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A novel approach to fabricating fuel compacts for the next generation nuclear plant (NGNP)

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ABSTRACT

The next generation nuclear plant (NGNP) is a combined complex of a very high temperature reactor (VHTR) and hydrogen production facility. The VHTR can have a prismatic or pebble bed design and is powered by TRISO fuel in the form of a fuel compact (prismatic) or pebble (pebble bed). The US is scheduled to build a demonstration VHTR at the Idaho National Laboratory site by 2020. The first step toward building of this facility is development and qualification of the fuel for the reactor. This paper summarizes the research and development efforts performed at Oak Ridge National Laboratory (ORNL) toward development of a qualified fuel compact for a VHTR.

1. Introduction

The very high temperature reactor (VHTR) is one of the reactor concepts identified in the Generation IV (Gen IV) International Collaboration. The VHTR utilizes a coated particle fuel that is combined with a mixture of graphite and resin and pressed into either a cylindrical or spherical shape. Cylindrical fuel compacts are used to power a prismatic core design, while spherical fuel balls are used to power a pebble bed reactor design.

The VHTR is cooled by helium and moderated by graphite. The hot helium that leaves the reactor core, which is made of graphite, can be used to do work, such as spin a turbine or provide process heat for a given reaction. As part of the next generation nuclear plant (NGNP) the VHTR provides the process heat needed to produce hydrogen, which can be used to fuel a hydrogen economy. NGNP, therefore, is the combined complex of a VHTR to generate process heat that is physically linked to a hydrogen production facility.

As part of the NGNP program, funded by the Department of Energy – Office of Nuclear Energy Science and Technology, fuel development and qualification for the VHTR was initiated. Helium cooled, graphite moderated reactors have been built in the past, but a high quality fuel and compact fabrication method were needed in order to achieve the outlet gas temperature of ~950 °C required for hydrogen production. Therefore, a new type of particle fuel and fuel compact were needed for the VHTR. The fuel particle and fuel compact development work was performed at Oak Ridge National Laboratory (ORNL) under a program titled advanced gas reactor (AGR). Phase one of the AGR program (AGR-1) involved fabrication of AGR-1 fuel compacts (also described as test articles) for irradiation in the Idaho National Laboratory (INL) advanced test reactor (ATR). The fuel compact development needed for fabrication of the AGR-1 test articles is described here.

2. Review of methods for making fuel compacts

Helium cooled, graphite moderated reactors, whether they are considered ‘very high temperature’ or not utilize either a cylindrical fuel compact, or spherical fuel pebble. A number of fuel compact fabrication processes have been developed over the years. Fig. 1 shows the major fabrication steps in the ‘admix’ process that was developed for the early fuel loads of the Dragon reactor. In the admix process the basic ingredients of the fuel compact matrix are a high char yield resin and a graphitized petroleum coke filler. The matrix mixture ultimately selected consisted of 88% filler and 12% resin binder [1,2]. The filler raw material was a calcined petroleum distillate (Shell H-100) with a needle coke structure and was ground to a particle size of less than 50 μm prior to graphitization at 2700 °C. The filler was well mixed into a dispersion of phenol formaldehyde resin (with a hexamine curing agent) in denatured alcohol. The solvent was then evaporated and the resin-coated granulated powder remaining was reground, sieved, and blended. In the last stage of the blending process the coated fuel particles were cold mixed with the granulated powder. Small amount of liquid paraffin spray were used to dampen the mixture and reduce segregation.

A fixed amount of the mixture was introduced into a cylindrical die and warm molded (pressed) at 180 °C at a pressure of ~7 MPa to form the annular fuel compacts. The ‘green’ compacts were

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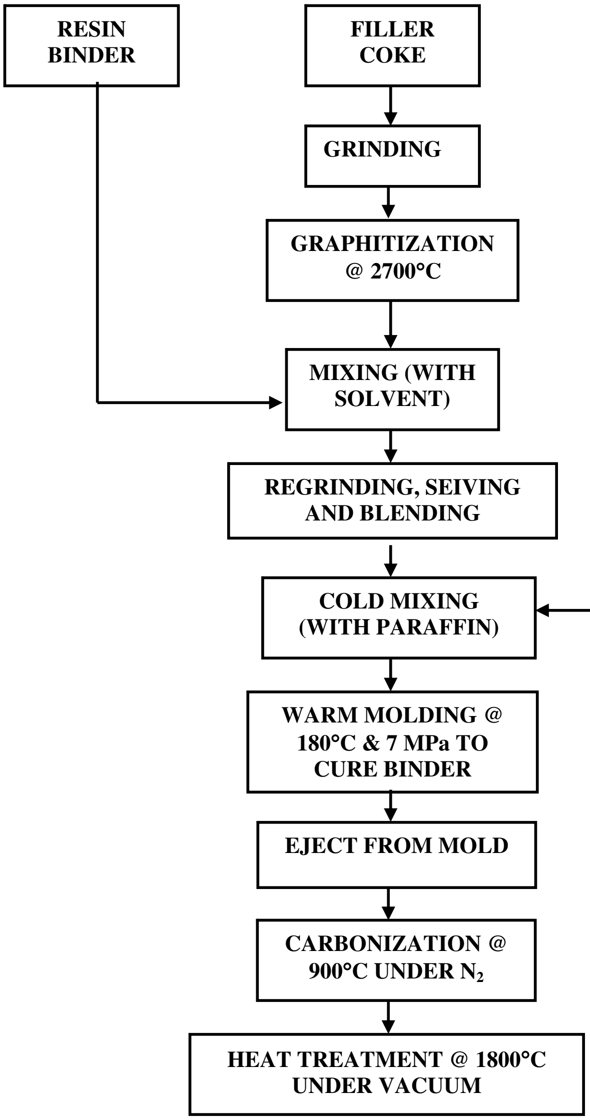


Fig. 1. The admix process for fuel compact fabrication as used by the Dragon project.

subsequently carbonized by heating in flowing N_2 at a rate of 5 °C/min. to 900 °C, followed by heating to 1800 °C under high vacuum (to degas the compacts). The thermosetting resin binder employed gave the compacts stability of shape during the freestanding carbonization, and resulting dimensional changes were small and predictable. Fuel particle volume fractions in this process were typically >25%. Some compacts with particle volume fractions of ~35% were made but there was an increased tendency for fuel particle coating damage to occur at these higher volume fractions. Consequently, the Dragon project developed an 'overcoating process' to reduce fuel particle-to-particle interaction and allow for higher volume fraction compacts to be manufactured. This approach is discussed subsequently.

A modification of the Admix process was used for the peach bottom reactor fuel compacts (Fig. 2). Hot-pressing an admixture of coated particle fuel, powdered graphite filler, and a pitch binder to about 30 MPa at 750 °C, followed by heat-treatment to 1400 °C in vacuum, produced a compact with a high fraction of graphite in the matrix (~90% of the green mix). About half of the ~10% pitch binder remained after heat-treatment giving a final matrix composition of 5% binder char and 95% of the more stable graphite [1]. Synthetic graphite flour was used (National Carbon Company Grade GP-38 with an average particle size of ~20 μm) and the binder was Barrett No. 30 medium pitch. An ethyl cellulose plasticizer (1 wt%) was added to aid granulation of the final mixture and trichloroethylene solvent was used to facilitate mixing (7 cm^3 per gram of pitch). The matrix components were well mixed with coated particle fuel and then granulated into 3–9-mm pellets and dried to remove the solvent. Segregation of the dense fuel particles and lighter graphite powder was avoided by use of the pelletization approach. The pellets were essentially clusters of overcoated particles, although the overcoating was much less uniform that those obtained for the Dragon overcoating process and subsequent German and Japanese overcoating methods.

The pelletized fuel mixture was compacted in a double-action graphite die at a pressure of ~7 MPa. The die was heated to ~250 °C and when the thermoplastic pitch was fully liquid the pressure was increased to ~30 MPa [3]; further heating to ~750 °C under pressure caused carbonization of the compact in the die. The compact was then removed from the die and given a final heat-treatment to 1400 °C under vacuum (to degas the compacts). High matrix densities (1.90–1.95 g/cm^3) and close dimensional control of the compact resulted from the in-situ carbonization. Fuel particle volume fractions were typically 25–35%.

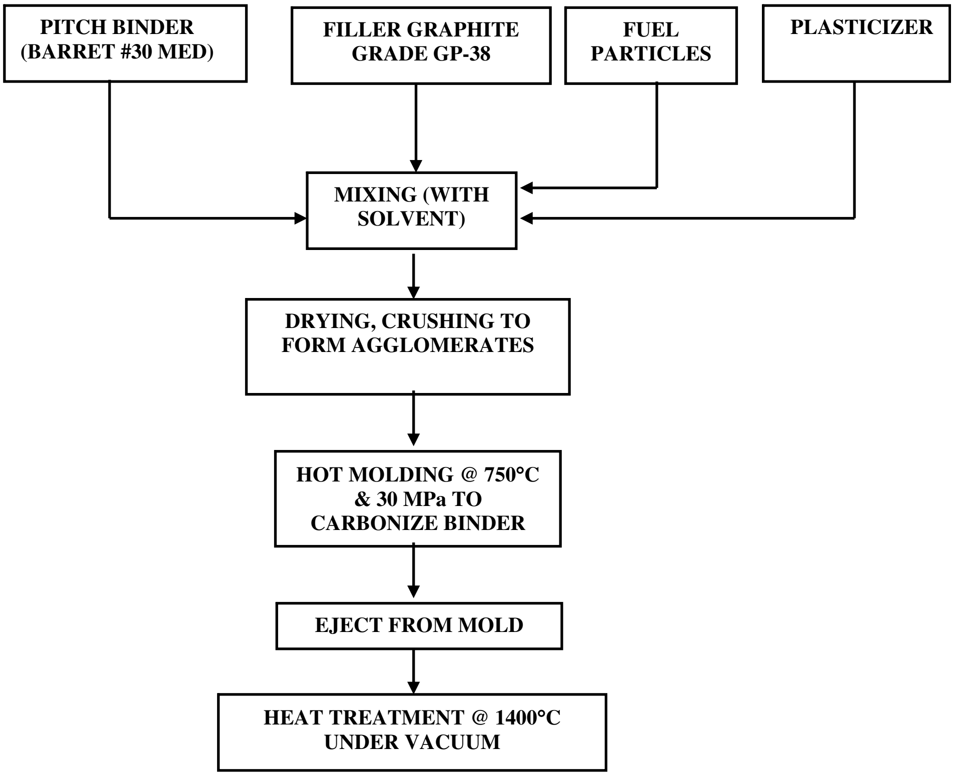


Fig. 2. The admix/agglomeration fuel compact manufacture process as used for the peach bottom reactor.

The need to increase the particle volume fraction in the compacts for later fuel loads led the Dragon project to develop the overcoating method [1,4]. In this process (Fig. 3) the coated fuel particles were overcoated with enough of the powdered matrix previously described to fill the interstices during the compact compression process (Fig. 1), thus replacing the cold mixing step. The fuel particles were tumbled in a slowly rotating drum and sprayed with alcohol while feeding in the powdered matrix mixture until a thick enough coating was obtained to provide all the matrix needed in the compact. The overcoated particles were then dried and loaded into compact dies, molded, and carbonized as previously described. By minimizing the overcoat thickness fuel particle volume fractions of ~50% were attained. However the resultant compacts had poor thermal conductivity and crush strength.

The German Pebble Bed reactor fuel pebbles were ultimately manufactured with a derivative overcoating process (similar to that in Fig. 3) [1,5]. The pebble consisted of a spherical fuel particle compact contained in a spherical shell of unfueled matrix material. The graphite powder and phenolic resin (diluted with methanol) were mixed, dried, milled, and homogenized. Part of this matrix mixture was then fed into a rotating drum along with the fuel particle and methanol. The overcoated particles were then dried, sieved, and isostatically pressed in a rubber mold at ~30 MPa (with additional matrix mix) to form the fuel compact. The remainder of the matrix mix was then molded around the fuel compact to form the fuel-free shell of the fuel pebble and the entire pebble subjected to high-pressure isostatic molding at ~300 MPa. The fuel pebble was then slowly heated to 800 °C in an inert atmosphere to carbonize the resin binder and in a subsequent heat-treatment heat-treated at temperatures up to 1950 °C in vacuum to degas the fuel pebbles [5].

The matrix mixture used in the German molding process was designated the ‘A3’ matrix and consisted of a mixture of 64 wt% natural flake graphite, 16 wt% graphitized petroleum coke, and 20 wt% phenolic resin [1], and in the carbonized condition it contained ~90 wt% graphite and only ~10 wt% of the less stable resin char. While this method is very similar to the Dragon method, a thinner particle overcoating is used in pebble production since a fraction of the matrix is introduced into the fuel compact after the overcoating step. The fuel particle volume fraction, including the fuel-free shell, in the German pebble process was typically rather low (~5% for the AVR and ~12% for the THTR).

The fuel compacts of the Japanese HTTR are also manufactured using an overcoating method [6]. A ‘resinated’ graphite matrix was prepared by mixing synthetic graphite powder, natural graphite powder, and phenolic resin (as the binder) in the ratio 16:64:20, followed by grinding the mixture to a powder (note this matrix composition was identical to the German ‘A3’ matrix). The coated fuel particles were then overcoated with the matrix to attain an overcoat of ~200 μm thickness, yielding a particle volume fraction of ~30% in the fuel compact. The overcoated particles were warm pressed in metal dies to form the green compact and cure the resin binder, followed by ejection from the die and carbonization of the compacts in flowing N_2 at 800 °C. The final step of manufacture was heat-treatment to 1800 °C in vacuum to degas the fuel compacts (Fig. 3).

The general atomics (GA) prismatic core design HTR, such as the fort saint vrain (FSV) reactor built for Public Service Company of Colorado, required much greater fuel particle volume fractions than were typically attained with any of the methods outlined above. Consequently, GA developed a new compacting method capable of achieving the ~60% particle volume fractions required [1,7]. Cylindrical molds were filled with coated particles in random close packing and a molten matrix material was then intruded into the particle bed and allowed to cool and harden forming the ‘green’ fuel rod. A disadvantage of this process was the need to limit the powdered graphite filler content in the matrix to <40% so that the matrix viscosity remained low enough to allow injection at reasonable injection pressures where particle coating damage was minimized. With this low level of graphite filler in the matrix it

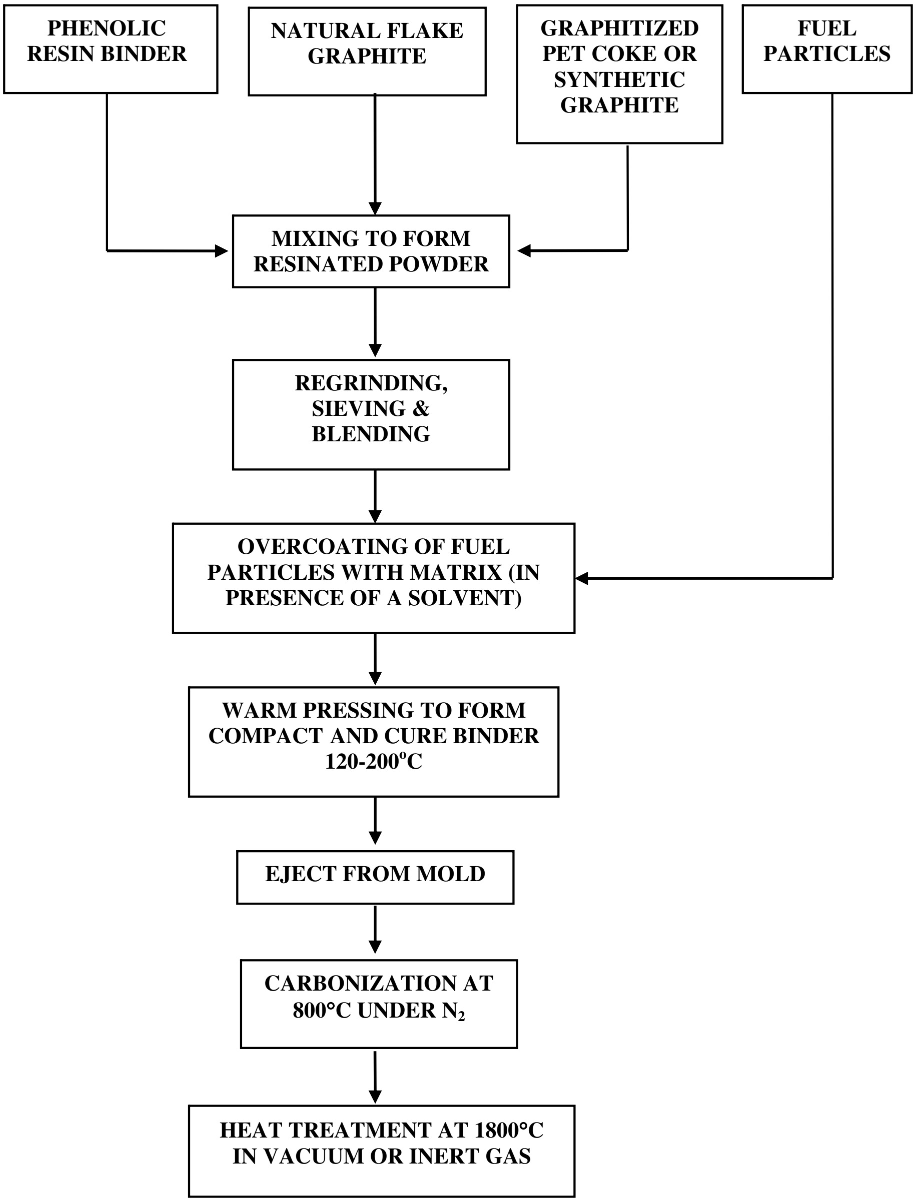


Fig. 3. A typical particle overcoating process for fuel compact manufacture as developed by the Dragon reactor and used in the HTTR and AVR/THTR programs.

became necessary to use a pitch binder, since this exhibited less dimensional shrinkage on carbonization than the resin-based binder systems. Similarly, a pitch-derived carbon would be expected to be more dimensionally stable under neutron irradiation than a resin-derived carbon. A thermoplastic (pitch) binder can also be reheated and softened in the forming equipment and maintained in a fluid state during the injection process. However, a major disadvantage of a thermoplastic pitch binder is the need to support the green compacts during carbonization when the binder softens prior to pyrolysis.

The GA process for FSV fuel compacts used a coal tar pitch binder (Allied 15 V) and natural flake graphite (Asbury grade 6353) as the filler. The graphite filler content of the matrix was ~28%. The matrix mixture was heated to ~200 °C to attain its maximum fluidity and injected at a pressure of 7 MPa into the close packed particle beds, and quickly cooled to form solidified green rods that were ejected from the molds. The green rods were vibrationally packed in fine alumina powder beds to support them during initial carbonization at 900–950 °C in flowing argon [8]. The compacts were then removed from the alumina beds and further heat-treated free standing in flowing argon to 1600–1850 °C in ~1 h with a ~30 min hold at peak temperature [1,8]. The rapid heating rates employed were designed to reduce the char yield of the pitch binder. Between heat-treatments the compacts were acid (HCl) cleaned to remove any metallic impurities emanating from the compact or furnace during carbonization.

In a later development of the GA injection process the use of thermosetting resins was demonstrated [7]. Again, a high fuel particle volume fraction (60%) was required in the thermosetting resin compacts. The optimum resin was Monsanto's commercial solvated phenolic SC-1008. The matrix filler was graphite flour (Lonza KS-15) with 95% of the grains having a size less than 15 µm. Early attempts at compact production via the injection process with a resin binder resulted in particle failures. The cause was identified as a combination of excessive binder/particle outer coating adhesion and large matrix shrinkage during carbonization. Since increasing the fraction of the more dimensionally stable graphite flour would result in a matrix that could not be injected, another solution had to be sought. The addition of a low char yield resin, such as polystyrene, proved to be an appropriate remedy. The binder shrinkage could be reduced according to a 'rule of mixtures' relationship by the addition of a low char yield resin at fractions up to 50%. Indeed the matrix shrinkage was reduced from ~50 vol.% with the phenolic binder alone to ~20 vol.% with 50% addition of polystyrene. The low char yield polystyrene resin virtually disappears on carbonization, thus providing a porous matrix similar to that observed with pitch-injected matrices. Moreover, the additional porosity weakens the particle-to-matrix bond, minimizing the chance of particle failure. The use of a 'fugitive' low char yield resin addition allowed for a thermosetting matrix mixture with low heat-treatment shrinkages comparable to the undiluted phenolic resin with about 65 wt% graphite filler, which themselves could not be injected (because the 65% filler content caused the matrix viscosity to be too high).

The dimensions of the cylindrical fuel compact for the VHTR reactor were set at approximately 0.5 in. diameter × 1 in. long, with a fuel loading of 35%. Based on the review of previous fuel compacting processes described here, the selected method of fuel compact fabrication for the VHTR was overcoating and compacting. This fabrication technique draws heavily on the overcoating work developed by the Germans and pressing process developed by the Japanese for the HTTR. The differences are that the Germans overcoated particles and then isostatically pressed them into spheres, in the presence of excess matrix, in order to form a fuel pebble with roughly 10% loading. The Japanese overcoated and pressed particles, but into an annular shape, as opposed to a cylindrical compact, and fuel loadings were slightly lower at 33%.

3. Experimental

3.1. Particle size distribution of graphite and matrix

The particle sizes of the natural graphite, synthetic graphite, and matrix made after combining the graphites with resin in order to make the A3 matrix were measured using a light scattering technique. A Horiba La-700 unit was used for measuring particle size. Hundred milliliter of de-ionized (DI) water was added to the sample well of the La-700 Horiba. Next, 1 drop of dispersant was added to the sample well. The agitator was then turned on in order to circulate the fluid through the measuring cell making sure to remove all bubbles from the wall of the scattering cell. The relative refractive index was then set to the appropriate setting based on the material being analyzed after consulting the unit's manual. The particle size reading for this blank sample was then recorded. The actual sample was then prepared by taking a quantity of the sample and adding it to a volume of DI water. A quantity of dispersant was added to the solution as previously described. The solution was then sonicated for a period of time such that the solid sample was well dispersed in the DI water. Next, the well dispersed sample was added to the sample well of the La-700 until the scatter meter indicated the proper concentration. The particle size was then measured. This procedure was repeated five times for each sample.

3.2. Glow discharge mass spectrometry (GDMS) measurements

GDMS measurements were obtained on the natural graphite, synthetic graphite, and resins used in this research. The samples were sent to Shiva Technology for GDMS testing. The basic principle of GDMS is the atomization of a solid sample by sputtering in a low pressure DC plasma. The atoms are then ionized and discharged to a mass analyzer for detection.

The procedures used by Shiva are considered proprietary, but involved forming approximately 0.10 g of the powdered solid sample into a cathode. The cathode (sample) is then placed in a discharge cell made of Tantalum. A potential of 1–2 kV is applied between the anode (discharge cell body) and cathode until glow discharge is achieved. Positive ions are then directed toward the sample. Upon impact with the sample neutral ions, which are predominately individual atoms, are sputtered off and attracted to the mass analyzer for detection.

3.3. Compact characterization by LBL

Compacts were analyzed using a multi-step process called leach-burn-leach (LBL). LBL is used to determine impurity content, defective SiC fraction, and exposed kernel fraction. The procedure consists of an initial electrolytic deconsolidation plus acid leach series to expose the particles in the compact matrix, followed by a burn to remove carbon, and then a final acid leach series.

Compacts are electrolytically deconsolidated to free the particles from the matrix. An anode is placed on one end of a compact and the other end is submersed in concentrated nitric acid containing a platinum cathode. As approximately 10 W is applied, the compact matrix material in contact with the acid breaks up and falls away from the OPyC of the particles. The compact is gravity fed into the nitric acid, maintaining about 5 mm of compact submerged until the entire compact is deconsolidated. The particles and matrix residue are leached twice in concentrated (~70%) nitric acid to dissolve any exposed uranium and/or impurities not contained by the particle coatings (i.e., in the compact matrix or broken particles). Inductively coupled plasma-atomic emission or mass spectroscopy (ICP-AES or ICP-MS) is used to determine the content of U and impurities (Fe, Cr, Mn, Co, Ni, Ca, Al, V, and Ti). The effective number of exposed kernels is obtained by dividing the amount of uranium leached by the average uranium content of one kernel. This effective number of exposed kernels may also include uranium from other sources (tramp uranium), but it is assumed that the U contamination is dominated by failure of individual particles in which the uranium in the kernel is exposed (e.g., particles broken by compacting).

The particles and residue from the deconsolidation-leach step are heated in air in a furnace at 750 °C for at least 72 h to remove the compact matrix carbon, the outer pyrocarbon (OPyC) coating layer as well as any inner pyrocarbon (IPyC) and buffer coatings that are exposed to air due to a through-defect in the SiC layer (e.g., a significant crack or hole through the SiC). The 'burned-back' particles and residue are acid leached twice to dissolve any exposed uranium and/or impurities. ICP-AES or ICP-MS is again used to determine the content of U and impurities. The number of particles with a defective SiC layer is determined by dividing the amount of uranium detected in this post-burn leach series by the average uranium content of one kernel. The impurity content results are added to the impurity content results obtained from the deconsolidation-leach test to determine total impurity content outside of the intact SiC-coated particles. Inspection of the particles after the final leach can sometimes be used to identify and further analyze individual failed particles.

4. Results and discussion

4.1. Matrix production method development

The matrix was made by mixing natural graphite, synthetic graphite, and a thermosetting resin in the ratio of 64, 16, and 20% (by weight), respectively. This is the A3 matrix formulation developed by the Germans, although the actual graphites used to make the matrix here are different. The reason for this breakdown in components stems from the irradiation behavior of carbons and graphites. Isotropic graphite bodies experience less structural damage, both on the microscopic (crystalline) and macroscopic scale, under irradiation than anisotropic or glassy carbons [9,10]. Therefore, the objective of the A3 matrix production process is to produce a graphite based matrix that has isotropic properties. An isotropic graphite body can be produced in either of two ways. First, a naturally occurring carbonaceous source can innately have a random orientation of structural units that will form into isotropic graphite upon heat-treatment. An example of such a naturally occurring isotropic graphite precursor is Gilsocarbon, which was widely used as a raw material for nuclear graphite production in the UK. However, today this type of naturally occurring isotropic graphite precursor is rare (Gilsocarbon is no longer available) so an isotropic graphite is typically formed by milling an anisotropic graphite into a fine particle size (less than 20 μm) and re-forming it into a graphite that exhibits isotropic properties on a macroscopic scale. This re-formed material can accurately be described as being microscopically anisotropic and macroscopically isotropic. Graphite that is microscopically anisotropic and macroscopically isotropic, which both Gilsocarbon and re-formed isotropic graphite are, is more resistant than anisotropic graphite to structural damage and subsequent dimensional change during irradiation, and is therefore considered superior for nuclear applications.

Natural graphite is highly anisotropic, but can be milled into a fine particle size and re-formed into a graphite that is macroscopically isotropic. Synthetic graphite is less anisotropic than natural graphite, but also can be milled and re-formed in order to produce an overall isotropic graphite. The reason for having a higher percentage of natural graphite versus synthetic graphite relates to the compressibility of each component. As mentioned, natural graphite is highly anisotropic, meaning there are large regions of aligned crystals and essentially zero cross-linking atoms between graphene layers and neighboring crystallites. The absence of these cross-links means that the structure is somewhat fluid and such graphite powders can be compressed into a shape without the presence of a resin binder because only van der Walls forces hold the graphene layers together. Synthetic graphite contains more cross-links and small regions of aligned crystals that produce a more rigid structure which cannot be pressed into shape without the presence of a binder. However, the presence of these cross-links and smaller graphene layers leads to a tougher graphite that is more resistant to crack propagation and failure. Conversely, natural graphite is weak and can easily be delaminated because only van der Walls forces hold the grapheme layers in place in the crystallographic *c*-direction. By mixing the natural graphite with the synthetic graphite a material with some compressibility (as a result of the natural graphite) and some toughness (as a result of the synthetic graphite) is achieved.

The resin used in the A3 matrix is added in order to provide some adhesion to the mixture and help it adhere to the TRISO particles during overcoating. Most importantly, though, the resin is present because it will harden and fuse the compact into a solid piece during the carbonization step. The ratio of resin to graphite used in the A3 matrix is much less than binder percentages used in commercially available graphites made with particle sizes used in this research (less than 30 μm). Typically, fine grained commercial graphite will contain greater than 50% resin binder. The A3 matrix contains 20% binder because the compressibility of the natural graphite allows the compact to form without the presence of a larger quantity of binder. This reduction in binder content also aids in resistance to dimensional change during irradiation because a thermosetting binder forms a glassy carbon during carbonization. Glassy carbon has poor stability under irradiation, so its presence is off-set by the natural graphite.

It must be noted that the A3 matrix is not a material, it is simply a recipe. The original raw materials used by the Germans to make the A3 matrix reported in the literature no longer exist. The matrix material made in the AGR program will have the A3 matrix recipe, but the raw materials used to make it will be different than what was used by the Germans. A considerable portion of the compacting process development was the selection and qualification of the three components used to produce the A3 matrix. At the time of this selection and qualification process a specification on the AGR-1 test articles had not been completed, so the matrix components could not be purchased to an AGR-1 material specification. From previous experience it was known that the graphites and resin should be as free of impurities as possible, and also be readily available in large quantities (e.g. enough for a large scale fuel fabrication facility should this work be commercialized). A number of natural graphites, synthetic graphites, and thermosetting resins were identified and purchased for this research. Table 1 lists the vendor, the type of material, and the material name. All of the materials were tested for impurities by glow discharge mass spectrometry (GDMS) at Shiva Technologies.

A low level of initial impurities was considered key to successfully producing a compact whose final impurities concentrations, after carbonization and heat-treatment, were within specification. Based on this requirement, the best natural graphite candidates were the Asbury Graphite Mills sample (RD 13371), and the Graftech natural flake milled grade (GTI-NFM). The synthetic graphites with lowest impurities content were the SGL sample (KRB2000) and the Timcal KS-15 grade. The resin candidates with the lowest impurities were the Borden Durite sample, and the Plenco P-800 resin.

With the two best natural graphite, synthetic graphite, and resin candidates, in terms of low level of impurities, selected, production of matrix was initiated. The matrix was produced by wet mixing the three raw materials in a jar mill. The two graphites were weighed out and poured into a 1000 mL container. The mass of graphite used was based on the A3 matrix (64% natural graphite, 16% synthetic graphite, and 20% resin by weight). For this size of container (1000 mL), 128 g of natural graphite and 32 g of synthetic graphite were mixed. Next, a quantity of ethyl-alcohol was added to the 1000 mL container such that the level of graphite and ethyl-alcohol was approximately 1 in. from the top of the container. The resin binder (40 g or ~30.8 mL) was then added to the nearly full container and the container was capped. The graphite–resin–alcohol mixture was then placed on a jar mill and spun for 1 h. After spinning, the contents of the container were poured into a large rectangular pan and allowed to dry for 48 h. After drying, the ‘cake’ of graphite and resin was broken into smaller pieces and charged to a Holmes pulverizer with a US Sieve 60 mesh screen (250 μm opening) in place. After pulverizing, the matrix production was complete. In the early stages of the research a particle size distribution analysis by light scattering analysis of the matrix was performed for quality control purposes. Table 2 shows the particles

Table 1
Candidate materials for A3 matrix

Vendor	Material	Material ID
Asbury Graphite Mills	Synthetic graphite	7105
Asbury Graphite Mills	Natural graphite	M890
Asbury Graphite Mills	Natural graphite	RD13371
Asbury Graphite Mills	Natural graphite	PG06
SGL carbon	Synthetic graphite	KRB2000
Graftech	Natural graphite	GTI-NFM
Graftech	Synthetic graphite	TG-652
Timcal	Synthetic graphite	KS-15
Superior graphite	Natural graphite	SGTP
Borden chemical	Resin	Durite SC1008
Georgia pacific	Resin	GP445D05
Plenco	Resin	P-800

Table 2
Graphite and matrix particle sizes

Material	Particle size (μm)
Natural graphite – GTI-NFM	6.52
Synthetic graphite – KRB2000	29.20
Matrix – GKrS	11.30

sizes of the natural graphite, synthetic graphite, and final matrix. The Graftech natural flake milled material, SGL KRB2000, and Durite resin were selected as the final matrix constituents based on low impurities (shown in Fig. 4) content and superior overcoating performance (described in Section 3.2). The matrix consisting of these components was identified at 'GKrS'. The particle size of the GKrS final matrix is most likely overestimated because the now 'resinated' graphite particles tended to agglomerate.

4.2. Overcoating method development

The next step in the compacting process involved overcoating the TRISO particles with a layer of matrix. The overcoating step is analogous to sugarcoating of pills, in that a hard circular object becomes encased in a malleable coating of powder. The overcoating process developed by the Germans involved slowly rotating the TRISO particles and matrix in a large steel drum. Methanol jets were also incorporated into the drum so that at the desired time a mist of methanol could be sprayed onto the TRISO particle/matrix bed. The methanol aided in the matrix adhering to the TRISO particle. The Germans rotated the steel drum slowly. The process is patented so no revolution per minute (RPM) data is available in the literature. However, in personal communications with individuals who have first hand knowledge of the process, the desired overcoating action was for the TRISO particles to climb approximately one-half of the way up the side of the overcoating drum before rolling down through the bed of matrix and particles. Here, the TRISO particle starts out with no overcoat layer of matrix, but begins to accumulate a thin layer of matrix after several rotations of the drum. The thickness of the overcoat layer is controlled by the number of revolutions of the overcoating drum, as well as the amount of matrix present in the drum.

In previous overcoating work performed by the Germans, Japanese, and Chinese, overcoating of TRISO particles was achieved by mixing particles and matrix in various concentrations and slowly rolling them together. The kernel size used by the Germans, Japanese, and Chinese was 500 μm, so the outer diameter of the TRISO particles was slightly larger than 1000 μm. The kernel size used in the AGR program (and for the surrogate and NUCO campaign described here) was 350 μm, and the outer diameter of the TRISO particle was nominally 780 μm, approximately 220 μm smaller than the previously used fuel particles. This reduction is diameter, and subsequent weight per particle, had dramatic effects on the overcoating process.

The first AGR overcoater utilized a top secured design shown in Fig. 5. As part of the AGR program plan, coating of 500 μm surrogate zirconia kernels (O.D. of ~1000 μm after coating) was initially performed in order to try to replicate the German coating process. The TRISO coated 500 μm particles produced as part of the first TRISO coating applications were available, so they were used to gain experience with the overcoating process and generate early data. The slow rolling method developed by the Germans was used to successfully overcoat these TRISO coated 500 μm surrogates in the top secured overcoater. Fig. 6 shows a particle overcoated by this method.

Coating of 350 μm kernels was initiated after completion of surrogate 500 μm kernel coating. TRISO coated 350 μm surrogates (OD of ~780 μm) were provided to the compacting group for

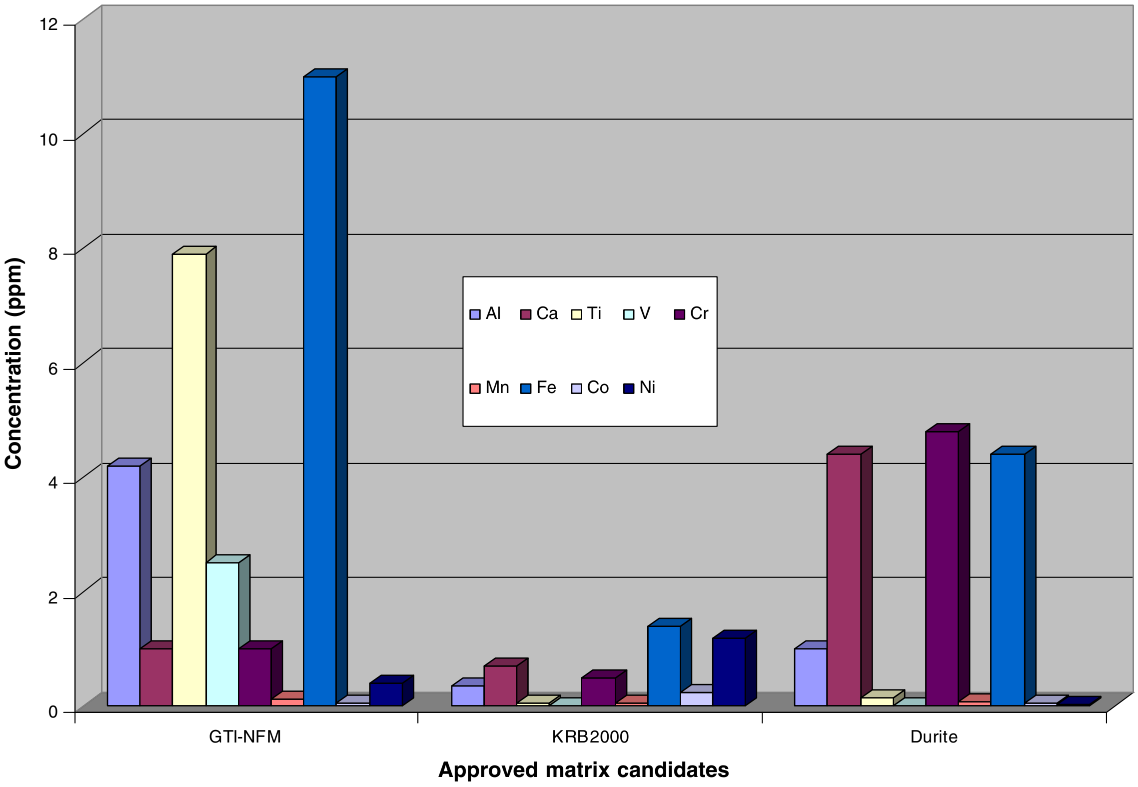


Fig. 4. Impurities content of natural graphite, synthetic graphite, and thermosetting resin used to make the matrix for the AGR-1 test articles. The impurities shown here were specifically called out by the AGR-1 test article specification.

Fig. 5. Top secured overcoater used in early development stages of AGR-1 test article development.

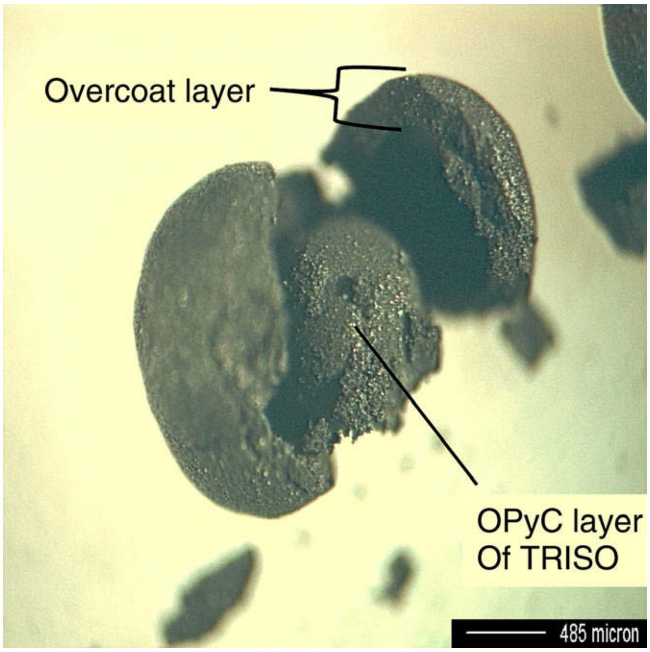


Fig. 6. Overcoated particle using top secured overcoater and slow rolling method, overcoat intentionally broken off to show TRISO particle.

further overcoating and compacting development work, as these surrogates would be identical in size to the eventual low enriched uranium oxycarbide (LEUCO) TRISO particles for AGR-1 test article fabrication. Interestingly, poor overcoating results were obtained in the top secured overcoater when switching from TRISO coated 500 μm particles to TRISO coated 350 μm particles. In fact, no overcoating was observed. The particles would discharge from the overcoater with essentially no matrix adhered to the particle, and the matrix would pour out still in powder form. Numerous additions and process changes were made to the top secured overcoater, including blowing argon through the stem that secured the overcoater lid to the motor in order to help mix the material together, and inserting a methanol drip through this stem as well, but a lack of overcoating persisted. It was then decided to secure the bottom of the overcoating chamber to the motor, thus allowing easier access to the particle/matrix mixture. Fig. 7 shows the bottom secured overcoater.

After redesigning the overcoating chamber such that its bottom was now secured to the motor, the slow rolling method was again tried. The open top of the overcoater allowed for matrix to be added incrementally (as opposed to the batch method needed for the top secured overcoater) and it appeared that this process change led to successful overcoating of TRISO coated 350 μm particles. However, upon inspection of the chamber's contents, it was found that no overcoated particles were present, i.e. the 'overcoated particles' were really just balls of matrix. The absence of

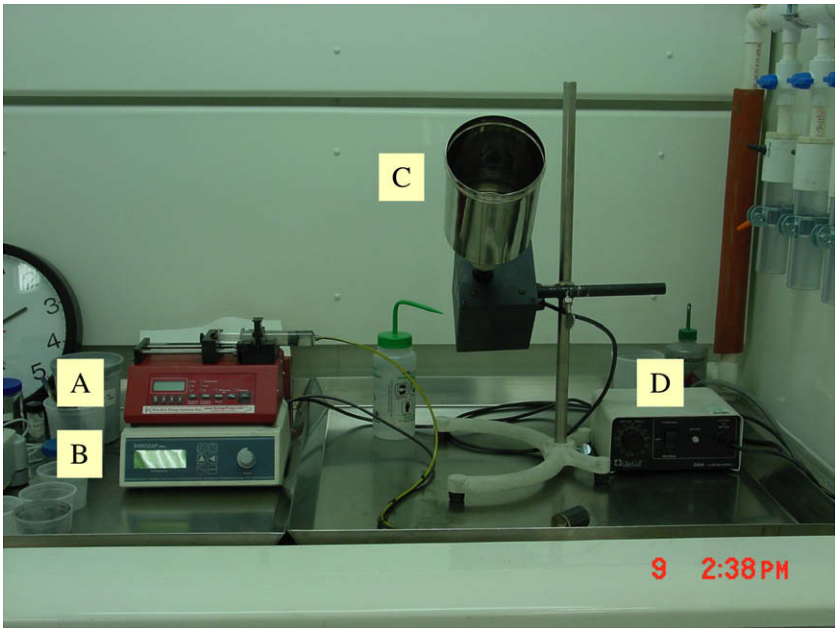


Fig. 7. Overcoater setup. Where 'A' is the syringe pump, 'B' is the ultrasonic atomizer, 'C' is the overcoating chamber, and 'D' is the motor.

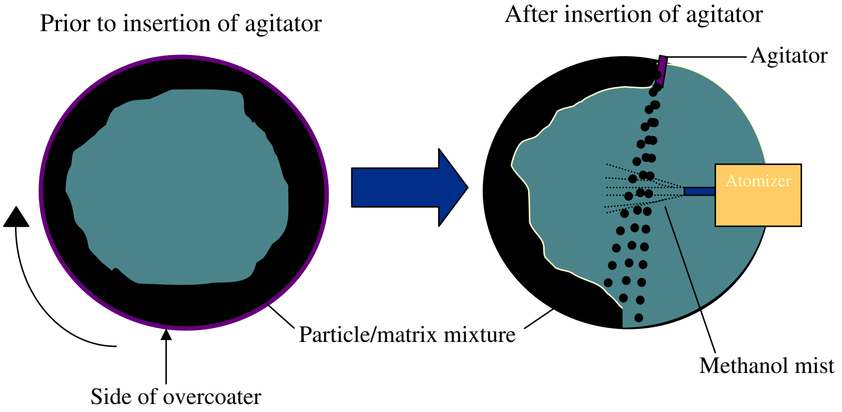


Fig. 8. Schematic diagram of the centrifugal overcoating process developed at ORNL. Development of this overcoating method was needed for the smaller LEUCO particles.

particles in the balls was confirmed by washing them with methanol. The methanol deconsolidated the matrix balls back into powder, showing that no particles were present.

Up until this point in the research project the use of methanol during overcoating had been unnecessary, or even detrimental. In overcoating the TRISO coated 500 μm particles methanol was not needed. The particles were mixed with a set amount of matrix, charged to the overcoater, and rotated slowly for a period of time. As previously stated, when switching to the TRISO coated 350 μm particles, this method was ineffectual. A methanol drip was added to the top secured overcoater, but it led to large clumps of matrix and particles, not individually overcoated particles. The formation of matrix balls observed while using the bottom secured overcoater (no methanol was used during this process) indicated that the matrix had a higher affinity for itself rather than the OPyC layers of the particles. In other words, the matrix wanted to adhere to itself instead of the particles, thus precluding any overcoating. Therefore it was determined that some additional agent was needed to help the matrix bond to the OPyC layer of the particles, and avoid the formation of matrix balls. Also, it was thought that keeping the particles and matrix well mixed, almost fluidized, would help prevent the formation of matrix clumps and balls. If the matrix was surrounded more by particles than by matrix, the formation of clumps should be hindered.

Through experimentation it was found that the best way to create the environment described above – well mixed particles and matrix with an added agent to aid in the adherence of matrix to particles – would be a centrifugal overcoating method. The key aspects of the centrifugal overcoating method are: (1) the particles and matrix are pre-mixed in a set ratio, (2) the particle/matrix mixture is spun at high enough speed to force the mixture to the walls of the overcoater, and (3) upon insertion of the agitator arm, the mixture comes off the wall of the overcoater as a spray which passes through a fine mist of methanol. The mist of methanol is achieved with a syringe pump and ultrasonic atomizer. A schematic of the centrifugal overcoating process is shown in Fig. 8.

It was found that these three key aspects create the best environment for overcoating. The pre-mixing of the particles and matrix in a set ratio allows for good contact between the matrix and the particles and a lesser probability that matrix clumps will form because essentially no excess matrix that could lead to clump formation is present. The spinning of the particle/matrix mixture ensures that the set mixture ratio will be maintained during overcoating, as the particles and matrix are not moving because they are pinned to the wall of the overcoater. The use of the agitator arm to spray the particles off the wall and through a mist of methanol ensures that the methanol is delivered at a time when the correct ratio of particles and matrix is present such that the matrix will adhere to the OPyC layers of the particles, and not itself. The mist of methanol helps to evenly coat the particles so that an even layer of matrix is deposited over the entire surface area of the particles. The centrifugal overcoating method proved effective and overcoated particles with the desired overcoat layer thickness were produced.

The initial outer diameter of the TRISO particles with 350 μm kernels was ~780 μm. In order to produce compacts with 35% packing fraction, an overcoat thickness of roughly 160 μm was needed, thus bringing the outer diameter of the overcoated particle to 1100 μm. Fig. 9 shows size and shape data for a batch of TRISO particles (350 μm kernel) used in this research. The mean diameter for these particles was 780 μm. The size of the overcoated particles was determined by an automated shadow scope. Fig. 10 shows the size and shape data for this batch of particles after overcoating via the centrifugal method. The mean diameter of the overcoated particles was 1105 μm, which was within 0.5% of the target overcoat thickness value for a 35% loaded compact.

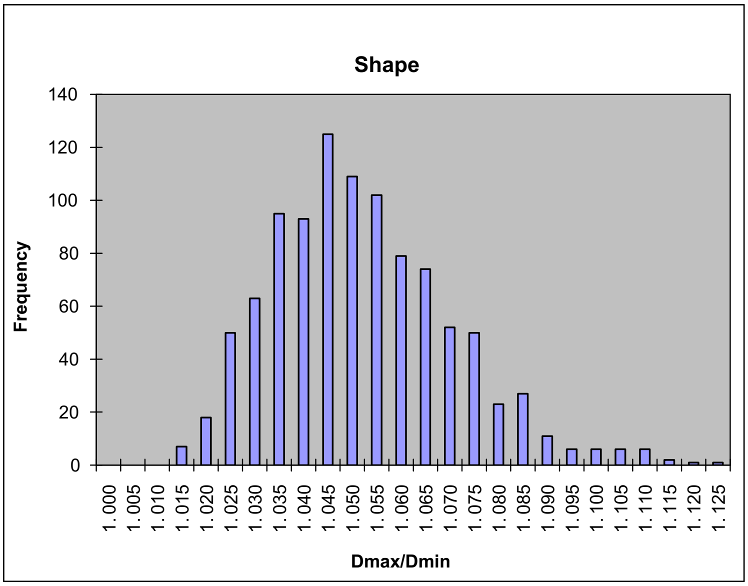
Overcoated particles of the desired outer diameter and overcoat layer thickness had now been successfully produced. The remaining tasks at hand were to increase the efficiency of the overcoating step and determine the best method for preparing the overcoated particles for compacting. A decision was made at this time concerning which combination of natural graphite, synthetic graphite, and resin was best for overcoating. It was found that the natural graphite GTI-NFM, synthetic graphite KRB2000, and resin Durite SC1008 produced the optimal matrix combination in terms of efficiency of overcoating. This matrix was identified as GKrS and the date on which it was made. For example, a matrix batch prepared on December 1, 2005 would be identified as GKrS 12105. In addition to final selection of matrix constituents, the process variables of quantity of TRISO particles, quantity of matrix, volume of methanol, angle of the overcoater, and speed of overcoating, were also determined experimentally. They are discussed in the Baseline and variant compact fabrication sections.

4.3. Compacting method development

Once the overcoating method had successfully produced overcoated particles with outer diameters required to meet the fuel particle packing fraction, compacting of those overcoated particles was initiated. The AGR-1 experiment called for four variants of the TRISO coating process and, therefore, four batches of compacts. The four batches of compacts were baseline (ORNL identification LEU01-46T), variant 1 (LEU01-47T), variant 2 (LEU01-48T), and variant 3 (LEU01-49T). The number of compacts needed for each

	Dmax/Dmin	Mean Diameter	St. Dev. In Diameter	Minimum Diameter	Maximum Diameter
Average	1.053	780	12	758	801
Standard Deviation	0.055	19	12	22	27
Minimum	1.011	728	2	569	743
Maximum	1.928	903	190	810	1124

Dmax/Dmin	Frequency
1	0
1.005	0
1.01	0
1.015	7
1.02	18
1.025	50
1.03	63
1.035	95
1.04	93
1.045	125
1.05	109
1.055	102
1.06	79
1.065	74
1.07	52
1.075	50
1.08	23
1.085	27
1.09	11
1.095	6
1.1	6
1.105	6
1.11	6
1.115	2
1.12	1
1.125	1
More	5



Mean Diameter	Frequency
730	3
740	23
750	45
760	71
770	121
780	203
790	249
800	176
810	79
820	35
830	3
840	1
850	0
860	0
870	0
880	1
890	0
900	0
910	1
More	0

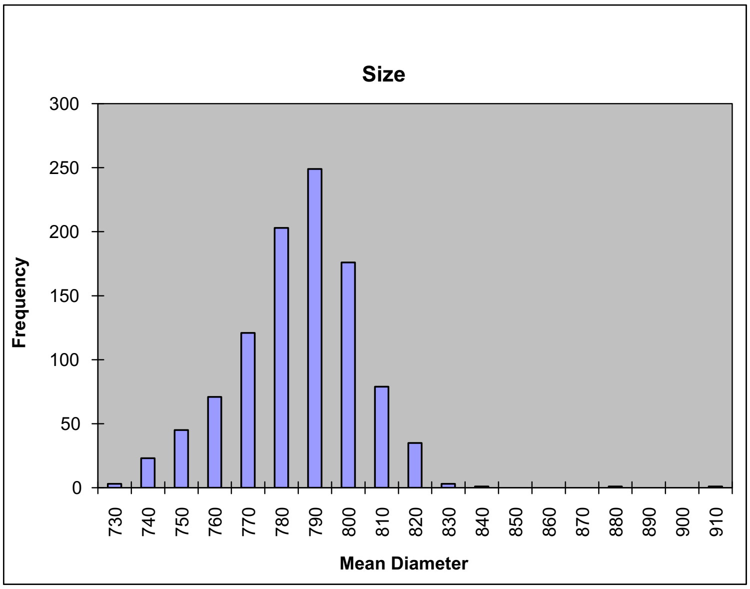


Fig. 9. Data set showing size and shape data for TRISO surrogates (all measurements in microns).

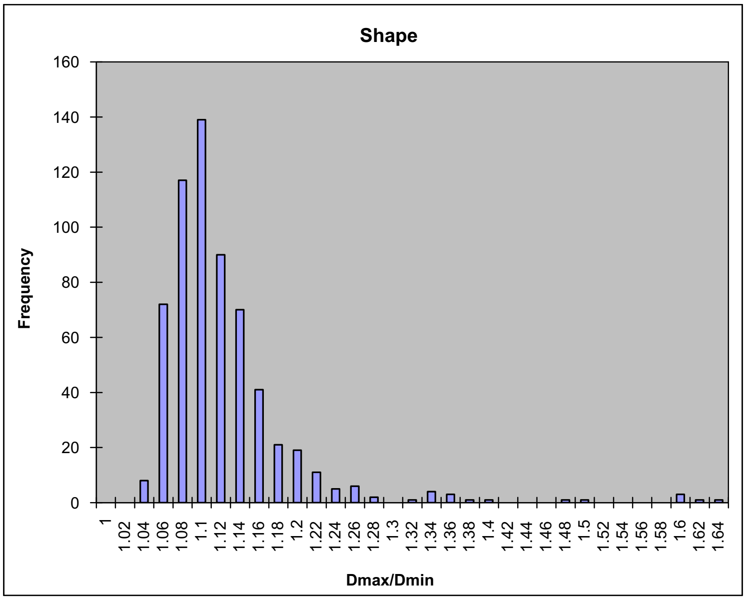
batch was 79, excluding variant 2, which required 67 compacts. The specification for uranium loading in the AGR-1 test articles was set at 0.905 ± 0.04 gU per compact. The method for determining the amount of uranium in the compact was LBL of a compact and subsequent ICP-AES or ICP-MS of the acid used in the leaching. The average mass of uranium per particle was known, but the number of particles being used to make a compact was not, as the number of particles being charged to a compact was not measured. The method for achieving a within specification uranium loading was a weight-per-overcoated particle approach, so a mass of overcoated particles could be weighed and used to form one compact.

4.3.1. Baseline fabrication

LEU01-46T TRISO (baseline) particles were received from the characterization group after having been characterized by performance inspection plan (PIP) 4, 'coated particle composites' [11]. Nineteen ~20g aliquots were prepared via riffling, and one aliquot

	Dmax/Dmin	Mean Diameter	St. Dev. In Diameter	Minimum Diameter	Maximum Diameter
Average	1.118	1105	36	1044	1171
Standard Deviation	0.103	61	29	61	93
Minimum	1.025	921	7	747	991
Maximum	2.477	1499	310	1218	2001

Dmax/Dmin	Frequency
1	0
1.02	0
1.04	8
1.06	72
1.08	117
1.1	139
1.12	90
1.14	70
1.16	41
1.18	21
1.2	9
1.22	11
1.24	5
1.26	6
1.28	2
1.3	0
1.32	1
1.34	4
1.36	3
1.38	1
1.4	1
1.42	0
1.44	0
1.46	0
1.48	1
1.5	1
1.52	0
1.54	0
1.56	0
1.58	0
1.6	3
1.62	1
1.64	1
More	3



Mean Diameter	Frequency
940	1
960	1
980	3
1000	5
1020	21
1040	38
1060	72
1080	86
1100	86
1120	79
1140	77
1160	63
1180	34
1200	22
1220	7
1240	11
1260	5
1280	2
1300	2
1320	1
1340	2
1360	0
1380	2
1400	0

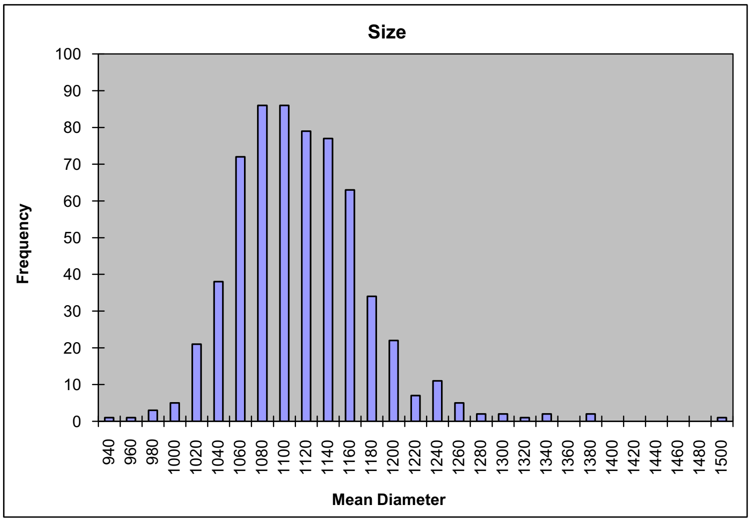


Fig. 10. Data set showing size and shape data for overcoated TRISO surrogates (all measurements in microns).

was used per overcoating run. Prior to overcoating, the particles were washed in methanol per procedure AGR-TRISOWASH-SOP-1 [12]. Washing of particles prior to overcoating was adapted in order to help reduce the amount of contamination on the particles that may have been acquired during processing or general handling. The washing procedure was adopted from general atomics particle washing procedures.

After washing, the LEU01-46T particles were overcoated using matrix batch GKrS 121405. All of the aliquots were overcoated producing 396 g of +18 particles. '+18' particles are those that pass through an ASTM E11 No. 16 sieve (1.18 mm) but do not pass through and ASTM E11 No. 18 sieve (1.00 mm). This quantity of +18 particles was insufficient (based on assumed weight-per-overcoated particle data and past overcoating experience) to produce 79 compacts, so −18 overcoated particles (those particles that passed through an ASTM E11 No. 18 sieve) were washed, in order to remove the overcoat, and subsequently re-overcoated. Prior to washing off the overcoat, overcoating of the −18 particles was performed in an attempt to increase the overcoat thickness enough such that +18 overcoated particles would be produced. However, the +18 overcoated particles achieved in this re-overcoating manner were different in color than the +18 overcoated particles achieved in a standard overcoating run, and were therefore considered unacceptable. Overcoated particle color is not specified in the overcoating procedure, but the author felt it was better to overcoat all the TRISO particles in the same manner in order to produce overcoated particles with as similar as possible properties. A standard overcoating run is therefore when TRISO particles (as opposed to -18 particles that have already been partially overcoated) are overcoated until +18 particles are achieved.

Overcoating of reclaimed TRISO particles (TRISO particles from washed overcoated particles) was continued until 537 g of +18 particles was produced. This quantity of +18 particles was then tabled and 410 g of 'Bin 3' +18 particles were recovered. Bin 3 particles are those particles that end up in the third bin of the tabler; these are the most spherical of the +18 particles. 410 g of Bin 3 +18 particles were needed because preliminary calculations showed that this would be a sufficient quantity to produce at least 79 compacts. A Bin 3 yield of 77% was observed, thus a total of 537 g of +18 particles was produced in order to achieve the 410 g quantity needed to fabricate 79 compacts.

Following tabling, the 410 g quantity of +18 Bin 3 overcoated particles was riffled into aliquots for weight per particle testing, via AGR-CHAR-DAM-22 [13], and compacting charges. Data Acquisition Method 22 (DAM-22) calls for ten aliquots of particles to be riffled out and for five of the ten aliquots to be counted and weighed. Combining the weight-per-particle data with the uranium content of each particle showed that 4.8156 g of +18 particles were needed to produce a compact with a uranium loading of 0.905 g. This value was checked by taking the five aliquots of particles that were not used in counting, combining them, and removing the overcoat by washing with methanol. The quantity of TRISO particles after washing was weighed, thus a grams-TRISO-per-grams-overcoated particles (gTRISO/gOvercoated particles) value was obtained. The gTRISO/gOvercoated particles value showed that 4.8668 g of overcoated particles was needed to produce a compact with a uranium loading of 0.905 g. The authors decided to select a charge on the side of a higher uranium loading (based on the weight-per particle approach), so the weight of overcoated particles needed to produce one compact was set at 4.8600 g. A greater overcoated particle charge is also useful during compacting as it increases the volume of overcoated particles in the mold, which allows an adequate compacting force to be achieved while making a compact of the specified length. An adequate compacting force is needed to produce a compact that is fully dense and smooth, but this need for force must be balanced with compact length, as a compact that has seen slightly too much force will be shorter than the lower length limit specified in engineering design file (EDF) 4380 [14]. The +18 particles were then riffled into aliquots of approximately 4.50 g. One jar of +18 particles with a mass of ~40 g was used to supplement each 4.50 g aliquot until the desired 4.8600 g charge was achieved.

Compacts were then made from the 4.8600 g aliquots. Each 4.8600 g aliquot of overcoated particles was placed in a sealed container rich in methanol vapor prior to being pressed into cylindrical compacts. The objective of this exposure to methanol vapors was the saturation of the overcoat layer with a (perceived) physisorbed monolayer of methanol. This monolayer of methanol aided in the pressing of the overcoated particles by increasing the malleability of the overcoat, thus allowing it to deform more easily into void spaces between particles. Fig. 11 shows an image of the outer surface of a compact that was formed without overcoated particle exposure to methanol prior to compacting. Notice the pits and open spaces between particles because the overcoat did not effectively flow under pressure into the inter-particle spaces. Fig. 12 shows a compact whose overcoated particles were saturated with methanol prior to compacting. The surface finish of this compact appears smoother and uniform because the overcoat was more malleable and able to fill the void spaces between particles.

The ease with which the overcoat could flow during compression was found to be crucial in avoiding TRISO particle touching and subsequent particle layer cracking. The rearrangement of TRISO particles and maximum achievable packing fraction are discussed by Morris and Pappano [15]. It was found that particle-particle touching was possible if the overcoat layer was not wetted with methanol, and that this touching led to particle cracking. Moistening of the overcoat layer essentially allowed the TRISO particle and overcoat layer to move independently of one another,

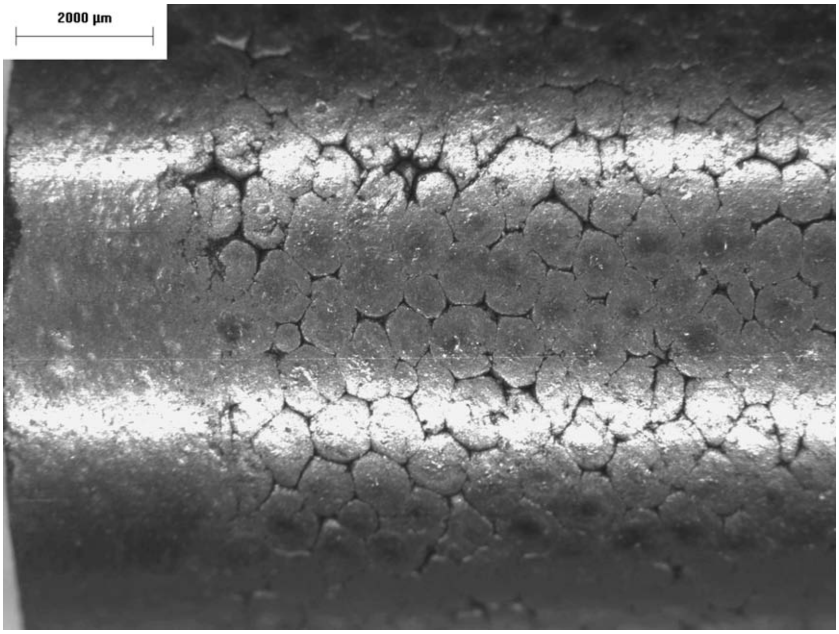


Fig. 11. Image of compact side showing incomplete pressing due to lack of methanol saturation in the original overcoated particles.

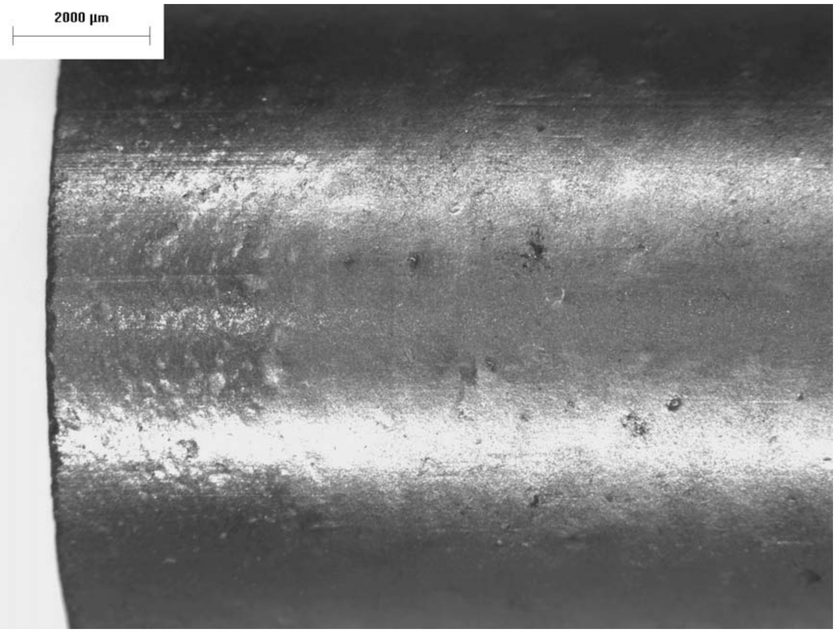


Fig. 12. Image of compact side showing complete compression due to proper saturation of overcoated particles prior to compacting.

which promoted the filling of voids between particles with overcoat. Morris and Pappano [15] also provides a mathematical model for determining the maximum packing fraction in a compact.

After saturation in methanol the overcoated particles were ready for compaction. Eight four baseline compacts were fabricated in total so that any compacts that may have been damaged during handling could be removed. All of the 84 green compacts were subsequently carbonized and heat-treated. Table 3 provides the carbonization and heat-treatment conditions. Seventy nine compacts were selected from this batch of 84 and delivered to the characterization group for testing. The selection of the 79 compacts for the characterization group was based on length measurements at each stage of compacting (green, carbonization, and heat-treatment), and visual inspection for surface defects. Compacts that were too small or had surface defects were not selected for characterization.

Table 3
Compact carbonization and heat-treatment conditions

Carbonization parameters	<350 °C/h in He
	Hold at 950 ± 50 °C for 1.0 ± 0.4 h
	Furnace cool
Heat-treatment parameters	20 °C/min in vacuum
	Hold at 1650–1850 °C for 60 ± 10 min
	Furnace cool at 20 °C/min to below 700 °C

4.3.2. Variants 1–3 fabrication

The fabrication process of variants 1–3 were performed in the same manner as described for baseline compacts in Section 3.3.1. In general, the coated particles were characterized and then released for compacting. The compacting group then washed the particles and overcoated them with GKrS matrix. The overcoated particles were then sized using sieves and tabled in order to extract the most spherical overcoated particles available. The sieved and tabled overcoated particles were then riffled into aliquots for compacting, where one aliquot was used to form one compact. The overcoated particles were saturated with methanol vapor prior to compacting in order to increase the malleability of the overcoat and thus allow it to more effectively migrate and fill inter-particle spaces during compacting. Table 4 provides a summary of the relevant overcoating and compacting data used in fabricating the AGR-1 test articles.

4.4. Characterization of baseline and variant 1–3 compacts

Sample compacts from four compacting campaigns were analyzed by LBL. Twelve compacts from each campaign were analyzed

Table 4
Overcoating and compacting summary table

Item	LEU01-46T (baseline)	LEU01-47T (variant 1)	LEU01-48T (variant 2)	LEU01-49T (variant 3)
g/TRISO	0.000727	0.000733	0.000724	0.000726
Diameter (µm)	799.7	804	798	795
Overcoating matrix	GKrS 121405	GKrS 030306	GKrS 030906	GKrS 051606
g of +18 particles, total	535	444	371	462
g of +18 particles, Bin 3	410	424	358	415
Tabler yield (%)	77	96	96	90
g/overcoated particle	0.00116	0.00112	0.00119	0.00121
Compacting charge wt/particle (g)	4.8156	4.6495	4.9401	5.0232
Compacting charge (gTRISO/gOvercoated particle)	4.8668	4.7474	4.9762	5.1083
Compacting charge used (g)	4.8600	4.7300	4.9400	5.0230
No. of compacts fabricated	84	85	71	82
No. of compacts needed for AGR-1	79	79	67	79

Table 5
Ninety five percent confidence prediction of the maximum mean impurity content for each compact lot, in μg/compact[a]

Measured impurity	Baseline	Variant 1	Variant 2	Variant 3
Fe	2.3	1.7	3.4	3.7
Cr	1.6	1.6	0.5	0.2
Mn	0.2	0.3	0.1	0.1
Co	0.6	0.7	0.2	0.1
Ni	1.3	1.3	3.2	1.3
Ca	10.2	9.4	18.3	21.3
Al	9.9	9.2	14.0	8.6
Ti	8.1	11.6	12.5	17.6
V	19.9	20.7	21.8	23.1

[a] Average compact mass = 5.5 g.

Table 6
Ninety five percent confidence prediction of the maximum defect fractions for each compact lot[a]

Property	Baseline	Variant 1	Variant 2	Variant 3
Exposed kernel fraction	$\leqslant 3.1 \times 10^{-5}$ (0/99470)	$\leqslant 4.1 \times 10^{-5}$ (0/74699)	$\leqslant 3.1 \times 10^{-5}$ (0/99110)	$\leqslant 3.1 \times 10^{-5}$ (0/99032)
Defective SiC coating fraction, after compacting	$\leqslant 1.3 \times 10^{-4}$ (2/49735)	$\leqslant 6.1 \times 10^{-5}$ (0/49799)	$\leqslant 9.6 \times 10^{-5}$ (1/49555)	$\leqslant 6.1 \times 10^{-5}$ (0/49516)
Defective SiC coating fraction, before compacting	$\leqslant 2.5 \times 10^{-5}$ (0/120688)	$\leqslant 4.0 \times 10^{-5}$ (1/121117)	$\leqslant 9.5 \times 10^{-5}$ (1/50265)	$\leqslant 4.0 \times 10^{-5}$ (1/120660)

[a] Values in parentheses are the actual measured defects over the number of particle in the analyzed compacts.

in sets of three. An additional twelve compacts (six for variant 1) from each campaign were deconsolidation-leached only, to increase the sample size for measurement of the exposed kernel fraction.

Table 5 gives the 95% confidence prediction for the maximum mean impurity content in each compact lot. This is calculated from the measured means and standard deviations using one-sided student-*t* statistics. Calcium content was abnormally high and was likely due to contamination from ceiling tile dust during fabrication and handling.

Table 6 gives the upper limit of the 95% confidence interval of the defect fraction for exposed kernels. This confidence interval is calculated using binomial distribution statistics. The values in Table 6 would be the lowest tolerance limits for which the compact lot would be deemed acceptable at 95% confidence based on the particular sample of compacts that was measured. Note that no exposed kernels were detected in any of the compact lots and there was no other indication that this low pressure compacting process will break particles. However, due to the limited sample size, the maximum defect fractions reported in Table 6 are the lowest that can be supported by this LBL data.

Table 6 gives the upper limit of the 95% confidence interval of the defect fraction for particles with defective SiC before and after compacting. No defective SiC was detected in variants 1 and 3. Baseline showed two defective particles and variant 2 showed one. The observed fraction of particles with defective SiC in variant 2 is as expected, given the defective SiC fraction determined for the particles before compacting. The baseline defective SiC fraction appears to have increased due to the compacting process. This may be due to cracking of the SiC during compacting on particles with abnormally thin regions of SiC (<15 μm as opposed to a normal thickness of 35 μm).

5. Conclusions

The AGR TRISO particle and compact fabrication program at ORNL successfully reinstated the US capability for producing particle fuel and compacts. AGR-1 compacts were fabricated via the novel overcoating and compacting method described here. The overcoating process involved fabricating a matrix of natural graphite, synthetic graphite, and thermosetting resin via a wet mixing method. Compacts were formed by a single acting compression molding press. The compacts were shown to be within specification, in terms of uranium loading and impurities content. The AGR-1 test articles are currently being irradiated in the ATR at INL. Results of this irradiation testing should be available in the spring of 2009.

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