

DEVELOPMENT OF A FLUIDIZED BED CVD SYSTEM FOR COATING UO₂ PARTICLES WITH TUNGSTEN. O. R. Mireles¹, J. W. Broadway¹, R. R. Hickman¹, ¹NASA Marshall Space Flight Center, ER24, MSFC AL 35812, omar.r.mireles@nasa.gov, ¹NASA Marshall Space Flight Center, EM32, MSFC AL 35812, jera-mie.w.broadway@nasa.gov.

Introduction: Nuclear Thermal Propulsion (NTP) systems are currently under consideration by NASA for development in support of deep space exploration concepts. This enabling technology allows for larger architecture envelope by taking advantage of a significant specific impulse (approximately 900 seconds) that will allow for a much larger range of destinations, shorter trip durations, and improved safety. As a result, the Nuclear Cryogenic Propulsion Stage (NCPS) project is under development to address specific concerns in or to mitigate risk.

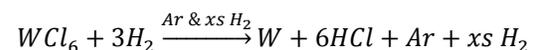
Although NTP is a mission enabling technology there are a number of concerns that require investigation. The current NTP effort is focused on risk reduction efforts in order to increase the maturity of NTP. One of the major objectives of the effort is to develop a tungsten-UO₂ CERMET fuel form. The reactor fuel must be capable of surviving at hydrogen exposure at temperatures approaching 3000 K. This aggressive environment requires a significant materials development program to ensure that CERMET fuel forms can meet mission service life requirements.

Previous efforts during the ANL CERMET and GE-710 programs found fuel loss to be excessive under thermal cycling conditions in a W-UO₂ cermet [1]. The CTE mismatch between W and UO₂ and the grain boundary structure of W particles (continuous path) were found to be responsible for the significant reduction of the UO₂ by hot hydrogen. In addition, UO₂ has a high vapor pressure and if exposed to hot hydrogen fuel material will be rapidly lost [2]. In efforts to minimize fuel loss during operation in a hot hydrogen environment, it was found that if particles are coated with a layer of high density tungsten, fuel loss is substantially limited [1].

Problem Statement and Objective: Previous programs utilized the H₂-WF₆ process to deposit tungsten coatings, which resulted in trace quantities of Fluorine (F) impurities. Since F will readily react with UO₂, the likely result is exacerbated fuel loss. Therefore, the application of the H₂-WCl₆ process is under development since Chlorine (Cl) does not react with UO₂ as readily as F.

Apparatus and Procedure: The basic design of the CVD system consists of a solid reagent delivery configuration coupled with a sublimation chamber. Ultra-high purity argon and hydrogen are supplied to the system two high precision mass flow controllers.

Gases are preheated before entering the reactor using a series of strip heaters. The sublimator consists of a small vessel partially filled with the WCl₆ reagent powder. The sublimator is heated by a heater band to approximately 200 °C in order to obtain reagent vapor, with careful emphasis to not accidentally melt or boil the reagent powder. Argon is then flowed through the sublimator and acts as a carrier gas for the WCl₆ vapor, which transports it from the sublimator to the reactor section. The reactor consists of a quartz tube concentrically aligned within an electrical resistance furnace. The nominal operating furnace temperature is in excess of 900 °C in order to provide the appropriate conditions to allow the reaction to occur. The furnace temperature is controlled through the use of a PID controller coupled to a type-K thermocouple located on the furnace inner diameter. When furnace and sublimator temperature have reached steady state operating conditions the spherical powder is then fed into the center-line of the reactor through a hopper mounted on the top of the reactor stack. The flow rate is adjusted to allow for the falling powders to achieve a fluidized state once in the reactor hot zone. Fluidization conditions are very sensitive to variation and require strict process control to allow for uniform coating production [3]. Hydrogen acts as the fluidization agent but also as the reducing agent, which reacts with WCl₆ vapor at temperature to form free W and HCl as shown in the basic chemical reaction:



The free W then deposits on the fluidized UO₂ particles, allowing for a uniform deposition to be grown. Coating thickness is controlled by varying the amount of time the reaction is allowed to take place. An expansion section at the aft end of the reactor section allows for the particles to lose momentum and fall back into the reactor, greatly aiding in powder retention. Exhaust gases flow out of the reactor expansion vessel through two small mesh screens and on to a 7 micron filter to prevent the loss of particles from the system. Once through the filter the exhaust gases travel through an acid trap to retain the HCl vapor, and then flow through another 0.3 micron filter to retain any trace of fine particles from the exhaust gases before they are directed to the hydrogen burn-off stack.

Once the appropriate amount of time has been met to deposit a coating of adequate thickness the sublimator inlet and outlet isolation valves are closed and the deposition process in the reactor terminates. The furnace is then shut-down and the particles continue to fluidize in an H_2 environment until the reactor temperature reaches ambient, at which point the H_2 flow to the reactor is terminated. The particles settle at the bottom of a conical reducer where they are then fed into a collection hopper. The collection hopper is disconnected from the CVD system, transported to an inert glove box, and the powders are further classified by size ranges. The CVD system is shown in Figures 1 and 2.

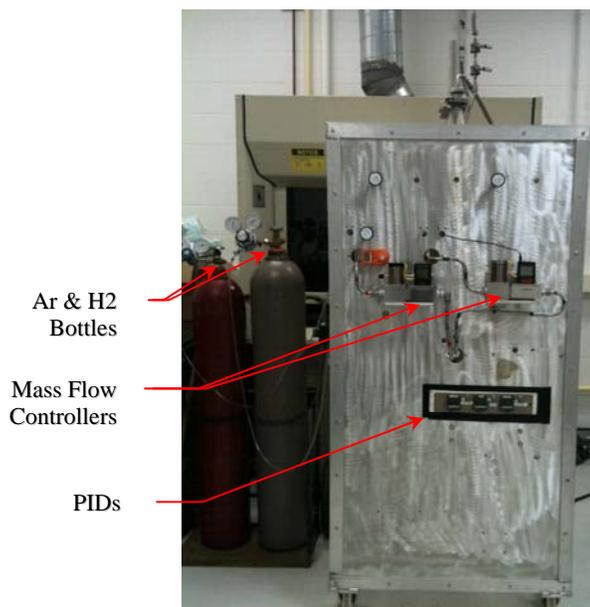


Figure 1: Fluidized bed CVD system

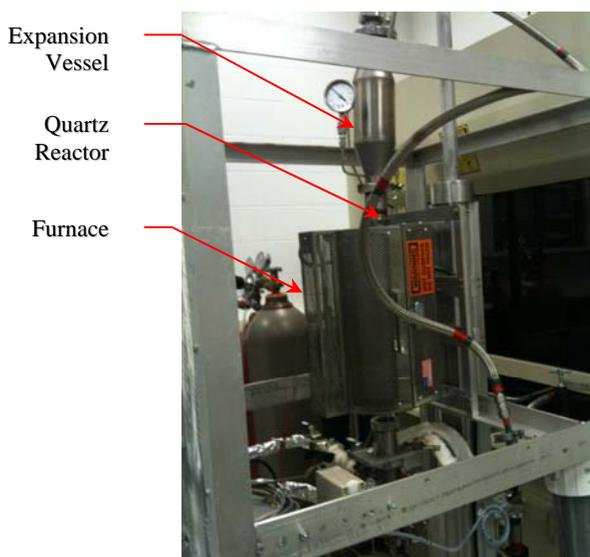


Figure 2: CVD reactor

Results: Ongoing efforts are underway to identify the flow conditions necessary to establish a fluidized bed. ZrO_2 spherical particles with the same size distribution as the UO_2 powders will have are used as a surrogate material in order to establish fluidization conditions without requiring additional controls. In addition, the process conditions for deposition of tungsten coatings onto ZrO_2 powder substrates are also underway. Once optimal fluidization and CVD process variables are identified and controlled then the process will be repeated using a CeO_2 surrogate powder and then onto depleted- UO_2 powder.

Conclusions: An iterative test process is essential to demonstrate the feasibility of coating UO_2 particles with tungsten with minimal impurities and with appropriate coating density, adhesion, and thickness. Successfully completing the test matrix will overcome a major obstacle to the successful fabrication and implementation of W- UO_2 cermet fuel forms.

Recommendations: The CVD system design is completed and efforts are ongoing to identify the optimal process conditions in order to obtain the desired particle deposition. Recommendations for future work include:

- Investigations into the effects of the CVD deposition columnar grain structure with the major axis parallel to the direction of growth and the affect on fuel loss.
- Develop more rigorous diffusion and kinetics relations to better understand behavior trends with regards to deposition quality.
- Identify design modifications to allow for high through put.

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