Materials for nuclear energy in the post-Fukushima era

An interview with John Marra

by Peter Wray

Fukushima Dai-ichi. Those two words are now synonymous with the earthquake and tsunami disaster in Japan on March 11, 2011, and the subsequent destruction of Tokyo Electric Power Company's nuclear power plant on the eastern coast of Japan. As time passes the name alone—Fukushima—will come to represent a sequence of events and responses in the same way that "Three Mile Island" and "Chernobyl" do.

John Marra spoke to GOMD attendees about the events that led to the Fukushima Dai-ichi accident and how the accident will impact materials selection in nuclear power plants.

Assuming that the Fukushima incident would have some tangible impact on nuclear engineering, the *Bulletin* recently was able to catch up with ACerS member John Marra, associate laboratory director of science and technology at the Savannah River National Lab. SRNL is the leading government facility within the Department of Energy for several nuclear technologies, including nuclear fuel storage and handling. At SRNL, Marra has been able to monitor the reports from DOE and other independent experts on the scene in Japan. He has been privy to many of the internal discussions about the lessons learned from Fukushima. And, after hearing what Marra has to say, it is hard not believe that Fukushima also may become synonymous with a rethinking of the properties and demands of key nuclear materials and, perhaps, the embrace of small-scale delivery of fission-based power.

The energy backdrop remains the same

The accident captured the world's attention, partly because of the human element of the disaster as it unfolded. Our collective hearts went out to Japan and its residents. But it also is human nature to personalize such events and project them into our immediate reality. What would be the impact on *my* business/industry if the nuclear contribution to *my* grid were lost? The United States had a brief glimpse of the economic cost of a large-scale electric power outage in 2003 when the East

Coast went dark for three days with an estimated economic impact in the range of $10 billion.

The timing of the Fukushima disaster, for better or worse, coincided with the growing acceptance among technical and political leaders in the US that nuclear energy would have to continue to be a big part of the nation's energy portfolio if CO_2 emission reductions and energy independence are to be achieved.

Despite what some might consider unfortunate timing, Marra said that although Fukushima will trigger many changes, the context remains the same. "The nuclear industry is still in the eye of a 'perfect storm.' Many fossil fuels are near record highs, energy demands are increasing and concerns about greenhouse gas emissions remain the same," he said. "There is no way to reach the national CO_2 and energy independence goals—you can run the scenarios any way you want—without a growth in the nuclear sector. You don't get to that 80 percent reduction in greenhouse gases unless you rely on some component of nuclear power, unless there are some types of miraculous developments in carbon capture (or sequestration) or efficiency gains in solar or wind that aren't out there," he said.

There is another technical issue that remains unchanged by Fukushima. Marra is quick to emphasize that even as wind, solar and geothermal power sources grow, the US electrical infrastructure will have to contend with variations caused by time, regional and seasonal considerations. "Energy storage has come a long way," said Marra, "and it may eventually smooth some of this out, but as long as the nation has a centralized grid system, there will be need for significant baseline power from large producers, such as nuclear power plants."

Marra did acknowledge that fears of dangers related to nuclear power among the public, real or perceived, were a contentious factor before Fukushima, and are probably worse now. Indeed, much of the world reacted swiftly in opposition to expanding this power source after the accident in Japan. For example, the German and Italian governments announced plans to "phase-out" nuclear power by canceling all new builds and decommissioning existing nuclear plants.

However, the same reaction didn't gel in the UK and the president of the Royal Society of Chemistry took the unusual step of publicly warning the government not to shut down its nuclear industry comprising 19 reactors producing around 23 gigawatts of power. Marra believes leaders in the US are taking a similar and pragmatic view of the situation.

Keep the old roadmap or find a new one?

When we last spoke with Marra, in 2010, DOE had just submitted a report to Congress, "Nuclear Energy Research and Development Roadmap" (see DOE's Nuclear "Roadmap"). The core of the document was a recommendation to pursue four pressing R&D objectives for the US nuclear industry (see DOE's Nuclear "Roadmap"), which were considered to be crucial to any effort to extend and expand its presence in the nation's energy portfolio.

Regarding the Roadmap in general, Marra didn't see much need for change but acknowledged that there is a new appreciation for need to understand correlated risks. For example, he said, the system designers understood the need to be able to maintain operations at a nuclear facility during individual low-probability, high-risk events, such as earthquakes and tsunamis. The lesson, he said, is that if there is a positive correlation between unlikely events, as was discovered in Japan, those scenarios must be planned for, too.

Beyond that, Marra said that the emphasis of two of the four goals — extending the life of current reactors and making it less expensive to open new reactors—likely would be modified.

For example, regarding extending the life of current reactors, Marra noted that during the next 20 years plant licenses are due to expire that represent 100 gigawatts, a huge amount of baseline power in the US. Already, the NRC has approved 20-year extensions to the original 40-year licenses at 66 plants. However, given the circumstances, systems and materials performances that occurred at Fukushima, Marra said life-extension programs likely would include reviewing site assumptions, cooling system designs and, perhaps, new fuel rod cladding systems.

Regarding the part of the Roadmap aimed at enabling new nuclear installations, Marra said that it was unclear at this point how much, if any, added costs would come from building new reactors with Fukushima-based safety features.

Marra predicted, however, that the events in Japan would give new impetus

Nuclear power facts

- Globally, there are 436 operational nuclear energy plants, with about 100 located in the US.
- Forty percent of the energy consumed in the US is in the form of electricity, and about 20 percent of US electricity is produced by nuclear power plants.
- Nuclear energy plants provide 70 percent of the non-carbon energy.

DOE's nuclear "Roadmap"

1. Develop technologies and other solutions that can improve the reliability, sustain the safety and extend the life of current reactors.
2. Develop improvements in the affordability of new reactors to enable nuclear energy to help meet the administration's energy security and climate change goals.
3. Develop sustainable nuclear fuel cycles.
4. Understand and minimize the risks of nuclear proliferation and terrorism.

Source: "Nuclear Energy Research and Development Roadmap," Report to Congress, US Department of Energy, April 2010.

to installations with a radically different scale of fission power: *small modular reactors*. Advocates and designs for SMRs have begun to appear in growing numbers, if for no other reason than the current price for a full-scale, multireactor power plant is a jaw-dropping \$5 to \$10 billion. To put that number in perspective, Marra pointed out that the price tag for a new build is larger than the equity market value of many small- and medium-sized utilities. They, their stockholders and other investors are loathe to assume that type of long-term financial risk.

SMRs, on the other hand, are only a fraction of the cost and size of current utility-scale reactors. A typical SMR is similar in size to a nuclear submarine or aircraft carrier power plant. Marra noted the submarine and carrier applications show that SMRs have actually been around in one form or another for decades. The new SMR proposals are an adaptation of current and advanced reactor technologies adopted to small-scale deployment.

"They could offer some significant advantages," said Marra. "They are cheaper to build—hundreds of millions of dollars, rather than several billion—and can use air cooling instead of water, standard forging and casting fabrication of components, and bring a smaller emergency planning zone." He estimated that an SMR could run a small city and would be an attractive energy option for rising economies or remote regions that do not have the grid capacity to support the output of a full-sized plant. "SMRs represent the distributed-power option for nuclear energy," he said.

Failures at Fukushima

As he turned to discussing the cascading series of emergencies that occurred in Japan in March, Marra first lauded the dedication of the TEPCO staff and other responders. "Those events," said Marra, "were extraordinary and exceeded all design contingencies. In the middle of numerous alarms and multiple system failures they worked furiously to triage the situation. They had to be torn by unimaginable concerns about their families, homes and communities, yet they returned the plant to a safe and stable condition."

The general sequence of events that led to the failure of four of the six reactors has been well documented in the mainstream and scientific press. But, Marra said it's important for ACerS and other materials societies to understand what happened to the materials in the reactors. He described the situation as a highly dynamic, nonequilibrium interaction of materials that were pushed well beyond their design specifications. He predicted that interesting forensic materials science is likely to be reported once the reactors can be entered and the fuel cores removed.

Marra recounted that as the primary and back-up cooling systems failed, water levels began to drop. Fuel rods became exposed ("Never a good case in any type of light-water reactor system."), and temperatures within the reactors began to rise. Then, he said, the limits of materials began to dictate the sequence of events.

"At the point where about half of the core was exposed, the cladding temperature exceeded 900°C. Keep that number in mind as we talk about materials advancement," he said.

Marra emphasized, "This was Zircaloy cladding. And at 900°C, the Zircaloy was losing a lot of its structural properties, and it started to balloon and break, and that begins the release of the fission products from the cladding gaps."

The ballooning and breaking Marra described is an artifact of the alloy's crystallography. At room temperature, Zircaloy alloys are an hexagonal close-packed alpha phase. During cold working, a preferred crystallographic orientation (or texture) develops, which allows properties to be directionally optimized. For example, Zircaloy manufacturer ATI Wah Chang's technical data sheet reports that the room-temperature yield strength of a 2-millimeter annealed strip of Zircaloy-4 is 80 megapascals in the longitudinal direction and 468 megapascals in the transverse direction. At about 810°C, body-centered cubic beta phase begins to form with complete transformation to beta phase at about 980°C. With the transformation, properties become isotropic and directional property advantages are lost. As temperatures in the Fukushima reactors climbed over 900°C, the cladding temperature was well within the hcp-to-bcc transformation range.

When the core reached three-fourths exposure, according to Marra, the cladding temperature rose to about 1200°C and began to react exothermically with steam in the reactor, producing hydrogen gas.

$$\boxed{Zr + H_2O \rightarrow ZrO_2 + H_2 \text{ (exothermic)}}$$

This caused the dramatic explosion (broadcast instantly around the world) and released the accumulated fission products into the atmosphere, which led to more heating of the core. As temperatures reached 1800°C, the cladding metal and steel structure of the containment vessels in Units 1, 2 and 3 melted.

"At 2,500–2,700°C, things get even worse," Marra said. "You begin to form [U–Zr] eutectics, which are tremendously corrosive and lead you to all kinds of problems."

In June, the Japanese government admitted that it is likely that the nuclear fuel rods in the three stricken reactors melted, breached the containment vessels and pooled in the outer steel containment vessels.

Ultimately, the reactor cores were drowned with seawater, and cooling water was restored to the reactor cores (within seven hours for two of the reactors and in 27 hours for the third).

The ceramics challenge: Mainly new uses for old materials

Marra predicted that the most obvious materials challenge is to find new cladding materials that can withstand much higher temperatures. He predicted that Zircaloy could be on its way to obsolescence as cladding and wryly noted, "Metals are just reduced ceramics, so there is an opportunity for [the ceramic materials community] to improve things."

He said there was a high likelihood that ceramic materials would be adopt-

Small modular reactors – Next gen nuclear?

The DOE defines SMRs as reactors fabricated in modules that are transportable by truck or rail and designed to generate up to 300 megawatts of electricity, although some companies have proposed 35–45 megawatts of electricity designs. SMRs are an adaptation of mobile nuclear power plant concepts, such as those found on aircraft carriers and submarines. Design work for stationary electric power generation has been underway since at least the early 2000s, although the agency's Office of Nuclear Energy Small Modular Reactor program was established only in FY 2011. The most likely to deploy first are light-water reactor designs (5–10 years out). Non-LWR designs, including a possible novel "traveling wave reactor," are not expected to deploy for 10–25 years. In the post-Fukushima era, materials that can perform in high-temperature, oxidizing environments will be sought.

Small

- 100–300 megawatts of electricity output
- 10–30 year fuel life
- 6 feet diameter reactors (instead of 20 feet for full-size)
- Less risk for capital investors (hundreds of millions versus several billions of dollars)
- Sized to match local demand or grid capacity

Modular

- Factory produced, fueled and sealed
- Transportable by truck or rail
- Add modules to increase power output
- Standard fabrication processes (casting, forging, etc.)

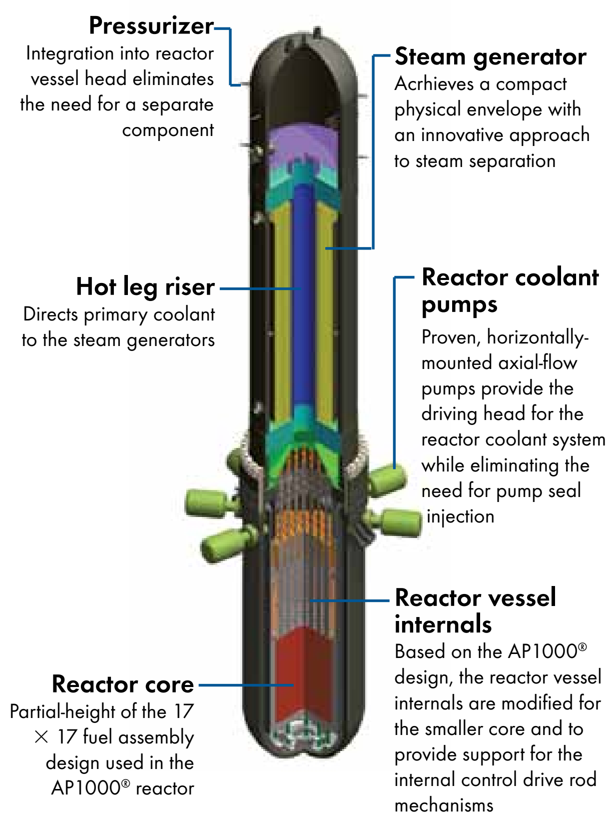
Reactors

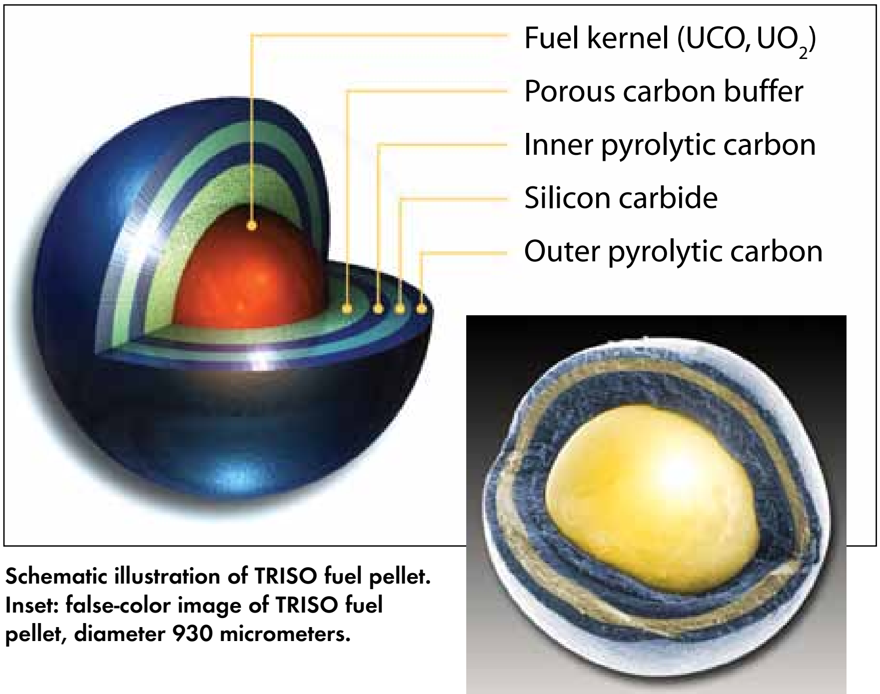
- Less fuel and smaller emergency containment zone
- High efficiency
- Passive air cooling (inherent safety)
- Underground construction possible
- Design simplicity and proliferation proof
- New materials developed for components

Active developers (partial listing)

- Babcox & Wilcox
- GE-Hitachi
- Hyperion Power
- NuScale Power
- Olgethorpe Power
- Sandia National Labs
- Tennessee Valley Authority
- TerraPower
- Toshiba Corp.
- Western Troy Capital Resources
- Westinghouse

Westinghouse SMR





(Credit: Idaho National Laboratory)

Schematic illustration of TRISO fuel pellet. Inset: false-color image of TRISO fuel pellet, diameter 930 micrometers.

ed in future nuclear designs to improve operating performance and safety. In his mind, if nothing else, a ceramic cladding would "buy time" in emergency situations. The example he used for a future cladding material is silicon carbide. With a nominal melting temperature of 3,245°C, SiC would be able to withstand reactor temperatures well beyond what zirconium alloys can tolerate. "In the event of an extreme incident like Fukshima," said Marra, "the ability of materials to withstand very high temperatures would give plant operators additional time before materials start to fail."

Marra offered that the claddings would be just the beginning. "For example, if SiC claddings are adopted, new glass-to-metal seal materials would be needed to seal endcaps to claddings and maintain the pressure barrier. And, while nuclear fuel is expected to be oxide-based for the foreseeable future, there are alternative fuel configurations designed and developed that use silicon carbide as a clad for tennis-ball-shaped fuel assemblies," he said.

The benefits of these ball-like pellets of fuel (known as TRISO or triple-coated isotropic fuels), according to Marra, is that they would self-encapsulate spent fuel and help prevent accidental release of fission products.

Returning to the topic of SMRs, Marra said materials such as SiC, SiC–SiC composites, pyrolytic carbon or carbon–carbon composites may find applications in these minireactors, perhaps for the reactor vessel itself.

And, while not totally new, Marra reminded us that waste containment continues to be a pressing materials problem and that ceramic and glass materials have long been used in this application.

Hurdles to qualifying new materials

The speed of the incorporation of any new materials is uncertain. Although economics and safety will be the primary drivers for adoption of new materials, Marra cautioned that the Nuclear Regulatory Commission can take up to 15 years to qualify new materials for wide-spread use as reactor components. Thus, "the first 'new' materials likely to be used are those about which much is already known like silicon carbide, silicon nitride and carbon–carbon composites," said Marra

In light of timelines like these and the added scrutiny and political pressure that the Fukushima incident will inevitably create, the Obama administration's goal of 80-percent CO_2 reduction by 2050 will make the next 39 years very challenging. But Marra thinks it will be an era when the talents of innovative ceramics and glass specialists will be in high demand.

"Ceramic materials have long played a very important part in the commercial nuclear industry with applications throughout the entire fuel cycle, from fuel fabrication to waste stabilization. As the international community begins to look at the next-generation nuclear technologies and advanced fuel cycles that minimize waste and increase proliferation resistance, ceramic materials will play an even larger role," Marra said.

The bottom line for Marra is that nuclear power generation will inevitably continue to be an important contributor to the energy portfolio of the US and the rest of the globe. He said the Nuclear Regulatory Commission is conducting a comprehensive review of the safety of domestic nuclear plants, and DOE's National Nuclear Security Administration is reviewing its efforts to plan and respond to nuclear emergencies. "Just as lessons learned from Three Mile Island improved design and practice," Marra said, "so, too, will the ones from Fukishima guide the future." ■

For more information:

• International Atomic Energy Agency's "Fact Finding Expert Mission of the Fukushima Dai-ichi Accident Following the Great East Japan Earthquake and Tsunami, www.iaea.org

• National Nuclear Safety Administration, www.nnsa.energy.gov

Nuclear Regulatory Commission, www.nrc.gov