Silicon Carbide TRIPLEX Materials for CANDU Fuel Cladding and Pressure Tubes

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ABSTRACT

Ceramic Tubular Products has developed a superior silicon carbide (SiC) material TRIPLEX, which can be used for both fuel cladding and other zirconium alloy materials in light water reactor (LWR) and heavy water reactor (CANDU) systems. The fuel cladding can replace Zircaloy cladding and other zirconium based alloy materials in the reactor systems. It has the potential to provide higher fuel performance levels in currently operating natural UO2 (NEU) fuel design and in advanced fuel designs (UO_2(SEU), MOX thoria) at higher burnups and power levels. In all the cases for fuel designs TRIPLEX has increased resistance to severe accident conditions. The interaction of SiC with steam and water does not produce an exothermic reaction to produce hydrogen as occurs with zirconium based alloys. In addition the absence of creep down eliminates clad ballooning during high temperature accidents which occurs with Zircaloy blocking water channels required to cool the fuel.

Introduction

Ceramic Tubular Products (CTP) has been developing and testing advanced Silicon Carbide (SiC) three-layered tubular products (TRIPLEX) as cladding for nuclear fuel in water cooled reactors, and channel boxes for Boiling Water Reactors. Test have been performed under simulated LOCA conditions including steam corrosion at temperatures up to 1600 ^{0}C, thermal shock resistance after quenching in water from 1000 ^{0}C, and mechanical shock conditions representative of handling and transportation loads. Results demonstrate 3 orders of magnitude increase in accident resistance as compared to traditional zirconium alloy materials.

The use of TRIPLEX cladding in place of Zircaloy can provide higher fuel performance and extend the life of the standard and advanced fuel designs being considered for CANDU reactors.

Based on evidence from both the Fukushima and TMI-2 accidents, the key to achieving accident resistance in commercial nuclear fuel is the fuel cladding, which is the primary boundary for containing the fuel and fission products against release during a severe accident. The capability of the zirconium alloy cladding to perform this function is lost when the cladding reaches temperatures above 500 ^{0}C. The material loses its strength and balloons, blocking core flow. This leads to an exothermic reaction between the zirconium and water releasing large amounts of heat and hydrogen. As long as zirconium is used a cladding material with its poor inherent properties above normal operating temperatures it will contribute to the severity of accidents. Other zirconium based core components such as channel boxes in BWRS andfuel end plates and pressure tubes in CANDUs also react exothermically with water during core overheating accidents, produce hydrogen and thus contribute to the severity of such accidents.

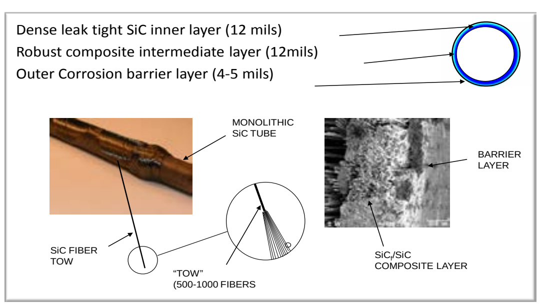
An alternative to zirconium alloys that does not exothermally react with water and release hydrogen is the ceramic material silicon carbide. Over several decades of research for fusion materials in Japan and the U.S, it was learned that some forms of silicon carbide are radiation resistant and are useable in fission reactors.

1.0 CTP TRIPLEX SILICON CARBIDE CLADDING

The CTP TRIPLEX^{TM} silicon carbide cladding is a three layer, all silicon carbide tube. It retains its strength to temperatures above 1400 ^{0}C, has at least 800 times slower reaction rate than Zircaloy during design basis accidents for light water reactors, and reduces the amount of hydrogen released during such accidents by a factor of 400.

As shown in Figure 1 each of the three layers fulfills a different design requirement. The inner high density monolith layer of stoichiometric beta phase SiC assures hermeticity and fission gas retention during normal operation and reactor transients. The central composite layer is made from stoichiometric beta phase SiC fibers and SiC matrix produced by the Chemical Vapor Infiltration (CVI) process. This composite layer reinforces the monolithic layer and assures a failure mode which is not severe even when the clad is subjected to high mechanical and transient thermal loadings thus avoiding the brittle failure mode of monolithic ceramics. The central layer assures that the clad tube retains its shape and solid fission product retention capacity during severe accidents. The third layer ensures longer lifetime.

Figure 1 TRIPLEX Fuel Cladding Construction



CTP's TRIPLEX clad incorporates an outer dense layer of SiC which serves as an environmental barrier protecting the central fiber layer assuring the cladding can achieve at least six years of exposure to PWR coolant without excessive corrosion. To demonstrate this capability, a special loop was constructed in the MIT Research Reactor to closely duplicate the actual coolant chemistry, temperature, flow and neutron flux conditions in the central region of a typical PWR. A variety of CTP's TRIPLEX SiC specimens were fabricated with different types of fibers, different types of matrices and different types of environmental coatings. Test specimens were exposed in this loop for periods of up to 480 full power days of operation. After 12 months in the loop the amount of weight loss was measured and material recession was calculated in microns. In the best combination of fiber type and composition a linear recession rate of 12 microns was achieved. This equated to about 54 microns of surface coating after 54 months of service or three full 18 month cycles of operation. The outer layer has a minimum of 100 microns thickness, therefore only 54% of the layer would be lost during the life of typical PWR fuel.

The superior behavior of TRIPLEX cladding during simulated severe accidents (simulated Loss of Coolant Accident, LOCA) has been demonstrated. For example, a quench test of SiC TRIPLEX clad was performed by emersion of a sample heated to 1000 ^{0}C in a room temperature water pool showed virtually no damage or loss of structure. Other tests of CTP's TRIPLEX clad in a 1200 to 1600 ^{0}C LOCA environment for 6 to 8 hours demonstrated a dramatic reduction in exothermic reaction and hydrogen release. These tests were performed at the CTP steam test rig. Weight loss of about 0.01% occurred after 6 hours of exposure to 1200 0C steam and about 0.04 to 0.05% after 8 hours of exposure to 1400 ^{0}C steam. Weight changes from these tests were converted to recession (depth of reaction) in microns and compared with values of recession and hydrogen evolution of Zircaloyfor typical 17x17 PWR fuel rod geometry. The recession and hydrogen evolution for TRIPLEX clad was two to three orders of magnitude lower than Zircaloy. This demonstrates an exceptional gain in safety margin for Silicon Carbide as compared to Zircaloy under LOCA conditions.

CTP has performed a variety of mechanical tests on the latest version of TRIPLEX clad to demonstrate the performance of the material for under typical PWR operating conditions. These include

- Mechanical shock test to demonstrate behavior under severe external shock that may occur during handling and shipping. TRIPLEX design incorporates a feature which increases shock resistance to avoid gross fracture in the event of an accidental drop during fabrication, shipping and handling. This shock design feature is able to absorb 2.5 to 5.5 times more energy as tubes without the feature.

- Measurement of the modulus of elasticity to demonstrate higher stiffness, enabling a reduction in fuel assembly distortion during operation and the option of reducing the number of axial zirconium alloy grids in LWR fuel bundles.

- A series of internal strength tests to determine the maximum plenum pressure that can be achieved in the cladding to allow for long fuel lifetime than that achievable with Zircaloy clad. An essential feature of the TRIPLEX clad is its ability to retain integrity at high internal pressure compared to Zircaloy. Plenum pressures exceeding 50 MPa with retained clad hermeticity have been demonstrated. The absence of creep behavior in TRIPLEX clad is able to compensate for increases in fuel temperature and fission gas release during normal operation. This feature ensures the absence of clad ballooning during severe accident conditions when the fuel channels can experience restricted coolant flow during severe accident conditions thereby preventing the blockage of flow and cooling during fuel overheating events.

2. TRIPLEX CLAD for CANDU FUEL

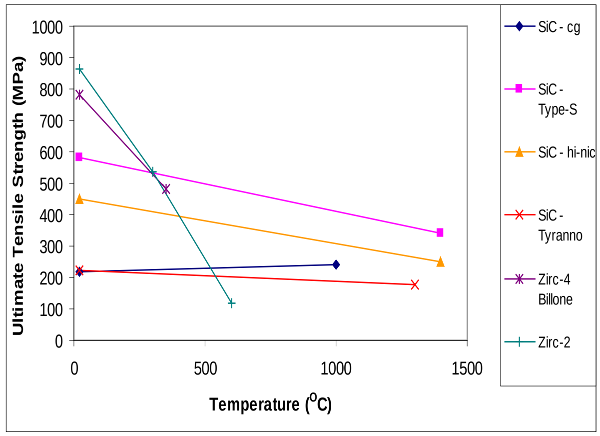
The standard CANDU fuel element is a 37 fuel rod bundle utilizing Zircaloy tubing and natural uranium dioxide fuel. The reference design for extended burnup fuel cycles is a 43 fuel rod design designated CANFLEX using Zircaloy tubing and slightly enriched uranium dioxide fuel. Extending the burnup of CANDU fuel to greater than 400 MWh/kgHE requires assessment of a number of performance issues. High fission gas release (FGR) may be observed , in fuel achieving power greater than 50kW/m and burnups greater than 500 MWh/kgHE and result in high internal gas pressures that cause stress corrosion cracking (SCC) in the cladding. Other performance parameters such as cladding strain, hydriding/deuteriding and corrosion, power ramp defect thresholds, CANLUB retention and pellet microstructural changes leading to swelling are also important to consider at extended burnup.

Fission Gas Release (FGR)

Inert gases are produced in the fuel matrix as a result of fission and these inert gases diffuse from the fuel matrix to the internal element free void space primarily as a function of temperature. The release of large quantities of fission gases to the free void space may result in high internal gas pressures that can lead to high clad stresses and strains and potential failure due to clad SCC.

The use of TRIPLEX SiC cladding has the ability to retain its integrity at high internal pressure compared to Zircaloy. With SiC cladding slightly higher temperatures may occur in the fuel, and potentially somewhat higher fission gas release. However, if the fuel to clad gap is minimized as in CANDU fuel FGR release may be minimized due to the absence of creep behavior of TRIPLEX clad. In addition, as shown in figure 2, SiC retains its strength to temperatures well above operating temperatures, as compared to Zircaloy which loses almost all its strength above about 400 ^{o}C.This provides a large increase of safety margin of fuels operating at extended burnups and higher power levels.

Figure 2 – Strength vs. Temperature for SiC Composites compared to Zircaloy



In CANDU fuel the Zircaloy clad is protected from SCC due to high stresses and strains by the use of CANLUB coating on the inside of the cladding tube. However with the use of TRIPLEX cladding the CANLUB application is not required for this purpose since SCC is not an issue at high cladding stresses and strains. The CANLUB coating be of value for other reasons and needs to be evaluated.

Corrosion Behavior

A critical requirement for successful behavior of the SiC cladding is that it must have acceptable corrosion during normal operation of the reactor. In contrast to Zircaloy, which forms and oxide layer that reduces the clad thickness available to support mechanical stress, SiC reacts very slowly with water in PWR conditions. The design stress basis for TRIPLEX SiC cladding is to carry the load completely in the first two layers, while allowing the third layer to act only as a corrosion protecting layer. For CANDU fuel operating to high burnups (>1000 MWh/kgHE) the effect of waterside corrosion under CANDU operating conditions is about 30 microns, which is comparable to that shown by SiC under PWR operating conditions for three 18 months cycles (the equivalent of 1200 MWh/kgHE). It should be noted that SiC cladding should be tested under CANDU operating conditions to make a clear comparison.

Cladding Strain Behavior

For CANDU fuel operated at burnups < 400MWh/kgHE mid-pellet strains of up to 0.5% can be observed and dependent on pellet geometry and density, full power and as-fabricated diametral clearance between the cladding and pellet. The observed maximum strain in extended burnupUO_2 increases as a function of burnup in the order of 1.5% at 750 MWh/kgHE. This is generally due to swelling of pellets due fission product buildup and associated pellet clad interaction.

An essential feature of TRIPLEX cladding is its ability to retain integrity as high internal pressure as compared to Zircaloy which requires long gas plenums in order to keep the fission gas pressure below the external pressure and thereby avoid clad creep out leading to failure of the clad. This important during normal operation, as it allows increased fission gas release and internal gas pressures during continued burnup of the fuel to high levels. It also has advantages during overheating accident situations where Zircaloy cladding will balloon at higher temperatures and block access to coolant flow. The absence of creep behavior in TRIPLEX clad is able to compensate for the increase in fuel temperatures and fission gas release.

Table 1 presents the results of hoop strength testing of TRIPLEX tubes and is based on the elastomer plug test protocol developed by ORNL to provide evidence of the strength retention of Zircaloy cladding containing mixed oxide fuel.

Table 1 – Internal Pressure at Failure of SiC Type S TRIPLEX Tubes

Tube ID	Internal Pressure (ksi)	Internal Pressure (MPa)
SL-1	7.234	49.2
SL-2	7.634	51.9
SL-3	7.140	48.6
SL-4	7.564	51.5
SL-5	7.422	50.5
SL-6	7.775	52.9

In order to apply this pressure containment feature to actual fuel design and licensing a safety factor of at least 20% must be applied to the measured data leading to an allowable ΔP across the wall of about 40MPa, allowing for the composite strength degradation lowers the allowable ΔP to about 35 MPa. For normal operation and operational transients, the external pressure will reduce the differential pressure across the cladding by about 15 MPa, leading to an acceptable internal fission gas pressure of about 50 MPa. The elastomer plug testing has demonstrated that the composite layer retains its shape even after the monolith has failed due to overpressure, hence ensuring retention of solid fuel fragments and solid fission products during severe accidents..

Neutronic Considerations

SiC cladding leads to better neutron economy than zirconium alloy. This is primarily based on the lower absorption cross-sections and therefore fewer parasitic captures in SiC compared to the isotopes present inzirconium based cladding. Consequently, SiC clad fuel can achieve the same cycle energy of fuel with zirconium cladding using a lower amount of initial fissile material. The better neutron economy of SiC cladding can lead to savings in fuel cycle costs. In perspective it can be anticipated that the burnup and enrichment limits can be extended and as a result, the benefits from the favorable neutronic characteristics of SiC will with its superior irradiation performance enable higher discharge burnup, fuel utilization and improved fuel cycle costs versus the current zirconium based cladding. Although these neutronic benefits have not yet been quantified for CANDU fuel, reference (9) reports a savings of $2 million per fuel reload for a typical 18 month cycle 3570 MWth PWR core, resulting from the reduced enrichment requirements for SiC cladding compared to Zircaloy cladding.

4. Severe Accident Resistance Using TRIPLEX Material

The progression of a severe core damage accident in a CANDU reactor is slow because the fuel is surrounded by a large quantity of light and heavy water. This constant blanket acts as a heat sink to remove the decay heat and decelerates the conditions leading to a severe accident and disassembly of the core. Severe accident phenomena occurring during core damage progression include

- Fuel bundle heat up and disassembly
- Fuel channel heat up sagging and perforation and melt-through
- Fuel and fuel channel debris separation (disassembly) from the remaining channel and formation of a suspended debris bed
- Suspended debris heat up, core collapse
- Terminal debris formation within the calandria vessel
- Calandria vessel failure
- Interaction of core debris with the calandria vault
- Failure of the containment structure

With the loss of moderator inventory surrounding the fuel channels and as a result of the exothermic zirconium-steam reaction, which generates hydrogen, the fuel channel temperature increases. The fuel channels on the top rows heat up and sag under gravity. The top row channels sags and contacts the next row of the lower uncovered channel and transfers the load and heat. During the sagging process the longitudinal total strain of the fuel channel increases. The total strain will be concentrated between the fuel bundle junctions as the sagging increases and lead to wall-thinning at the junction region between the fuel bundles. The calandria tube will perforate as a result which will allow steam to enter the gap between the pressure tube and the calandria tube. The fresh Zircaloy surfaces in the gap between the pressure tube and the clandria tubes are exposed to steam and as a result the fuel channel temperatures will rapidly increase from the zircaloy-steam exothermic reaction and produce hydrogen.

The use of TRIPLEX SiC material for both the fuel cladding and the pressure tubes reduces the inventory of Zircaloy in the reactor system. This reduction of zirconium inventory has the following major benefits

- The amount of hydrogen produced during a severe core damage accident is essentially zero in comparison
- mechanical property benefits of TRIPLEX SiC over Zircaloyat accident temperatures can lead to a large reduction in fuel failures releasing fission products
- Ballooning of the fuel cladding is minimized with less fuel channel flow blockage
- Sagging of the pressure tubes can be minimized

3. Alternative CANDU Fuels

Natural UO_2 fuel (NU) which is presently utilized in operating CANDU reactors typically achieves a burnup approximately 200 MWh/kgHE and excellent performance continues to be demonstrated at this level. AECL has developed technologies to extend burnups in order to increase fissile material utilization and reduce spent fuel quantities. These advanced fuels include slightly enriched uranium (SEU) (burnup 400-600 MWh/hgHE), MOX and thoria (up to 1000 MWh/kgHE). The reference design for extended burnup fuel cycles is the 43 fuel rod CANFLEX bundle which provides increased operating margins over the 37 fuel rod design bundle. Over the past twenty yearsa significant number of CANDU fuel bundles have operated to burnups in the range of 400-1200 MWh/kgHE, including UO_2, MOX and thoria fuels. High fission gas release and pellet swelling has been observed in the various fuels achieving > 50kW/m and burnups >500MWh/kgHe and in some cases leading to cladding failures due to SCC and high clad strain. This can lead to an incentive to reduce maximum fuel element power in extended burnup fuel.

The use of TRIPLEX SiC cladding has the potential to mitigate the performance issues related to advanced CANDU fuel (NU, SEU, MOX and thoria) at higher powers and extended fuel burnups.

4. CONCLUSIONS

A great deal of progress has been made in the development and testing of the CTP TRIPLEX SiC material. The product can meet the basic requirements for operating conditions in current LWR plants with reliability that matches or exceeds that achievable with Zircaloy. Tests performed at Oak Ridge National Laboratory and Massachusetts Institute of Technology demonstrates a superior performance during a LOCA event compared to Zircaloy tubing.

The application of the TRIPLEX material to the fuel and reactor pressure tubes in CANDU reactor plants provide major benefits to current fuel and extended fuel performance and mitigate many dire effects as a result of a severe accident.

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