Coated particle fuel: Historical perspectives and current progress

Paul A. Demkowicz [a, *], Bing Liu [b], John D. Hunn [c]

[a] Fuels and Materials Division, Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID, 83415, USA
[b] Institute of Nuclear and New Energy Technology, Collaborative Innovation Center of Advanced Nuclear Energy Technology, Tsinghua University, Beijing, 100084, China
[c] Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN, 37831-6093, USA

HIGHLIGHTS

- Coated-particle fuel has matured over six decades of development.
- Early evolution involved a wide variety of a different coating configurations.
- Modern TRISO particle design can maintain coating integrity under extreme conditions.
- Fission products are largely retained at their source in the fuel kernel.
- Several major programs on TRISO fuel qualification are currently in progress.

ARTICLE INFO

ABSTRACT

Coated particle fuel concepts date back some 60 years, and have evolved significantly from the relatively primitive pyrocarbon-coated kernels envisioned by the first pioneers. Improvements in particle design, coating layer properties, and kernel composition have produced the modern tristructural isotropic (TRISO) particle, capable of low statistical coating failure fractions and good fission product retention under extremely severe conditions, including temperatures of 1600 °C for hundreds of hours. The fuel constitutes one of the key enabling technologies for high-temperature gas-cooled reactors, allowing coolant outlet temperatures approaching 1000 °C and contributing to enhanced reactor safety due to the hardiness of the particles. TRISO fuel development has taken place in a number of countries worldwide, and several fuel qualification programs are currently in progress. In this paper, we discuss the unique history of particle fuel development and some key technology advances, concluding with some of the latest progress in UO_2 and UCO TRISO fuel qualification.

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* Corresponding author.
  E-mail addresses: paul.demkowicz@inl.gov (P.A. Demkowicz), hunnjd@ornl.gov (J.D. Hunn).

1. Introduction

The modern coated fuel particle is a robust, carbon and ceramic fuel form based on the objective of encapsulating small spherical kernels of fissile material to retain fission products at their source. The fuel has evolved over the last 60 years from relatively modest beginnings and performance expectations to a design that can be manufactured with very high quality and excellent performance at extremely high temperatures. The fuel is primarily used in high-temperature gas-cooled reactors (HTGRs), which have graphite cores and are cooled by pressurized helium with coolant outlet temperatures in the range of 700—950 °C [1,2]. However, the fuel is also considered for other reactor designs, such as the fluoride salt-cooled high-temperature reactor (FHR) [3,4], and fuel concepts such as fully ceramic microencapsulated (FCM) fuels [5,6].

The state-of-the-art, tristructural isotropic (TRISO) coated fuel particle generally consists of a uranium-bearing kernel (either UO_2 or UCO, a heterogeneous mixture of uranium oxide and uranium carbide [7,8]) with a diameter in the range of 350—600 μm. This is surrounded by four different coating layers (Fig. 1). The first is a porous (approximately 50% of theoretical density) pyrocarbon buffer layer, which provides a void space for the fission gases released from the kernel and accommodates fission recoils. The next layer is a dense, highly isotropic inner pyrocarbon layer (IPyC).

This layer acts as a seal layer to protect the kernel from the HCl byproduct of SiC layer deposition, provides a deposition surface for the SiC layer, and contributes to the retention of fission gases in the particle. The SiC layer provides the main structural strength of the particle and is the primary barrier to the release of non-gaseous fission products not sufficiently retained by pyrocarbon or within the kernel itself. Finally, the outer pyrocarbon layer (OPyC)—with properties similar to the IPyC layer—protects the SiC layer during handling, acts as a surface for bonding to the graphitic fuel matrix, and provides an additional barrier to fission product release (especially fission gases). Coating layer properties and thicknesses can vary depending on the specific particle design and the fabrication processes.

There are several methods for fabricating fuel kernels. The sol-gel process is the most commonly used process today. Sol-gel spheres can be made using internal or external gelation methods [10,11]. Typically, some form of uranium, other actinide (e.g., Th), or actinide mixture (e.g., Th/U) is dissolved in nitric acid and internal or external sources of ammonia are used to precipitate a metal oxide from the sol (e.g., UO_3) and form a gel sphere as the droplets pass through a heated liquid medium. The gel spheres are washed, dried, and heated in appropriate atmospheres to drive off byproducts of the sol-gel conversion and form the final sintered kernel (e.g., UO_2). Carbon can be dispersed in the sol to support
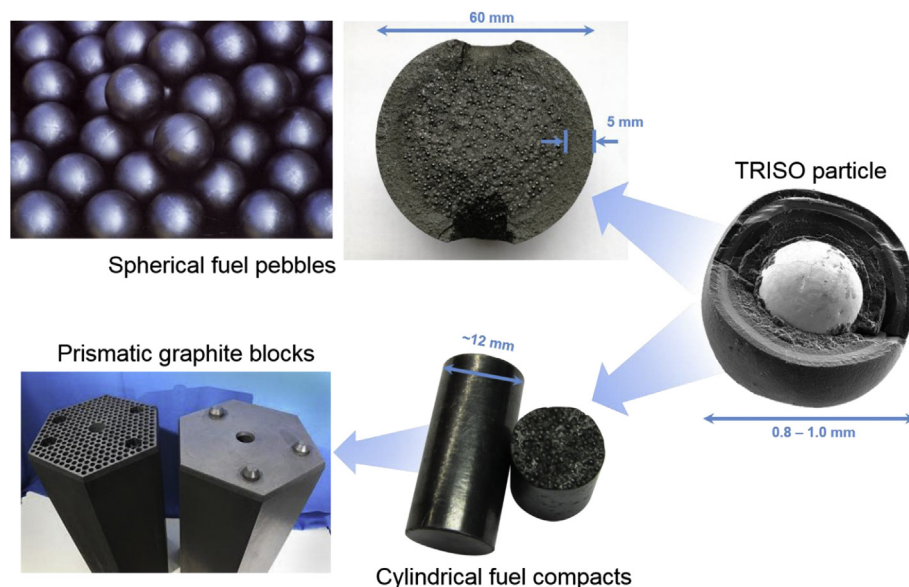

Fig. 1. Micrograph of a coated particle with exposed kernel and coating layers [9], examples of spherical (pebble) and cylindrical (compact) fuel forms, and prismatic graphite blocks. Images supplied courtesy of the US DOE, INET, and X Energy, LLC.

chemical conversion of some or all of the uranium oxide to carbide or nitride.

The TRISO coating is typically applied using a fluidized-bed chemical vapor deposition system (FB-CVD) [12]. In a commonly used FB-CVD process, kernels are fluidized with argon and reactant gases are added at optimized coating gas fractions and temperatures for the deposition of each layer [13]. Acetylene (C_2H_2) is used for deposition of the low-density buffer. Propylene (C_3H_6) is mixed with the acetylene and argon to deposit the higher-density IPyC and OPyC layers. Methyltrichlorosilane (CH_3SiCl_3) mixed in hydrogen provides the source for SiC deposition and can be used with or without the argon fluidization gas. Fig. 2 shows cross sections of modern TRISO particles.

TRISO particles are commonly consolidated into two types of fuel forms depending on the reactor in which they will be used (Fig. 1). In all cases, the surrounding "matrix" material is primarily made from graphite and carbonized resin. The matrix is formed by molding a conglomeration of TRISO particles, graphite flake, and a thermoplastic or thermosetting resin under pressure to achieve a sufficient density and then heating the pressed form to cure and carbonize the resin. In most modern-day fuel forming methods, TRISO particles are overcoated with the graphite/resin blend prior to pressing in order to prevent particle-to-particle contact during the molding process, which may damage the particles [14—16]. Spherical fuel pebbles are used in pebble-bed HTGRs, with the most common design consisting of around ten thousand particles in a 60-mm diameter sphere. A 50-mm diameter central region contains the fuel particles, while a 5-mm thick non-fueled region comprises the outer perimeter [17]. Particles may also be formed into cylindrical compacts (either solid cylinders or annular cylinders), which are placed into holes in the graphite blocks in a prismatic HTGR core.

Coated particle fuel has a long development history that is represented by an expansive publication record in the scientific literature. As the fuel form evolved and the experience base broadened, modifications to the particle design were implemented that addressed specific shortcomings and improved overall fuel performance. Given the relative technical maturity of TRISO fuel, a number of excellent reviews have been published in the new millennium, covering all areas of the technology including the evolution of the fuel form, fabrication methods and quality control, irradiation performance, high-temperature accident testing, particle failure mechanisms, and computational modeling of fuel performance [1,2,18—21]. In addition, numerous publications are available that focus on specific past fuel development efforts. These include many references summarizing the history and successes of the extensive German program in the 1980s and early 1990s to develop and test the performance of LEU UO_2 TRISO fuel [17,22—29], which has been influential on many subsequent endeavors across the globe. Consequently, the authors' intent is not to provide a comprehensive review of TRISO fuel or its behavior. Rather, this article will provide a concise look back at the history of coated particle fuel development, and then turn to a description of the current major efforts in TRISO fuel development and qualification in the last decade.

2. Coated particle fuel development history

2.1. Origins, early demonstrations, and coating evolution

The concept of nuclear fuel microspheres with refractory coatings to contain fission products during operation in HTGRs can be traced to the early years of the nuclear age, during the initial stages of the Dragon Project in the 1950s [30]. While the earliest versions involved fuel kernels coated with only a single pyrolytic carbon layer intended to protect the kernel during fabrication, this rapidly evolved in the following decade into more complex and more effective particle designs, as it was recognized almost from the beginning that the coating layers could provide some degree of advantageous fission product retention when properly designed [31,32]. A number of different particle designs are apparent in the early literature and reflect the innovations that took place in the 1960s [33—36] (Fig. 3).

Fracturing of the pyrocarbon layer in these particles during irradiation was found to be a concern, and analysis revealed a number of potential causes, including kernel swelling, radiation-induced pyrocarbon dimensional change, damage from fission recoils, fission gas pressure, and chemical reaction of fission products with the coating [35]. One of earliest design modifications was the introduction of a sacrificial layer of pyrocarbon between the kernel and the dense, outer pyrocarbon layer. The inner layer was fabricated with lower density and compliant mechanical properties such that it could accommodate fission gases and kernel swelling, while the outer layer would perform the function of fission gas retention [35]. Some iterations of this two-layered particle design were termed "BISO" (for Buffer-isotropic [33,37] or Bistructural isotropic [1]) and exhibited significantly improved performance over the more primitive pyrocarbon-coated particles [33] (Fig. 3(a)).

An early concept to limit crack propagation in the coatings was the use of several pyrocarbon layers with a "laminated" structure, sometimes with the various layers deposited with different
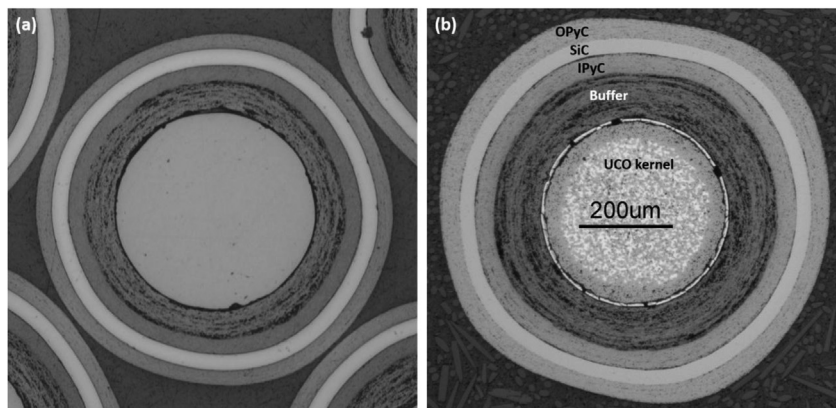

Fig. 2. (a) UO_2 TRISO particle with 500 μm diameter kernel. (b) UCO TRISO particle with 425 μm diameter kernel.
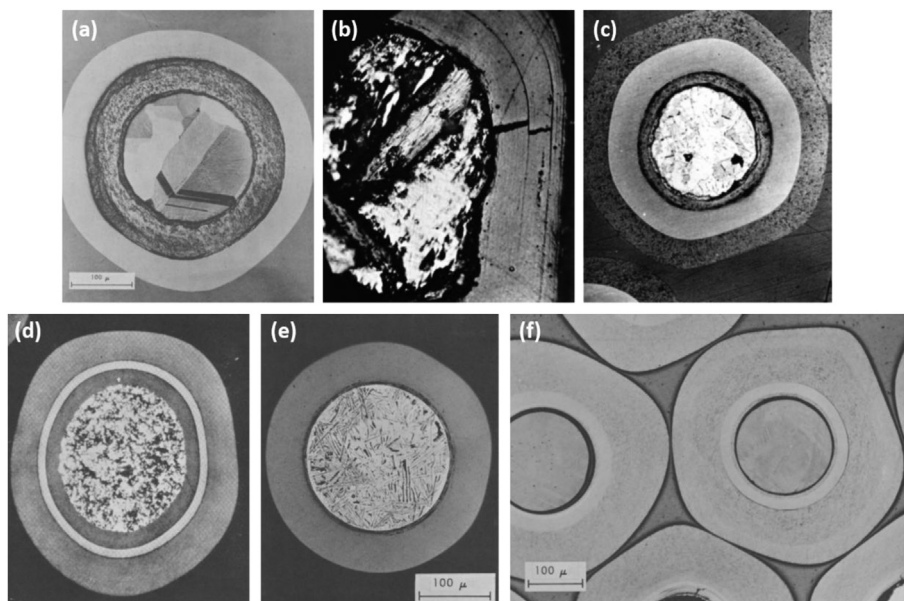
Fig. 3. Coated particles from the first decade of development. (a) Early example of a BISO particle. (b) Particles with "laminated" pyrocarbon layer structure demonstrating arresting of crack propagation at the lamination interfaces. (c) Particle with "Triplex" structure (porous buffer layer followed by laminar and columnar pyrocarbon layers). (d) Fertile $(Th,U)C_2$ particle used in Dragon first charge, consisting of PyC-SiC-PyC structure. (e) Carbide particle with single PyC coating layer used in Peach Bottom first core. (f) "Duplex" $(Th,U)C_2$ particle used in AVR first core load. Images from References 33,35, used by permission.

microstructures and properties, such that a crack in one layer would be halted when encountering the next [35] (Fig. 3(b and c)). However, irradiation testing demonstrated that pyrocarbon layer properties—in particular the level of crystalline anisotropy—could have a significant impact on irradiation performance. Pyrocarbon with significant anisotropy is much more susceptible to asymmetric dimensional change due to fast neutron irradiation, which can result in unacceptably high stresses in the layer and ultimately fracture. Particle development involved experimentation with a wide variety of PyC coating parameters, including various coating gases, gas concentrations, and deposition temperatures [38,39]. Ultimately, fabrication conditions that promoted isotropic pyrocarbon were pursued to improve radiation stability [38,40], and the earlier laminar coatings were abandoned.

The notion that refractory ceramics, including metal carbides, could provide significantly improved particle strength and retention of fission products such as cesium and strontium compared to pyrocarbon alone was suggested near the very beginning of coated particle development [30,31]. This led to the addition of a silicon carbide layer placed between two layers of dense pyrocarbon. This innovation, along with the porous buffer layer of BISO particles, formed the basis for the modern TRISO coating design, which is used in all contemporary HTGRs.

Burgeoning development of the fuel form in the 1960s involved improvements to fabrication methods, extensive irradiation testing of particles and fuel elements in materials test reactors (MTRs) and demonstration reactors, and the development of computational models to predict coating behavior (see, for example, References [37] and [41]). The major early developers included the UK (as part of the OECD-sponsored Dragon project), Germany, and the US. Coated particle fuel experienced its first practical demonstrations starting in the 1960s in the Dragon Reactor in the UK, the Arbeitsgemeinschaft Versuchsreaktor (AVR) in Germany, and Peach Bottom Unit I in the US. These reactors served as test beds for various types of coated particle fuel throughout their lifetimes.

The Dragon reactor was a 20MWt prismatic-core HTGR that achieved first criticality in 1964 [30]. The first fuel charge included fissile particles consisting of several consecutive layers of pyrocarbon with different properties, and fertile particles with both PyC and SiC coating layers[1] [33] formed into annular cylinders. A large variety of experimental particle types were inserted into the first charge core and subsequent charges [38,39]. Among the coating variations that were part of this experimentation was a version with a very low-density buffer layer with a reported density similar to that in modern TRISO fuel (~1 g cm^{-3}).

Peach Bottom was a 115 MWth prismatic HTGR. The reactor first went critical in 1966, with the first core utilizing pyrocarbon-only coated particles [33] (Fig. 3(e)). Widespread (~80%) particle coating failures were expected in the first core fuel, with a gaseous purge system installed in each fuel element to limit release to the coolant [42]. After experiencing widespread failure of fuel elements in the first core due to swelling associated with particle failures, the use of BISO particles started with the second core loading in 1970, continuing until final shutdown in 1974 [43]. The reactor also contained fuel test elements in which a large number of different types of particles were irradiated, including TRISO particles with various kernel types, and post-irradiation examination of these elements yielded a large amount of data on fuel performance [44,45].

The 46 MWth AVR was the first pebble-bed reactor design to be constructed, and started operation in 1967. The first core loading utilized $(Th,U)C_2$ kernels with coatings referred to as Duplex (Fig. 3(f)) in machined graphite spherical elements. The Duplex coatings had two pyrocarbon layers: a thin, sacrificial inner layer and a higher-density outer layer to retain fission products [33,46,47]. The particles were mixed with graphite and thermosetting binder, and then injection molded into a spherical inner cavity machined in the spherical fuel element. The fuel element manufacture process soon changed to an isostatic molding method,

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[1] Although these first-charge particles contained a PyC-SiC-PyC structure, micrographs of the fertile Dragon particles initially loaded into the core and presented in the literature (e.g., [33,38]) differ in appearance from modern TRISO fuel. In particular, the particles did not utilize distinct buffer and IPyC layers (see Fig. 3(d)).

with the first such spheres inserted into the core in 1969. While BISO fuel particles dominated the core inventory for roughly the first decade of operation, core reloads containing TRISO fuel began in earnest in the late 1970s, and this fuel type became the standard for the remainder of reactor operation into the late 1980s [44].

During the 1970s, TRISO coatings were becoming more widely used based on superior structural integrity and fission product retention, and a significant body of experimental data was becoming available on coating performance during irradiation. A special issue of the journal *Nuclear Technology* in 1977 included over 3 dozen articles focusing on the state of coated particle fuel development, including advances in fabrication technology, SiC and pyrocarbon coating properties, coated particle irradiation performance, fuel performance modeling, and fission product release observations [48]. Experimental results helped to elucidate important phenomena in the particles that could influence coating integrity and result in particle failure, such as kernel migration, pressure vessel failure, fission product attack of the SiC layer, and radiation-induced changes in the pyrocarbon. Improvements in particle design included: (1) a shift from pyrocarbon layers deposited at high temperatures (HTI, for high-temperature isotropic) to pyrocarbon deposited at lower temperatures (LTI, for low-temperature isotropic) that exhibits greater dimensional stability under irradiation and (2) kernel composition modifications to address thermochemically driven phenomena that could threaten coating integrity (discussed further in the next section).

The accumulating experimental database in this timeframe was revealing various aspects of fission product transport within fuel particles. This included the observation that silver can transport through intact TRISO coatings and result in relatively high release from the fuel [49,50], a phenomenon that has subsequently spurred a significant amount of research, in particular in the last several years [51–57]. Yet another important observation was the migration of fission product palladium from the kernel to the SiC layer, where reaction to form palladium silicides could threaten the structural integrity of the layer [58–61] (Fig. 4(a)).

The first three demonstration reactors were followed by two prototype HTGR power plants: Fort Saint Vrain (FSV) in the US (an 842 MWt prismatic core design) and the Thorium High-Temperature Reactor (THTR) in Germany (a 750 MWt pebble bed core design), both constructed and operated in the 1970s and 1980s [1]. THTR utilized $(Th,U)O_2$ BISO fuel throughout its lifetime, from first criticality in 1983 to shut down in 1988 [44]. FSV utilized HEU fissile $(Th,U)C_2$ and fertile (ThC_2) kernels with TRISO coatings. The reactor operated from 1976 to 1989. The fuel experienced temperatures exceeding 1300 °C, a maximum burnup of 16% FIMA, and maximum fast neutron fluence of 4×10^{25} n/m². The fuel performed better than expected; while some particle failures were detected based on fission gas levels in the coolant, they occurred at levels lower than predicted by fuel performance codes [64].

2.2. Kernel composition

Coated particle fuel allows a great degree of flexibility in kernel material, since the porous buffer layer mechanically decouples the kernel from the load-bearing, dense outer coating layers to a certain extent, thereby accommodating some degree of kernel swelling [32]. As a result, in addition to the evolution of the coating layers, a variety of fuel kernels have been employed throughout the development of coated particle fuel, including both high- and low-enriched uranium, fissile and fertile fuels, and both carbide and oxide phases. A number of considerations have influenced these variations, including fabrication concerns, kernel behavior under irradiation (including fission product retention capabilities), reactor design (including peak fuel temperatures and power

densities), and the desired type of fuel cycle. Virtually all of the early work in coated particle fuel development favored fertile (Th) fuel designs, and carbide kernels were originally used because of the high-temperature compatibility with the surrounding graphite fuel elements. However, these kernels were susceptible to hydrolysis during fabrication due to reaction with moisture. While a simple pyrocarbon layer was added to the kernels to stabilize them during fabrication, it was recognized that oxide kernels could simplify the overall fabrication process, as they are not susceptible to oxidation [32]. Oxide kernels also had the advantage of maintaining better chemical stability during the very high temperature heat treatment of spherical fuel pebbles during manufacture [32].

A challenge identified with oxide kernels, however, was the gradual increase in oxygen potential in the fuel due to uranium fission and the reaction with the surrounding carbon layers at high temperatures to form CO(g). The CO(g) can contribute to the overall gas pressure inside the particle (along with inert fission gases released from the kernel), increasing the stresses on the coating layers. One of first coated particle failure mechanisms identified was rupture of the coatings due to this internal pressure ("pressure-vessel" failure) [2], as demonstrated in Fig. 4(b). Another consequence of CO(g) buildup in the particle is the kernel migration phenomenon ("amoeba" effect) (Fig. 4(c)). This effect is dependent on the magnitude of the temperature gradient across the particle. Mass transport of carbon as CO(g) from the hot side of the particle to the cold side results in displacement of the kernel in the opposite direction [1,65,66]. If the process proceeds for sufficient duration that the kernel penetrates the IPyC layer on the hot side of the particle and comes into contact with the SiC layer, it can result in rapid degradation and ultimately failure [1,66]. Both of these effects are largely addressed in modern fuel by particle design (e.g., sufficient buffer volume to accommodate gas pressure or oxygen gettering) and operation of fuel within allowable limits (including burnup, temperature, and temperature gradients).

Early work in the US retained the carbide kernel design, because the large HTGR designs pursued at that time involved fuel conditions (including high peak fuel temperatures and high power densities) which would result in excessive kernel migration in oxide fuel. Elsewhere, fuel development in the UK and Germany adopted oxide fuel kernels in the 1960s and early 1970s, as the less severe thermal conditions in their reactor designs prevented detrimental levels of kernel migration.

By the mid-1970s, it was becoming increasingly clear that rare earth fission product elements migrating from carbide kernels during irradiation could chemically attack the SiC layer of TRISO particles and compromise the structural integrity [60,63,67,68] (Fig. 4(d)). Oxide kernels are much less susceptible to this phenomenon, because these rare earth elements tend to be strong oxide formers and therefore more stable in oxide kernels. The desire for a more chemically stable kernel that could operate beyond the CO-related limitations of UO_2 led to the suggestion of a kernel comprised of a mixture of both oxide and carbide phases. The approach was to include sufficient UC_2 to getter the excess oxygen produced by UO_2 fission, thereby avoiding formation of CO, while still taking advantage of the superior retention of the metal oxide form for most rare earths in the predominantly UO_2 kernel [63,69], with Eu and Sr—which form carbides in the UCO system—being notable exceptions. The strong thermochemical basis and favorable experience with the UCO kernels by the US program lead to the adoption of UCO fuel as the reference fissile particle design by the early 1980s [32], with an approximate UO_2/UC_2 mixture of 80%/20%. Other programs, including those based on pebble bed reactor designs and exemplified primarily by the work in Germany, maintained UO_2 as the preferred kernel type given that the fuel performance was acceptable because of the smaller
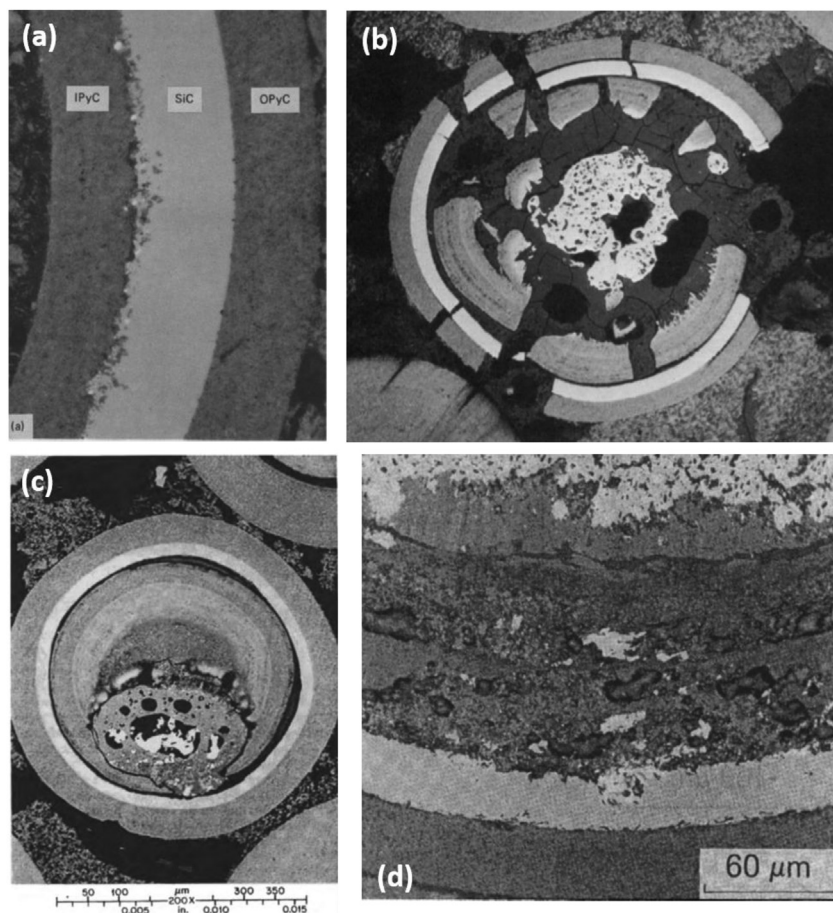
Fig. 4. Examples of particle failure mechanisms and coating corrosion. (a) Pd corrosion of SiC in UCO particle [61]. (b) Pressure vessel failure in UO$_2$ particle [62]. (c) Kernel migration in UO$_2$ [63]. (d) Predominantly rare-earth corrosion of SiC in UC$_2$ particle [60]). Images used by permission.

temperature gradients in the sphere. In either case, whereas the focus had traditionally been highly enriched uranium (HEU) fuel in the 1960s and 70s, this had shifted by around 1980 in both the US and Germany to low-enriched uranium (less than 20% ^{235}U) because of proliferation concerns, and LEU remains the standard today for reactor designs utilizing coated particle fuel [1,2].

2.3. Alternate particle designs

While SiC was one of the earliest refractory ceramics considered for the primary structural layer in coated particle fuel and remains today the de facto standard, other materials have been considered and developed. The most commonly studied alternate coating material has been ZrC, with experimental work dating to the first decade of TRISO fuel development and carried out since that time primarily in the US, Japan, and Russia [44,70]. ZrC has been pursued as a direct replacement for the SiC layer because of several potential benefits, including improved high-temperature performance (owning to the very high melting point of 3540 °C compared to the thermal decomposition of SiC at T > 1900 °C) and a greater resistance to chemical attack by fission product palladium. A detailed review of ZrC coating development is provided in an IAEA technical report [44]. Some experimental results have been obtained demonstrating improved high-temperature performance over conventional SiC-TRISO [71]. However, concerns persist over the oxidation resistance of ZrC compared to SiC and the potential for higher release of certain fission products [19], as well as changes in mechanical properties during high-temperature irradiation [72].

Additionally, ZrC is a more complex material to fabricate because of the relatively large range of Zr/C stoichiometries for the carbide phase. Consequently, stoichiometry and microstructure of ZrC are very dependent on processes conditions and will impact coating properties and performance [72], including fission product retention [73]. At this stage there is limited and insufficient irradiation data on CVD ZrC coated particles to adequately compare performance with conventional SiC [72,74].

Another notable use of nonfissile carbide phases (e.g., ZrC, SiC) in TRISO fuel is as a sacrificial, oxygen-gettering layer applied to an oxide kernel or as a separate phase within the kernel itself, with otherwise conventional buffer-IPyC-SiC-OPyC TRISO coatings applied on top [69,75,76]. Some UC$_2$-alternative getters like ZrC and SiC offer the advantage of allowing Eu and Sr to form oxides in the kernel [77]. However, UC$_2$ has the advantage of not diluting the actinide density. In all cases, the intent is to encourage reaction of the carbides with oxygen in the kernel and avoid the formation of CO, thereby eliminating one of the negative behaviors of UO$_2$ kernels. Irradiation and post-irradiation performance of UO$_2$ TRISO fuel with a ZrC coating on the kernel (with a thin pyrocarbon layer between the kernel and ZrC) was very promising in screening irradiations [75,78], however, the results did not become available until after UCO had been selected as the US reference fuel [79].

2.4. Irradiation testing and accident performance evaluations

Irradiation testing is an essential step in the evaluation of fuel performance, and in-pile experiments of coated particle fuel were

performed from the earliest stages of development. This included testing both in the first demonstration HTGRs as well as accelerated tests performed in MTRs. Testing in the prismatic reactors Dragon and Peach Bottom Unit 1 involved use of fuel test elements with experimental fuel varieties. In the pebble-bed AVR, reloads consisting of a few thousand to as many as ~25,000 pebbles were inserted into the core periodically. While irradiation in HTGRs offers the advantage of a prototypical neutron spectrum and a realistic accumulation of burnup and fast fluence over time (avoiding potential artifacts that can result from an excessive degree of acceleration [80]), irradiation in MTRs offers the advantages of independent control and monitoring of the test fuel in dedicated capsules and completion of experiments more rapidly, decreasing the overall time for the performance evaluation and design improvement cycle. A large number of irradiations have been performed on coated particle fuels in many countries and reported in the scientific literature. A 1968 article indicates that "well over 100" irradiation tests had been conducted on coated particle fuel in the US at that time [37]. A more recent compilation lists 61 US and 22 German irradiation capsules containing HEU fuel (many of these taking place after 1970), and an additional 9 US and 15 German irradiations after the switch to LEU fuel [81]. Additional irradiation tests on coated particle fuel have been performed in Japan, Russia, and France [44,81].

A key metric in evaluating coated particle fuel irradiation performance is the level of coating failure induced from various causes. In high-quality fuel operated within its design performance envelope, these failures occur in relatively low fractions, with the acceptable failure fraction dictated by reactor design and safety analyses. For example, the design specification for the allowable in-service particle failure fraction may be on the order of 2×10^{-4} for modular HTGR designs. Actual observed failure rates during irradiation testing can be significantly less, however. Therefore, it is impractical to quantify failures based on post-irradiation examination of the fuel.

As a result, particle failures are typically evaluated by monitoring radioactive fission gas release from the fuel during irradiation, which is a function of in-service particle failures, initially defective particles with exposed kernels, and uranium contamination residing outside of the retentive coatings in the fuel compact or sphere. The release-rate-to-birth-rate ratio (or R/B) is determined by comparing the measured release of short-lived isotopes of Kr and Xe to the inventory generation rate. Historically, measured R/B values decreased as particle fuel development progressed and improvements were seen in as-fabricated fuel quality and in-pile performance. As examples, ^{85m}Kr R/B values in the Peach Bottom coolant and purge gas streams during operation with the second core BISO fuel were between 10^{-4} and 10^{-3}, whereas values for FSV TRISO fuel were approximately $0.3–1.3 \times 10^{-5}$ and high-quality LEU UO$_2$ TRISO fuel tested in the German program routinely demonstrated ^{85m}Kr R/B values in the range of 10^{-9} to 10^{-6} in MTR irradiations [44].

Post-irradiation examination (PIE) and accident simulation testing constitute another important evaluation of fuel performance. PIE allows measurement of metallic fission product release (e.g., ^{110m}Ag, ^{137}Cs, ^{154}Eu, and ^{90}Sr) and observation of the state of fuel kernels and coatings. High-temperature testing to assess particle integrity and the fission product retention capabilities of the fuel under accident conditions has been performed in dry helium (to simulate conditions in the core during a depressurized loss of forced cooling) and in atmospheres containing air or moisture (to simulate conditions during ingress of these oxidants into the core). Testing of this nature has been undertaken in a number of countries, including Germany, the US, and Japan [2,21,44,82]. Some of the key isotopes evaluated in these types of tests and their

significance for evaluating fuel performance are listed below:

- ^{85}Kr: Long-lived Kr isotope that serves as an indicator of exposed kernels (either from pre-existing exposed kernels or failure of all TRISO layers during the test), as release tends to be very low in their absence.
- ^{137}Cs, ^{134}Cs: Main indicators of a loss of integrity in the SiC layer, since LTI pyrocarbon is not retentive of cesium.
- ^{90}Sr, ^{154}Eu: Generally well-retained inside of intact TRISO particles, but greater diffusive release through the coating layers occurs at higher temperatures. Once outside of particles, exhibits significant retention within the fuel matrix, with release increasing with temperature.

2.5. Experience with LEU TRISO fuel from the 1980s onward

2.5.1. Germany and EU

The TRISO particle design in Germany continued to evolve in the 1970s, with one of the notable progressions being a switch to the use of LTI pyrocarbon coating by the end of the decade [1]. Fuel development in the 1980s demonstrated the high as-manufactured fuel quality and excellent in-pile performance that can be accomplished with LEU UO$_2$ fuel [23–25]. Efforts involved refinement of fuel fabrication and quality control capabilities, irradiation testing of fuel spheres both in MTRs and in AVR, and post-irradiation examination and heating tests to assess performance in-pile and under accident conditions. The results demonstrated low as-manufactured particle defect fractions and low particle failure fractions during irradiation and during post-irradiation heating tests at postulated accident temperatures. The nominal dimensions of the reference particle design that resulted from this work are shown in Table 1 [23]. These basic coating dimensions have been adopted by a number of other programs (either with UO$_2$ or UCO kernels).

This effort culminated in the large-scale fabrication campaign of the so-called GLE-4/2 fuel for AVR (16.8% ^{235}U) and the small-scale fabrication of the proof test fuel for the HTR-Modul 200 MWt modular reactor design (10.6% ^{235}U), both with very low defective particle fractions ($\leq 2.0 \times 10^{-6}$ and $\leq 5.3 \times 10^{-6}$, respectively, representing the upper bound at 95% confidence) [23]. Fuel fabrication efforts ceased in 1988 concurrent with the shutdown of THTR, but irradiation testing in MTRs continued through 1994, finishing with the proof test fuel irradiations (designated HFR-K5 and HFR-K6) in HFR-Petten [23]. The two final proof test irradiations involved a total of 8 spheres at temperatures 800–1140 °C and burnup of 8–11% FIMA. Cumulative fission gas release (^{85m}Kr R/B ratios were $1.7–9.0 \times 10^{-7}$) was attributed to uranium contamination and as-manufactured particle defects (one defect in one pebble and two in another) with no radiation-induced particle failure, giving a calculated particle failure fraction of $\leq 2.6 \times 10^{-5}$ (upper bound at 95% confidence). Taken as a whole, German irradiation testing of 60-mm-diameter spherical fuel elements in MTRs (totaling

Table 1
Nominal dimensions of the German reference LEU UO$_2$ TRISO particle design.

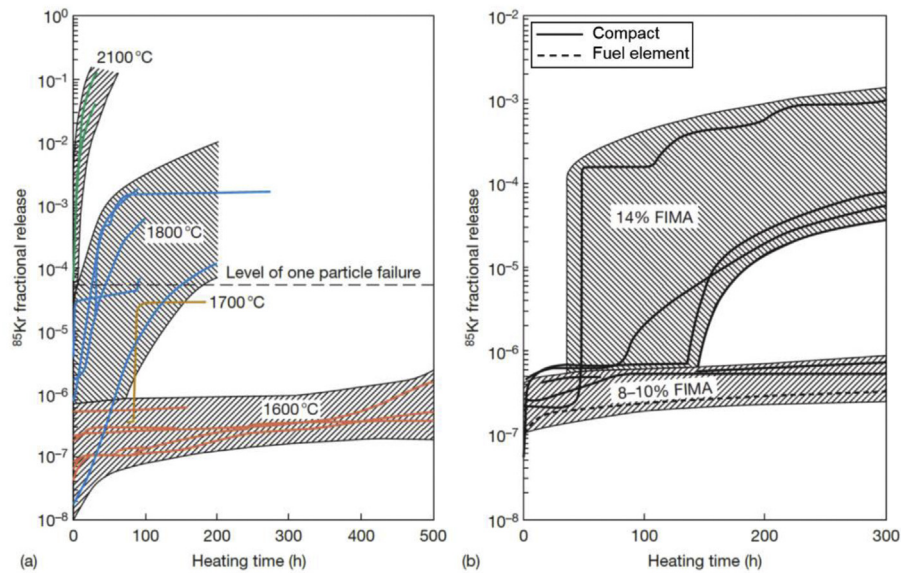| Component	Dimension
Kernel diameter	500 μm
Coating thickness	
Buffer	95 μm
IPyC	40 μm
SiC	35 μm
OPyC	40 μm

Fig. 5. Summary of ^{85}Kr release fractions during post-irradiation heating tests from the German program (adapted from Ref. 21).

approximately 277,000 particles) was determined to have produced no particle failures [23], giving a failure fraction of $\leq 1.1 \times 10^{-5}$ at 95% confidence.

German fuel elements (including both standard spherical fuel elements as well as smaller cylindrical compacts containing ~1,600 particles in a central spherical fueled zone) with burnup $\leq 11\%$ FIMA also exhibited no failures during 1600 °C isothermal accident tests in dry helium (based on ^{85}Kr release fractions $\leq 2 \times 10^{-6}$),[2] and cesium release fractions were below $\leq 1 \times 10^{-4}$, indicating intact, retentive SiC layers. However, at reported burnups $\geq 14\%$ FIMA[3] or temperatures ≥ 1700 °C during post-irradiation heating tests, particle failures began to manifest as higher ^{85}Kr releases (see Fig. 5). Cesium release also increased at the higher temperatures, reaching ~10^{-2} to 10^{-1} from fuel spheres at 1800 °C (burnup <11% FIMA) and >10^{-1} for fuel compacts (burnup 12% FIMA) [28]. Based on these results, it has been suggested that if the fuel is pushed to a burnup of ~15% FIMA, accident temperatures should be limited to 1600 °C, but for fuel with peak burnup of 11% FIMA the allowable accident temperature limit may be higher than 1600 °C [23]. Additional post-irradiation heating under oxidizing conditions, performed on a more limited scale, demonstrated that additional particle failure can occur after prolonged exposure (several hundred hours) in air above 1300 °C, and 800 °C exposure to steam can result in increased release of fission gas from exposed kernels. For additional reading on the results of the German irradiation and safety-testing program, the reader is referred to References 17, 22—29.

Additional irradiation testing of German TRISO fuel was performed from 2004 to 2010 in HFR-Petten using previously manufactured fuel spheres of the GLE-4/2 type and sponsored by the European Commission, with the intent of demonstrating the UO$_2$ fuel performance at temperatures and burnup beyond the conventional fuel performance envelope for modular pebble-bed HTGRs [86—88]. Burnups achieved in these irradiations were

approximately 11% and 14% FIMA in the EU1bis and EU1 irradiations, respectively (both falling somewhat short of the originally targeted values [89]), and some post-irradiation examination results from the EU1bis experiment have been reported [90]. While the AVR spheres in the EU1 irradiation (sphere surface temperatures reported to be 950 °C) exhibited relatively low fission gas R/B ratios indicating no failed particles [87], the higher-temperature EU1bis irradiation (sphere center temperatures were reportedly maintained at 1250 °C [91]) had ^{85m}Kr R/B of 4×10^{-6}, suggesting that some particle failure occurred [88]. Post-irradiation heating of several spheres from these irradiations resulted in low release of ^{85}Kr, indicating no full TRISO coating failures. However, ^{134}Cs release at 1600 °C reached $1-2.5 \times 10^{-3}$ for EU1bis spheres[4] and 6×10^{-4} for an EU1 sphere [83,93,94], all of which are significantly higher than observed in historic tests of German LEU UO$_2$ TRISO fuel and indicate release through the SiC layer of the particles, suggesting the onset of degradation and/or layer failure. It therefore appears that these irradiation tests may have challenged an upper limit for acceptable performance for LEU UO$_2$ fuel.

In a compilation of German irradiation and accident testing data, Kania et al. [23] have summarized the performance of spherical fuel elements during 1600 °C isothermal heating tests and transient-temperature tests that simulate the time-varying peak fuel temperature in the reactor during a depressurized loss of coolant flow accident with a maximum temperature of 1620 °C. This includes spheres irradiated in AVR as well as fuel from proof test irradiations and the more recent EU irradiations. Based on a total of 5 observed failures out of 287,480 particles tested, the reported upper bound for the failure fraction at 95% confidence is $\leq 3.7 \times 10^{-5}$.

2.5.2. United States

Even while the focus in the US was shifting to UCO TRISO fuel, coated particle fuel development in the 1970s and into the 1980s still involved a wide variety of different kernel types (including UCO, UO$_2$, UC$_2$, ThO$_2$, ThC$_2$, and (Th,U)O$_2$), uranium enrichments,

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[2] Note that more recent heating tests on proof test spheres from the HFR-K5 and -K6 spheres has resulted in somewhat higher ^{85}Kr release fractions [83,84], although still falling below 1×10^{-5}, indicating no particle failures.

[3] Several methods were used to empirically measure the burnup of the fuel compacts; the reported values are the highest among the various methods, indicating the possibility that burnup could be overestimated by ~10–20% [85].

[4] Subsequent analysis of the testing system by Seeger et al. [92] indicates that the temperature of some of the EU1bis tests was most likely lower than originally reported by approximately 100 °C.

and coating designs, and supported a two-particle, fissile-fertile approach [1,80]. A large number of test irradiations were performed in this timeframe. Development efforts culminated in the fuel fabricated and irradiated for the Modular High Temperature Gas Cooled Reactor (MHTGR) and New Production Reactor (NPR) programs in the early 1990s, which included additional pyrocarbon "seal" coating layers designed to serve a number of postulated functions (so-called "TRISO-P" fuel) [95]. Fuel performance results from these tests were overall much less satisfactory than comparable results from the German program [80], and as a consequence the US TRISO fuel development effort was halted in 1993.

Extensive subsequent analyses of these fabrication and irradiation efforts were undertaken in 1993 and again in the early 2000s, along with a comparison to the German fuel fabrication and irradiation experience [80,96], in an effort to understand the causes of the poor performance and the relationship between key particle characteristics and in-pile performance. The results of these analyses suggested a number of causes for the poor performance related to fuel manufacture and the manner in which irradiation tests were conducted. Particularly highlighted were the poor properties of the pyrocarbon layers and the use of the additional pyrocarbon layers in the TRISO-P fuel, which were deemed unnecessary and ultimately detrimental to particle performance. The injection-molding process used to make compacts was also suspected to have led to high particle defect fractions and considered less favorable to the German-adopted overcoating method. In 2002 a new TRISO fuel development program was initiated by the US Department of Energy (DOE) that focused on LEU UCO TRISO particles manufactured with coating processes based on the historic German fuel experience and with similar coating and matrix properties [97]. The UCO kernel remained the primary choice because of the continued focus on prismatic reactor designs and the desire to take advantage of UCO performance capabilities at high burnup [98]. This program is continuing today, with the fourth and final fuel irradiation test currently in progress. Fuel performance to date has been very good to a maximum, compact-average burnup of nearly 20% FIMA. Current progress in this program is described further in Section 3.2.

2.5.3. China

Beginning in the early 1970s and into the 1980s, HTGR fuel development in China was focused on the basic design and essential research for kernels and coatings. During this first stage of development, fuel examination methods were established, and various techniques were implemented at the laboratory scale. Several fuel configurations, including the spherical and block type fuel elements and both BISO and TRISO-coated particles, as well as the related manufacturing technology were studied at the experimental scale.

The second stage of effort consisted of development of the HTR-10 experimental reactor (a 10MWt pebble bed design) and its fuel (LEU TRISO particles in spherical fuel elements) from 1983 to 2003. The coated particles consist of nominally 500-μm-diameter UO_2 kernels (17.1% ^{235}U) with coating dimensions roughly similar to those of German LEU TRISO fuel (Table 1) [99]. The 60-mm-diameter fuel spheres contain approximately 8,300 particles. From 1995, equipment from the NUKEM plant in Germany (including equipment for kernel, coated particle, and spherical fuel element fabrication, as well as fuel element characterization) was installed in the labs at the Institute of Nuclear and New Energy Technology (INET). Some components and instruments for fuel manufacture and quality control were supplemented by INET.

In this period, an integral experimental fabrication line was established in the lab. The experimental work and characterization of as-fabricated fuel properties in production scale were completed before the end of 1998. The fabrication of the first loading fuel for the HTR-10 started in February 2000, and a total of 20,541 spherical fuel elements were produced. HTR-10 achieved first criticality in December 2000.

Two irradiation tests of HTR-10 fuel spheres in MTRs have been performed. The first was a qualification test for the HTR-10 first loading in which four spherical fuel elements were selected at random from the first and second production batches and irradiated in the Russian IVV-2M reactor [100,101]. The irradiation test started in July 2000 and ended in February 2003. The sphere center irradiation temperature was kept at $1000 \pm 50\,°C$ and the burnup of fuel elements SFE 5, SFE 12 and SFE 7 reached 11.9, 13.1, and 11.6% FIMA, respectively. The fast neutron fluence reached 1.10×10^{25}, 1.31×10^{25} and 1.06×10^{25} n/m^2 (E > 0.1 MeV), respectively. During the irradiation test, the temperature of element SFE 12 was increased to $1200\,°C$ (i.e. the maximum fuel temperature under HTR-10 accidents) for 200 h and subsequently to $1250\,°C$ for 200 h, when its burnup reached 4.7 and 7% FIMA, respectively. Due to experimental complications in Capsule 4 containing element SFE 8, fission gas release rates were only recorded up to 4.5% FIMA. However, the operation of Capsule 4 continued without fission gas measurement. The final burnup and fast neutron fluence of element SFE 8 reached 12.5% FIMA and 1.30×10^{25} n/m,^{2} respectively. The R/B ratio did not increase significantly during irradiation, with end-of-irradiation values in three capsules ranging from 10^{-6} to 8×10^{-5}. However, an in-pile heating test of element SFE 7 performed at the end of the irradiation led to a failure of approximately 6% of the coated particles. After the test it was estimated that the fuel temperature had very likely been much higher than the intended $1600\,°C$.

The second irradiation test of HTR-10 fuel was performed in HFR Petten as part of the HFR-EU1 experiment from September 2006 to February 2010. The experiment consisted of two INET spheres randomly taken from the latest production batch and three German AVR pebbles. The objective of the HFR-EU1 experiment was to irradiate the fuel pebbles to burnup exceeding the specifications of current HTR designs with pebble bed cores (e.g., HTR-Modul, HTR-10, HTR-PM and PMBR) to demonstrate that pebble bed HTR are capable of enhanced performance in terms of sustainability (further increased power conversion efficiency, and improved fuel use) and thus reduced waste production. The original target peak burnup for the INET pebbles was 17% FIMA [89].

The experiment was performed in two campaigns from September 2006 to February 2008 (12 reactor cycles of 28 days) and continued from October 2009 to February 2010 (4 reactor cycles). Up to the end of irradiation, the experiment had accumulated 16 reactor cycles totaling 445 EFPD. The calculated burnup was 9.3% FIMA (pebble INET 1) and 11.6% FIMA (pebble INET 2) and the maximum fast neutron fluence (E > 0.1 MeV) was about 4.95×10^{25} n/m^2. The surface temperature of pebble INET 2 during irradiation was approximately $940\,°C$ [99]. The ^{85m}Kr R/B ratio for the INET capsule was approximately 8×10^{-8}. The second irradiation test results are much better than the first. Based on the Booth Model and assuming a capsule-average temperate of $900\,°C$, the calculated ^{85m}Kr release fraction from a single coated particle would be 3.26×10^{-3}, and from a single failed particle in the capsule with two INET fuel spheres (~16,600 particles) 1.96×10^{-7}, which is higher than the observed R/B. This suggests that no complete particle failure occurred during the irradiation and that the measured fission gas release originates from uranium and thorium impurities in the matrix graphite of the pebbles and in the graphite cups used to hold the pebbles in place.

Starting in 2004, work began on development of fuel for the HTR-PM reactor and continues to the present time. The current progress is summarized in Section 3.1.

2.5.4. Japan

Research on HTGRs in Japan began in the late 1960s, with early fuel development activities focused on fuel fabrication and characterization capabilities, irradiation testing, and high-temperature accident simulation tests [102,103]. This led in the late 1980s to a focus on developing the High Temperature Test Reactor (HTTR), with construction initiated in 1991 [102,104]. HTTR is a 30MWt prismatic-core research reactor that reached full power in 2000 [105]. The fuel consists of LEU UO_2 TRISO particles (multiple ^{235}U enrichments between 3.4 and 9.9% are used; the average is ~6%) formed into hollow cylindrical (annular) fuel compacts and placed in graphite sleeves which in turn are inserted into graphite blocks [106]. Irradiation performance of the HTTR fuel, which has a relatively low target peak burnup ($\leq$3.6% FIMA) and a relatively high maximum fuel temperature ($\leq$1495 °C) [107], has been very good based on fission gas release rates, both during operation in HTTR and in accelerated irradiation testing in an MTR up to 6% FIMA and 1200 °C [105].

A modified coated particle design was developed to allow burnups of approximately 10% FIMA in HTTR. Preliminary testing in MTRs at ~7−9% FIMA indicated good performance, albeit with several particle failures that were postulated to be due to as-fabricated SiC defects based on fuel performance models [108]. This fuel notably involved a change in the specification for kernel diameter (from 600 to ~550 μm), buffer layer thickness (from 60 to 90 μm), and SiC layer thickness (from 25 to 35 μm) compared to the initial HTTR fuel particles. Based on these results, additional modifications in the particle design were implemented with a specified kernel diameter of 500 μm (9.9% ^{235}U enrichment) and buffer thickness of 95 μm [102], such that it closely resembled the standard TRISO particle design from the German program. This fuel was recently irradiated to >9% FIMA in the WWR-K reactor (Kazakhstan). A few exposed kernels were observed based on R/B data, but these were again postulated to be related to particles with as-fabricated SiC defects [109], suggesting that no in-pile particle failures occurred.

Research and development has also been performed in Japan dating back to the early 1970s on TRISO fuel with ZrC replacing the SiC layer, in order to improve upon the high-temperature stability and provide a reduced tendency to react with fission product palladium [102,110], although such particles are not considered sufficiently mature for deployment.

2.5.5. Others

Other countries that have been involved in coated particle fuel development include South Africa, France, Russia, and the Republic of Korea [1,18,19,44]. Between the mid-1990s and 2010, South Africa pursued development of HTGR and TRISO technology as part of the Pebble Bed Modular Reactor (PBMR) project [1,111], with LEU UO_2 TRISO particles and fuel elements similar to German design. This involved fuel fabrication and QC capability development based on historic German experience, and irradiation testing of TRISO particles in the US AGR-2 irradiation experiment in the Advanced Test Reactor as a collaboration initiated under the Generation IV International Forum [112]. The PBMR project was terminated in 2010, however, due primarily to economic factors [113].

2.6. Current progress on TRISO fuel development and qualification

2.6.1. China

The high-temperature gas-cooled reactor pebble-bed module (HTR-PM) demonstration power plant is a small modular nuclear reactor under development in China. It is a Generation IV reactor partly based on the earlier HTR-10 prototype reactor. Two separate reactors with 250 MWth output are connected to a single steam turbine to generate 210 MW of electricity. The HTR-PM is expected to be the first Generation IV reactor to enter operation. Began in December 2012 at the Shidao Bay Nuclear Power Plant, the pressure vessels of the two reactors were installed in 2016. Installation of the graphite block, control rod and all the equipment in one of the reactors was completed 2017. The sphere fuel handling system installation was completed in 2018 and the first loading of fuel into the reactors is expected to start in 2019.

HTR-PM fuel development was conducted at INET starting around 2004 and has been based closely on the development of HTR-10 fuel technology. The manufacturing technology and facilities were enhanced to the industrial scale and a demonstration line was established with the capability to produce 100,000 pebbles per year. The HTR-PM fuel uses the same TRISO-coated particle design as HTR-10, but the U loading increased from 5 to 7 g per pebble, corresponding to an increase in particles from 8,000 to 12,000 per pebble. At the same time, the free uranium fraction in the pebbles decreased from 5×10^{-4} to 6×10^{-5}. After establishing the technology, a batch of spheres was fabricated, with several being selected at random for an irradiation qualification test in HFR Petten.

Irradiation testing of five HTR-PM fuel spheres was performed in HFR Petten [114]. The irradiation was designed so that the upper four pebbles would reach a burnup higher than 12.3% FIMA with center temperatures of 1050 ± 50 °C. The irradiation lasted 355 full power irradiation days and was completed in December 2014. Based on neutronics calculations, the total fast fluence levels were between 3.79 and 4.95×10^{25} n/m² (E > 0.1 MeV). Burn-up estimates are 11.1% FIMA for Pebble 5 and between 12.6 and 13.7% FIMA for the other pebbles. The central temperatures remained in the target boundaries of 1050 ± 50 °C. The five spheres were irradiated in a single test rig, with online fission gas measurement capturing the cumulative release from all five spheres. R/B ratios were very low throughout the irradiation, as shown in Fig. 6. The calculated R/B from a single failed particle (out of 60,000 particles) at 1050 °C is 1.1×10^{-7} for ^{85m}Kr. Measured ^{85m}Kr R/B values during the last cycle were between 2.4 and 3.3×10^{-9}, indicating zero manufacturing defects and no in-pile particle failure.

PIE of the HTR-PM irradiated fuel specimens has been performed at Petten and Karlsruhe separately. The initial PIE at Petten shows that the dimensional shrinkage in all 5 pebbles is between 0.88% and 1.25%, and further PIE has been performed at the European Commission Joint Research Center in Karlsruhe, including simulated accident temperature heating tests, the deconsolidation of a fuel sphere, and microstructural examination of the fuel. There the irradiated fuel pebbles have been exposed to heating tests in pure helium in the KÜFA facility that simulates high-temperature accident conditions in the reactor [115]. The final PIE results will be reported at a later date.

Following production of the qualification fuel, the fabrication technology and facilities were transferred to China North Nuclear Fuel Company (CNNFC) where the HTR-PM fuel manufacturing line was designed and constructed in Baotou, Inner Mongolia, China. The fuel line was designed with the capability of 300,000 sphere fuels per year, and the goals for the fuel plant include expanding overall fabrication capacity and stabilizing the mass-production process to provide consistency in the fabricated fuel. The plant will provide fuel for the initial HTR-PM first-core loading and produce sufficient elements for the operation of the demonstration plant, using similar technology, materials, and fabrication principles as those established at INET. The first pebbles were produced on this fabrication line in 2016, and more than 500,000 fuel pebbles for the HTR-PM first core loading have been produced to date.
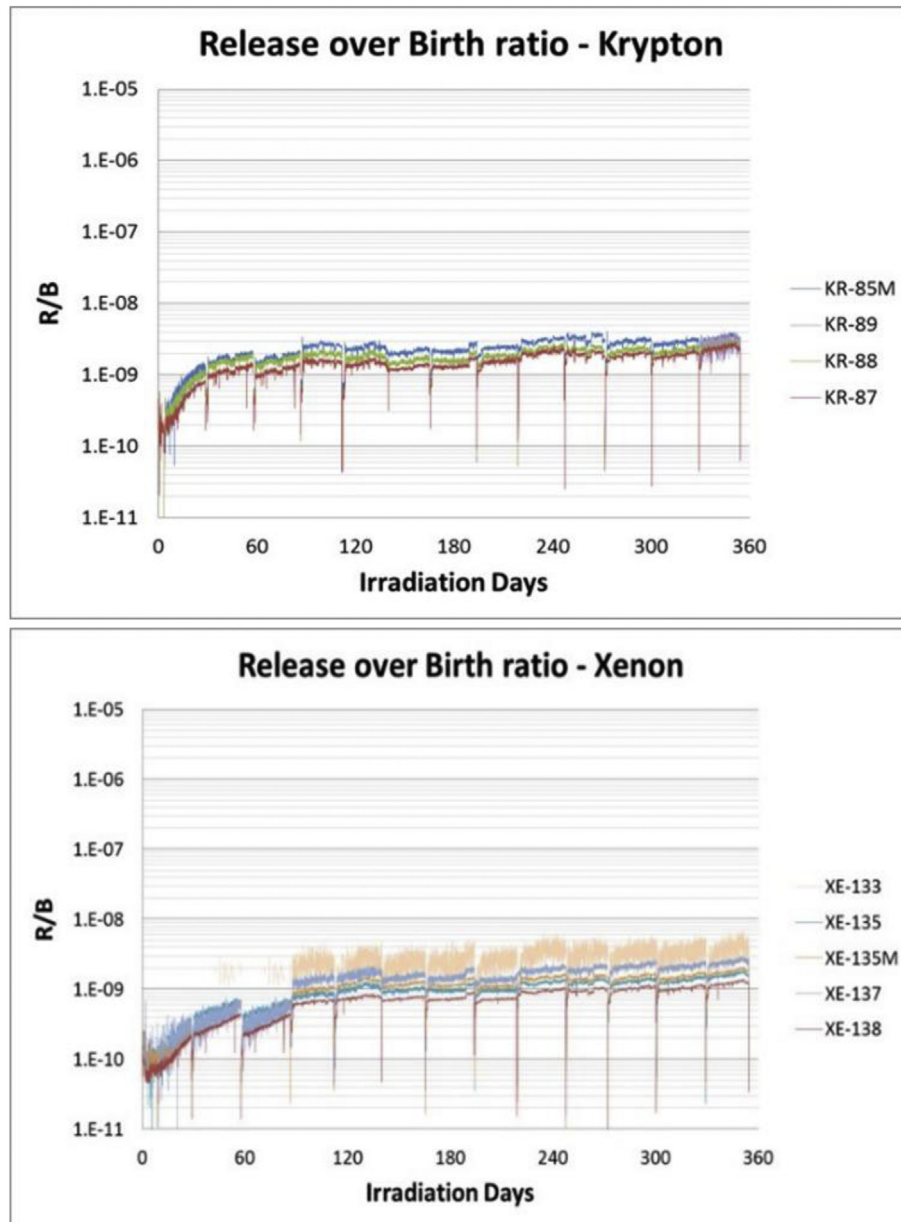
Fig. 6. R/B ratio of Kr and Xe isotopes in the HTR-PM fuel irradiation in Petten.

2.7. US

2.7.1. Fuel program description

The US DOE Advanced Gas Reactor (AGR) Fuel Development and Qualification program was initiated in 2002. The objective is to generate fuel qualification data to support licensing of an HTGR through fabrication, irradiation, post-irradiation examination, and accident safety testing of high-quality, TRISO-coated fuel. While both UO$_2$ and UCO fuel have been fabricated and irradiated, the program has focused primarily on UCO fuel to take advantage of performance benefits at high operating temperatures and burnups as high as 20% FIMA. Four irradiation experiments have been initiated in the program. Three of these (AGR-1, AGR-2, and AGR-5/6/7) involved intact TRISO fuel particles in cylindrical fuel compacts to assess fuel performance. The fourth (AGR-3/4) was an irradiation dedicated to evaluating fission product transport in fuel matrix and core graphite materials, involved both intact driver fuel and a 1%

fraction of designed-to-fail fuel [116], and has been discussed recently by Collin et al. [117].

The UCO kernel diameter and ^{235}U enrichment has changed slightly as the program has progressed. Table 2 presents summary data on the fuel particles and compacts used in the three fuel performance irradiations. Nominal coating dimensions for all particles produced in the program are very similar to those used in the German program (Table 1), with the exception of a target buffer thickness of 100 μm. For all three irradiations, the particles were formed into right cylindrical compacts with nominal dimensions of approximately 25.0 mm long × 12.3 mm diameter. Particle packing fraction for the UCO compacts has varied between 25 and 38%. While most of the fuel irradiated has contained UCO particles, the AGR-2 irradiation experiment included a capsule with twelve compacts containing UO$_2$ particles.

The AGR-1 experiment contained coated particles and compacts produced at the laboratory scale at Oak Ridge National Laboratory

Table 2
US AGR program fuel parameters.

Experiment	Kernel (diameter, μm)	Enrichment (% ^{235}U)	Particles per compact (packing fraction, %)	gU per compact
AGR-1	UCO (350)	19.7	4140 (37)	0.91
AGR-2	UCO (427)	14.0	3180 (37)	1.26
	UO$_2$ (508)	9.6	1540 (23)	0.99
AGR-5/6/7 [a]	UCO (426)	15.5	2240 (25)	0.89
			3430 (38)	1.36

[a] AGR-5/6 irradiation capsules contained both 25 and 38% packing fraction compacts; AGR-7 capsule consisted entirely of 25% packing fraction compacts.

(ORNL) using a 50-mm coating furnace [14,118]. The objective was to confirm performance of the UCO fuel particle and to act as a shakedown test for the irradiation methods. The AGR-2 coated particles were produced at BWX Technologies (BWXT) using a 150-mm furnace and the particles were compacted on laboratory scale equipment at ORNL [8,118,119]. The experiment objectives were to confirm the performance of the particles fabricated on an industrial-scale coater, compare the performance of UCO and UO$_2$ fuel, and to assess the performance of UCO fuel at temperatures significantly higher than those expected during normal operation. The AGR-5/6/7 fuel particles were coated at BWXT using the 150-mm coater and the compacts were fabricated at BWXT using a pilot-scale compacting process. Two different particle packing fractions were used in the experiment (25 and 38%) to accommodate the different power density and temperature requirements in the various irradiation capsules.

The irradiations have all been performed or are in progress (AGR-5/6/7) in the Advanced Test Reactor at Idaho National Laboratory and have consisted of multiple capsules with independent temperature control and sweep gas monitoring (with associated fission gas R/B measurements). Table 3 provides a summary of the fuel used and the irradiation conditions for each experiment. The AGR-2 irradiation included one capsule (Capsule 2) containing UCO fuel that was operated with higher fuel temperatures than expected during normal operation of a modular prismatic HTGR (fuel compact time-average maximum temperatures ≤1360 °C). The AGR-1 and AGR-2 irradiations were completed in 2010 and 2013, respectively.

AGR-5/6/7 consists of five separate capsules to achieve two main test objectives in a single irradiation test train [120]. Four capsules comprise the AGR-5/6 portion of the irradiation test train, which constitutes the formal fuel qualification test for the UCO fuel. A key requirement is to irradiate at fuel temperatures that represent a prototypical distribution that would be experienced by the fuel in a prismatic, modular HTGR core (the temperatures provided in Table 3 represent the approximate distributions expected based on established experiment objectives) and to accomplish this with a statistically significant number of particles. The fifth capsule comprises the AGR-7 experiment, which is a fuel performance margin test to evaluate fuel performance under conditions much more severe than those expected in a modular HTGR (and also more severe than those experienced in AGR-2 Capsule 2). The AGR-7 goals are to irradiate the fuel to time-average peak temperatures as high as 1500 °C and at peak burnup of approximately 18% FIMA. The irradiation began in February 2018 and is expected to complete in 2021.

Post-irradiation examination of the fuel and irradiation capsules is focused on quantifying the level of fission product release and observing radiation-induced changes to the kernels and coatings. This includes analysis of fission product inventory in the coated particles, in the fuel compact matrix, and in the capsule components, as well as microstructural analysis of compacts and particles. Post-irradiation, high-temperature safety testing of the fuel is also being performed to assess fuel behavior during accidents. The PIE and safety testing for the AGR-1 experiment is complete [121,122], while work is still in progress for the AGR-2 experiment [123].

2.7.2. Fuel irradiation performance

Fuel performance data has been collected from the AGR-1 and AGR-2 irradiations and subsequent PIE and safety testing. AGR-1 ^{85m}Kr R/B values for the six individual capsules were between 2×10^{-8} and 2×10^{-7} at the end of irradiation, indicating no particle failures, with a peak burnup of 19.6% FIMA. This corresponds to a calculated in-pile particle failure fraction of $\leq 1.1 \times 10^{-5}$ at 95% confidence. Interpretation of the fission gas R/B data from the AGR-2 experiment was significantly complicated by experimental issues during the irradiation, including unintentional mixing of gas effluents from various capsules (a result of suspected gas-line

Table 3
US AGR program fuel performance irradiations.

Experiment	Fuel type	Compacts/particles	Irradiation conditions (averaged over each compact)			
			TAVA Temp. [a] (°C)	TA Peak Temp. [b] (°C)	Burnup (%FIMA)	Fast fluence [c] (10^{25} n/m^2)
AGR-1 [d]	UCO	72/298,000	955–1136	1069–1197	11.3–19.6	2.2–4.3
AGR-2 [d]	UCO	36/114,000	987–1296	1080–1360	7.3–13.2	1.9–3.5
	UO$_2$	12/18,500	996–1062	1072–1105	9.0–10.7	3.1–3.5
AGR-5/6	UCO	170/516,000	Time-average temperature range [e]		7.4–18.6 [f]	2.2–7.2 [f]
			600 ≤ T < 900 (30%)			
			900 ≤ T < 1050 (30%)			
			1050 ≤ T < 1250 (30%)			
			1250 ≤ T < 1400 (10%)			
AGR-7	UCO	24/54,000	TA Max 1500 ± 50		17.4–18.2 [f]	6.8–7.4 [f]

[a] Time-average, volume-average temperature.
[b] Time-average peak temperature.
[c] Fast fluence is provided for E > 0.18 MeV.
[d] AGR-1 and AGR-2 temperature ranges are based on calculated individual compact temperatures.
[e] AGR-5/6 and AGR-7 temperature ranges are approximate and based upon the irradiation test goals. Numbers in parentheses are the percentage of particles experiencing this range of time-average temperature.
[f] Burnup and fluence values given for the AGR-5/6 and AGR-7 capsules are predictions based on pre-test physics calculations.
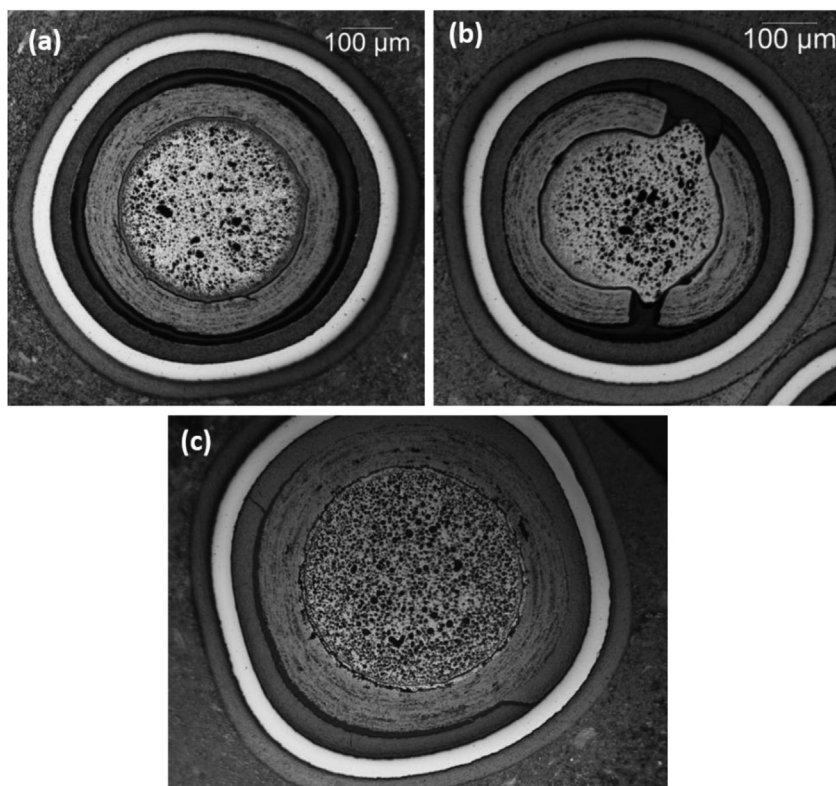
Fig. 7. Irradiated UCO particles from the AGR-1 experiment. Particle taken from compacts with an average burnup of 13.7% FIMA ((a) and (b)) and 15.7% FIMA (c).

ruptures due to experiment handling), such that definitive conclusions about particle failures during the latter portion of the experiment cannot be made [124]. The ^{85m}Kr R/B values at the beginning of the irradiation were in the range 10^{-6} for the three capsules containing UCO fuel and 10^{-7} for the US UO$_2$ fuel. Ongoing PIE of the fuel is expected to shed additional light on the possibility of any in-pile particle failures.

Analysis of irradiated AGR-1 and AGR-2 particle cross-sections has revealed that the dominant coating morphology was IPyC and buffer layer separation as the buffer densified, with all coating layers remaining intact, along with noticeable void formation inside the UCO kernels (Fig. 7(a)). Buffer fracture was not uncommon, but the frequency varied considerably depending on irradiation temperature and fast fluence [125,126], and the UCO kernels tended to expand into the gap formed as a result of volume expansion (Fig. 7(b)). However, this appeared to have no detrimental effect on the outer dense coating layers.

While no in-pile TRISO failures[5] occurred during the AGR-1 experiment, a limited number of particles experienced failure of the SiC layer while the OPyC remained intact (four particles from a total of 298,000 are believed to have experienced this failure mode). These particles retained fission gases but exhibited elevated cesium release. Observation of such particles and investigation of the causes of coating failure is improbable when relying on microscopy of random particle cross sections because of the relatively low rate of occurrence and the limited likelihood of observing specific regions where localized coating degradation and failure

occurred in the plane revealed in cross section. Several advances in coated particle post-irradiation examination that were implemented in the AGR program have enabled a focused study of these particles to better understand coating failures. This involves identifying specific compacts that may contain one or more particles with failed SiC, and then gamma counting all of the particles to locate the specific particle(s) with failed SiC based on their diminished cesium inventory. This is then followed by nondestructive, X-ray examination of the interior coating microstructure of the particle to isolate the region related to the SiC failure, and finally mounting the particle and employing traditional metallography techniques to examine the degraded region in detail [127].

The results revealed that the dominant cause of SiC failure in these particles was fission product (primarily Pd) corrosion. However, a precursor for this attack was mechanical fracturing of the IPyC layer, allowing concentration of fission products at the inner SiC surface in a small region of the particle. The IPyC failure appeared to be driven by strong adhesion to the buffer layer, which underwent substantial dimensional shrinkage during irradiation (Fig. 7(c)). Similar behavior has been observed for the irradiated AGR-2 fuel [123].

During irradiation, cesium release from the fuel was relatively low. In four of six AGR-1 capsules, total fraction of cesium released from the fuel compacts was $<3 \times 10^{-6}$. In the remaining two capsules, release fractions were ~10^{-5}, with the higher values resulting from several particles experiencing SiC failure and Cs release. Consistent with historical observations, silver release from particles and fuel compacts was often relatively high, depending on irradiation temperature. In the AGR-1 irradiation, the aggregate fuel compact ^{110m}Ag release in each capsule ranged from around 1% to nearly 40% [121].

Europium and strontium release from the AGR-1 fuel compacts was $<5 \times 10^{-4}$ and $<3 \times 10^{-5}$, respectively, for all capsules.

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[5] The term "TRISO failure" is used to denote failure of all three dense coating layers resulting in loss of fission gas retention, and is distinguished from "SiC failure", which denotes failure of the SiC layer while at least one of the pyrocarbon layers remains intact.

However, higher fractions were found in the matrix, indicating modest release through intact coatings as well as significant retention in the compact matrix [121]. The AGR-2 UCO compacts in Capsule 2, operated at irradiation temperatures exceeding those expected during normal HTGR operation (time-average peak temperatures around 1350 °C), demonstrated that the inventory residing in the compact matrix could be as high as 10^{-2} to 10^{-1} [123].

2.7.3. Safety test performance

Post-irradiation safety testing in dry helium has been performed on 18 AGR-1 compacts [122,128] and 7 AGR-2 compacts [123] at temperatures from 1600 to 1800 °C. Some of the major observations from work completed to date are summarized below.

- No TRISO failures have been observed at 1600 or 1700 °C for nominal hold times of 300 h in either the UCO or UO$_2$ fuel. A small number of failures have been observed during 1800 °C tests of UCO fuel based on Kr release (no 1800 °C UO$_2$ test have been performed because of excessive Cs release at 1700 °C). Based on these data, the upper bound at 95% confidence for the TRISO failure fraction for UCO fuel at 1600 °C was determined for the AGR-1 fuel to be $\leq 6.6 \times 10^{-5}$. This is approximately a factor of 10 lower than the design specifications for allowable particle failures during accidents for US and German modular HTGR designs [121]. Similar data will be compiled for the AGR-2 tests once completed.

- The ^{134}Cs release from UCO during the tests has been relatively low: $\leq 2 \times 10^{-4}$ after 300 h 1600 °C and $\leq 2 \times 10^{-3}$ after 300 h at 1800 °C (AGR-2 peak values are lower than those from AGR-1 [123]). Cesium release was dominated by a small fraction of particles that suffered SiC layer failure and subsequent release of a significant portion of their inventory. Fig. 8 shows a summary of ^{134}Cs release data from AGR-1 safety tests.

- Cesium release from US UO$_2$ fuel from the AGR-2 experiment (burnup of approximately 10.6% FIMA) has been appreciably higher by comparison to UCO, with peak release fractions reaching ~10^{-2} and 10^{-1} at 1600 °C and 1700 °C, respectively. This has been found to be due to degradation of the SiC layer caused by CO corrosion, with the effect increasing appreciably at higher temperatures [123]. Thus while all particles maintain at least one intact layer and retain krypton, the loss of retention in the SiC layer results in relatively high cesium release.

- The release of silver, europium, and strontium during 1600 °C UCO heating tests appears to be dominated by inventory that resided in the compact matrix at the end of the irradiation, which was released from intact particles in-pile. Additional release through intact coatings is only observed after ~100 h at 1800 °C. In UO$_2$ fuel, the release of these elements during heating tests is also likely affected by the particles suffering SiC failure due to CO corrosion.

- The dominant SiC failure mechanism in UCO fuel is the same as that observed in-pile (i.e., localized fission product corrosion of the SiC layer precipitated by IPyC fracture), with the severity of the degradation increasing with increasing safety test temperature.

2.7.4. Summary

Both the UCO and UO$_2$ fuel tested to this point in the AGR program have performed well in-pile, with low observed TRISO failure fractions. The results to date also indicate excellent performance of the UCO fuel during high-temperature, post-irradiation
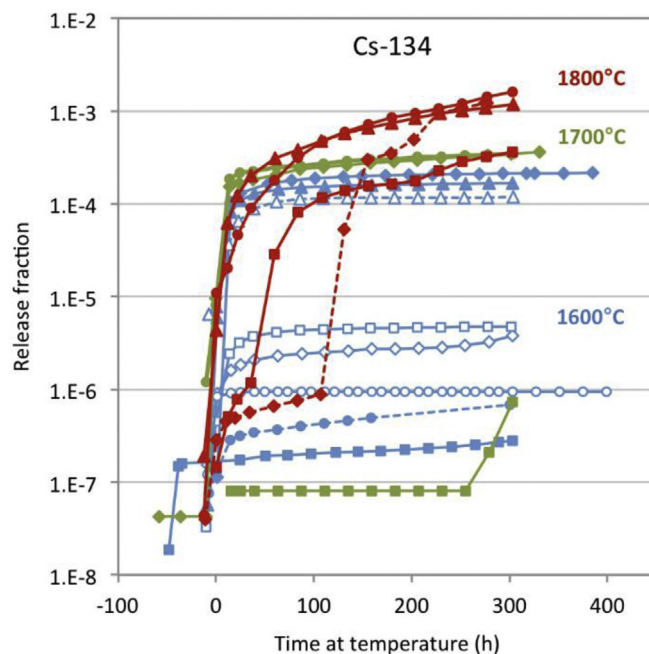

Fig. 8. Combined results of ^{134}Cs release during AGR-1 UCO fuel safety tests in dry helium at 1600−1800 °C. Burnup of the fuel specimens ranged from 13.4 to 19.1% FIMA [121].

heating tests, and demonstrate several differences in behavior from UO$_2$ TRISO fuel. Among these is the virtual elimination of SiC attack due to CO corrosion, which enables operation to high burnup (up to 19.6% FIMA in this program). The result is a far lower incidence of SiC layer failure and cesium release, particularly at T $\geq$ 1600 °C. Additional post-irradiation, high-temperature testing of the AGR fuel is planned in atmospheres containing O$_2$ and H$_2$O to explore fuel behavior in oxidizing environments, including those experienced during air or steam ingress into the reactor core.

3. Concluding remarks

Through several decades of development, testing, and improvements, fuel particle coatings have matured into what is in practice an international consensus design, with two types of kernels (UO$_2$, UCO) pursued in current development programs. Fabrication and quality control methods have been demonstrated that are capable of producing fuel with low manufactured defect fractions and low residual contamination. High-quality fuel exhibits very low particle failure rates during irradiation, with failure fractions approximately 10^{-5} (upper bound at 95% confidence) demonstrated for UO$_2$ fuel up to ~11% FIMA and UCO fuel up to ~20% FIMA. This performance translates into low fission product release from the fuel under normal conditions, and is a major factor controlling the radiological source term released from the reactor.

The particles exhibit remarkable durability under severe accident conditions, which can include temperatures up to 1600 °C in modern modular HGTR designs. In the German and US programs, testing has demonstrated failure fractions of $\leq 4 \times 10^{-5}$ and $\leq 7 \times 10^{-5}$ UO$_2$ and UCO, respectively, at 1600 °C for hundreds of hours, with good retention of most fission products within the fuel spheres and compacts.

The fuel performance database continues to expand through several ongoing R&D and qualification programs. Important efforts in this regard include exploring the performance margin of the fuel

in terms of temperature and burnup to establish the limits on the fuel capabilities, testing in oxidizing environments to bound air and moisture ingress incidents, and improving fuel performance models to better understand and predict fuel behavior.

In addition to the construction and pending start of operations at the HTR-PM reactor in Shandong Province, China, numerous companies in North America and elsewhere are currently pursuing high-temperature reactor designs utilizing TRISO fuel particles to take advantage of the high-temperature performance capabilities [129]. It is anticipated that these developments may usher in a new era of particle-fueled reactor operating experience and the opportunity to accumulate additional data on the performance of modern TRISO fuel in demonstration and prototype modular reactors.

Acknowledgements

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Dr. Paul A. Demkowicz is a Distinguished Staff Scientist at the Idaho National Laboratory, focusing on development and characterization of fuels for advanced nuclear reactors. Dr. Demkowicz is the technical director for the US DOE Advanced Gas Reactor fuel development and qualification program. This program is fabricating, irradiating, and performing post-irradiation examination and safety testing on tristructural isotropic (TRISO) coated particle fuel, with the goal of qualifying this fuel form for use in high-temperature reactors.

Bing LIU PhD, professor of nuclear fuel and fuel cycle, now is the director of Division of Nuclear Fuel Element, Institute of Nuclear and New Energy Technology, Tsinghua University. The main research work is the coated particle manufacture technology with fluidized bed chemical vapor deposition (FB-CVD), the irradiation behavior of coated particle and the calculation model of coated particle in HTGR. As the engineer in charge of China HTR-PM demonstration project, responsible for the HTR-PM demonstration Nuclear fuel elements design, manufacture technology and fuel safety analysis, and responsible the technical work of the HTR-PM demonstration fuel manufacture factory design, construction and manufacture fuel for HTR-PM.

John D. Hunn, Ph.D. ORNL Distinguished Scientist, Nuclear Science and Engineering Directorate Fusion and Materials for Nuclear Systems Nuclear Fuel Materials. Dr. Hunn has over 25 years of materials and fuels research experience at the Oak Ridge National Laboratory and has been part of the DOE - Advanced Gas Reactor (AGR) Fuel Development and Qualification Program since 2002. Dr. Hunn manages the ORNL Coated Particle Fuel Development Laboratory, which was used for development, fabrication and acceptance testing of the AGR-1, AGR-2, and AGR-3/4 irradiation test fuel. Since 2007, Dr. Hunn has been the ORNL Program Manager and Technical Lead for AGR coated particle fuel development, post-irradiation examination (PIE), and safety-testing activities at ORNL.