Is silicon carbide a solution to safer light-water reactor fuel?

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On Friday, 11 March 2011, a nuclear facility that was up to that moment rather unknown to the public all over the world was forced into the limelight by an earthquake of a magnitude of 8.9 on the Richter scale and a subsequent tsunami. This facility, a boiling-water reactor complex in the Fukushima prefecture on the north-east coast of Japan, sustained considerable damage to three of its operating units, as well as one unit that was shut down. For many years, designers have designed reactor structures to withstand the forces created by site-specific earthquakes of certain magnitudes. The reactor at Fukushima was no exception and responded reliably to the forces created in the structures due to the accelerations. The three operational reactors were made subcritical by the earthquake-sensing devices, as required under such circumstances. Up to that point, everything went well, but just more than an hour later, it was as if the curtain on the nuclear centre stage was raised when a tsunami wave of 14 m above normal sea level struck the northeast coast of Japan. It drowned the essential emergency cooling and power systems of the reactors. The nuclear drama that was set to commence then started playing its overture by shedding the protective cladding on the structures of the reactors in a spectacular number of explosions.

The current safety objective

The safety analyses performed for reactors worldwide have to provide answers to three main questions. These questions are whether the reactor will, following an upset event, shut the nuclear chain reaction down, whether the fuel elements will be provided with sufficient cooling to protect them from releasing radioactivity, and whether the radioactivity present in the plant will not be released into the environment.

If the safety analyses of the reactors at Fukushima have provided sufficient proof that these questions could all be answered satisfactorily, why did the event unfold in such a spectacular way? In order to provide an answer, it is necessary to first introduce a number of important concepts. A reactor core that has been operating for some time does not cease to produce heat when the nuclear chain reaction is shut down. It behaves like a car when the engine is turned off. The engine structure is hot due to the heat stored in it. After the engine has been turned off, it slowly cools down due to natural or temperature-activated cooling processes. In a reactor, the heat after shutdown behaves in a similar way, but the heat that is stored in the structures, as in a car, is not the only heat that must be removed. There is also the additional energy that must be removed from the decay of the highly radioactive products that were created while the reactor was operating.

As part of the safety objective to contain the radioactivity in a nuclear facility, the fuel material in a reactor core is surrounded by cladding material that keeps the radioactivity produced from being released and protects the fuel material from reacting chemically with the cooling medium. The heat generated by the fuel material during operation is transferred to the cooling medium, which in turn transports it to the power conversion system during normal operation. The design of the cooling system attempts to provide the most economical transfer of heat from the heated fuel elements to the power conversion system. The transfer of heat requires the temperature of the heated surface to be optimised for the maximum thermodynamic efficiency of the power conversion process. The high temperature on the surface of the cladding material must, however, be kept sufficiently low to prevent a reduction in heat transfer due to bulk boiling occurring. Such a reduction in heat transfer can cause the cladding to overheat and consequently lose some, or all of its capability to retain radioactivity.

The design of the cladding and heat removal system is optimised to have a sufficient margin against a degradation of its integrity during normal operation, and also following upset events that may cause the cooling to be reduced.

Figure 1 shows a generic image of the fuel element of a pressurised water reactor. The detail in the image shows the fuel rods that consist of the fuel material, typically uranium dioxide in a tube of zircaloy, an alloy of zirconium.

The tubes are sealed on both ends by zircaloy end plugs. This material has been under development during the last five decades of reactor operation worldwide.

If the forced cooling in watercooled reactors is interrupted by some event, it can be shown that the decay heat immediately after shutdown is sufficient to boil the reactor dry, causing the fuel cladding to be grossly degraded unless replenishment of the water inventory is provided. The provision of water replenishment systems is therefore a safety requirement for all current-day water-cooled reactor designs. If this water replenishment should fail, however, the fuel cladding may become exposed due to the boil-off of the water



inventory in the reactor vessel. When the hot zircaloy cladding material becomes exposed, an exothermic oxidation reaction takes place between the zirconium and the steam that is generated. This reaction produces hydrogen.

The rate of energy released in this reaction was calculated for a reactor with a thermal power of 3 800 MW. The results are plotted in Figure 2. Also shown in this figure is the heat produced by the decay of the fission products. It can be seen that when this reaction takes place, its rate of heat production is approximately four times the rate of the heat produced by the decay of fission products. This has a compounding effect on the rate of total heat released and consequently accelerates the rate at which the cladding degrades.

When the above progression of a typical cladding degradation event is compared with the chronology of the Fukushima event, it is not difficult to imagine the entire sequence of reactions that took place, with the hydrogen eventually being released and dramatically exploding to signal the commencement of the core meltdown sequence. By just looking at this explosion and knowing that an open path was somehow created from the fuel to the environment, it was not difficult to predict that at least part of the radioactivity in the fuel would find its way into the environment – an event that eventually cost more than US\$20 billion to clean up.

Improvement of reactor fuel to enhance safety

Some years before the accident at Fukushima, the reactor designers and regulators were developing a roadmap towards making reactors safer and more economical, reducing the production of waste, as well as making reactors more (weapons) proliferation-resistant. This roadmap is now being rolled out with the main focus currently on the improvement of the safety features, and specifically towards

 \rightarrow Figure 1: Construction of a typical pressurised water reactor fuel element.



→ Figure 2: Power produced by the decay of fission products and the zirconium – steam reaction following a loss of cooling accident in a 3 800 MW (thermal) reactor core.

enhancing the passive safety features of all reactors. Naturally, the immediate objective is to start with the fuel and focus on the cladding material. The lessons learned from the high-temperature reactor programmes with the excellent fission product retention barrier that silicon carbide (SiC) provided in those designs are now being considered as a possible solution.

In an international collaborative development, which started in

1959, fuel was developed for hightemperature reactors that utilised SiC as the main fission product barrier. This material has many superb characteristics compared to the metallic fuel used in light-water reactors. It has shown its predictable behaviour in many fuel irradiation tests and high-temperature reactor operations worldwide since the start of its development. The fuel testing included conditions that would be prevalent in reactor accidents caused by a loss of cooling capability.



 \rightarrow Figure 3: Temperature behaviour of a composite SiC (Triplex®) compared to that of other current core materials.

In the fuel geometry used in hightemperature reactors, the cladding geometry is a very small spherical pressure vessel with an outside diameter of ~0.92 mm, of which the wall consists of a composite structure of pyrolytic carbon/silicon carbide/ pyrolytic carbon. This small pressure vessel surrounds the actual fuel, which is typically a small ~0.5 mm diameter particle of a uranium compound, such as uranium dioxide. Of the composite structure, the SiC is generally regarded as the main fission product barrier. SiC is a brittle material and, although it functioned very well in a small particle, it cannot be seen to be directly applicable in a tubular geometry of much larger dimensions.

SiC fuel cladding

One of the companies in the USA at the forefront of the development of SiC tubes for light-water reactor cladding is Ceramic Tubular Products (LLC), which is developing a dense leak-tight composite tube called Triplex[®]. This tube consists of an inner layer of dense leak-tight SiC and an outer SiC layer called a corrosion barrier. Embedded between these two layers is a composite intermediate layer of SiC fibres. The wall thickness of the Triplex[®] composite is ~ 0.3 mm. Testing is currently underway to determine its characteristics in comparison to zircaloy. The initial results of the mechanical testing so far have shown a fundamentally different behaviour pattern, as indicated in Figure 3.

The results of other development testing carried out thus far are as follows:

- The inner layer holds the fission gases up to 54 MPa.
- There is no shrinking (creep) of the tube, as occurrs in the zircaloy tubes, where it shrinks onto the fuel pellet during irradiation.
- In case of a severe power excursion, the inner layer may start leaking, but the central composite layer retains the tube in shape up to strains in excess of 8%.

 No ballooning due to hightemperature and high-fuel burnup (high fission gas pressure) occurs and there is no possibility that an exothermic cladding/ steam reaction would take place.

Testing is currently being carried out on un-irradiated and irradiated specimens of the SiC cladding at the Massachusetts Institute of Technology (MIT), the Idaho National Laboratory and other proprietary laboratories in the USA. The University of Pretoria is engaged in SiC production research and the study of heat transfer from the surface of SiC cladding tubes.

Benefits of changing to SiC cladding

The temperature of the cladding is dependent on the power produced in the fuel pin and the heat transfer coefficient on the surface of the fuel cladding. If the SiC cladding should exhibit a higher heat transfer coefficient than zircaloy, the surface temperature will be lower for the same power produced in the fuel tube. If the designer of the fuel chooses to keep the cladding temperature the same, the power produced in the fuel pin can be raised, which means that the reactor can operate at a higher power level. This has an economic advantage to the operator of the plant. As can be seen in Figure 3, SiC can operate at a much higher temperature than zircaloy before any degradation of mechanical properties should become a problem, so the temperature of the fuel may even be allowed to be higher than that of the zircaloy, which means that the power can be raised even more.

Other benefits of using SiC in lightwater reactor fuel may come from its smaller neutron absorption characteristics, which means that the enrichment of the fuel in the core can be lowered. If the enrichment of the fuel stays the same, a higher burnup can be achieved, which can have a sizeable economic benefit. Due to its excellent thermal properties, the power density in the core can be raised.



ightarrow The flow loop for testing heated fuel pins.

These two aspects will positively impact on the cost of power production.

In addition to the normal operational benefits gained from using SiC cladding, the ultimate benefit will come from its safety characteristics.

This option should, however, not be seen as a short-term "fix". There are challenges, such as the production of the SiC tubes to exacting specifications and the joining of the sealing end plugs to the tube body. Furthermore, although SiC has been qualified as a nuclear material in high-temperature reactor programmes, regulators will require qualification and demonstration of safe operation before a licence can be issued for the general use of this revolutionary fuel. An estimate of the time scale until final approval is granted can be anything between 22 and 24 years from now.

In order to conduct the heat transfer studies, the Department of Mechanical and Aeronautical Engineering at the University of Pretoria has designed a flow loop where electrically heated fuel pins can be studied in three orientations relative to the flow direction of the water coolant. The flow loop was constructed using funding from the Institutional Research Theme (IRT) on Energy. The photograph above shows the loop when commissioning testing was being performed.

With all of this in mind, the question that has to be answered is whether SiC is a solution for safer reactor fuel. The answer is most probably a very positive "yes". But, to get to the point where it is generally accepted by regulating authorities and the general public, a huge amount of research and development is still required.

The University of Pretoria is proud to be part of this initiative. Θ

Reference

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