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A US perspective on fast reactor fuel fabrication technology and experience. Part II: Ceramic fuels

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ABSTRACT

This paper is Part II of a review focusing on the United States experience with oxide, carbide, and nitride fast reactor fuel fabrication. Over 60 years of research in fuel fabrication by government, national laboratories, industry, and academia has culminated in a foundation of research and resulted in significant improvements to the technologies employed to fabricate these fuel types. This part of the review documents the current state of fuel fabrication technologies in the United States for each of these fuel types, some of the challenges faced by previous researchers, and how these were overcome. Knowledge gained from reviewing previous investigations will aid both researchers and policy makers in forming future decisions relating to nuclear fuel fabrication technologies.

1. Introduction

There has recently been renewed interest in fast reactor fuels in the US, leading to the development of US Department of Energy Programs, such as the Generation IV Initiative [1,2], the Advanced Fuel Cycle Initiative [3], and the Global Nuclear Energy Partnership [4]. Fast reactors are poised to effectively address a number of factors facing the world today. These factors include the consequences of the greenhouse effect and subsequent possibility of a carbon tax, production of hydrogen and hydrogen-rich fuel cells for transportation, increased demand for potable and irrigation water, proliferation concerns associated with separated plutonium, and finding an alternative to a permanent spent-fuel repository for minor actinides and long-lived fission products. Fuel fabrication of metal fuel alloys and associated fast reactor assembly design and fabrication were the basis for Part I of this review. In Part II, the ceramic-based fuel fabrication technologies in the US are reviewed, particularly those associated with oxides, carbides, and nitrides.

Oxide fuels are one of the most popular choices for fast reactor fuel systems, metal fuels being the other. The basis for this popularity can be largely attributed to the great successes in fabrication and operation of oxide fuels for light water reactors (LWRs). However, for next generation fuel systems that contain minor actinides, certain modifications will be required in order to minimize waste generation, maximize safety, and maintain operation economics. In order for future researchers to build upon the great successes of the LWR industry and fast reactor campaigns to date, a sufficient review of the technologies utilized seems relevant.

Furthermore, while carbide and nitride based fuel systems have not enjoyed the same successes as metal and oxide fuel systems, they are potential and viable candidates for much longer-term fuel designs. The carbides and nitrides have mainly been investigated in other parts of the world, but some of the technology has been adopted for use in the US for specific fast reactor applications, such as the space nuclear programs. The review represents the US's perspective on ceramic fuel fabrication technologies. Although countries such as the United Kingdom [5], France [6], the Russian Federation, India, and Japan have continued to refine and expand on similar fuel fabrication technologies. From this standpoint, new questions arising from researchers and policy makers, both present and future, relating to the growth of nuclear technology through fuel fabrication and assembly are addressed.

A brief review of the terminology used in this paper will be helpful, especially since efforts has been made to keep the terminology used here consistent with that of previous researchers, although the terms may be slightly different from those used today. The term "pellet" is defined as an un-encapsulated, as-fabricated piece of pellet-type *ceramic* fuel. The term 'fuel rod' is defined as the fuel capsule, also sometimes referred to as the fuel pin or fuel element. The term 'fuel column' is defined as a stack of fuel pellets; the fuel column height defines the active core height for a fast reactor driver fuel.

2. Mixed oxide fuel fabrication technology

2.1. History and experience

Mixed oxide (MOX) fuel, $UO_2–PuO_2$, is often selected because of its excellent burnup potential, high melting points, and relative

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ease of commercial fabrication and because LWR fuel fabricators already have extensive experience with UO_2 fuel fabrication, irradiation stability, and the proven safety response using a negative Doppler coefficient that mitigates overpower transients [7]. These advantages must often be weighed against the disadvantages of oxide fuel, such as lower thermal conductivity that leads to fuel restructuring and enhanced swelling [8], reduced compatibility with sodium [9–11], low fissile atom density, and the presence of two moderating atoms for each metal atom.

Development of the MOX fuel program in the US has culminated in over 20 years of performance information, leading to an extensive fuel database with more than 63,500 driver and test fuel rods irradiated, mainly through the use of commercially fabricated fuel irradiated in the Fast Flux Test Facility (FFTF) and, to a lesser extent, in the Experimental Breeder Reactor II (EBR-II) [12,13]. MOX fuel has had similar success in other reactors throughout the world, including Rapsodie and PHENIX (Liquid Metal Fast Breeder Reactor Demonstration Plant) in France, BOR-60 in the former Soviet Union, and PFR (Prototype Fast Reactor) in the United Kingdom. In Germany, MOX fuel was fabricated and planned for use in the SNR-300 and the KNK II had been MOX fueled [7].

2.1.1. EBR-II

A number of test designs for oxide fuel rods were irradiated in the EBR-II. These designs provided a limited amount of fuels performance information because EBR-II allowed irradiation of only short, small bundles. Irradiations conducted in EBR-II, however, did provide a sufficient foundation of information for designing the driver fuel elements used in FFTF (Series I and II), which were used throughout its lifetime.

2.1.2. FFTF

One of the most significant advantages of FFTF irradiation designs was that FFTF used components that were near prototypic in size to many commercial scale Liquid Metal Reactors. Six different designs were tested for oxide driver fuel use in FFTF, including austenitic stainless steel-clad $(U, Pu)O_2$ fuels, as well as $(U, Pu)O_2$ and UO_2 fuels clad with ferritic–martensitic (HT9) steel in wire-wrapped fuel rod bundles housed in a hexagonal duct [14]. Table 1 summarizes these designs. Over 48,000 Series I and II full-length driver fuel rods have been irradiated in FFTF, all using commercially fabricated fuel pellets.

Over 16,000 test rods were irradiated, comprising the Series III.a, Prototype Oxide and Core Demonstration Experiments. The Series III.a design that used the lower-swelling D9 stainless steel improved operation economics because this cladding allowed longer fuel assembly lifetimes. The Prototype Oxide Experiment and Core Demonstration Experiment used 169 fuel rods per assembly, rather than the typical 217, to reach the full power rating of FFTF, on par with power levels attained using metal fuel. However, use of 169 rods per assembly significantly lowered the power level (i.e., 300 MWt rather than 400 MWt) because of limits on fuel rod power to avoid fuel melting. It was later observed that initial fuel melting was not a deleterious effect and design constraints could be relaxed somewhat with this particular design; however, use of 217 smaller-diameter fuel rods per assembly was the most likely solution to attain full power rating. The annular pellet design produced less transient-induced cladding strain compared to the solid pellet design because the central hole allowed an effective pathway to the plenum alleviating pressure buildup in the fuel during overpower transients. The FFTF reference fuel design consisted of a MOX fuel pellet He bonded to a 20% cold-worked SS 316 cladding and wire wrap with a 1.0 fuel-to-plenum volume ratio and smeared fuel density of 85% [12,15].

All fuel pellets for the Series I and II driver fuel rods were fabricated by two different commercial vendors using a different manufacturing technique [16]. Kerr–McGee fabricated the fuel pellets employing a high-pressure, pre-slugging method that resulted in fuel structures with regions of high- and low-density separated by interconnected porosity. Slugging is a commonly used technique for dry powder granulation to improve the freely flowing nature of the powders through generation of powders with desired particle characteristics (e.g., density, size distribution, etc.). Babcock and Wilcox/Nuclear Materials Division fabricated the fuel pellets employing a low-pressure, pre-slugging method and using a pore former that resulted in a fairly uniform fuel matrix with varying sizes of closed spherical pores. Babcock and Wilcox-fabricated fuel rods tended to fuse together in the fuel column with increased cracking as exposure was increased, particularly in the upper portion of the fuel columns where operating temperatures were higher. Kerr–McGee-fabricated fuel rods showed very little pellet-to-pellet interaction along the length of the fuel column, but some gaps were observed in the upper portion of the column. The insulator pellets where typically cracked, hypothesized to result from Cs fission product migration to the upper portion of the fuel column with increased exposure and subsequently higher cladding temperatures. Nevertheless, the differences in as-built fuel microstructure resulting from the two fabrication methods had minimal influence on overall fuel restructuring during irradiation. Both fuel types showed the formation of continuous central voids that extended axially once peak beginning-of-life fuel rod powers were reached. The central voids increased in size radially as both fuel rod power and exposure increased, which was primarily due to the inward movement of void volume from the fuel-to-cladding gap generated by irradiation-induced swelling of the 20% cold-worked SS316 cladding.

Design parameters of MOX fuel rods were adjusted to increase lifetime. One such adjustment was reducing fuel-smeared density to reduce the fuel-cladding mechanical interaction; the initial fuel porosity was also reduced to compensate for lower smeared density and to prevent slumping [17]. Fuel porosity has important impacts on fuel behavior in that pores influence fission gas transport and in-reactor densification [7]. Another design change considered to decrease fuel-cladding mechanical interaction was to coat the

Table 1
Selected design parameters of FFTF oxide driver and test fuel elements [14].

Series	I	II	III.a	POE[a]	III.b	IV	CDE[b]
Fuel type	$(U, Pu)O_2$	$(U, Pu)O_2$	$(U, Pu)O_2$	$(U, Pu)O_2$	U–Zr	UO_2	$(U, Pu)O_2$
Pellet form	Solid	Solid	Solid	Solid	Solid slug	Annular	Annular
Cladding diameter (mm)	5.84	5.84	5.84	6.99	6.86	5.84	6.86
Fuel smeared density (%)	85.5	85.5	85.5	88	75	80	80
Maximum burn-up achieved (MWd/kg)	100	100	108	122	95	17	194
Cladding thickness (mm)	0.38	0.38	0.38	0.37	0.56	0.38	0.56
Number of fuel rods per assembly	217	217	217	169	169	217	169
Cladding material	SS316	SS316	D9	D9	HT9	D9	HT9
Approximate number of fuel rods irradiated	>48 000		6076	5790	1026	1357	2996

[a] POE = prototype oxide experiment.
[b] CDE = core demonstration experiment.

inner cladding wall with a soft metal – a process common to the current boiling water reactor (BWR) fuel fabrication technology [7]. In an attempt to produce more acceptable O_2 levels and reduce fuel-cladding chemical interaction (FCCI), coating the cladding with Ti- or Cr-reactive metal was attempted and successfully accomplished [7]. A similar reduction in FCCI was observed by reducing the O_2-to-metal ratio of the fuel.

2.2. Mixed oxide fuel fabrication process

2.2.1. Process overview

Fabrication of MOX fuel for use in fast reactors is similar to the existing commercial process used to fabricate oxide fuels for use in light-water reactors. The process involves mixing feedstock powders in the desired ratios, followed by milling and sorting the blended powders. Slugging, which involved feeding the dry particles into a compression machine, compacting at high pressures into large tablets or 'slugs', and milling the slugs into an appropriate powder size and distribution using a separate sizing machine, was performed to improve the freely-flowing nature of the powders. The powders of desired size were formed into compacts and sintered under ideal conditions to produce fuel pellets. The sintered pellets are post-processed to ensure correct dimensions, and characteristics are held to tight quality assurance standards. Oxide fuel pellets are then loaded into fuel rods and closure welded, as is typical with all fast reactor fuels. A schematic showing the salient steps associated with processing and fabricating oxide fuels is provided in Fig. 1.

2.2.2. Feedstock preparation

Plutonium dioxide preparation methods are very similar in nature to methods used for UO_2 preparation. Feedstock for powder preparation can be in the form of solutions, such as Pu nitrates derived from the dissolution of irradiated fuel material in nitric acid, or metallic form, such as Pu pits recovered from weapons systems. The most well known solution conversion method is the PUREX (Pu–U extraction) process used to reprocess spent fuel [18], although other precipitation methods were employed as well. Conversion of Pu metal to PuO_2 typically includes direct oxidation or conversion to an intermediate species followed by conversion to dioxide.

Precipitation methods offer improved purification and more adequate handling properties of the powder product than other techniques [19]. Plutonium nitrate is conditioned with H_2O_2 before

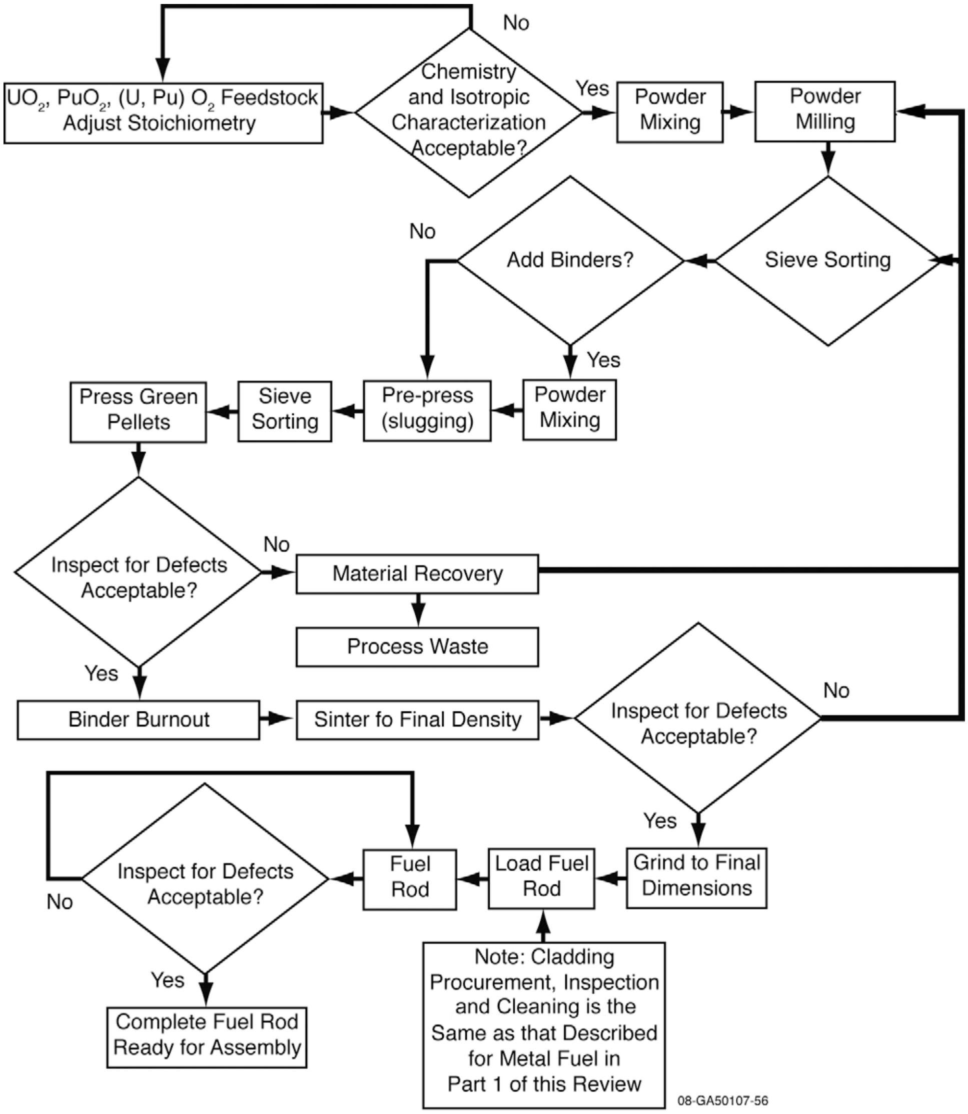

Fig. 1. Process flow diagram for fast reactor MOX fuel fabrication.

precipitating a Pu oxalate with $H_2C_2O_4$, after which it decomposes to an oxide form. The precipitation process is represented by Eqs. (1) and (2).

$$Pu(NO_3)_4 \cdot 6H_2O + 2H_2C_2O_4 \rightarrow Pu(C_2O_4)_2 \cdot 6H_2O + 4HNO_{3(l)} \quad (1)$$

$$Pu(C_2O_4)_2 \cdot 6H_2O + Heat \rightarrow PuO_2 + 2CO_{(g)} + 2CO_{2(g)} + 6H_2O_{(g)} \quad (2)$$

Particle characteristics are controlled by the Pu, nitric and oxalic acid concentrations, and decomposition temperature. Typical decomposition temperatures are in the range of 600 °C, although small variations can result in significant particle specific surface area characteristics. In addition, complete conversion of the oxalate to oxide is necessary because residual C impurities can complicate the ensuing pellet fabrication process. Specifically, C can give rise to solarization during sintering (i.e., fuel swelling rather than densification).

Thermal denitration is an alternative, straightforward method of converting a nitrate into an oxide, which is shown in Eq. (3) for $Pu(NO_3)_4$.

$$Pu(NO_3)_4 \cdot 6H_2O + Heat \rightarrow PuO_2 + 4NO_{2(g)} + O_{2(g)} + 6H_2O_{(g)} \quad (3)$$

However, this method typically produces powder with poor particle characteristics (e.g., coarse, non-uniform particles that are highly agglomerated), even though the process has the fewest steps and produces a low volume of liquid effluent. Ammonium nitrate can be added to the $Pu(NO_3)_4$ in order to form a mixed salt and control the decomposition path to reduce particle coarsening.

Co-precipitation methods can be employed to produce solid–solution oxide feedstock from mixed aqueous streams rather than dry blending oxides prior to sintering that would then rely on solid-state diffusion. Co-precipitation is carried out with ammonium diuranate, or ADU, and Pu hydroxide formed from mixed nitrates with ammonia gas in a N_2 carrier gas. This feedstock preparation method is complicated by the fact that precipitation of Pu and U occur at different pH values [20,21]. This has been overcome by feeding an ammonium co-precipitation slurry into a fluidized bed reactor where thermal decomposition occurs, referred to as the COPRECAL, or co-precipitation and calcination method [22].

Conversion of Pu metal to oxide powder might be necessary when a pyrometallurgical reprocessing scheme is employed rather than an aqueous reprocessing scheme, or when weapons grade Pu is being used as a feedstock. Plutonium metal oxidizes in an uncontrollable manner if exposed to an oxidizing environment (pyrophoric in powder form). Roasting or calcining the metal is the most common method of producing oxide powder feedstock from a metal [23]. Weapons grade material often contains Ga as a stabilizing metal and, therefore, may have to be removed prior to irradiation because Ga can react with many metals [24,25].

Regardless of how powder feedstock is produced, the proper particulate geometry(ies) is required to produce a properly sintered pellet. During granulation, it is challenging to obtain a uniform distribution of Pu in the product [26], as well as adequate distribution of binder and/or pore former agent; in general, the surface area of the oxides should be similar [27]. As a result, milling of the PuO_2 feedstock is often required because agglomeration of the particles is relatively common. Appropriate chemical compositions of fine UO_2 and PuO_2 powders are blended using a typical blending apparatus and milling media that further enhance particle size characteristics for optimized sintering.

Oxide stoichiometry (i.e., adjustment of the fuel oxide-to-metal ratio) is extremely important in minimizing FCCI phenomena during fuel operation. Additional sintering agents, such as binders, lubricants and pore formers (e.g., oxalic acid diamide), are added and blended with the chemical constituents during this time. The primary blending step produces a well-mixed powder with fine-particle size characteristics for typical fast reactor oxide compositions (i.e., 20–30 wt% PuO_2). Additions of up to 40-wt% PuO_2 are achievable but result in a significant decrease in sintered density [28]. The advantages of mechanical mixing of the UO_2 and PuO_2 powders include an approximate 20% reduction in fabrication costs and reduced criticality problems as less handling of the PuO_2 is required [29]. Conversely, coprecipitation methods offer the advantage of slightly lower sintering temperatures, making it possible to achieve an acceptable level of density due to the enhanced ability to reach an almost complete solid solution formation.

Nuclear fuel fabrication methods have involved sphere or kernel forming techniques for a long time. These early methods were based on more traditional powder metallurgical techniques that produced non-spherical particles with non-uniform product characteristics and undesirable by-products, such as fines and dust. More recently, new methods to produce micro- and macro-sphere particles have been investigated and applied to fuel and target manufacturing. These methods belong to the broad-ranging solution–gelation (sol–gel) technique of spherical particle formation. Sol–gel applications for actinide sphere formation (through internal or external gelation processes) were originally developed in the 1960s in the Netherlands [30] and at Oak Ridge National Laboratory [31,32]. They were further developed in the 1970s for specific applications at Oak Ridge National Laboratory, Harwell [33], Jülich [34], Mound, and other institutions.

The typical sol–gel methods used for heavy elements are generally classified as either internal or external gelation processes. Both processes involve the use of a heavy metal, nitrate solution that is induced to form a colloidal suspension (solution) of particles by the addition of an ammonia source. With the external gelation process, the ammonia is supplied externally as either a solution or gas while with the internal gelation process, the ammonia is generated in situ. With external gelation methods, the droplets originate and retain a spherical shape due to the high surface tension of the solution. The amount of initial solidification can be controlled by varying the reaction time and concentration of ammonia in the precipitation solution. Fabricated particles are washed to remove ammonia salts formed during gelation, dried in air, and calcined under hydrogen to improve particle density. This particular method is viewed as more easily accomplished remotely (simple droplet formation) and can be done using solutions readily available from typical reprocessing operations that may also lead to a diversion resistant reprocessing scheme. The problems associated with external gelation result from the high percentage of binder addition, which requires more difficult processing after sphere formation. The binders may also be susceptible to damage from radiation sources. Internal gelation methods are capable of eliminated the use of corrosive ammonia gas and or ammonia solutions (since the ammonia is generated in situ). Metal oxide powders are suspended with water in a temporary binder, for which alginates are the most conventionally used. The suspension is molded employing a vibrational dropping process into an aqueous solution where the microspheres solidfy, followed by washing, drying, and decomposition of the binder via calcination. Internal gelation involves easy preparation of solutions and methods of producing a wide variation in particle sizes, but the solutions are often unstable, contain more and varied polymer materials to deal with in washing steps, and, therefore, usually create more waste products that ultimately increases the cost of the process. However, the internal gelation method is still being investigated for next generation actinide fuel fabrication, mostly due to the improved product microhomogeneity and reduced process steps compared to the external gelation method.

Fuel microspheres consisting of $(U, Pu)O_2$ fuel can be pressed and sintered into conventional pellets as an alternative fabrication method [35]. The microspheres are fabricated employing a sol–gel microsphere pelletization route, thus avoiding the complexity and dust hazards associated with powder metallurgy while also remaining robust enough that the technology can be applied to conventional pellet loading. Experimental results employing this process have been encouraging, producing fuel pellets with controlled microstructures and densities upwards of 96% theoretical [36].

2.2.3. Sintering

The blending apparatus was joined with a pellet press sieve to separate improperly sized particles. Particles that pass through the sieve, typically less than 44 μm, were transferred into a press feed hopper where a double-acting cam hydraulic press produced one or more pellets simultaneously by compacting the powder. Pressed pellets were typically 50–60% of theoretical density and had dished ends to compensate for radial variation in the thermal expansion at reactor temperatures, an inherent effect associated with oxides due to a high centerline temperature caused by lower thermal conductivity. A larger powder-size distribution required higher compacting pressures as coarser particles needed to be fractured during pressing in order to make acceptable compacts. A distribution of finer oxide particles required much lower compacting pressures. Particles that did not pass through the sieve (i.e., coarse particles in the [−140 + 300] mesh range), could be used to fabricate MOX fuel pellets employing sintering followed by a swaging process.

The pressed pellets were placed into Al_2O_3 sintering crucibles for densification. The used binders were removed from the pressed samples by heating well above the decomposition temperature of the binder for several hours in a resistance furnace with an inert or slightly oxidizing atmosphere [37]. Pressed pellets have been typically sintered at 1600–1700 °C for 4–10 h in a H_2–Ar or H_2–He gaseous atmosphere. Above 1200 °C the heating rate was held below 100 $°C\,h^{-1}$ in order to maintain adequate dimensional stability [27]. Sintered MOX pellets were typically 6.4 mm long, 3.8 mm in diameter, and 95–96% of theoretical density with pore sizes typically ranging from 5 to 10 μm in diameter. Longer pellets could be produced but were typically found to result in greater abrasion and galling of the pressing dies. The sintering furnace is operated in a batch process mode with a programmed time-temperature profile based on well-defined quality specifications.

2.2.4. Swaging

One early fabrication method of fuel rods employed swaging. Coarse particles obtained from the sieving process were sintered in a furnace at 1400 °C for a few hours under H_2–He cover gas [38]. The appropriate amount of powder was loaded into stainless steel cladding, with one end cap and an insulating UO_2 pellet of the same dimensions in place, and vibratory compacted. The tube was then swaged to produce a compacted fuel of approximately 75% TD. Swaging is not used today to produce fast reactor fuel rods.

2.2.5. Pellet preparation and quality assurance

Sintered MOX pellets were transferred to a grinder feed hopper and continually processed through a center-less grinder to produce a pellet diameter within specification. Confirmation of the pellet diameter was obtained using a linear variable displacement transducer or laser micrometer and sintered pellets were visually inspected and sorted. Scrap pellets segregated during the visual inspection were collected into a crusher box, crushed into an appropriate particle size by a jaw crusher, and re-introduced into the material feedstock.

2.2.6. Fuel rod fabrication

Pellets that passed through the sorting process were stacked to prepare a column for one fuel rod. The fuel rod jacket, traditionally constructed of stainless steel alloys 304, 316, or 347 (though newer D9 and HT9 alloys were also used) consisted of a cladding tube attached to a bottom end cap using an orbital GTAW (gas tungsten arc welding) technique. Typically, the elements were created with an open plenum volume equal to that of the fuel itself to accommodate fission gas release and control the level of pressure-induced stresses that are created in the cladding tube.

Construction of the element began by inserting a 90% theoretically dense UO_2 insulator pellet[1] into the bottom of the jacket to minimize heat loss through the bottom of the fuel column and to protect the lower end plug from a high-temperature pellet in the fuel column. The fuel rod jacket slid over the fuel column through a hood enclosure at one end of an inert atmosphere fabrication glovebox and into a He gas purge chamber. A plenum spring was inserted into the cladding to prevent the fuel column from sliding into the void section of the element and the jacket end from becoming contaminated, and then an end cap was installed. Lastly, the fuel element was sealed using an orbital GTAW or pulse magnetic weld to seal the upper end plug. An example schematic of a Series I and II FFTF fuel rod is provided in Fig. 2.

All sealed fuel rods were inspected for contamination and verified prior to removal from the glovebox. X-radiography analysis was performed to ensure that the end caps were properly welded, and gamma scanning was carried out to verify Pu concentration of the pellets. Leak testing was carried out employing one of three methods: spray probe testing, vacuum chamber testing, or sniffer probe testing [39]. Spray probe testing involved evacuating a test piece attached to a mass spectrometer leak detector (MSLD) and spraying the surface of the test piece with a tracer gas, such as Xe or He, from a spray probe. Vacuum chamber testing involved detecting leakage from a He-bound fuel rod placed in a vacuum chamber. Leakage was determined by sensing He with an MSLD attached to the test fixture. Sniffer probe testing was conducted by backfilling or pressurizing a test piece with a tracer gas and sniffing the surface of the test piece with a specially prepared probe attached to an MSLD.

Once the fuel rod was confirmed as sealed, the tag gas capsule (Fig. 2) was broken to distribute its contents throughout. The tag gas capsule contained Xe and Kr gas in an isotopic ratio that was the same for each fuel rod in a single assembly, but was different for each assembly loaded into the core. Thus, if a fuel rod began to leak during irradiation, the fuel rod was identified by analyzing the Xe–Kr isotopic ratio in the cover gas above the level in the sodium coolant.

3. Carbide fuel fabrication technology

3.1. History and experience

There is not an extensive fabrication and irradiation database for carbide fuels established in the US compared to oxide and metal fuels. Other than in support of the irradiation experiments carried out in EBR-II and FFTF in the 1980s, carbide fuel fabrication activities have mainly been pursued in Japan, France, India, and the Russian Federation. Carbide fuels are candidate materials for transmutation of the actinides due to high atomic densities [40]. In addition, carbide fuels can be fabricated with either a small, He-bonded fuel/cladding gap or a large, sodium-bonded gap that, in many regards, combines the best attributes of both metal and oxide fuels [41]. For example, if higher reactor/fuel temperatures are desired that result in a softer fuel at reactor temperatures, fuel pellet diameter can be increased and a He bond used, while lower

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[1] Uranium dioxide insulator pellets could be composed of either natural or depleted U.

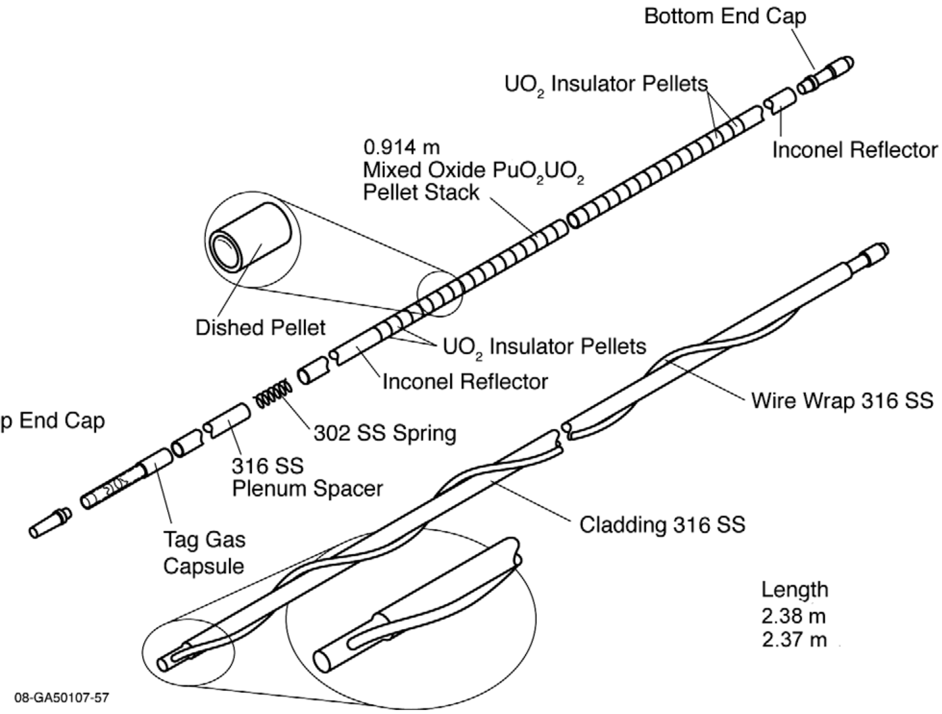

Fig. 2. Schematic of Series I and II FFTF fuel rod showing pertinent features of a design typical for a commercial type Liquid Metal Reactor [14].

reactor/fuel temperatures would result in a harder fuel with more gas retention and warrant use of a smaller fuel pellet diameter and a sodium bond that also accommodates swelling of the fuel.

3.1.1. EBR-II

Over 470 (U, Pu)C fuel rods were irradiated in EBR-II to obtain information on bonding (Na or He) and cladding [42,43].

3.1.2. FFTF

Over 200 carbide-fueled UC–PuC rods were irradiated in FFTF, including both Na-bonded and He-bonded types [12,43–46]. The AC-3 test irradiated both pellet and sphere-pac fuel [45].

3.2. Carbide fuel fabrication process

Carbides are attractive due to high thermal conductivity, less restructuring of the microstructure compared to oxide or metal, and a fissile density that permits superior breeding performance and high specific power operation, which combine to increase Pu production and reduce fuel cycle and power costs [46].

3.2.1. Process overview

Uranium carbide fuel has been researched for possible use in reactors since the early 1960s. At that time, there were three well-known and established fabrication routes: melting and casting, traditional pressing and sintering, and reaction sintering. These basic initial processes have since been combined and expanded upon leading to more efficient modern fabrication techniques.

3.2.2. Melting and casting

Originally, melting and casting technology was heavily relied upon because it had several advantages over more conventional powder metallurgy processes. These advantages included products that were almost fully dense and had a higher attainable purity level. Of the several methods developed to melt and cast UC, laboratory-scale drop casting was the most straightforward. Drop casting involved arc melting feedstock, either UO_2 or U metal chips, with graphite into a button. The button was only partially melted, and as such required repeated rotation in order to homogenize the melt. After homogenization was complete, the button was set on the mold opening, fully melted, and dropped into the mold cavity by gravity. A thin graphite barrier could also be placed over the mold opening so that once the molten button reacted with this layer, the product dropped into the mold. Fuel pellets as large as 1.8 cm in diameter and 15 cm long were cast employing this technique. For larger fuel pellets, a skull melt, non-consumable vacuum arc furnace was designed that could cast approximately 8 kg of UC into six 2.3-cm-diameter by 18-cm-long pellets. The skull melt process takes advantage of hollow copper fingers that rapidly remove heat from the melt resulting in a thin crust of solid, termed a skull, formed on the outer periphery of the melt; thus permitting the material to serve as its own container and minimizing the amount of contamination. Despite the early interest and reasonable success of melting and casting, the technique ultimately proved largely uneconomical and produced coarse–grained products that were not as desirable as the fine–grained powder metallurgy products [47].

3.2.3. Sintering

Carbide powder feedstock for sintering was typically produced via carbothermic reduction. UC powders were produced by combining UO_2 and C powders in the desired ratio followed by compaction into a pellet. The pellet was placed in a furnace under vacuum (approximately 10^{-5} Pa) and heated to approximately 1700 °C for 2 h. The reaction product was UC even though UC_2 was likely formed as an intermediate reaction product, according to Eqs. (4) and (5).

$$0.5\ UO_2 + 2C \rightarrow 0.5\ UC_2 + CO(g) \tag{4}$$

$$0.5\ UO_2 + 1.5UC_2 \rightarrow 2UC + CO(g) \tag{5}$$

The UC reaction product was crushed and ground to a fine powder suitable for the sintering process.

Similarly for (U, Pu)C production, $UO_2(NO_3)_2$ and $Pu(NO_3)_4$ were converted to stable oxides (UO_2 and PuO_2) with a simple heat treatment to decompose the nitrates, thereby off-gassing water and nitrous oxide(s). A mixture of UO_2 and PuO_2, along with carbon black or graphite, was blended using ball milling. Fuel enrichments were, nominally, 20% total Pu in the U–Pu mixture. Prior to carbothermic reduction, the mixed powders were consolidated by briquetting, which promotes formation of a homogeneous product. Typically, the actual carbothermic reduction step was carried out under vacuum at 1600 °C according to the reaction in Eq. (6) [43].

$$3UO_2 + 3PuO_2 + 16C \rightarrow (U,Pu)C + (U,Pu)_2C_3 + 12CO(g) \tag{6}$$

The resulting carbide product was mechanically crushed and vibratory-ball milled in order to obtain powders of the desired particle size, less than 44 μm.

The carbide powders were combined with a binder and pressed to approximately 410 MPa. The resultant pellets were sintered under flowing Ar cover gas at 1800 °C (though temperatures as low as 1550 °C have been used) for several hours, and then underwent center-less grinding to achieve the desired pellet diameter [46]. Sintered densities of approximately 80% of theoretical suitable for He bonding have been achieved through varying powder particle size and sintering temperature. Sintered densities of 90% theoretical were achieved at a temperature of 1900 °C for 20 h [36,43]. All steps were carried out in gloveboxes with an inert, high-purity Ar atmosphere. The press and sinter process was employed to fabricate several thousand UC pellets with 10–15% of U_2C_3 that met requirements for irradiation in FFTF. Sintering aids, such as Ni, were also investigated and appeared to have developed a thermally stable dense microstructure with discrete pores controlled through pore-former additions. It should be pointed out that sintering under a vacuum typically resulted in increased sintered densities, but also resulted in a low density of larger pores caused by volatility of impurities at the sintering temperatures [47].

3.2.4. Reaction sintering

Reaction sintering involved combination of UO_2 and C pressed into a pellet followed by heating to 1125 °C at a rate of approximately 300 °C $\min^{-1}$. An exothermic reaction was initiated through reduction of the oxide by C at approximately 800 °C that raised the local temperature above the melting point of U. The U was present as a molten metal for several minutes, and after approximately 2 h at 1125 °C, only 0.25 to 1% of the U remained in a metallic state, in addition to excess C. The localized high temperature and presence of liquid U enhanced sintering of the UC product. Excess C was removed by increasing the furnace temperature to 1400 °C [47]. Effectively this route is similar to the powder fabrication route for the press and sinter process, less the mechanical crushing and grinding step.

3.2.5. Microsphere fabrication

An alternative, newer technique for carbide fuel fabrication is the internal (wet) gelation method, similar to that discussed earlier for oxide powder feedstock fabrication. This method offers the advantage of integrating the final nitrate-to-oxide conversion step in reprocessing by using a co-conversion process with colloidal carbon to produce spherical carbide particles for direct loading into fuel pellets [46]. Blending of an acid-free, concentrated feed solution of $UO_2(NO_3)_2$ and $Pu(NO_3)_4$ was carried out in the liquid phase, which produced a very homogeneous solution of Pu in U. As a result, the mechanical milling, crushing, blending, and grinding steps associated with carbide powder feedstock preparation were eliminated altogether. The liquid solution was mixed together with hexamethylenetetramine, urea, and dispersed carbon black. The solution mixture was chilled and injected or dropped directly into hot Si oil, which resulted in an immediate temperature rise and decomposition of the hexamethylenetetramine. The decomposition formed ammonia, which precipitated ammonia-diuranate or corresponding plutonate within droplets, forming microspheres. A washing step removed the Si oil and solvents. The reaction products were dried in air at 110 °C. Volatiles were driven off during calcination under an Ar/H gaseous mixture. Adjustment of the U–Pu to desired oxidation states was also carried out during this time. High-density carbide microspheres were produced by heating the microspheres at 1950 °C under flowing Ar cover gas for 8 h, resulting in reduction of the oxides and formation of carbide microspheres. A sieving process was used to remove oddly sized particles. Clean, dust-free spherical particles with densities of greater than 95% theoretical were then loaded directly into fuel rods. The microsphere fabrication technique and associated process equipment were easily accommodated in a glovebox. A process flowchart comparing the typical press and sinter process and wet internal gelation fabrication process for carbide fuel fabrication is provided in Fig. 3.

3.2.6. Pellet preparation and quality assurance

Final pellet preparation (after sintering/hot pressing) for mixed carbides can be very similar to that of MOX fuels. One must ensure that correct conditions for pellet fabrication have resulted in pellets with specified density/porosity, grain size, stoichiometry, and impurity content.

The difference occurs where the pellets will be sodium bonded. In this case, the pellet-cladding gap can be much larger than when a He bond is used. Mixed carbides are relatively low-swelling fuels; thus, this increased gap allows relaxation in tolerances for pellet diameter. In turn, this allows elimination of the final center-less grinding step, an advantage especially when considering fabrication of these fuels for transmutation applications, using reprocessed fuel containing minor actinides in a hot cell. In this case, the hot cell interface makes operations like grinding more difficult because grinding is a potential source of contamination and for the generation of waste products containing minor actinides and requires more precise dimensional checks. The disadvantage is that a sodium-bonding step must be added to the fuel rod fabrication and radiography added to evaluate the quality of the bond.

3.2.7. Fuel rod fabrication

Carbide fuels were never used to fuel an entire reactor for an extended period in the US. Furthermore, because no US reactors (Clinch River Breeder Reactor, FFTF, PRISM [power reactor innovative small module]) were designed for carbide fuel, the experiment database for this type of fuel is much more limited than what is available for oxide and metallic fuels. As such, the US's carbide fuel rod fabrication experience is not as extensive, nor are methods as thoroughly developed, as they are for oxide and metal fuels. It can be assumed that fabrication techniques would be similar to those for MOX fuel for He-bonded versions and to techniques for metal fuel fabrication for Na-bonded versions of carbide fuels. Note that there are some special fabrication requirements, such as the need to keep the environment free of O_2 and water vapor due to potential performance effects [44], as is also the case for nitride fuels [48].

4. Nitride fuel fabrication technology

4.1. History and experience

Nitride fuels do not have as extensive a fabrication and irradiation database in the US as oxide and metal fuels. Interest in nitride fuels for space power applications, such as the SP-100 space reactor program [49–51], has been the main driving force behind

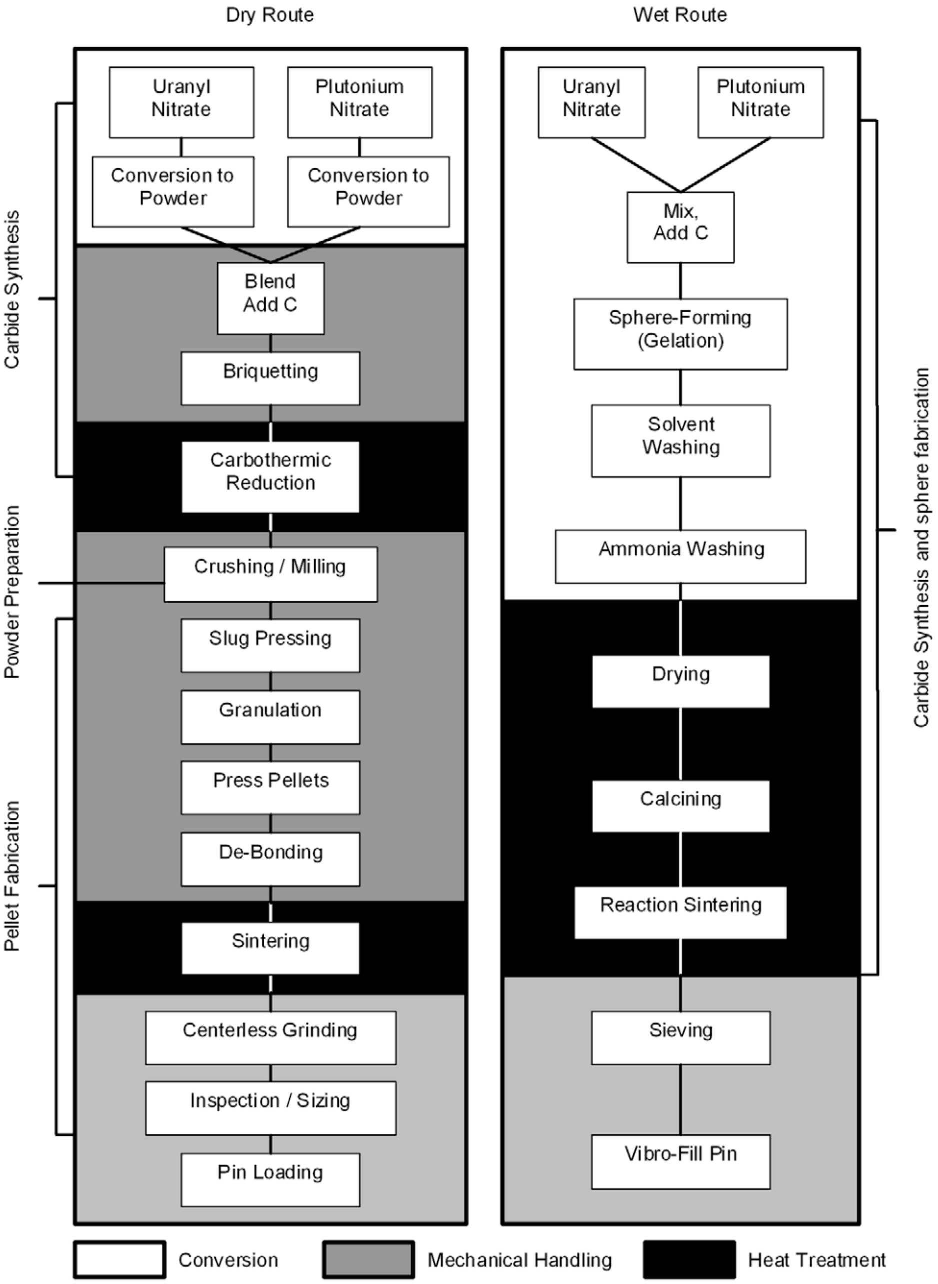

Fig. 3. Comparison of process flow charts for carbide fuel fabrication employing a carbothermic reduction route (left) and an internal gelation route (right) [46].

improving fabrication technology. Similar to carbide fuels, nitrides are candidate materials for transmutation of the actinides, due to their high atomic densities [40], and can also be fabricated with either a small, He-bonded fuel/cladding gap or a large, sodium-bonded gap [41].

4.1.1. EBR-II

Approximately 100 (U, Pu)N fuel rods were irradiated in EBR-II, mainly to obtain information on bonding (Na or He) and cladding [42,43]. These early investigations in the EBR-II program revealed a potential for nitride use in fast reactors [42,52].

4.1.2. FFTF

Over 54 UN rods were irradiated in FFTF. These rods contained small fuel columns approximately 10–15 cm long and were clad in a refractory metal alloy [12].

4.2. Nitride fuel fabrication process

Nitride fuel is an attractive option because it has similar desirable characteristics as metallic fuel, such as high heavy metal atom density and exceptional thermal conductivity. Moreover, nitride fuels have higher melting temperatures. Compared to oxides, nitride fuels have only one moderating atom per molecule and are compatible with existing fabrication and reprocessing methods established for oxide fuels [12].

However, nitrides have moderate disadvantages, in that fuel compositions sublimate or decompose at temperatures lower than the congruent melting point if N_2 overpressure is not maintained, and the absorption cross-section of N_2 for thermal neutrons is high enough to reduce the breeding ratio [7]. In addition, there is concern over the reprocessing of nitride fuels fabricated using natural N_2 due to the production of biologically hazardous ^{14}C [53]. Both the biological and absorption cross-section concerns can be addressed by enriching the ^{15}N component in natural N_2 used for fabrication, but this affects the desirable economics of fabrication technology. Conversely, nitride fuels have very similar irradiation performance, better chemical stability, and easier dissolution in the head end of the PUREX process in comparison to carbide fuels [54].

4.2.1. Process overview

Similar to carbide pellet fabrication, there are two common techniques used to fabricate nitride fuel samples in the US. The first technique is carbothermic reduction, and because this is explained in detail for carbide fabrication, only the salient points and deviations from the previous description will be discussed in the following section. The second method is a hydride–dehydride–nitride process, which will be discussed in following section.

4.2.2. Nitride feedstock preparation

Uranium nitride fuel fabrication employing carbothermic reduction involves, first, converting UO_2 to UC through blending and milling of UO_2 with C and then pelletizing it into coarse briquettes heated to 1600 °C under vacuum. Further reduction was carried out by soaking the UC in N_2–6% H_2 gas that formed UN and removed free C in the form of methane gas, followed by a final crushing and milling step to create feed powder [55,56]. The process is summarized in Eqs. (7) and (8).

$$UO_2 + 3C \rightarrow UC + 2CO(g) \tag{7}$$

$$UC + 0.5N_2(g) + 2H_2(g) \rightarrow UN + CH_4(g) \tag{8}$$

A C/UO_2 mole ratio of less than 2.20 was required to maintain low C levels at the reaction temperature of 1600 °C [48]. This reaction temperature was found to be necessary in order to keep O_2 levels low because O_2 levels increased with decreasing C content. Both O_2 and C residual contents were found to be extremely sensitive to the batch size and flow rate of the reducing N_2–6% H_2 gas [48].

The hydride–dehydride–nitride process requires a high-purity metal feedstock (i.e., from electrorefining) similar to requirements for metallic fuels, typically less than 0.001 mass percent of detectable metallic impurities [57–59]. High levels of O_2 and C were undesirable in the fuels as these promoted cladding attack and coolant interactions.

Several hundred grams of nitride were fabricated in a batch process using fine U turnings. Turnings were washed in CCl_4 to remove residual machine oil from the turning process and were then rinsed in acetone. Surface oxides were removed from the dried turnings by washing them in a 10% aqueous solution of nitric acid. Turnings were placed in high-purity alumina boats with UN liners. The boats were placed in a stainless steel or impervious ceramic tube inside a furnace enclosed in a high-purity Ar atmosphere glovebox. The system was initially purged with high-purity H_2 and then a prescribed flow rate of H_2 was established. The temperature of the reaction vessel was raised to 230 °C at a prescribed heating rate. The temperature was maintained for a few hours to perform the hydride step.

Dehydriding was performed by evacuating the reaction vessel as the temperature was increased to 400 °C at a prescribed heating rate. UH_3 decomposes under atmospheric pressure at 300 °C [60]. The temperature was maintained for a few hours, during which time the metal hydride(s) decomposed and H_2 was removed from the atmosphere, along with any additional impurities such as C and O_2, while the pressure inside the reaction vessel decreased. The reaction vessel was cooled to approximately 200 °C under vacuum to remove all of the H_2 and impurities from the hydriding process. High-purity N_2 gas was introduced to the reaction vessel while the temperature was increased to 800 °C at a prescribed heating rate. An increase in N_2 flow was necessary to maintain the reaction vessel pressure from 500 to 600 °C because the rate of reaction between U metal and N_2 increased. The temperature was maintained for a few hours during which time no further N_2 was absorbed. The furnace temperature was further increased under vacuum to 1120 °C at a prescribed heating rate for a few hours in order to decompose U_2N_3 to UN and reduce the surface area of the product [61]. The mononitride powder was allowed to cool rapidly to room temperature under vacuum followed by removal of the product to the inert glovebox. Fabrication of PuN is similar to, though not as complex as, PuN produced by direct nitridation of metallic Pu at 1000 °C with no formation of sesquinitride [62,63].

4.2.3. Sintering

As-synthesized UN powders were typically vibratory ball-milled to reduce particle size in a reasonable amount of time, slugged, granulated, pressed into green pellets, and sintered under various conditions. Other consolidation methods that were used were hot pressing and hot isostatic pressing, although conventional press and sinter methods were the most commonly used [48,62]. Specimens to be sintered were isostatically pressed into right cylindrical specimens of specified dimensions with a typical green density of 65% theoretical.

Sintering was carried out under a N_2 and/or Ar gaseous atmosphere. Specimens sintered at low temperature and pressure for 2 h achieved higher densities than those sintered under low temperatures and high pressures. Sintering temperatures greater than 1800 °C with low N_2 pressures typically resulted in dissociation of residual U_2N_3 to UN and N_2 and further dissociation of UN into liquid U and N_2 gas, and the resulting gas pressure caused the pellets to crack. Either sub-stoichiometric UN powders were used ($UN_{0.98}$) to minimize U_2N_3 phase formation, or a N_2 sintering atmosphere of at least 10^{-3} Pa for sintering temperatures above 1600 °C was used to prevent further dissociation of UN [48,64].

Higher sintering temperatures (approximately 2200 °C) could be used in conjunction with low N_2 pressure, with the balance of Ar being used to maintain an increased overall pressure. As such, dense UN compacts (approximately 92% of theoretical) were fabricated [64]. On average, sintered UN fuel pellets were 87% of theoretical density, had uniform, small grains approximately 5–20 μm, and had irregularly shaped open pores that tended to manifest themselves within the grains rather than at grain boundaries. Hot pressing UN at 1400 °C and 44 MPa pressure was found to be an effective densification technique. An increase in temperature to 1450 °C allowed pressure to be reduced by half while maintaining sintered densities greater than 90% of theoretical [62].

4.2.4. Pellet preparation, quality assurance, and fuel rod fabrication

Methods employed for nitride fuels in terms of pellet preparation, quality assurance, and fuel rod fabrication are very similar to those for carbide fuels, as far as US experience goes. These methods have been discussed in the previous sections for carbide fuels, and there are only minor differences in methodology, if any.

5. Summary

The US's experience with ceramic fast reactor fuel fabrication technology is relatively extensive. This experience has been gained through the diligent efforts of numerous organizations, such as the government, national laboratories, industry, and academia. It is imperative that the experiences gained by these organizations over the last six decades be well documented and understood so that future researchers can easily access and understand this information.

A critical path toward and in support of the nuclear energy renaissance is and will continue to be fuel fabrication. Technologies that improve the quality, enhance the safety, and reduce the cost associated with nuclear fuel fabrication should continually be sought and should build upon the established technologies and traditions of previous researchers documented in this paper.

6. US Department of Energy disclaimer

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