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SPOUTED BED DESIGN CONSIDERATIONS FOR COATED NUCLEAR FUEL PARTICLES

Fluidization XV

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Abstract:

High Temperature Gas Cooled Reactors (HTGRs) are fueled with tristructural isotropic (TRISO) coated nuclear fuel particles embedded in a carbon-graphite fuel body. The TRISO coatings consist of four layers of pyrolytic carbon and silicon carbide that are applied on uranium ceramic fuel kernels by chemical vapor deposition in a spouted bed reactor. The TRISO coatings are the primary containment structure for radionuclides in the HTGR reactor and must have very high integrity and uniformity.

The four TRISO coatings are deposited in a concatenated series of batch operations at temperatures ranging from 1230°C to 1550°C. As the coatings are deposited, the mean particle diameter doubles, bed volume increases 8-fold, and particle envelope density drops from 11 g/cm^3 to 3 g/cm^3.

All depositions are accomplished without the aid of sight ports or internal instrumentation that could compromise the integrity of the layers.

The converging section and gas injection nozzle were designed to prevent bed stagnation zones and premature decomposition of reactive coating gases. The equipment designs and operating methods have yielded very good equipment reliability and product reproducibility.

Background Information

Nuclear reactor designs for High Temperature Gas-Cooled Reactors (HTGRs) employ small kernels of uranium ceramic (425µm - 500µm diameter) that are hermetically sealed in four shells of pyrolytic carbon and silicon carbide and dispersed in a matrix of graphite and vitreous carbon. The layers in the tristructural isotropic (TRISO) particle constitute the primary containment for highly-radioactive fission products and must be deposited in a manner that ensures chemical purity, mechanical integrity, and narrowly distributed layer thicknesses, densities, grain sizes, and orientation of graphene planes (dense pyrocarbon layers only).

Each layer has important functions; Keeley (1). The inner most layer (1 in Figure 1) is a 100 µm buffer layer of low-density pyrolytic carbon (pyrocarbon) that absorbs the recoil energies from atomic fragments after a fission event and provides a volume for accumulating gaseous fission products, thus avoiding excessive internal pressures that could induce fractures in the outer three layers.

The second layer (2) is the dense inner pyrocarbon layer (IPyC, ~40µm), which provides a gas-tight seal to protect the fuel kernel during coating and the silicon carbide (SiC) layer from carbon monoxide and metallic fission products during irradiation.

The penultimate layer (3) is composed of SiC (~35µm) and serves as the structural member and a barrier to metallic fission product diffusion.

The outer layer (4) is the dense outer pyrocarbon layer (OPyC, ~40 µm) that protects the silicon carbide from mechanical damage during handling and is the final barrier to fission product diffusion.

Both dense pyrocarbon layers shrink during irradiation and take some of the tensile load from the SiC layer. The outer three layers constitute the primary containment structure against fission product migration into the graphitic fuel bodies and the helium cooling system.

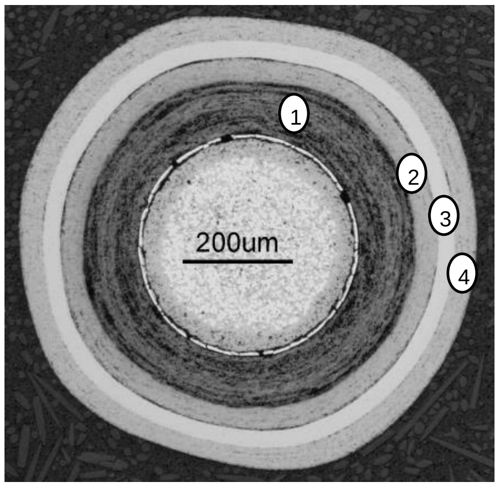

Figure 1. Sectioned TRISO fuel particle.

Each nuclear reactor may contain billions of TRISO particles embedded in the fuel form, so high integrity TRISO coating layers are essential to keep fission product releases at acceptable levels during normal operations and during postulated accident scenarios involving the loss of helium coolant flow. By design, the peak centerline core temperatures, during postulated accident scenarios, safely remain 200 – 300°C below fuel fabrication temperatures, so minimal damage occurs to the fuel form.

The need for energy from safe, high-temperature, and low-carbon emitting sources to replace fossil fuels has stimulated a renewed, worldwide interest in HTGR technology for process heat, high-temperature hydrogen production processes, and co-generation of electricity.

Design Challenges

Constraints

Multiple and often competing performance objectives and constraints must be considered during the design of chemical vapor deposition (CVD) reactors for the production of TRISO coated nuclear fuels; some of which are unique to the nuclear industry. For instance, to prevent an unplanned, uncontrolled nuclear criticality in the fuel fabrication equipment, fissile material enrichment and mass, equipment geometry, and neutron moderation are constrained within safe limits to provide multiple, independent defenses “in depth.” Equipment designed for depositing TRISO coating layers of low-enriched uranium (LEU) kernels have had retort diameters up to 24 cm; Noren (2), Kasten (3). The system design discussed in this paper is housed in a facility licensed for highly-enriched uranium (HEU) and is constrained to an internal diameter of 15.2 cm to ensure criticality safety should HEU be used inadvertently.

The relatively small retort diameter, required to ensure the system is critically safe by geometry, introduces more design caveats. The first caveat relates to matching of the Glicksman dimensionless scaling relationships; Glicksman (4). Having similar superficial gas velocities and gas residence times would be preferred, but not possible. Gas flow rates were established in lab-scale equipment to achieve the desired linear deposition rate for each layer and to ensure fluidization as particle dimensions change. The linear rate of deposition is controlled because it influences the layer isotropy, grain size, and density. Isotropy is essential to promote uniform pyrocarbon shrinkage to preclude excessive stress accumulation in the layers; Jellison (5).

At constant linear deposition rates, the volumetric gas flow rates are proportional to the bed mass, which is proportional to the cube of the retort diameter. The superficial gas velocity is a function of volumetric flow and the retort cross sectional area, so it increases proportionally to the retort diameter. Gas residence time decreases with the increase in gas velocity, which adversely impacts coating efficiency. Coating efficiency can be improved with a well-designed gas distributor nozzle that encourages percolation of coating gases through the descending annular bed.

Secondly, the narrow retort also makes gas distribution important to preclude accretions forming on the walls. Lifting the bed in the bottom of the retort can expose the retort wall to condensing species and causing accretions. Equally important is to avoid direct impingement of reactive coating gases on the retort wall that would form accretions and impact bed circulation.

A third consideration is the fluidization mode. Bed slugging is avoided and bed circulation is enhanced by maintaining a relatively shallow bed and intentionally operating the coater in a spouting bed mode. The uranium ceramic fuel kernels have narrowly distributed particle diameter of either 425µm or 500µm and particle envelope densities near 11 g/cm^3. The final products, after the TRISO coating layers are applied, are 860 – 950 µm in diameter with particle densities near 3 g/cm^3. The particles roughly double in both mass and diameter during the coating process and the bed expands to approximately eight times the original volume. These changes happen too quickly, during pyrocarbon deposition, to allow the bed to operate in steady state conditions. An example of the dimensional changes is given in Table 1.

Table 1. Nominal TRISO particle layer dimensions with a 425 μm kernel.

Material	Layer Thickness (μm)	Layer Density (g/cm^3)	Particle Diameter (μm)	Particle Density (g/cm^3)	Relative Mass	Relative Volume	Δ Time (min)
Kernel	---	---	425	11.1	1.00	1.00	---
Buffer	100	1.00	625	4.18	1.20	3.18	5
IPyC	40	1.90	705	3.49	1.43	4.56	12
Silicon carbide	35	3.20	775	3.42	1.87	6.06	100
OPyC	40	1.90	855	3.03	2.22	8.14	13

The TRISO coating furnace retorts are fabricated from purified, nuclear grade, graphite to prevent chemical contamination of the coating layers that would compromise layer integrity and fission product retention during irradiation. Transition metal contaminants can form molten carbide phases that can migrate along thermal gradients to create pathways for carbon monoxide and fission product escape through the pyrocarbon that can corrode the SiC layer; Minato (6), Tiegs (7).

Instrument penetrations through the graphite retort wall are precluded because they are problematic to seal, may trap partially coated particles, and refractory instrument sheaths are susceptible to breakage. The radial position of thermocouples hung from the top of the coater cannot be controlled or maintained. The radial temperature profile within the coater, due to the influx of cool carrier gases and the exothermicity of coating gases, makes futile any attempt to use an internal thermocouple for process monitoring or control. Another concern is that nascent coating layers may be cracked or broken when a spouted particle impinges on the an instrument probe.

An opaque, black smoke is generated during the deposition of the pyrocarbon layers that prevents the use of optical pyrometers for direct bed temperature monitoring or any visual observation of bed dynamics. In consideration of the limitations on instrumentation, the coating depositions are accomplished without internal instrumentation.

The TRISO coater retorts are externally heated and the temperature is controlled by external thermocouples set off from the retort wall by a few millimeters. The ratio of retort wall surface area to the volumetric gas flow rate decreases with increasing retort diameter and bed mass. The net result is that process temperature setpoints established in a laboratory-scale coating furnace are inaccurate for larger diameter furnaces and additional process development is often needed to achieve the desired coating layer material properties.

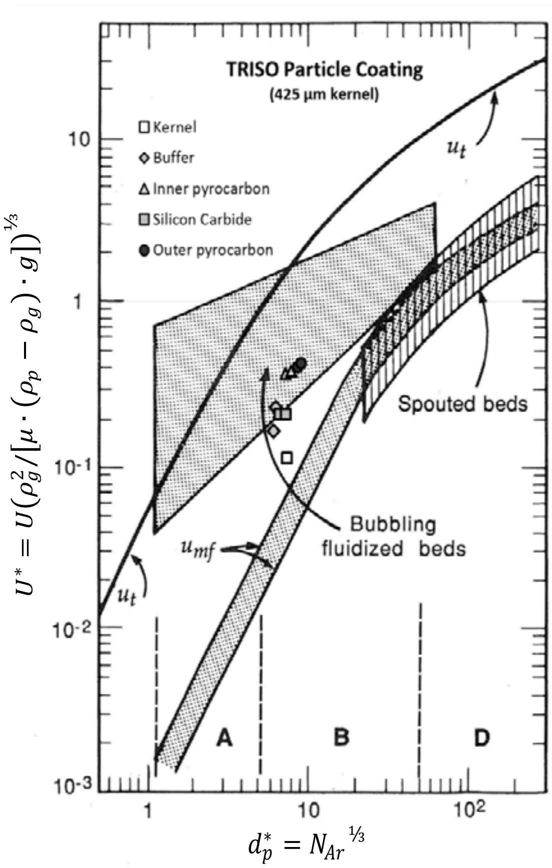

Figure 1. Mapped TRISO layer start and end points for each layer.

The particle and fluidizing gas properties are such that the entire coating process falls outside of the region mapped for spouted beds (Figure 2); Grace (8), Kunii (9). High process temperatures (approximately 1230°C to 1550°C) and small particle diameters decrease the dimensionless particle diameter and dimensionless gas velocity during coating.

Stable (coherent) spouting is not achieved, but the bed fluidizes in a pulsatory spouted bed mode; Epstein (10), Pianarosa (11). Pulsatory spouting was verified by sensing pressure fluctuations transmitted upstream through the gas inlet and by modelling conditions with CPFD Barracuda VR® software.

Spouting is achieved with a mixture of argon and hydrogen when silicon carbide is deposited and argon and organic precursors (propene and/or ethyne) for deposition of the pyrocarbons.

As one might anticipate, published correlations for minimum spouting velocities, which were developed under less extreme conditions, do not yield reliable estimates of the minimum spouting velocity for the TRISO coating conditions.

Gas Injector

The gas injector nozzle was designed to resist excessive accretion formation at the orifice(s) by precluding premature decomposition of precursor gases and shielding the nozzle surface with inert gases. Furthermore, the nozzle configuration diffuses the gas jet to prevent bed stagnation zones while precluding direct impingement of coating gases on the retort wall. Laboratory testing has demonstrated that product uniformity was improved using a gas distributor nozzle in preference to the single gas inlet commonly used in spouted beds; Noren (2), Kasten (3), Lackey (12). Moreover, a single gas orifice in a 5-cm conical-cylindrical coater, formed volcanoshaped accretions around the inlet that trapped fuel particles between the accretion vent and the retort wall.

Design objectives were met using a domed gas distributor nozzle with multiple gas inlet orifices and a serrated flange (Figure 3).

The orifice sizes were selected to ensure sufficient gas velocity to prevent particle weeping and sufficient back pressure to ensure balanced gas distribution and prevent instantaneous gas flow reversals in individual orifices.

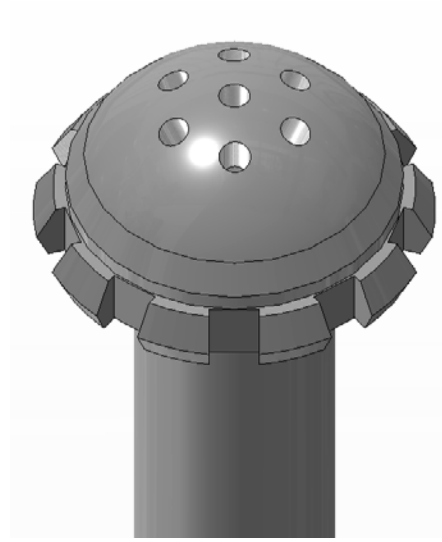

Figure 2. Graphite gas distributor nozzle.

The serrated flange engages a conical port in the bottom of the retort converging section. The serrations reduce heat conduction from the retort by limiting the contact area and ensure sufficient off set to form an annular orifice between the base of the dome and the converging section through which an inert gas is passed. The inert gas fluidizes particles in the bottom of the bed, convects heat away from the nozzle, displaces reactive gases away from the nozzle face, and dilutes the precursor gases to the desired concentration within the bed.

Although orifice accretions were not entirely eliminated, all orifices remain open for the duration of the coating depositions and none of the accretions trap fuel particles. Each distributor nozzle is a single use component that is replaced after each fuel coating run.

Converging Section

The converging section of a spouted bed encourages bed circulation by eliminating regions that would be stagnant in a cylindrical configuration. Common spouted bed designs have conical converging sections with included cone angles ranging from 45° to 60°. While many factors may be considered in the choice of the cone angle, the driving factors for nuclear fuel coating are maintaining particle sphericity and achieving a narrow distribution in coating properties.

When a narrow cone angle (~45°) is employed, the injected gases may lift the entire bed away from the vessel bottom; causing localized slugging and possibly exposing the walls to the reactive coating gases. A narrow converging section, however, does promote more bulk-flow behavior in an annular bed of spherical particles and less drag on bed movement along the walls. When the cone angle is wide (~60°), the spout is more easily stabilized and bed depth is more shallow, but bed circulation along the walls is impaired and some localized bed stagnation may occur, especially with spherical particles, which readily exhibit funnel-flow behavior in a spouted bed. Any bed stagnation or impairment of circulation leads is undesirable.

Designs of some bubbling and spouted beds incorporate narrow included cone angles at the top of the converging section and transition step-wise to broader angles at the bottom; Epstein (10), Flamm (13), Uhlemann (14). Stepped changes in the cone angles can adversely impact bed circulation where the conical frusta meet. A gradual transition in the slope is deemed preferable. Whereas TRISO coating furnace retorts have diameters generally less than 24cm and are lathed from graphite billet, the converging section profile is not constrained by economics to conical geometries, as might a larger metal vessel.

The objectives of encouraging spout stability with a broad cone-angle base and facilitating good bed circulation without exposing the converging section walls to reactive coating gases or risking bed stagnation zones were met by designing a curvilinear converging section with a continuously variable slope, (Figure 4); Marshall (15). The local slope at the intersection with the cylindrical section equates to an included angle of 22° and approximately 116° at the base.

Figure 3. Rendered cross-section of a curvilinear converging section.

Inspections of the converging section and gas injection nozzle after each coating run show no evidence of bed stagnation zones or significant accretions. Metallographic mounts of the TRISO coated particles reveal that the variability in coating thickness is low and that particle sphericity is acceptable.

Carbonaceous “soot” accumulates above the fountain and on the wall of the cylindrical section during the deposition of the carbon buffer layer. Ejecta from the particle bed will, occasionally, impact the soot and take a loosely adherent soot layer back into the bed where it becomes encapsulated within the layer being deposited. These soot inclusions (Figure 5) are undesirable as they interfere with physical properties and function of the layers. Although frequently observed in mounted TRISO particles coated in a laboratory-scale (5-cm) conical-cylindrical spouted bed with a single gas inlet port at the cone apex, soot inclusions are infrequently observed in product from the 15-cm coater with the curvilinear converging section and multiport gas distributor nozzle.

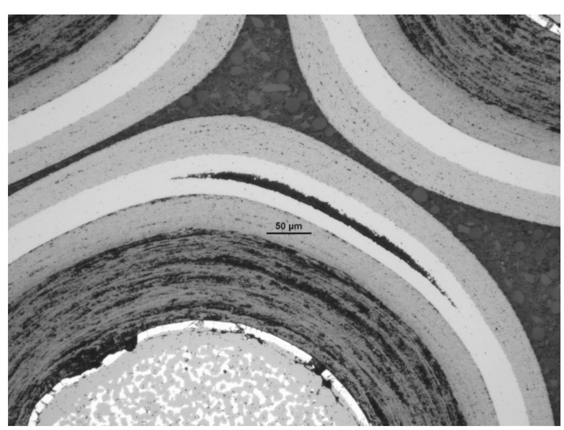

Figure 4. Soot inclusion within the SiC layer.

Dual Gas Paths

The coater is designed with an annular orifice between the base of the converging section and the outer margin of the gas distributor nozzle. A portion of the inert diluent gas is routed through the annular orifice to fluidize fuel particles below the elevation of the orifice ports on the distributor nozzle, to cool the nozzle, and to limit accretion formation by displacing reactive coating gases near the nozzle face.

Laboratory tests were conducted in a 2-D physical model of the 15-cm coater using similar geometries. The tests showed that diverting a portion of the inert diluent gas to the annular orifice widens the spout diameter and decreases the height of the spouted bed fountain. Diversion of some inert gas to the annular orifice in concert with the converging section and multiport nozzle configurations and the larger retort diameter are thought to be the contributing factors in reducing the incidence of soot inclusions in the product.

Conclusions

Design and scale up of a TRISO coating retort requires careful consideration of several competing design parameters and a measure of creativity in order to achieve high quality coatings on nuclear fuel kernels. Fully developed bed circulation must be assured without excessive fluidization that could result in soot inclusions or abrade the layers.

The design objectives were met by incorporating a multi-orifice distributor nozzle, diverting a portion of the inert gases through an annular orifice at the base of the coater, and using a curvilinear converging section. High quality TRISO coated fuel particles with narrowly distributed layer properties, infrequent indication of soot inclusions, and no observed evidence that bed stagnation has occurred suggest a successful design has been achieved.

This coating furnace design is in use by the Advanced Gas Reactor Fuel Development and Qualification program funded by the United States Department of Energy.

Nomenclature

d_p	particle diameter
d_p^*	dimensionless particle diameter
g	gravitational constant
N_{Ar}	Archimedes number
U	superficial gas velocity
U^*	dimensionless superficial gas velocity
μ	gas viscosity
ρ_g	gas density
ρ_p	particle envelope density

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