

Thorium Sintering Flow Calculations

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We are planning to sinter a mixture of thorium metal and thorium dioxide in a tube furnace at 1500°C in an inert atmosphere of flowing, 5N (99.999% pure) argon. The last time we did this, a significant amount of radioactive thorium was carried out of the tube furnace system and contaminated the exterior of the bucket through which the argon was being bubbled.



Figure 1: Picture from Jacob Baron of the tube furnace. Note the flow meter and the HEPA filter on the outflow hose, which were installed after the initial run released thorium onto the exterior of the outflow bucket.

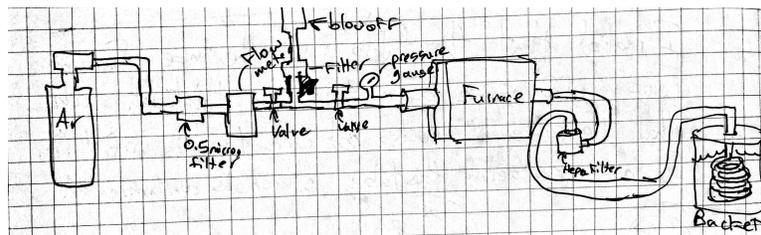


Figure 2: Schematic by Jacob Baron of current tube furnace setup with a 5N argon supply, a flow meter, and a blowoff valve installed on the upstream side of the furnace, and a HEPA filter installed on its downstream side followed by a bucket of water with a coil of copper tube. This setup is designed to trap condensibles either by filtration or by diffusion to a solid or liquid surface.

In light of this incident, we have added a HEPA filter to the downstream side of the tube furnace to trap thorium before it escapes the system. See Figures 1 and 2 for a picture and diagram of the new setup. We would also like to run the argon at the lowest practical flow rate to reduce the amount of radioactive material entrained in the gas in the exit lines. This document outlines some conditions on the argon flow rate.

1 Pumpout and Diffusion Timescales

We would like for a thorium atom to diffuse to the wall of the tube’s outflow hose and stick there long before the time it takes for an atom entrained in the argon flow to escape the system. The time-scale for bulk argon flow to carry an atom out of the system is given by:

$$t_{\text{flow}} = (\text{Length of tube}) \times (\text{Cross-sectional area of tube}) / (\text{Argon flow rate}) \quad (1)$$

$$t_{\text{flow}}^{\text{tube}} \approx \frac{20 \text{ cm} \times \pi(3 \text{ cm})^2}{f_{\text{Ar}}} \quad t_{\text{flow}}^{\text{hose}} \approx \frac{200 \text{ cm} \times \pi(0.5 \text{ cm})^2}{f_{\text{Ar}}} \quad (2)$$

$$t_{\text{flow}}^{\text{tube}} \approx \frac{570 \text{ cm}^3}{f_{\text{Ar}}} \quad t_{\text{flow}}^{\text{hose}} \approx \frac{160 \text{ cm}^3}{f_{\text{Ar}}}. \quad (3)$$

The first terms on the right in Eq. (2) and (3) give the flow time through the cold part of the alumina tube protruding from the furnace, while the second terms give the flow time through the ID 1 cm hose. Therefore, the atoms spend only about 22% of their total traverse time in the narrow hose.

The diffusion time to the wall is given by

$$t_{\text{diff}} = \frac{x^2}{6D}, \quad (4)$$

where x is the distance to the nearest cold wall, and D is the diffusion coefficient. The distance x is approximately the diameter of the alumina tube (6 cm) for the first 78% of their trip and the diameter of the hose (1 cm) for the last 22% of the trip. The diffusion constant is given approximately by (see [3] Eq. (30)):

$$D \approx \frac{3}{16n_{\text{Ar}}\sigma} \left(\frac{2\pi kT}{m_{\text{Ar}}} \right)^{1/2}, \quad (5)$$

where n_{Ar} is the number density of argon atoms in the system, σ is the collision cross section between thorium and argon, m_{Ar} is the atomic mass of argon, k is the Boltzmann constant, and T is the temperature. We can find the number density of argon at standard temperature and pressure using the ideal gas law. The result is $n_{\text{Ar}} \approx 2.5 \times 10^{19} \text{ cm}^{-3}$. We estimate the collision cross section to be $\sigma \approx 10^{-14} \text{ cm}^2$. We take the temperature to be 300 K. The diffusion constant is then

$$D \approx 0.05 \text{ cm}^2/\text{s}. \quad (6)$$

The diffusion time to the wall in the alumina furnace tube is therefore $t_{\text{diff}}^{\text{tube}} \approx 2 \text{ min.}$, and the diffusion time to the wall in the hose is $t_{\text{diff}}^{\text{hose}} \approx 3 \text{ s}$. Since the diffusion time is so much shorter in the narrow hose, the probability of a thorium atom depositing in the hose is about 10 times the probability of it depositing in the tube in spite of the relatively short amount of time spent in the tube, assuming a uniform distribution of thorium throughout the system. We therefore neglect the tube for the remainder of this comparison.

We would like the flow rate to be such that the entrained thorium molecules spend several diffusion times t_{diff} in the hose, i.e., $t_{\text{flow}}^{\text{hose}} \geq 10 \text{ s}$. (We want several diffusion times both because x is only a *mean* travel distance and because the sticking probability at a surface is probably not exactly unity.) Solving Eq. (3) for f_{Ar} sets an upper limit on the flow of

$$f_{\text{Ar}} \leq 1000 \text{ sccm}. \quad (7)$$

We would, however, like to run in the upper part of the permissible range, at least while the tube is heating up and desorbing reactive contaminants near the target, in order to carry away these impurities as quickly as possible. Furthermore, it should be fine to run a little (order unity) above this flow range since most particles (and, I believe, free atoms of species like thorium that have low vapor pressure at room temperature) will be trapped by the HEPA filter, anyway.

2 Target Contamination Timescales

I have done some additional calculations showing that if a significant fraction of the outgassed impurities—or indeed, even a significant fraction of the impurities in the 5N argon—that impinge on the target were to react with it, the top layer of grains would be consumed in a matter of seconds. These calculations are described below. However, I imagine that either this effect does not occur (i.e., a very small fraction of contaminants are able to react with thorium), or it is unimportant since other groups seem to have managed to fire thorium at these temperatures with only moderate precautions against contamination (see, e.g. [1, 2]).

With 5N argon at atmospheric pressure, the minimum partial pressure of impurities is $760 \text{ Torr} \times 0.00001 \approx 10 \text{ mTorr}$. Let’s make the extremely conservative assumption that all of these impurities are reactive with thorium metal at the sintering temperature of 1500°C . From kinetic gas theory, the rate of collisions per unit area of a gas at pressure P and temperature T with a wall of the container is

$$A = \frac{P}{\sqrt{2\pi kTm}}, \quad (8)$$

where m is the atomic mass of the gas. We assume that the gas in the hot part of the tube is in mechanical equilibrium with the cold gas outside so that the partial pressure of contaminants is $P \approx 10$ mTorr. We take the temperature to be 1500°C , and for m we assume the atomic mass of molecular oxygen, the “worst-case” contaminant. Therefore, the number of collisions per unit area of contaminant gases with the target and tube surface is

$$A \approx \frac{10 \text{ mTorr} \times (101300 \text{ Pa}/760 \text{ Torr})}{\sqrt{2\pi(1.381 \times 10^{-23} \text{ J/K})(1800 \text{ K})(0.032 \text{ kg/mol})/(6.02 \times 10^{23} \text{ molecules/mol})}} \quad (9)$$

$$= 1.5 \times 10^{22} \text{ molecules/m}^2\text{s}. \quad (10)$$

The first thorium atoms that these impurities encounter will be in the top layer of grains. The size scale for a grain is about $50 \mu\text{m}$ (see this lablog entry estimating the grain size: https://bussle.rc.fas.harvard.edu/lablog_EDMgraduate_2009-Elizabeth_Petrik/2010/01/thorium_metal_powder_1.html). We now calculate the number of thorium atoms per unit area in this top grain layer:

$$\text{No. thorium atoms per unit area} = (\text{Thickness of layer}) \times (\text{Mass density of thorium})/(\text{Atomic weight of thorium}) \quad (11)$$

$$= (50 \mu\text{m}) \times (1.17 \times 10^4 \frac{\text{kg}}{\text{m}^3}) \times \frac{6.02 \times 10^{23} \text{ atoms}}{0.232 \text{ kg}} \quad (12)$$

$$= 1.5 \times 10^{24} \frac{\text{atoms}}{\text{m}^2}. \quad (13)$$

By dividing Eq. (10) into Eq. (13), we see that if the contaminants in the argon atmosphere are highly reactive with thorium at the sintering temperature, the top layer of grains will be consumed in a matter of ~ 100 s or so. In order for a significant fraction of the top layer of grains to be preserved for the entire sintering time of ~ 10 h, less than 0.1% of collisions with impurities must result in a reaction.

References

- [1] R. Benz. Thorium-thorium dioxide phase equilibria. *Journal of Nuclear Materials*, 29:43–49, 1968.
- [2] M. Boivineau, H. Colin, J. M. Vermeulen, and Th. Thevenin. Thermophysical properties of solid and liquid thorium. *International Journal of Thermophysics*, 17:1001–1010, 1996.
- [3] Nicholas R. Hutzler, Hsin-I Lu, and John M. Doyle. The buffer gas beam: An intense, cold, and slow source for atoms and molecules. *Chemical Reviews*, 112(9):4803–4827, 2012.