary susceptor. It has the lowest microwave susceptibility of the recommended options, but good thermal conductivity limits the danger of hot spots.

Option 2: Yttria-Stabilized Zirconia

Of the recommended options, yttria-stabilized zirconia is the strongest absorber of microwave energy—so much so that it may not require a secondary susceptor. This would be a good option if the simplicity of the crucible installation is critical. An erbia coating is required. Higher levels of Y-doping, such as 8 mol% Y, lead to higher ionic conductivity, which should lead to higher microwave susceptibility (microwave susceptibility data[4] presented in this report are for unknown dopants). The low thermal conductivity of zirconia, combined with the steep increase in microwave susceptibility, can lead to hot spots and thermal runaway. This effect can be limited by a more uniform microwave field, or by a reduced heating rate.

Option 3: Alumina/Zirconia Composite

A composite of alumina with yttria-stabilized zirconia combines some of the benefits of each material. The zirconia phase will absorb microwaves strongly, while the higher thermal conductivity of the alumina phase will act to spread out the generated heat and prevent hot spots. This may allow for higher heating rates. Many examples of alumina/zirconia composites have been studied. The volume percent of alumina should be at least 50% to ensure good heat conduction. Yttria-stabilized zirconia could be incorporated as particles or fibers, at 8 mol% Y as discussed above. Such composites may have higher mechanical strength than pure alumina. An erbia coating is required. A secondary susceptor may still be required depending on the susceptibility of the composite.

Option 4: Aluminum Nitride

Aluminum nitride is the only recommended option that does not require an erbia coating, because it is more stable than the nitrides of uranium and molybdenum. It also has very high thermal conductivity, which could enable high heating rates. Its microwave susceptibility is higher than that of alumina, but lower than that of yttria-stabilized zirconia. A secondary susceptor would still be required.

3.3.2 Secondary Susceptors, Geometry, and Materials

Secondary susceptors can be employed in various geometries: a powder bed for the crucible, a composite material with the crucible material itself, a cylinder that completely surrounds the crucible, or in the picket fence arrangement. A powder bed would make processing quite complicated. Composite materials have the potential to interact with the melt. A surrounding cylinder must be chosen carefully for thickness and would require production of a new cylinder for each experiment. The picket fence arrangement offers the simplest and most easily adjustable option.

In the picket fence arrangement, rods of the chosen material should extend vertically for the full height of the crucible. The rods should be fixed and supported by the surrounding insulation, and spaced evenly around the crucible. Eight rods would be a good starting point for experimentation. With these large crucibles, more rods may be required. Faster heating will be obtained the closer the secondary susceptor rods are to the outside of the crucible.

SiC is the most common secondary susceptor. While silicon impurities have been seen in the U-Mo product, these may be coming from the SiO_2 in the mullite crucible and not from the SiC. SiC is the recommended secondary susceptor for the picket fence arrangement.

$MoSi_2$ is another option, but it is also a potential source of silicon impurities. Graphite rods may also be used as secondary susceptors. These may lead to plasma formation in the furnace. If graphite rods are used, the ends should be smoothly rounded to avoid sharp points in the electric field that can lead to plasma formation.

Fibrous yttria-stabilized zirconia can be employed as an insulating material that also absorbs microwaves as a secondary susceptor. Thickness and packing density can be adjusted to adjust absorbance. Depending on the furnace geometry, this may be an option.

3.3.3 Recommended Experiments

Experiments should be conducted to determine the heating rate and chemical compatibility of the chosen materials. The exact properties of the microwave melting furnace at Y-12 are not known. Because each microwave cavity is unique, testing for heating rates should be done in the actual furnace to be used; small-scale testing may be of little value. This will require procurement of a full-sized crucible and any secondary susceptor materials. Coating the crucible with erbia would not be necessary for these tests. The empty crucible can be heated and monitored in the melting furnace to determine if heating can be accomplished in a reasonable time. Secondary susceptor geometry (rod diameter, number of rods) can be adjusted during these experiments, if required.

Once adequate heating has been demonstrated, a representative melt can be processed with a depleted U-Mo alloy to demonstrate chemical compatibility. Small-scale testing may be useful here, possibly in a conventional furnace. An erbia coating should be applied (except for AlN crucibles) before melting is attempted. Standard analytical techniques used for quality control of the U-Mo product should be employed to look for any impurities.

The amount of development required to demonstrate microwave melting with these materials is estimated to be low (<\$500K). The melting furnace is already in place, and the recommended materials are all widely available.

3.3.4 Suppliers

The first contact for a crucible supplier would be Hadron Technologies (formerly Microwave Synergy) in Arvada, Colorado. Hadron supplied Y-12 with the 24 kW microwave melting furnace and will have insight into crucible composition and secondary susceptor geometry.

CoorsTek in Golden, Colorado has a wide range of custom ceramic capabilities.

Custom carbon and SiC products are available from MWI in Rochester, NY.

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