Screening of Advanced Cladding Materials and UN-U₃Si₅ Fuel

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Abstract

In the aftermath of Fukushima, a focus of the DOE-NE Advanced Fuels Campaign has been the development of advanced nuclear fuel and cladding options with the potential for improved performance in an accident. Uranium dioxide (UO₂) fuels with various advanced cladding materials were analyzed to provide a reference for cladding performance impacts. For advanced cladding options with UO₂ fuel, most of the cladding materials have some reactivity and discharge burn-up penalty (in GWd/t). Silicon carbide is one exception in that the reactor physics performance is predicted to be very similar to zirconium alloy cladding. Most candidate claddings performed similar to UO₂-Zr fuel-cladding in terms of safety coefficients. The clear exception is that Mo-based materials were identified as potentially challenging from a reactor physics perspective due to high resonance absorption. This paper also includes evaluation of UN-U₃Si₅ fuels with Kanthal AF or APMT cladding. The objective of the U₃Si₅ phase in the UN-U₃Si₅ fuel concept is to shield the nitride phase from water. It was shown that UN-U₃Si₅ fuels with Kanthal AF or APMT cladding have similar reactor physics and fuel management performance over a wide parameter space of phase fractions when compared to UO₂-Zr fuel-cladding. There will be a marginal penalty in discharge burn-up (in GWd/t) and the sensitivity to ¹⁴N content in UN ceramic composites is high. Analysis of the rim effect due to self-shielding in the fuel shows that the UN-based ceramic fuels are not expected to have significantly different relative burn-up distributions at discharge relative to the UO₂ reference fuel. However, the overall harder spectrum in the UN ceramic composite fuels increases transuranic build-up, which will increase long-term activity in a once-thru fuel cycle but is expected to be a significant advantage in a fuel cycle with continuous recycling of transuranic material. It is recognized that the fuel and cladding properties assumed in these assessments are preliminary, and that additional data are necessary for these materials, most significantly under irradiation.

Introduction

Over the past decade, BNL has developed and benchmarked a methodology to perform an initial screening of the performance and safety of candidate fuel concepts in support of the Advanced Fuels Campaign (AFC) and its predecessors, recognizing that the fuel and reactor are an intimately linked system. The resultant methodology provides the capability to screen candidate fuel concepts, to address the desired objective of ensuring that a proposed concept has at least equivalent performance and safety characteristics relative to current uranium dioxide (UO₂) fuel and zirconium alloy (Zr) cladding.

After the Fukushima accident, a major focus of the AFC has been on fuels with enhanced accident tolerance, a.k.a. “Accident Tolerant Fuels (ATF)”. The key objectives that an attractive and viable ATF must satisfy include:

“First do no harm” → at least comparable performance under all conditions, and improved performance in accident scenarios

Preserve/Improve Characteristics for Normal Operation, e.g.:

- Burnup/Cycle length (criticality, fuel performance)
- Operations (power distribution, peaking factors, margins, etc.)
- Reactivity coefficients and control (shutdown margin, rod worths)
- Handling, transportation, storage (isotopics, dose)
- Compatibility with infrastructure (economics)

To warrant the label of “ATF”, the proposed concept must have improved response in the full spectrum of transients/accidents:

- Anticipated Operational Occurrences (AOOs)
- Design Basis Events (DBE), e.g., LOCA, RIA
- Beyond Design Basis Events (BDBE), e.g., Fukushima

To address these objectives, a key activity has been the development of metrics that can be used to evaluate candidate ATF fuel concepts (Bragg-Sitton et al., 2014). While many of the metrics address specific fuel and cladding characteristics and performance requirements, there is an explicit recognition of the need to consider performance of advanced concepts in the reactor and holistically within the entire system (Bragg-Sitton et al., 2014):

“An assessment of the potential beneficial impact or unintended negative consequences of candidate ATF concepts must address the obvious “fuel-specific” characteristics of the concept but, perhaps more importantly, the assessment must address how implementation of the concept will affect reactor performance and safety characteristics.”

The search for attractive and viable ATF candidates requires consideration of a broad spectrum of potential fuel and cladding options that are targeted at satisfying the requirements specified in Bragg-Sitton et al. (2014). To be attractive to utilities and vendors, proposed concepts should offer performance and safety benefits under normal operating conditions and AOOs and DBEs, as well as BDBEs. A screening methodology of candidate concepts is therefore essential to optimize the allocation of time and resources since not all proposed concepts can be pursued through the required analytic and experimental assessment and validation process. This is further exacerbated by the ultimate objective of testing of prototypic ATF concepts in commercial reactors, which will involve acceptance by a utility, and review and approval by the U.S. Nuclear Regulatory Commission (NRC).

The methodology developed, benchmarked, and utilized by BNL for screening of advanced concepts, described below, achieves these goals by considering the key analyses that need to be performed, and the required level of fidelity. The key elements of the methodology (as shown in Figure 1) are:

Initial Screening Analyses: Infinite lattice calculations for an explicit model of a fuel assembly to provide an estimates of the impact of the new fuel relative to the reference UO₂-Zr fuel-cladding on cycle length/burnup, reactivity and control coefficients, etc.

Three-Dimensional Core Analyses: Core analyses utilize a full three-dimensional model of the reactor core including thermal-hydraulic and temperature feedback. These can be used for fuel cycle analyses (cycle-by-cycle or equilibrium cycle) and time-dependent accident analyses such as for a rod ejection accident.

Transient Analyses: The analysis of selected transients is performed with reactor systems analysis tools using point kinetics to represent the reactivity response, or coupled to a three-dimensional core analysis model for reactivity driven transients.

Each of these synergistic elements represents a key component of the answer to the question: *what is the reactor performance impact for a particular concept?*

Figure 1. A Synergistic View of Reactor Performance and Safety Analyses for Candidate ATFs

The analyses described in this paper focus on implementation scenarios that would require minimal changes to the fuel assembly and/or operations to facilitate the ability to incorporate the ATF into current LWRs. We refer to these as “drop-in” because the hydraulic diameter, cladding outer diameter, and assembly geometry are preserved allowing implementation of a proposed concept in the cores of operating reactors for testing and eventual incorporation as partial or full-core loadings. We considered that the cladding thickness may be modified in a potential “drop-in” configuration, with the radius of the fuel pellet increased accordingly. Cladding thickness and pellet outer diameter were variable, but gas gap thickness and cladding outer diameter were not modified.

This paper considers several advanced cladding options, with potential benefits including improved oxidation kinetics with steam and enhanced high temperature strength. In addition, uranium nitride-based ceramic composites fuels are considered. Potential benefits of nitride-based ceramic composite fuels include significantly increased uranium

density (~40%), enhanced thermal properties, and the potential to integrate burnable absorbers into the fuel (Brown et al. 2014). Uranium nitride reacts with water, but in these ceramic fuels the primary nitride phase is “shielded” from water by a secondary phase, which would allow the potential benefits of nitride fuels to be realized. Secondary phases, such as uranium silicides (U_3Si_2 and/or U_3Si_5) and/or uranium borides (UB_2 and/or UB_4) among other options, might potentially “shield” the nitride phase. Efforts to identify the most promising UN-based ceramic fuel options are presently underway within the AFC. The focus of this paper is on UN- U_3Si_5 fuels, which are being studied by Los Alamos National Laboratory.

Screening of the impacts of advanced fuels/ATF can be accomplished under two basic “boundary conditions”:

In the “reference/baseline” case, accommodate the differences in fuel-cladding characteristics such that performance and operations are unchanged. For the fuel and cladding combinations considered:

- The higher heavy metal density of UN vs. UO_2 allows a reduction in the enrichment required for the same cycle length, but will result in reduced burn-up (in GWd/t) due to the increased initial heavy metal mass.
- Fe- or Mo- based cladding would need to be thinner than zirconium alloy because of the neutronic penalty due to the higher absorption. This allows the option of a larger fuel pellet. The higher heavy metal density and larger fuel pellet enables UN-based fuels to offset the reactivity penalty of Fe-based cladding materials and match the cycle length of UO_2 -Zr.
- The harder neutron spectrum with some fuel and cladding options reduces the worth of thermal absorbers (e.g., boron) and affects some reactivity coefficients. However, the impact on reactivity coefficients is similar to that with U-Pu mixed-oxide (MOX) fuel.

An alternative approach would take advantage of the advanced fuel/cladding characteristics to potentially improve performance and/or operations while still preserving many/most of the “drop-in” benefits:

- An obvious benefit with the nitride-based fuels is the potential for longer cycle lengths (or increased number of batches) for the same enrichment due to the higher heavy metal density. However, the burn-up (in GWd/t) will be reduced for most concepts.
- The option of smaller batch sizes for refueling (for example four-batch versus conventional three-batch for PWRs) may have operational and economic benefits

If the above constraints are removed/relaxed, there are additional opportunities for optimization, e.g., optimizing the fuel-to-moderator ratio within the current rectangular fuel assembly envelope. However, modifying the fuel rod outer diameter could significantly impact implementation and licensing due to the mechanical/thermal-

hydraulic testing that would likely be required. Changing to different lattice geometry entirely (e.g., hexagonal) would entail additional issues.

Assembly-Level Analysis of Candidate Fuel and Cladding Materials

We analyzed several ATF concepts in previous efforts and we have applied lessons learned from previous assessments of candidate ATFs to this work (Carmack, 2006; Brown et al., 2013 a, b, c; Brown et al. 2014). The assembly calculations described in the present paper were used to inform on key operational and safety parameters, like cycle length, discharge burn-up, and reactivity coefficients. Unless stated otherwise, the assembly calculations assumed a three-batch fuel management scheme at the assembly power of a reference PWR. The linear reactivity model was used to estimate the cycle length, assuming a 3% core leakage and no burnable absorber loading or soluble boron in the coolant. It is noted that the fuel management scheme, core leakage, absorber loading, and soluble boron all impact the achievable cycle length. The objective of this approach is to compare the reactor performance of ATF options to the reference UO₂ fuel and Zr cladding on a common basis. The majority of calculations in this document assumed an enrichment of approximately ~4.9%. The average power per pin was maintained for all cases and an 18 month cycle length was maintained.

The primary lattice physics tool utilized for analysis of the proposed composite fuels on the assembly level and for the generation of few-group constants for subsequent 3-D core analyses was TRITON/NEWT, from the SCALE 6.1 package (Oak Ridge National Laboratory, 2011). ENDF/B-VII.0 cross sections were utilized for the majority of the analyses in this report. TRITON/NEWT has been widely used by the U.S. NRC for independent regulatory analyses and therefore can be considered a regulatory-grade reactor physics tool.

On the full-core level, the Purdue Advanced Reactor Core Simulator (PARCS) was utilized as the core simulator in this study (Downar et al., 2013). PARCS is state-of-the-art and the official U.S. Nuclear Regulatory Commission (NRC) core simulator.

For many of the analyses we utilized a somewhat simplified assembly and core design based on the Westinghouse AP1000 (Westinghouse, 2009) as the reference core for the initial assessment of the operational and safety performance of the composite fuel concepts. The key parameter maintained for the composite fuels is the power per pin. The basis for some of the evaluation parameters in this document is the Final Safety Analysis Report (FSAR) for the reference PWR (Westinghouse, 2009). A visualization of the reference PWR fuel assembly and 1/4-core is shown in Figure 2.

Figure 2. Planar Slice of an LWR Core Model Showing Fuel Assembly Detail

Analysis of Advanced Candidate Cladding Materials with UO_2 Fuel

In an effort to better understand the potential neutronic penalty associated with various alternative claddings (Kanthal AF, Kanthal APMT, SiC, Mo), we performed assembly calculations for UO_2 fuel with Zr and alternative claddings in two different configurations (one standard, and one with thinner cladding and a bigger fuel pellet). These configurations are shown in Figure 4. The gas gap (not shown explicitly) was retained at a constant thickness for all of these options. The “thin cladding geometry” is based on more than a decade of operational experience with Fe-based cladding in LWRs (primarily SS304). It is noted that SS304 experienced very low failure rates, especially in PWRs (Terrani et al., 2014). Due to this operating experience, an assumed cladding thickness of 0.0419 cm can be considered reasonable for the Fe-based cladding materials. It is vital to note that some of the advanced cladding materials could have significantly reduced thickness due to favorable properties, including high temperature strength. The assumed thicknesses are may be conservative, and are a tool to compare the potential reactor physics impacts of various cladding materials.

Results indicated penalties for AF, APMT and Mo and similar performance (with respect to Zr) for SiC, as shown in Table 1. Cycle lengths are reported in Effective Full Power Days (EFPD). These calculations with advanced cladding materials and UO_2 fuel provide reference cases for the impact of cladding materials on reactor physics performance. Most of the advanced claddings will have *some* burn-up performance penalty (exception is SiC). Thinner cladding generally equates to better performance, but even for thin cladding Fe- or Mo- based options may have some penalty vs. Zr. At a fuel rod radius of 0.4249 cm and a cladding thickness of 0.0419 cm the achievable burn-up with UO_2 -AF and UO_2 -APMT is 84% and 81% of the nominal UO_2 -Zr values. An appropriate and

acceptable thickness for cladding needs to be determined for each advanced cladding based on mechanical and thermal characteristics to have better confidence in the trade-offs associated with these alternate cladding materials. It is also vitally important to understand the impact of irradiation on mechanical and thermal characteristics of potential cladding materials. Key material characteristics, like strength, are a function of not only temperature but introduction of cold working and neutron fluence (especially fast fluence). This is also significant due to the hardened neutron spectrum with some of these candidate cladding materials, which will increase fast fluence.

Figure 4. Fuel Geometries Used for Advanced Cladding Scoping Calculations (Gas Gap is Not Shown)

Table 1. Three-Batch Cycle Length for Various Advanced Cladding Materials with UO_2 Fuel

4.9%-enriched UO_2 fuel - 0.4096 cm pellet radius – 0.0572 cm cladding thickness					
Fuel - Clad	$\text{UO}_2\text{-Zr}$	$\text{UO}_2\text{-SiC}$	$\text{UO}_2\text{-AF}$	$\text{UO}_2\text{-APMT}$	$\text{UO}_2\text{-Mo}$
Discharge burn-up (GWd/t)	61.6	63.2	48.1	47.2	37.6
Cycle length (EFPD)	533	547	416	408	326
4.9%-enriched UO_2 fuel - 0.4249 cm pellet radius – 0.0419 cm cladding thickness					
Fuel - Clad	$\text{UO}_2\text{-Zr}$	$\text{UO}_2\text{-SiC}$	$\text{UO}_2\text{-AF}$	$\text{UO}_2\text{-APMT}$	$\text{UO}_2\text{-Mo}$
Discharge burn-up (GWd/t)	61.8	62.9	51.9	49.9	42.9
Cycle length (EFPD)	576	586	483	465	400

Assembly level reactivity coefficients were also calculated for the candidate cladding materials as a function of burn-up. The differences in the reactivity coefficient results are primarily due to resonance absorption in some of the proposed advanced cladding materials. All calculations assume a three-batch fuel management scheme with 3% core neutron leakage and no burnable absorber or soluble boron in the coolant.

To illustrate the differences driven by the resonance absorption, the moderator temperature coefficient, including both spectral and density components, is shown in Figure 6 for $\text{UO}_2\text{-Zr}$, $\text{UO}_2\text{-SiC}$, $\text{UO}_2\text{-AF}$, $\text{UO}_2\text{-APMT}$, and $\text{UO}_2\text{-Mo}$. Most of the cladding options are very similar to the reference $\text{UO}_2\text{-Zr}$ system, but the clear outlier is Mo. The reason for this is that Mo is a very strong resonance absorber. This is illustrated

in Figure 8, which shows the absorption reaction spectra for several elements proposed as components of advanced cladding. It is noted that Si and C are not shown, but they are relatively neutron transparent like Zr and Al. The elemental order of the cumulative absorption rate is: Mo > Ni > Cr > Fe >> Zr > Al. Figure 10 shows the neutron capture cross section for several Mo isotopes, and the strong resonances which impact the moderator temperature coefficient so drastically. These resonances indicate why Mo-based claddings are unattractive cladding materials from the reactor physics perspective. It is noted that such a strongly negative moderator temperature coefficient may impact shutdown margin. Reducing the thickness of Mo would reduce this impact, but resonance absorption in Mo cannot be mitigated entirely because of the underlying physics. It is also recognized that isotopic enrichment is a potential way to mitigate the impact of thermal neutron absorption with Mo cladding. However, almost all of the isotopes of Mo are strong resonance absorbers. Mo is not a light element, and the economic impact of isotopic enrichment might be significant.

Figure 6. Moderator Temperature Coefficient as Function of Burn-Up (GWd/t)

Cumulative absorption rate: Mo > Ni > Cr > Fe >> Zr > Al

Figure 8. Absorption Reaction Spectra for Several Components of Candidate Advanced Cladding Materials

Figure 10. Neutron Capture Cross Section for Mo Isotopes

The strong neutron absorption rates of some advanced cladding materials gives rise to a key question: *how is the cladding material transmuting due to neutron absorption?* These assembly level calculations can be leveraged to provide an answer to this key question. The results in Figure 7 focus on APMT cladding, with the atom density of various cladding materials shown as a function of irradiation time in EFPD. The key transmutation reactions include the transmutation of ^{54}Fe to ^{55}Fe , ^{56}Fe to ^{57}Fe , and ^{95}Mo to ^{96}Mo . One important result is that hydrogen is generated in the cladding (in very small amounts).

Figure 12. Transmutation of Kanthal APMT Cladding in a Light Water Reactor with UO_2 Fuel

Impact of Fe-Based Cladding on Fuel Assembly Neutronic Performance with UN- U_3Si_5 Fuel

Several advanced iron-based claddings are under consideration due to increased oxidation resistance versus zirconium-based cladding. We analyzed these claddings to inform the planned ATF irradiation test of UN- U_3Si_5 fuel. In this subsection, the UN phase constitutes 80% of the fuel volume, the U_3Si_5 phase 10% of the fuel volume, and the porosity is 10%. It is important to note that differences in neutronic performance for UN- U_3Si_5 composite fuels relative to conventional UO_2 fuel have a first-order dependence on the respective effective heavy metal densities. We also analyzed SS304 and SS316, both of which have a significant operational history in reactors. In addition, we considered two commercial alloys, Kanthal AF (referred to as AF) and Kanthal APMT (referred to as APMT). The cladding thickness for the predominantly iron-based cladding materials is 0.0419 cm versus 0.0572 cm for zirconium-based cladding. The fuel pellet thickness is increased, maintaining the cladding outer diameter and the gap thickness but increasing the fuel pellet diameter and therefore the heavy metal loading. The reduced cladding thickness is based on the significant operational experience with iron-based cladding, such as SS304. Our analyses show that there is a large reactivity penalty due to these advanced claddings as shown in Figure 13. The reactivity coefficients are shown in

Figure 15 - Figure 19. It is notable that the behavior of all of the Fe-based options are similar.

However, in terms of cycle length, with three-batch fuel management and nitride-composite fuels this reactivity penalty is compensated via the higher heavy metal loading and increased fuel pellet thickness, as shown in Table 3. The increased heavy metal density of UN-based fuels coupled with the increased pellet radius hardens the neutron energy spectrum due to a lower moderator-to-fuel ratio. One consequence of this is decreased worth of soluble boron in the coolant, as shown in Figure 11. The increased heavy metal loading acts to significantly decrease the discharge burn-up per unit heavy metal mass in a three-batch scheme. In addition, it is noted that UN-U₃Si₅ with zirconium alloy cladding can nearly match UO₂-Zr three-batch cycle lengths in a four-batch fuel management scheme, but that the proposed advanced cladding materials eliminate this potential fuel management and economic advantage.

Table 3. Three- and four-batch cycle length and discharge burn-up for various advanced fuel/cladding options at 4.9%-enriched ²³⁵U.

Fuel	UO ₂ - Zr	UN - U ₃ Si ₅ - Zr	UN - U ₃ Si ₅ - AF	UN - U ₃ Si ₅ - APMT	UN - U ₃ Si ₅ - SS304	UN - U ₃ Si ₅ - SS316
Three-batch						
Discharge burn-up (GWd/t)	61.6	60.9	52.2	51.5	49.2	48.5
Cycle length (EFPD)	533	656	604	597	570	562
Four-batch						
Discharge burn-up (GWd/t)	65.7	64.3	54.7	54.0	51.5	50.8
Cycle length (EFPD)	426	519	475	469	448	442

Figure 13. Infinite Multiplication Factor as a Function of Burn-Up (EFPD) for Fe-Based Cladding and UN-U₃Si₅ Fuel

Figure 15. Fuel Temperature Coefficient as a Function of Burn-Up (GWd/t) for Fe-Based Cladding and UN-U₃Si₅ Fuel

Figure 10. Moderator Temperature Coefficient as a Function of Burn-Up (GWd/t) for Fe-Based Cladding and UN-U₃Si₅ Fuel

Figure 11. Soluble Boron Coefficient as a Function of Burn-Up (GWd/t) for Fe-Based Cladding and UN-U₃Si₅ Fuel

Figure 19. Control Rod Worth as a Function of Burn-Up (GWd/t) for Fe-Based Cladding and UN-U₃Si₅ Fuel

Scoping Analysis of UN – U₃Si₅ Fuel at Various Phase Fractions

The objective of this section is to summarize studies performed in support of the development of “drop-in” ATF candidates that minimize the impact on reactor operations and safety. Another constraint is to stay within the form factor of existing assembly designs in terms of cladding outer diameter, hydraulic diameter, assembly layout, and dimensions. Several approaches are possible to develop “drop-in” fuels - we focused on adjusting the volume percent of the constituent phases. It is noted that these ceramics may not be viable from the perspective of materials science. The focus of the effort was to inform the fuel designers about the range of phase fractions that, if viable, would most closely match the reactor performance of the UO₂-Zr system.

Another way to optimize a “drop-in” fuel would be to adjust enrichment. Reducing the enrichment of a high-density UN-based ceramic fuel can be used to match the operational cycle length of UO₂. However, the neutron energy spectrum will remain hard due to the increased fuel density, which impacts reactivity control and burnable absorber worth. It is important to note that increased heavy metal density has potential advantages, but also may require redesign of control rods, enrichment of soluble boron, and other changes to the operation of the reactor.

Perhaps most importantly, it is unknown what, if any, volume percent of U₃Si₅ phase is adequate to “shield” the UN phase from water. This analysis is informative in the sense that it describes performance over a range of phase contents and shows that many options

over a range of phase contents have performance similar to the UO_2 -Zr fuel-cladding system. This acts as a boundary condition for fuel designers to advise on the applicable design space.

A secondary reason for the focus on phase fractions was that our efforts have shown a significant spectral impact of high-density ceramics with UN as the primary phase. UN has a heavy metal density 40% greater than UO_2 . By contrast, the secondary phase of the fuels considered here, U_3Si_5 , has a heavy metal density 25% less than UO_2 . By increasing the volume percent of the U_3Si_5 phase, the spectrum hardening impacts of the UN phase are obviated and the reactor physics behavior of the fuel can enable close matching to that of UO_2 . However, higher heavy metal density may enable potential benefits; increased fraction of a secondary phase is one approach we considered in order to have a fuel that is “most similar” to UO_2 for retrofittability in existing reactors.

Potential “Drop-In” Fuel-Cladding Combinations

Several fuels were considered within these scoping studies. The UN- U_3Si_5 volume percent of each phase was varied with the constraint of 10% porosity. This porosity was selected to maintain a consistent basis for comparison versus existing studies. The volume of the UN phase was varied between 50% and 80%, with the U_3Si_5 phase varied between 40% and 10% (constituting the remainder of the available volume), respectively. These fuels were studied with zirconium alloy, Kanthal AF, and Kanthal APMT cladding. In addition, admixture of 0.25% UB_2 was considered as a third phase for reactivity control.

The cladding thickness for the AF and APMT cladding materials is 0.0419 cm versus 0.0572 cm for zirconium alloy cladding, and the fuel pellet diameter is increased accordingly to 0.4249 cm. The assembly power was assumed constant based on values for the reference PWR. The consequence of this is that the UN- U_3Si_5 fuels have reduced specific power density due to high heavy metal density, and the specific power density is further reduced by the increased fuel pellet diameter with AF and APMT cladding. Enrichment was assumed constant at approximately 4.9% for all cases and 100% enrichment in ^{15}N was assumed for these cases.

The three-batch cycle length (EFPD) is shown as a function of UN phase percent in Figure 13. Nearly all of the options studied are within 20% of the nominal UO_2 -Zr fuel-cladding system. The reactor performance behavior of AF and APMT cladding are very similar, with a small penalty for the APMT cladding due to small Mo content. If the objective is to closely match the UO_2 -Zr fuel-cladding system cycle length at a constant enrichment, the applicable phase contents are approximately 55% - 65% for UN in UN- U_3Si_5 with AF or APMT cladding or UN- U_3Si_5 - UB_2 with Zr cladding, or approximately 75% - 85% for UN in UN- U_3Si_5 - UB_2 with AF or APMT cladding. The reactivity impact of including a small percent of UB_2 phase is clear, as is the impact of AF or APMT cladding. It is notable that the impact of the Fe-based cladding is more significant than the impact of 0.25% of UB_2 phase content with natural boron. This illustrates the significant penalty due to parasitic absorption in these potential cladding materials.

Figure 13. Cycle Length in Effective Full Power Days as a Function of UN Phase Content (vol%) for UN-U₃Si₅ Fuel and Several Candidate Cladding Materials

Another significant result, the discharge burn-up in GWd/t versus the UN phase content in volume %, is shown in Figure 15. This illustrates, assuming a constant enrichment, the burn-up or resource utilization impact of various fuel and cladding combinations. It is evident that UN-U₃Si₅ fuel with zirconium alloy cladding can match the discharge burn-up in GWd/t of UO₂ with zirconium alloy cladding. Although it is possible to match UO₂ cycle lengths with UN-U₃Si₅ fuel and AF or APMT cladding, the burn-up penalty incurred is significant. The behavior of UN-U₃Si₅-UB₂ is interesting, given the positive slope of the trend lines. The reason for this is that as the UN content is increased, the reactivity worth of the UB₂ phase is decreased due to the hardening of the neutron spectrum. This can be further explained by observing the trends in the soluble boron coefficient shown in Figure 17 for UN-U₃Si₅-UB₂ with Zr cladding and Figure 19 for UN-U₃Si₅-UB₂ with APMT cladding. Although UB₂ is within the fuel, the physics is similar to the soluble boron coefficient. As the UN content is increased, the neutron energy spectrum hardens, and the reactivity importance of ¹⁰B, a thermal absorber, decreases.

Figure 15. Discharge Burn-Up in GWd/t as Function of UN Phase Content (%) for UN-U₃Si₅ Fuel and Several Candidate Cladding Materials

Figure 17. Soluble Boron Coefficient as a Function of Burn-Up (GWd/t) for Zr Cladding and UN-U₃Si₅-UB₂ Fuel

Figure 19. Soluble Boron Coefficient as a Function of Burn-Up (GWd/t) for Kanthal APMT Cladding and UN-U₃Si₅-UB₂ Fuel

The assembly infinite multiplication factor and assorted reactivity coefficients were calculated for the various candidate materials as a function of burn-up. The infinite multiplication factor is shown in Figure 21 for UN-U₃Si₅ fuels with AF or APMT cladding that have similar physics performance to UO₂-Zr. The UN volume percent is shown in parentheses, with U₃Si₅ making up the remainder of the volume (assuming 10% porosity). The differences in the reactivity coefficient results are primarily due to the differences in the resonance absorption within the various composite fuels and advanced cladding materials. The fuel temperature coefficient (Figure 23) is similar and negative for all cases. As the UN content increases under-moderation increases, and the moderator temperature coefficient (including both density and spectral effects) becomes more negative (Figure 25). The resonance absorption in AF or APMT and/or UB₂ phase will also act to make the system more under-moderated. Similarly, the soluble boron coefficients become less negative due to the increased resonance absorption and spectral hardening (Figure 27).

Figure 21. Infinite Multiplication Factor as a Function of Burn-Up (EFPD) for Kanthal AF and APMT Cladding and UN-U₃Si₅ Fuel

Figure 23. Fuel Temperature Coefficient as a Function of Burn-Up (GWd/t) for Kanthal AF and APMT Cladding and UN-U₃Si₅ Fuel

Figure 25. Moderator Temperature Coefficient as a Function of Burn-Up (GWd/t) for Kanthal AF and APMT Cladding and UN- U_3Si_5 Fuel

Figure 27. Soluble Boron Coefficient as a Function of Burn-Up (GWd/t) for Kanthal AF and APMT Cladding and UN- U_3Si_5 Fuel

Key conclusions and observations from this effort include:

It appears feasible, solely from the perspective of the reactor physics, to closely match UO_2 -Zr fuel-cladding reactor physics behavior with UN- U_3Si_5 and AF or APMT cladding

Closely matching UO_2 -Zr fuel-cladding performance may or may not be a vital objective, but UN- U_3Si_5 exhibits similar reactor physics performance over a relatively wide range of UN and U_3Si_5 phase fractions

Adjusting fractions of a secondary phase informs fuel designers about the possible envelope for engineering a “drop-in” replacement for UO_2

Higher heavy metal loading of UN- U_3Si_5 fuels can compensate for the significant reactivity penalty associated with Fe-based cladding options in terms of the cycle length in EFPD (but not the discharge burn-up in GWd/t)

These scoping calculations assume a constant enrichment and assembly power, but adjusting enrichment as a parameter is important to consider as part of future studies

“Most similar” to UO_2 may not be best in terms of operational attractiveness, e.g. higher density fuels with zirconium alloy cladding could enable four-batch operation with an 18-month cycle length

Impact of ^{14}N (n,p) on Neutronic Performance

A similar approach was taken to study the impact of ^{14}N content on reactor performance. Nitrogen-14, which constitutes the bulk of natural nitrogen, is a thermal neutron absorber, producing ^{14}C via an (n,p) reaction. This cross section is shown in Figure 29. The (n,p) reaction is not a concern for ^{15}N . If natural nitrogen is used, the impact on the reactor performance, and probably fuel performance, is significant, as shown in Figure 31. This shows that, with natural nitrogen, the phase content of UN is on the order of $\sim 80\%$ if the objective is to match UO_2 cycle length. However, natural nitrogen is not credible in an LWR fuel. The penalty is clearly very significant, and some enrichment in ^{15}N must be performed. The use of ^{14}N would result in hydrogen generation in the fuel, as shown in Figure 33.

Figure 29. Cross Section for (n,p) in ^{14}N and ^{15}N

Figure 31. Cycle Length in Effective Full Power Days as a Function of UN Phase Content (vol%) for UN-U₃Si₅ Fuel with Natural Nitrogen and 100% Enrichment in ¹⁵N

Figure 33. Fuel Transmutation and Hydrogen Generation Due to (n,p) Reaction in ¹⁴N

Analysis of the Rim Effect and Transuranic Build-Up in High Density UN-based Ceramic Fuel

In a light water reactor, due to self-shielding within a fuel pin, the highest burn-up is achieved in the “rim” of the fuel rod adjacent to the cladding. This is primarily due to the softer local spectrum, which increases the local fission density in this region. The increased local fission density yields proportionally increased local fission gas concentration, resulting in significantly increased porosity in this region. In addition, the build-up of ^{239}Pu is enhanced. As demonstrated in (Brown et al., 2014), UN-based ceramic fuels have significantly enhanced heavy metal density and similarly enhanced self-shielding. The objective of this section is to quantify the impact of the increased self-shielding, both on the burn-up distribution in the fuel pin and also on the build-up of transuranic elements. The build-up of transuranics is an issue in terms of fuel performance and fuel cycle impact. This is also a significant function of spectral hardness; as the neutron energy spectrum hardens (as it does with some candidate advanced cladding and UN-based ceramic fuels), the transuranic build-up will increase.

To accomplish this, five pin cell models were used to assess the relevant sensitivities. The pin cell models are: a reference UO_2 with zirconium alloy cladding and 5% porosity; a UN-based ceramic with 80% UN phase (and 100% enrichment in ^{15}N), 10% U_3Si_5 , 10% porosity, and zirconium alloy cladding (pellet outer radius = 0.4096 cm); a UN-based ceramic with 80% UN phase, natural nitrogen (mostly ^{14}N), 10% U_3Si_5 , 10% porosity, and zirconium alloy cladding (pellet outer radius = 0.4096 cm); a UN-based ceramic with 80% UN phase (and 100% enrichment in ^{15}N), 10% U_3Si_5 , 10% porosity, and Kanthal AF cladding (pellet outer radius = 0.4249 cm); and a UN-based ceramic with 55% UN phase, 100% enrichment in ^{15}N , 35% U_3Si_5 , 10% porosity, and Kanthal AF cladding (pellet outer radius = 0.4249 cm).

The pin cells are divided into fifteen concentric rings of equal volume, as shown in Figure 35. This enables capturing the edge effects due to the self-shielding in the pin. The temperatures and geometry are fixed throughout the simulation. Therefore, this simulation isolates the spectral and self-shielding impacts on local burn-up and plutonium build-up

Figure 35. Pin Cell Geometry Used to Study Rim Effect

The rim effect is illustrated for UO_2 in Figure 37, with power density contour plots at 0 EFPD and 1500 EFPD. Orange shades represent higher power density and yellow shades lower power density. Similarly, lighter shades of blue represent higher thermal flux and darker shades of blue lower thermal flux.

Figure 37. Formation of High-Burnup Rim: Power Distribution Contours at (a - left) 0 EFPD and (b - right) 1500 EFPD

This is further illustrated in Figure 39 for each of the options considered here. It is notable that the burn-up at the rim is highest (in GWd/t) for UO_2 fuel. However, UO_2 fuel also has the lowest heavy metal loading. The relative burn-up is shown in Figure 41. This indicates that the rim effect (in terms of relative burn-up) is not substantially enhanced for the proposed high density ceramic fuels, and that overall the burn-up distributions are very similar, even with the enhanced self-shielding in UN-based ceramics.

In contrast to this is the ^{239}Pu build-up, as shown in Figure 43 in weight percent at 1500 EFPD. The ^{239}Pu build up is markedly higher in all of the analyzed UN-based fuels, with zirconium alloy and AF cladding, and even at low UN phase content (55%). This is largely a function of the harder neutron spectrum with these fuels, but it has significant implications in terms of the fuel cycle. The increased transuranic content will act to increase the long-term specific activity in a once-thru fuel cycle, but could have significant advantages in a fuel cycle with continuous recycling or transuranic (TRU) material. The pin average ^{239}Pu build-up is shown in Figure 45 and the pin average total TRU build-up is shown in Figure 47, both as a function of effective full power days. The behavior is explained via the capture per unit lethargy plot shown in Figure 49, which shows the increased neutron capture above 1×10^{-6} MeV with the higher density UN- U_3Si_5 ceramic fuel. The main driver of the difference is increased heavy metal density. Note that this finding is consistent with Brown et al. (2014) where the resonance escape probability was shown to be significantly smaller for UN-based ceramics versus UO_2 fuel.

Figure 39. Burn-up Distribution (in GWd/t) at 1500 EFPD for Considered Fuel-Cladding Combinations

Figure 41. Burn-up Distribution (relative) at 1500 EFPD for Considered Fuel-Cladding Combinations

Figure 43. ^{239}Pu Build-Up Distribution (weight percent) at 1500 EFPD for Considered Fuel-Cladding Combinations

Figure 45. ^{239}Pu Build-Up (weight percent) as a Function of EFPD for Considered Fuel-Cladding Combinations

Figure 47. Total TRU Build-Up (weight percent) as a Function of EFPD for Considered Fuel-Cladding Combinations

Figure 49. Neutron Capture per Unit Lethargy

Core Analysis of UN-U₃Si₅ Fuel with Iron Based Cladding

Core analyses with thermal feedback play a vital role in understanding reactor performance and fuel management. Core analysis provides realistic boundary conditions for evaluation of fuel performance. The analyses reflect power peaking, burn-up histories, and reactivity control. Core analyses act as initial conditions or boundary conditions for transient/accident analyses, and for selected transients (such as a rod ejection accident) are a key component of coupled three-dimensional reactor kinetics analyses.

PARCS was utilized with two-group parameters generated by TRITON to calculate full-core reactivity coefficients and kinetics parameters. The few-group parameters were extracted from the lattice physics results. Each few-group parameter set included 20 branch cases that track the variation of cross sections as a function of fuel burn-up and relevant parameters, including moderator density and temperature, fuel temperature, soluble boron concentration, and control rod presence. The relevant fuel material properties were integrated into the PARCS thermal hydraulic feedback module. The PARCS multi-recycle capability was utilized to calculate equilibrium cores with a discharge burn-up convergence criteria of 0.1 GWd/t, and a fixed fuel shuffling scheme. Full details of the core calculation methodology are presented in (Brown et al., 2014).

Thermal Properties in the Core Simulator and Transient Analyses

Example thermal properties used in the core analyses, namely thermal conductivity and volumetric heat capacity (ρC_p), are shown in Figure 51 and Figure 53 for UO₂ and UN-U₃Si₅ with 80-vol% UN and 10-vol% U₃Si₅ as well as 55-vol% UN and 35-vol% U₃Si₅. Similarly, these properties are shown in Figure 55 for AF and APMT cladding. The sources for the properties are Hayes et al. (1990 a,b,c,d) for UN, White et al. (2014) for U₃Si₅, and Kanthal (2012, 2013) for AF and APMT. Thermal properties are the best available and are at zero-burnup conditions. Properties for UN-U₃Si₅ are volume-weighted based on the constituent phase content (rule of mixtures). The rule of mixtures was shown to be a very good approximation for UN-U₃Si₅ in White et al. (2014).

Figure 51. Thermal Conductivity of UO_2 and $\text{UN-U}_3\text{Si}_5$ as a Function of Temperature (K)

Figure 53. Volumetric Heat Capacity of UO_2 and $\text{UN-U}_3\text{Si}_5$ as a Function of Temperature (K)

Figure 55. Thermal Conductivity and Volumetric Heat Capacity of Kanthal AF and Kanthal APMT

Methodology for Core Calculation

The equilibrium core configurations were calculated with a single burnable poison configuration in a multi-cycle calculation. An identical burnable absorber configuration was utilized for all fuel types, with the caveat that the worth of the burnable absorber is slightly different for each fuel. The burnable poison utilized in all but one case was standard Westinghouse ZrB_2 coating (Simmons et al., 1988), e.g. IFBA. In the full-core calculations presented here we did not consider other PWR burnable absorber options, such as borosilicate glass rods or wet annular burnable absorbers (WABA) rods. This burnable absorber configuration is simplified and was utilized for the purposes of these initial scoping studies. The assembly geometry used in the generation of the few-group parameters is shown in Figure 57. The cases presented here for the equilibrium cycle core analysis are: the reference UO_2 case, the UN- U_3Si_5 composite case with AF cladding (with 55vol% or 80vol% of the UN phase), and the UN- U_3Si_5 composite case with APMT cladding (with 55vol% or 80vol% of the UN phase). A lower-bound gap conductance is assumed for each case (5,000 W/m-K) with a constant gas gap thickness. It is recognized that gap conductance, thickness, and pellet-cladding interaction are all very significant parameters, and further study of the impact of this assumption is needed.

Figure 57 Assembly Geometry Utilized for Generation of the Few-Group Parameters

Core Analysis of UN-U₃Si₅ Fuel with Kanthal AF and APMT Cladding

The objective of this section is to present core-level analyses of UN-U₃Si₅ fuel with AF and APMT cladding. The analyses were conducted at two volume contents of the UN phase, 55% and 80%, with 35% and 10% of U₃Si₅, respectively, and 10% porosity. The same fuel shuffling scheme is used that was described in Brown et al. (2014) and the other assumptions, such as the equilibrium cycle convergence criteria (0.1 GWd/t), are consistent as well. The change in critical boron concentration during the equilibrium cycle is shown in Figure 59, the radial assembly power peaking in Figure 61, and the local power peaking in Figure 63. It is notable that the 55% UN cases dubbed “drop-in” candidates have nearly the same soluble boron concentration throughout the cycle as the UO₂-Zr case. This demonstrates what is intended by a “drop in” ATF: one that minimizes the impact on reactor physics and fuel management behavior vs. UO₂-Zr fuel-cladding. The radial power and burn-up distributions at BOC and EOC are shown in Figure 65 to Figure 69. The reference UO₂-Zr values are shown in Figure 71.

Figure 59. Critical Boron Concentration as a Function of Cycle Burn-Up (EFPD) – UN-U₃Si₅-AF/APMT

Figure 61. Radial Assembly Peaking Factor as a Function of Cycle Burn-Up (EFPD) – UN-U₃Si₅-AF/APMT

Figure 63. Local Assembly Peaking Factor as a Function of Cycle Burn-Up (EFPD) – UN- U_3Si_5 -AF/APMT

Figure 65. Equilibrium Core Power and Burn-Up Distribution for UN (55) – U_3Si_5 – AF

Figure 40. Equilibrium Core Power and Burn-Up Distribution for UN (55) – U₃Si₅ – APMT

Figure 41. Equilibrium Core Power and Burn-Up Distribution for UN (80) – U₃Si₅ – AF

Figure 69. Equilibrium Core Power and Burn-Up Distribution for UN (80) – U₃Si₅ – APMT

Figure 71. Equilibrium Core Power and Burn-Up Distribution for UO₂

Simulation of a Rod Ejection Accident with UN-U₃Si₅ Fuel

This section presents three-dimensional kinetics simulations of a rod ejection accident in a PWR fueled with various UN-based ceramic fuels. The value of these simulations is that they can inform transient testing of candidate ATFs at the Transient Reactor Test Facility (TREAT) facility. This facility at Idaho National Laboratory is undergoing planning for an anticipated restart (Versluis 2014). In addition, these demonstrate the impact of control rod worth on the progression of a rod ejection accident and show that the potential optimization or design of control elements and poisons for candidate ATFs is also an important component of the viability assessment of candidate fuels. Transient fuel performance simulations also require this type of high-fidelity three-dimensional kinetics simulation of a rod ejection accident as a boundary condition.

This example shows results for ejection of a control rod bank for UO₂ fuel with zirconium alloy cladding and UN (55-vol%) - U₃Si₅ (35-vol%) fuel with AF cladding. This is a sub-prompt critical rod ejection with moderate worth (approximately 44 cents of reactivity) in 0.2 seconds. The reactors are at a hot full power condition, and it is notable that initial conditions such as power, temperature, and flux distribution are not the same for the two cases. In addition the critical boron concentration is different, 411 ppm for UO₂ and 463 ppm for UN-U₃Si₅. The delayed neutron fraction is also different, 0.00610 for UO₂ and 0.00625 for UN-U₃Si₅. This illustrates the difficulty in achieving an “apples-to-apples” comparison for this event, because the events are fundamentally different for the two fuel types, driven by the different physics, including neutronic and thermal properties. Nevertheless, the reactivity in dollars is very similar for this test case. The core average power for the rod ejection is shown in Figure 44, the control reactivity in dollars is shown in Figure 46, and the average fuel temperature *increase* in Figure 48.

Figure 44. Predicted Reactor Power During Rod Ejection Accident

Figure 46. Ejected Control Worth in Dollars

Figure 48. Average Fuel Temperature Increase During Rod Ejection Accident

The “drop in” ceramics, e.g. the UN (55) cases, have similar control worth to UO_2 . Qualitatively, the RIA response is very similar to that of UO_2 , with the rapid power increase mitigated by the resultant increase in the fuel temperature. All of the fuel temperatures are much lower than would be expected in a similar event for UO_2 fuel. However, it is known that the important quantity in this context is not the fuel temperature but the local energy deposition. This energy deposition may be used to inform a fuel performance simulation or an eventual TREAT test. The difference in response between UO_2 and UN- U_3Si_5 fuels is driven primarily by the difference in the volumetric heat capacity. The volumetric heat capacity of the UN- U_3Si_5 fuel assumed in this simulation (and consistent with available data at zero burn-up) is less than for UO_2 and therefore the similar power pulse in the early phase of the transient (< 0.2 seconds) drives the UN- U_3Si_5 fuel temperature to initially increase more rapidly than UO_2 , which terminates the transient more rapidly due to fuel temperature feedback. As the accident progresses the increased ability of the UN- U_3Si_5 to conduct heat will also play a role in the response. This significantly decreases the net energy deposition in the fuel during the event, although the peak power of the pulse is similar for both cases. This simulation is just an example and further work is needed in this area to improve the fidelity of the prediction.

Summary and Conclusions

In the aftermath of Fukushima, a focus of the DOE-NE Advanced Fuels Campaign has been the development of advanced nuclear fuel and cladding options with the potential for enhanced performance in an accident. Evaluation of the performance of these fuel and cladding options is vitally important, because it identifies whether the options have at least equivalent performance as the present UO_2 -Zr fuel-cladding system under nominal conditions and improved performance in accident scenarios.

Several potential cladding materials were studied with UO_2 and UN-based ceramic fuel and compared to zirconium based alloys. Other than SiC, which performs very similarly to zirconium alloys, all of the cladding materials have some reactivity and discharge burn-up/resource utilization penalty (in GWd/t). It is notable that the results here regarding burn-up and cycle length for Fe-based cladding and UO_2 fuel are in general agreement with other recent studies, including George et al. (2015) and Terrani et al. (2014). Most candidate claddings performed similar to UO_2 -Zr fuel-cladding in terms of safety coefficients. The clear exception is that Mo-based materials were identified as potentially challenging from a reactor physics perspective due to high resonance absorption, which will alter moderator temperature coefficients via the underlying reactor physics. Although this can be mitigated, to some extent, by very thin cladding, it is an undesirable impact. It is also notable that the high absorption rates in Mo means that the material will “burn out”, especially ^{95}Mo . An appropriate and acceptable thickness for cladding needs to be determined for each advanced cladding based on mechanical and thermal characteristics (including irradiation performance) to have better confidence in the trade-offs associated with these potential cladding materials.

Assuming a constant enrichment, the higher density of UN-based fuels can compensate for the parasitic absorption in Fe-based cladding in terms of cycle length in EFPD relative to the UO_2 -Zr system. This is true even for the potentially conservative cladding thicknesses utilized in this work. However, there will still be a marginal penalty in discharge burn-up in GWd/t.

UN- U_3Si_5 fuel paired with Kanthal AF or APMT cladding can match UO_2 -Zr fuel-cladding reactor physics and fuel management performance over a wide parameter space of phase fractions. There will be a marginal penalty in discharge burn-up (in GWd/t). The sensitivity to ^{14}N content in UN is high and significant amounts of ^{14}N isotope will result in a reactivity penalty and hydrogen generation in the fuel. Core analysis with thermal feedback shows that the “drop in” options with 55vol% UN, 35vol% U_3Si_5 , and 10% porosity have very similar performance to UO_2 -Zr fuel-cladding in terms of the boron letdown and the cycle length in EFPD for a fixed fuel management scheme.

The rim effect, which is important from the perspective of fuel performance, is not significantly enhanced for the UN-based composites versus UO_2 fuel when considering the relative burn-up distribution within the pin. However, the hardened neutron energy spectrum and increased self-shielding will result in increased overall build-up of transuranic elements in terms of weight percent of discharged fuel, which has cascading implications throughout the fuel cycle.

The response of a UN-U₃Si₅ fueled core with AF or APMT cladding has a qualitatively similar response to a rod ejection accident when compared to UO₂-Zr fuel-cladding. The rod ejection accident is very sensitive to the control worth and volumetric heat capacity of the fuel. It is important to consider realistic control rod designs for potential ATFs, where the rod worth may be different. In addition, the simulations in this paper are an example of studies that could be used in the future to inform fuel performance simulations, and eventually, transient testing in TREAT.

Significant additional work is necessary to determine whether the proposed fuel and cladding candidates are viable. However, this screening process has determined that the fuels are potentially promising in spite of some identified challenges. It is also important to note that these analyses focused on “drop in” replacement of UO₂ fuel. These fuels, which feature enhanced thermal conductivity and heavy metal density, may enable other performance benefits not explored in this paper.

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