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Ceramic Oxide-Based Nuclear Fuel Materials: Critical Considerations and Advancements in Thermal Processing

Advanced thermal processing technology can help nuclear fuel producers meet increasing industry demands.

By Prasad Apte and Peter Witting

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Thermal processing of advanced materials like ceramic oxide pellets requires equipment that can reliably balance high-quality product output, safety, and economic production. If processing demands operation within a controlled atmosphere, the equipment must further ensure that the gases inside are not mixed with air and do not leak into the atmosphere.

In the case of processing uranium compounds, equipment requirements are even more stringent. Uranium is a radioactive material. Thermal processing is performed in hazardous atmospheres, such as hydrogen (flammable) 5/2/2014 Ceramic Oxide-Based Nuclear Fuel Materials: Critical Considerations and Advancements in Thermal Processing | 2014-05-01 | Ceramic Industry

and hydrogen fluoride (corrosive). In some cases, the quantity of uranium has to be controlled to avoid criticality, and the material has to be handled such that nothing can be permitted to fall out of the equipment on the ground as waste. Instead, all waste must be collected and reprocessed. Processing temperatures are high and can reach up to 1800°C. Operator safety is the highest of priorities due to the radioactivity and the toxicity of uranium and the hazardous gases used in the processing. In addition, uranium compounds have high specific gravity.

These considerations dictate that processing equipment must be low maintenance, robust in construction, and reliable in quality and operation. Processing equipment must also have excellent atmosphere control, high-quality process monitoring and control devices, few moving parts, and be operable safely with little operator interaction.

With these necessities in mind, manufacturers are discovering the application of advanced <u>thermal processing</u> technology and techniques to support various critical nuclear fuel material processes. The next evolution of this technology is likely to support the nuclear market's ever-changing needs.

Thermal Processes for Nuclear Fuels

The thermal processing steps for making nuclear fuel for use in reactors are as follows:

- Reduction of uranium trioxide (UO₃) to uranium dioxide (UO₂)
- Hydrofluorination of UO₂ to uranium tetrafluoride (UF₄)
- Oxidation of UO₂ to triuranium octoxide (U₃O₈)
- Sintering of UO₂ pellets for making reactor elements

Reduction of UO₃ to UO₂

This step is accomplished in a hydrogen atmosphere at temperatures ranging from 600-900°C. The raw material and the product are both powders, and the reduction process requires intimate mixing with hydrogen.

The reaction can be performed in batch-type processes or batch continuous processes (i.e., a pusher <u>furnace</u>); however, a rotary furnace is best suited for this process. The rotation of the tube raises the powder, which then falls through the flowing hydrogen gas, giving excellent mixing and rapid reactions. Uranium trioxide is yellow, and uranium dioxide is blackish brown.

Hydrofluorination of UO_2 to UF_4

This process involves the reaction of uranium dioxide with hydrogen fluoride (HF) to form uranium tetrafluoride, and is carried out between 600-900°C. The oxide is blackish brown, and the fluoride is green. Once again, the oxide and fluoride are both powders. Intimate contact with the gaseous hydrogen fluoride is essential.

Batch processes are feasible, but a rotary calciner is best suited for the process. In this application, the materials of construction are particularly important because gaseous HF is very corrosive to the materials of construction. Further, the sealing requirements on the furnace are extremely stringent because of the hazardous nature of gaseous HF.

Considerations of worker safety are paramount in the design and construction of these units. The furnaces are operated remotely from an atmosphere-controlled room. This process is part of the mechanism for making enriched UO₂. It is not needed for reactors that use natural-grade UO₂.

Oxidation of UO_2 to U_3O_8

This process is used for recovery and reutilization of scrap and waste in a fuel processing plant. U_3O_8 is the most stable oxide of uranium. It is used in making fuel pellets, especially from enriched UO_2 . In the manufacture of nuclear fuel pellets, broken pellets can be found before or after sintering.

Sintered fuel pellets must be perfectly cylindrical. This is achieved by centerless grinding of the sintered pellets. The grinding process generates powders referred to as "swarf." Broken pellets and swarf are converted to U_3O_8 in many fuel production plants. Here, the process is oxidation by air. Once again, rotaries are best suited for this process to ensure excellent mixing.

Thermal process systems have also been used to dilute highly enriched nuclear material into non-enriched material. U-235 is fissile (i.e., it can sustain a fission chain reaction). Some thermal processing systems have been used to convert highly enriched UO_2 to U_3O_8 . This enriched U_3O_8 is blended with natural-grade U_3O_8 material to dilute the concentration of U-235. This dilute material is then reprocessed into nuclear fuel.

Sintering of UO₂ Pellets

Uranium dioxide is pressed in dies to make small cylindrical shapes, called pellets. The pellets have to be sintered in order to densify them and give them strength. The sintering process requires a high temperature between $1600-1800^{\circ}$ C, and has to be performed in a reducing atmosphere. The reducing atmosphere is provided by hydrogen. Hydrogen accelerates the sintering process and prevents the UO_2 from converting to U_3O_8 .

Rotary Furnaces

Rotary systems are appropriate for several nuclear fuel applications, such as oxidation, reduction, and hydrofluorination, and offer advantages for the tight atmosphere seals required to contain the potentially flammable atmosphere, as well as dust containment measures. More advanced rotary systems include baffles and other features to reduce end tube radiation loss to improve energy efficiency. Rotary systems often provide great energy efficiency, as there is no carrier load that requires energy to heat and cool the boat or product tray.

The rotary system also offers a number of mixing techniques that can be applied for nuclear fuel materials processing. Riffle flights provide axial mixing for processing at a fixed average composition and are excellent for moderating exothermic reactions or continuous in-line mixing. Riffle flights enable similar benefits as standard continuous stirred tank reactors (CSTR), but in a continuous rotary tube format. Helical flights convey material without back-mixing and are used for nuclear materials that require narrow residence time distributions.

The absence of moving parts in the rotary's tube support system provides a simple, reliable, robust design that enhances scalability. In addition, the tumbling action of the product within the tube results in high degrees of temperature uniformity and gas-solid contact, producing a more homogenous product, reducing processing times and increasing production rates.

Pusher Furnaces

Pusher furnace systems are often a fit for nuclear fuel sintering processes, as they provide the high-temperature capability required, along with hydrogen atmosphere and dewpoint control. These sintering furnace systems are typically molybdenum element heated with high-alumina oxide-based refractory.

The most advanced systems on the market include enhancements that consider the delicate pressure control within the system to provide accurate direction of the atmosphere flow path in the kiln. This facilitates evacuation of volatiles and optimizes atmosphere uniformity. Another innovative design feature in the more cutting-edge furnace systems is stripping chambers for optimal isolation of the internal tunnel chamber environment from ambient, as well as efficient purging of ambient atmosphere entrained within the load entering the furnace without the use of mechanical doors and seals.

The Future of Nuclear Fuel Materials Processing

The demand for electric power continues to increase, placing a strain on supply. This demand also results in continued growth in the demand for nuclear power. Nuclear power is one of the cleanest energy sources, and worldwide expansion is anticipated.

For the market to capture this demand, it must consider how to improve its existing plant investments and deliver more efficiencies in its processes. Implementing well-developed business procedures, such as proactive, preventative maintenance programs and onsite or offsite/vendor-managed spare parts programs, can assist nuclear fuel producers with minimizing production downtime and helping meet the increasing demands placed on the industry.

Many existing plants are nearing time for refurbishment. The changing requirements of these plants often require upgrades and modifications of the production facilities. Many weapons-grade nuclear materials are planned for reprocessing, and these sensitive materials require special attention. Well-executed refurbishments to extend the life and safety of existing production equipment will be integral to stable production.

Turnkey solutions, including integrated gas treatment and handling and fully integrated control systems, are also

helping to support the needed improvements in process efficiencies. As a specific design example, a cascading rotary system where each rotary delivers a different process step in one integrated system is an effective tool for producing nuclear fuel materials more efficiently.

For additional information, call (716) 684-7400 or visit www.harperintl.com.

Prasad Apte, Ph.D., is Director of Technology at Harper International. Peter Witting, Ph.D., is Senior Process Technology Engineer for Harper International.