Metallic Fuels for Fast Reactors
Dr. Steven Hayes, INL, USA

Berta Oates: Good morning. Welcome, everyone. Today's presentation for the Gen IV International Forum webinar series is on metallic fuels for fast reactors. Doing today's introduction is Dr. Patricia Paviet. Patricia is the Director of the Office of Materials and Chemical Technologies in the Office of Nuclear Energy in DOE, and she is also the Chair of the GIF Education and Training Task Force.

Patricia Paviet: Thank you, Berta. Good morning, everybody. It's a pleasure today to have Dr. Steven Hayes with us. He's a Fellow of the Nuclear Science and Technology Directorate at the Idaho National Laboratory. During his career, he has been engaged in the development, testing, and modeling of a variety of nuclear fuels, including metallic, oxide, and nitride fuels for liquid metal reactors and high-density dispersion fuels for research reactors.

He led numerous fuels and materials irradiation experiments in the Experimental Breeder Reactor II prior to its shutdown, and today he maintains an active fuel testing program in the Advanced Test Reactor.

Dr. Hayes is a national leader in the development and testing of metallic fuels for the US DOE’s Advanced Fuels Campaign and in the development of multiscale, multiphysics fuel performance codes for the US DOE’s Nuclear Energy Advanced Modeling and Simulation program.

I would like to thank you again, Steven, for volunteering to give this presentation, and I give you the floor. Thank you, Steven.

Steven Hayes: Thank you, Patricia, for this invitation, and I appreciate the forum to make this webinar presentation on metallic fuels. So we'll be talking about metallic fuels for application to fast reactors, specifically sodium fast reactors, but in general, fast reactors.

And outline on my presentation so you know sort of what I'll be covering this morning. I'm going to start with a little background. Motivation. Certainly metal fuels have a number of different applications for which they can be used, not all of them necessarily include actinide transmutation, but to be honest, the work that we've been doing to advance metallic fuels technology in the last ten or 15 years has been with a view to an actinide transmutation mission. So I'll start by talking about why that might be a good thing to do and the reasons for it.

And then for those who are new to metallic fuels perhaps, I'll step back and give a little bit of history on who has used metallic fuels down through the nuclear world and what we perceive as their benefits.

I'll talk about casting process development. So in line with the actinide transmutation mission, there are some issues that have arisen with the historical way that metallic fuels have been fabricated using casting that creates some difficulties and some effects that we'd rather not have. A lot of the fabrication development work we've been doing lately has been to address those issues. I'll explain the issues and talk about what we're doing to address them.

A little bit of an overview on the irradiation testing that we've been doing on metallic fuels in recent years. Again, specifically looking at the issues associated with actinide transmutation.

And then before I end, I'll spend a little bit of time on future directions, and these are some things we're doing to enhance or to advance metallic fuels technology that don't necessarily have anything to do with actinide transmutation but broader issues related to improvements to metallic fuel, and that's what I'll end with, and then provide a summary, conclusions, and hopefully be able to answer some questions.

Okay, moving into it, the background section.

This is a slide many people have probably seen over the years that sort of sets the stage for why actinide transmutation might be something you'd want to consider doing. If you plot some parameter – and you can use different parameters – some parameter that talks about the hazard associated with spent nuclear fuel after it's discharged with the reactor and compare it to some metric like natural uranium ore, what you find is that if you were to just directly dispose of spent fuel, say, in a geological repository, the hazard associated with that material would remain well above the natural uranium ore that you started with for tens of thousands or perhaps many more years than that. So you're faced with licensing a repository of some kind that you understand and can control its performance for tens of thousands of years, and again, perhaps much longer than that.

As a human race, we don't have a lot of experience doing that. Well, we have none. So people who have looked at this have realized that if you did nothing more than remove the actinides, all the actinides, from your spent nuclear fuels, that helps you in your long-term hazard associated with that quite a bit. When you're just disposing of fission products and activation products, that material will decay below your metric in just a few hundred years, and so that creates the motivation for an actinide transmutation mission, a prospect of being faced with licensing an ultimate geological repository and controlling its performance for a dramatically shorter period of time.

Now, if you're going to achieve this benefit, you really have to get as close to this line as possible. So if during your recycle and your refabrication of fuels you are losing materials to various waste streams and those waste streams eventually come into the stream of material that will go to the geological repository, what you'll find is this ideal curve starts to move out pretty quickly, and so the benefits you can derive quickly begin to evaporate on you.

So when I talk about later of the steps or the effort we're going to to minimize waste streams in our fabrication and recycle processes related to metallic fuel, it's for this reason. It's to get the real curve as close to this ideal curve as possible so that maximum benefit could be derived.

Now let me just say quickly, the US as a nation has not taken the position today that actinide transmutation and removal of all actinides from any waste stream that goes to repository is what we're planning to do. That's not a decision that's been made yet, and yet the Department of Energy is funding long-term research into these areas to more or less keep the option on the table, and when a decision is made at some future time, hopefully there will be technology available to perform the actinide transmutation mission if that's the decision on the direction our country wants to go.

Now metallic fuels, I will make the case, will be very helpful in an actinide transmutation mission, but the real key to actinide transmutation is fast reactors, and in the US we've had a couple recently, EBR-II and FFTF. The fast reactors are an indispensable part of an actinide transmutation mission if you want to undertake it. You really can't pull it off with just thermal reactors. You need fast reactors for a couple of reasons.

One, you need a lot of excess neutrons because a reactor has got to stay critical and yet then have a lot of extra neutrons available to do your mission of transmuting actinides in his case.

It's also extremely valuable, if not necessary, to have neutrons of very high energy. Of course you have high energy fission neutrons in a thermal reactor, but they're quickly slowed down, whereas in a fast reactor you maintain a large popular of neutrons with very high energies, and that's helpful in many threshold reaction that you can take advantage of for an actinide transmutation mission.

So these two features of a fast reactor really allow you to look into a variety of actinide management, strategies, approaches, schemes, one of which could be breeding, and historically that's been of course the motivation for fast reactors. You use those excess neutrons to convert fertile material to fissile material for resource extension. Not so much of interest today. Today the interest is more related to number two, waste management. Use those excess neutrons to transmute and eventually destroy your actinide in an actinide transmutation scheme.

If you're going to undertake an application like that, actinide transmutation, you're going to use a sodium fast reactor to do it. The fuels could look at little bit differently, a little bit different than what they've historically looked like, especially related to minor actinides and rare earths.

So some of the unique features of transmutation fuels could be the plutonium content. Depending on what conversion ratio, what transmutation rate you want your reactor to achieve, you might want to drive that conversion ratio down to maximize the destruction rate, and if you do that, you might want to increase the plutonium content. So we've been looking at plutonium contents in metallic fuels well above the historic levels.

Of course the mission is to transmute actinides, and so you're going to have actinides in your fuel, including transuranics, the minor actinides, americium, neptunium, even curium may be present in significant quantities. And then, because an actinide transmutation mission necessarily would be a scheme where you'd be interested in continuous recycle, you might have fission products being carried over from your recycle processes. Or metallic fuels, they typically use electrochemical recycling, and that tends to bring some small quantity of the rare earth fission products over from recycles into the next refabrication step, and so that's something that is a bit different.

And all of these features give rise to some challenges that will need to be addressed and some unknowns that will have to be resolved. With the high quantity or the higher quantity of minor actinides and potentially some fission product carryover, you're probably going to be faced with the need to remotely fabricate your fuel, and so by that I mean fabricate inside a hot cell, perhaps the very hot cell you're using to do your recycle step.

You may need, you'll likely need, either new fabrication methods or some enhancements to existing fabrication methods due to the unique features of these kinds of fuels. Two that come to mind would be, the elevated quantity of americium could pose an issue, especially in metallic fuels for using high temperature casting processes. Americium is a very volatile constituent that will evaporate under certain conditions, so you'll need to address that in your fabrication.

And then, as I mentioned before, just an overall strategy of waste minimization, you really need to keep as many of the actinides out of your waste streams as possible to derive the goals that you're trying to achieve overall with the actinide transportation mission.

And then, lastly, and perhaps most importantly, it's just effects on fuel performance. These are changes to historical fuels and we have to understand what changes in performance they might bring.

So that's the background and the motivation for an actinide transmutation mission and the need to develop some fuels that would fit that mission. Now I'll step back and do just a few slides on the history and benefits of metallic fuels in general for those who might not be as up to speed on metallic fuels as others.

Metallic fuels have really been around since the earliest days of the nuclear reactor industry. Metallic fuels were used in the experimental breeder reactor in the US, the first one, which eventually became to be known as EBR-I. It used a variety of fuels in its lifetime; all of them were metallic alloys.

Over in the UK, the Dounreay Fast Reactor used metallic fuels. Back in the US we actually had for a period of time, back in the '60s, a commercial power reactor, a sodium fast reactor that was a commercial power reactor, and it used metallic fuel. But then, the real volume of information that we have on metallic fuels and the platform used to really develop the technology to the point where it is today was the Experimental Breeder Reactor II here in the US. And it started with fissium, U-5f fuel. These were fuels that just naturally came out of a melt refining process. We used those for many years. And then when recycle was detached from the reactor, the reactor eventually moved to the metallic fuel that's probably most familiar to people, U-10Zr and then U-20Pu-10Zr, these are the main metallic fuel alloys in modern times. And EBR-II operated on metallic fuels for over 30 years.

Now FFTF here in the US was a much bigger sodium fast reactor that operated in the '80s, early '90s. It was fueled by MOX and yet there was a lot of metallic fuel testing performed in FFTF, and in fact, U-10Zr, all the qualification assemblies were completed in FFTF to allow the reactor to be converted to metallic fuel, and had it continued operating, that was likely to be the case.

So that's sort of the history of metallic fuels down through the last 50 or 60 years.

I personally think there are many benefits to metallic fuels, but for those less familiar with the technology, let me just summarize sort of the key features of metallic fuels.

Again, in modern times, the alloy base is either uranium-zirconium or uranium-plutonium-zirconium, as the starting point. Many years of experience with this alloy system has shown it just has really good irradiation stability, and its effects are very repeatable and understandable.

Now one thing associated with metallic fuel is it does swell quite a bit in the early stages of irradiation. Within the first couple percent burnup, metallic fuel will swell 30% quite quickly. After that, the swelling rate decreases dramatically and it's not so much of a problem, but you have to be able to accommodate that initial swelling that you get with metallic fuel, and typically that's done with controlling smeared density.

Early metallic fuels used much higher smeared density and therefore weren't capable of going to very high burnup, and the early reputation of metallic fuels was that it didn't have burnup capability. But over the years, the smeared density was decreased and eventually settled in at about 75% of smeared density, so that provides plenty of void volume to accommodate that initial swelling, and then after that, metallic fuels can go to quite high burnup. That was a huge step forward in metallic fuel technology.

Now with that big of a gap in metallic fuels, you have to put a liquid metal bond in the fuel cladding gap, and typically that's been sodium. So liquid sodium in the fuel-cladding gap is the bond that keeps the fuel temperatures low, provides a lot of other benefits. It's just generally a good thing, but it's an added complication in fabrication.

Metallic fuel, because of the large amount of swelling that happens early on and those gas bubbles interconnect very early in life, and then the metallic fuel will begin releasing most of its fission gas on the order of 80%, so you need a large fission gas plenum to accommodate the high gas release. That's not really any different than MOX fuel fast reactors.

And then we've evolved to marrying this fuel technology up with low-swelling ferritic/martensitic cladding, cladding and ducts, which allow it to go to quite high burnup.

Here's a little sketch of what I've described of metallic fuel looking like.

Now this technology has some historic benefits, like I said, once the lower smeared density approach was undertaken and when metallic fuels had proven to be capable of operating reliably to very high burnups. The EBR-II driver fuel was approved to 10% burnup and never experienced failures at that burnup level. Experimental fuels and experimental subassemblies went to much higher burnups, 20% or even above.

The natural complement to metallic fuels is to use electrochemical recycling if you're going to do recycling, and that comes with certain benefits in and of itself. One of the great benefits of metallic fuel has been just the ease and the compactness of the fabrication processes that are typically used to make metallic fuels. They're simple, they're compact, they are readily deployed in a remote environment if need be, and they tend to keep the costs associated with fabrication much lower than, say, an oxide fuel fabrication cost.

And then lastly, although I'm not going to talk about it really during our presentation today, but metallic fuels have certain natural characteristics that marry well with an overall passive safety approach to reactor safety.

Now when you take these features of metallic fuel, and especially the historical benefits of them, they tend to lend themselves well to an actinide transmutation mission. In the area of fabrication, as I mentioned, the historic ease of fabrication, even on large scale, in a remote environment, is a plus because with an actinide transmutation mission you're probably faced with fabrication in a hot cell – that has been done with metallic fuels. I'll say more about that later.

The fabrication process is not that sensitive to fuel composition, and if you're doing a continuous recycle approach, there are going to be some swings in your fuel composition. Metallic fuel fabrication typically handles those quite readily, the exception, the major exception, being the americium. Something is going to have to be done to address the volatility associated with americium.

And sodium bonding, while it's an extra step in the fabrication process that costs a little, brings some benefits with it. Because there's excellent heat transfer between the cladding and the fuel, you don't have to worry about tight tolerances on fuel diameters needed to maintain a very small gap, and it just gives you a lot of thermal margin.

In the irradiation performance area, I'll show a little bit on this later, but what we've shown over decades of experience with metallic fuel is the performance tends to be very consistent and quite similar over quite a range of compositions and deviation in a metal fuel alloy, so you don't have to keep content controlled within a very narrow band in order to know how your fuel is going to perform.

Metallic fuel is entirely compatible with sodium. In fact, there's bond sodium on the inside of the pin so it makes for the possibility to have failure of running beyond cladding breach in a very benign way.

And as I mentioned before, the demonstrated high burnup reliability of metallic fuels is a huge plus in your actinide transmutation as well because if you're worried about controlling losses during fabrication or losses during recycle, then the more times you have to recycle and refabricate, those are just added multipliers to your mass going out in waste stream, so if you can drive the burnup higher, you can drive the waste stream actinide content in them even lower.

Okay, so that's the history and benefit of metallic fuels in general and some issues related to actinide transmutation. We're now getting into a little more of the technical details, starting with fabrication, specifically casting.

Casting is the approach that has historically been used to fabricate metallic fuels and specifically counter-gravity injection casting. So the way this is performed is we typically use a graphite crucible. Now metallic fuel is not really compatible, molten metallic fuel, is not compatible with graphite, so the graphite crucible has to be coated, typically with an yttria wash, so a coated crucible is used to heat the metallic fuel charge up to a molten state, initially under an inert atmosphere, but once it's achieved its molten temperature, it's melted with a proper superheat, the furnace chamber is evacuated, now historically to a very hard vacuum.

And then you bring in your molds from the top, and these are quartz or silica, silicon oxide molds, metal fuel is not very compatible with quartz either so the quartz molds have a wash on them as well, zirconia typically, and then after you've plunged the molds into the molten metallic fuel, the furnace is rapidly pressurized, and it injects that molten material up into the quartz molds. It solidifies almost instantaneously. And then you remove your mold palette, and you just shatter, you just mechanically break the quartz molds off of the metallic fuel pins, typically called slugs. You section them to length and then you use them to fabricate your metallic fuel pins.

The crucible is generally reusable a number of times, but it does have a reaction product on it that has to be removed by typically wire brushing. It has to be recoated, but the crucibles typically are used repeatedly.

I said previously that this is a process that is very amenable to deployment in a remote or hot cell environment. In fact, that's not just a theoretical statement; this was in fact done back in the late 1960s at EBR-II. Over 39,000 metallic fuel pins were fabricated in the hot cell, introduced back into EBR-II to demonstrate their performance over a three-year period, and there were no issues associated with that.

In fact, Berta, if I could get you to advance the slide for me.

Now the traditional process of metallic fuel fabrication is extremely consistent and repeatable. It's been used for decades with great success, but there are some issues associated with it that create problems or limitations if you were to use this for an actinide transmutation mission, and this slide reviews some of those.

The first thing to note is the counter-gravity approach to injection casting. It's just not possible to use the entire molten charge of metallic fuel alloy when you're casting in a counter-gravity mold. You're just necessarily going to be left with a pretty large heel in that crucible, something like 30 to 40% of your initial charge will be left behind. Now you can recycle it, but with each recycle it picks up impurities, and that's not a great thing.

You also have some other waste fuel loss streams. The yttria coating that you use with the crucible will react with the melt and you'll have some actinide or fuel loss associated with that. The quartz molds and the zirconia wash used with them will react with the fuel as well prior to its solidification, and you'll have an actinide loss stream there, some of which is recoverable but not without difficulty.

And then as I mentioned, the traditional approach takes the furnace chamber down to a hard vacuum prior to casting, and that can exacerbate loss of any volatile constituents that may be in the melt, and specifically, it does exacerbate the loss of americium if your fuel has a significant quantity of that.

So there are some significant loss mechanisms associated with the traditional approach in casting. Then beyond that, you do have graphite crucibles that eventually cannot be reused and take with them some fuel material into the waste stream. And even more of a concern are all these shattered quartz molds that take with them into the waste stream.

So in an actinide transmutation mission where you're not wanting actinides of any significant quantities in your waste streams, these represent problems or issues you would like to eliminate. There are also just some handling difficulties associated with crucibles, the way they're reused historically that make the process labor-intensive. You really wouldn’t want to do it in a hot cell.

So with all that in mind, what we've been looking at over the last decade or more are some enhancements to the casting process that would address some of these issues. We still like casting, although admittedly we're looking at some other fabrication approaches for metallic fuel, but we still like casting because it is very amenable to a remote environment and it is very cost effective if you can make it work for you.

Some of the things we've been looking at to address the issues are very rapid cycle times, just to cut down on evaporation and reaction by minimizing the time available for it, as well as developing advanced crucible and mold materials to address the reaction issues as well. So if you can minimize the fuel losses and coatings and make the molds reusable in and of themselves as well, that can help you minimize the waste dramatically. It can also help you eliminate these time and labor-intensive processes typically associated with crucible preparation, cleaning, recoating, that sort of thing.

The other major thing that we've done is we are looking at a bottom casting process. So here is an experimental furnace that we developed early on and used for testing and optimization of various things. And you'll see, here is the crucible, it's still a crucible that's heated by induction, but now we're going to put the mold palette down below the crucible, and that benefits you in a couple of ways. First, since you're not bringing in a molten palette from above, it allows you to put a lid on the crucible and cut off loss by evaporation that way.

But it also, since you're draining the crucible from the bottom in the mold palette below, it allows you to in theory utilize 100% of the charge of every casting batch. So these things have greatly improved casting and minimized the waste associated with the traditional approach.

So we've done a lot of testing along these lines to make sure that they do in fact bear out in practice and not just in theory. We started actually with the drawing of the furnace I showed in the previous slide. We fabricated one of these units and used it for a number of years just in a hood, testing that goes that much more efficiently. We did cast with uranium in the hood, but we used various surrogate materials to simulate other constituents. We used manganese as a surrogate material for americium having a very similar issue with volatility, and we proved out a lot of the ideas we had in the hood and then we went to the next generation of furnace, which was installed in a glove box, and validated much of what we'd learned using surrogate materials in a glove box using actual materials, actual actinide materials, so casting with uranium, plutonium, americium.

What we learned is these changes to the approach did two things for us. One, they did greatly improve melt utilization by being able to drain the entire crucible in every casting run, and they really did drive the americium loss during each fabrication pass down to essentially zero.

So that's what we've done in the area of fabrication to address issues. How do these metallic fuels perform?

Well, we've been looking at their performance primarily in the advanced test reactor here in Idaho, and about 15 years ago we started in a test series to look at the performance of these new fuels. AFC-1 was a series of experiments we did in ATR. Initially we were looking at both metallic fuels and nitride fuels. We started looking at the baseline composition and what effects were observed when we introduced minor actinides.

The AFC-II test series, we were still looking at metallic fuels, and now we were looking at some oxide fuels as well. Again, the approach was to test baseline materials and then start adding minor actinides to see what the effect was, and then even adding rare earths to simulate what might be coming over with carryover and look at performance effects that could be caused by that.

Those are two test series that are over now, and we're in the AFC-3/4 test series now. Here we're looking almost exclusively at metallic fuels and not just the actinides transmutation mission, but when I get to a little bit later near the end, what we call the advanced metal fuels concept, the additional design features that are being incorporated into metallic fuels for various reasons, the AFC-3/4 test series is exploring the performance of a lot of those as well, but also with the minor actinides situation also being looked at.

So we're in the middle of the 3/4 test series now, and hopefully next year we'll be starting a new experiment called IRT. This is the Integrated Recycle Test, and this is the test we're doing in collaboration with the Korea Atomic Energy Research Institute, specifically looking at performance effects associated from not simulated but genuine recycled fuel. So part of this project is an actual recycle of fuel, taking that recycle feed, using it to fabricate metallic fuel in the hot cell, and then introduce it back into the reactor, in this case ATR, to determine how it performs. That should be a test starting next year.

Now as I mentioned, we're doing this testing in the ATR, so these are fuels designed to fuel a fast reactor, but we've done most of our testing in the ATR, which is a thermal spectrum materials test reactor. It's not entirely, it's not at all a prototypic environment the way it starts out, although we've modified the way we do these tests to make it a bit more prototypic.

So here's a cross-section of the ATR core. Initially we were testing in the east flux trap. We've moved now to these positions called outboard A. Here's one of the outboard As, but there are actually four of them, one in each four quadrants. And within each one of these outboard A positions, we can test five rodlets. So rodlets are miniature rods. In the diametral dimension, they are essentially prototypic but they are much shorter rods.

And, as I said, we have four testing positions, and in each position we can test up to five rodlets, so we can have up to 20 different rodlets under irradiation at any given time, and most of the time we do have most all of those positions populated by fuels that we're testing.

The test vehicle that we've developed, it's a double encapsulated testing approach, so your miniature rodlet is then put inside a secondary steel capsule with a very small gas gap between the capsule and the cladding that's used to raise the temperature of the cladding up into a temperature range you'd really like it to be. With this testing approach we've been able to test at linear powers up to about 500 W/cm. You get the cladding temperature as high as 650 – that's not where we typically test it. Typically, it's more about 550, but it's not exposed to the water coolant; it's just exposed to inert capsule gas.

Now this approach comes with some complications associated with neutron energy. So you can see from this graph here, this is what the neutron spectrum really should look like if these were being tested in a real fast reactor. You'd essentially have no epithermal, very few epithermal and no thermal neutrons in a genuine fast reactor. The yellow curve is the ATR spectrum. So you have your fission, your high-energy fission spectrum, a lot of epithermal neutrons associated with slowing down, and then a large thermal peak. This would be a neutron energy spectrum entirely unacceptable for testing these fast reactor fuels, and so what we've done is we've come in and we've built into our test vehicle a cadmium shroud, so the rodlets are entirely shrouded by cadmium, and this dramatically alters the spectrum that the test fuels see. So that's what you see here in the blue. You still have your high energy fission spectrum. You have a lot of epithermal neutrons. The thermal neutrons below the cadmium cutoff are essentially entirely removed from the test fuel. We were able to achieve greater than 99% removal of thermal neutrons seen by the test.

And what that altered spectrum does is, it certainly doesn't change the test spectrum into a prototypical fast neutron spectrum. It doesn't do that. But what it does is it eliminates the thermal neutrons. Most of the fissions are being driven by epithermal neutrons, which are much more penetrating in these high density fuels than thermal neutrons, and what we've done is eliminated the gross self-shielding that these fuels would experience if we just put them into a thermal spectrum.

And these curves attempt to show what we're achieving. So this is local to average power plotted versus fuel radial position. So this here zero is the fuel center line, and out here at 1 would be the fuel surface. And if we just put our test fuels into an unaltered thermal spectrum, you would see massive peaking of the power at the fuel periphery and serious depression of the power in the fuel center. And what that would to you temperature-wise would be something not very good, and so this is a temperature plot, fuel temperature versus radial position of the fuel.

Now I've done this plot for one of our oxide fuel tests just because the effects are exaggerated in oxide fuel, which has a very low thermal connectivity, but a similar effect is achieved in metallic fuel. And so this flat, essentially flat power profile that you would expect in a fast reactor because the main free path of a fast neutron is so much larger than the fuel pin dimension, the power profile will be flat in a fast reactor and will result in a fuel temperature profile that looks like this blue curve.

But if you have this depressed, this power profile depressed in the center and peaked in the periphery, what you do is you shift your power production out to the outer portion of the fuel, and you bring your temperature profile down dramatically in the center of the fuel. And of course this would be a non-prototypical temperature profile that you'd be testing under, but what we've been able to achieve with the cadmium shrouding is a change that looks like these yellow and red curves here.

So it still does have some peaking at the periphery. It still has some depression in the fuel center, but these have been dramatically reduced, and when you translate this power profile into a temperature profile in the test pins, what you see is a profile that is very close to what you would expect in a genuine fast reactor. It's certainly not the dramatically altered profile that you would have in a thermal spectrum.

And so while we haven't achieved a prototypic neutron spectrum inside the test articles, what we have achieved is a near-prototypic temperature profile, and so for those processes, those fuel performance processes that are primarily driven by temperature, temperature and temperature profile effects, we should be capturing those pretty well.

Berta, if I could get you to advance the slide for me.

So the next slide is entitled metallic fuel alloys tested in the ATR. So we started this test program back in about 2003. We've been doing the testing now for almost 15 years, and over that period of time we've tested a wide variety of metallic fuel alloys in the ATR, in this cadmium shrouded testing configuration. In fact, I haven't updated the bottom of the chart here. There are some additional alloys related to the advanced metallic fuel concept that could be added, but you're certainly looking at all the alloys we've tested that have been looked at relative to actinide transmutation.

We've looked at some alloys in the early days that were an accelerator driven system, as alloys, these alloys without uranium wouldn't be of interest to reactor or critical systems. But for critical systems we've even tested a wide spectrum of metallic fuels, some with quite high contents of plutonium, which would be capable of driving down the conversion reactor, the conversion ratio of a reactor maximizing minor actinide destruction, some significant quantities of minor actinides. We've looked at tests with simulated rare earth or lanthanide carry-over, and we've taken some of these fuel tests to very high burnups.

Now, I'm not suggesting that we demonstrated fuels like this are capable of 30% burnup necessarily. We are testing in this non-prototypic ATR environment. We don't have the fast neutrons, the high quantities of fast neutrons that typically grade cladding performance and can limit burnup capabilities, so we're not capturing every effect we need to capture to demonstrate high burnup. But we're capturing many of the fuel behaviors and things are looking good so far.

What do I mean by, things are looking good so far?

What we've found is even when you vary compositions of metallic fuels dramatically with minor actinides or without, with a variety of minor actinides, concentration levels, with plutonium going significantly above the level of 20%, which is historically what's been studied, what you find is when you plot the performance parameter, whatever it may be, in this case fission gas release, versus fission density, and you do have to plot it versus fission density instead of versus percent burnup because what has changed is all these different alloys, especially alloys with different levels of zirconium, they have different densities, so their performance doesn't correlate directly with percent burnup the way it does when the density remains constant. But when you plot it versus fission density, the curves all tend to line up on the same trajectory.

And so you tend to get very consistent performance. And what I mean by that is, we've tested this base metal fuel alloy, uranium, plutonium, americium, neptunium, zirconium, and potentially with rare earths added to simulate recycle carryover. We've tested zirconium between 10 and 40%, plutonium up to almost 35%, americium and neptunium up to 10% or more, as much as one-and-a-half percent rare earth carryover, and even with these big changes in composition to the fuel, the performance characteristics, whether it be fuel swelling, fission gas release, fuel cladding interaction, it tends to be what we call typical. And we say metallic fuel behaves like metallic fuel with very consistent performance irrespective of really some pretty big changes in composition.

So that's been positive for our work relative to actinide transmutation. Actinide transmutation would bring with it a significant swing in composition, depending where you are in your equilibrium cycle, and that doesn't pose a big problem for metallic fuels. They tend to be able to handle that and accommodate it without too much issue.

Now just a few slides on future directions.

So the actinide transmutation work that we have been doing continues, but we're looking at some other things relative to metal fuel too.

A number of years back, I think it was about 2010, we were challenged to take a step back not just with metallic fuels but with all the fuels the DOE was looking at, and say, or ask the question, what innovations or what changes could be brought to current technology that would offer the potential for some sort of multifold advance in fuel reliability and performance? And we took a hard look at metallic fuels and we decided there are a number of things we'd like to look at that might fit that bill.

One of the features was decreasing the fuel smeared density. Metal fuel smeared density was reduced to about 75%, and that gave the ability to get well over 10% burnup, but it doesn't allow you to go to ultra-high burnups that some people are now interested in. For ultra-high burnup metallic fuel, we'd look at lowering the smeared density even more. As you drive smeared density lower, it becomes questionable as to whether or not you still want your metallic fuel slug to be a solid cylindrical slug, and so we wanted to look at annular fuel form, specifically with the lower smeared density as well.

Looking at coatings and liners on the cladding inner diameter to deal with the fuel cladding-chemical interaction issue.

I'm not going to talk anything about vented fuel pins, but for those concepts that want to take fuels to ultra-high burnups, 30 and 40%, you are almost certainly faced with having to look at vented fuel.

We wanted to look at optimizing the alloy base. We've been using just straight zirconium for many decades. Are there some changes to the alloy system that could bring some improvements overall?

And then very specifically we wanted to look at some target fuel allow additions, some minor alloy additions, that might help with a very specific issue, which I will talk about in the next slide or two.

If you're going to decrease your fuel smeared density and look at annual fuel, that raises the interesting possibility of fabricating fuel by extrusion perhaps being a better approach.

And then lastly, this is not an innovation per se but it's more of a guiding principle, we didn't want to introduce any innovation or change to metallic fuel technology that would affect its compatibility with the standard electrochemical recycling scheme that's historically been used.

Okay, I'm just going to highlight a couple of these features. I won't talk about all of them. I'm going to highlight decreasing the fuel smeared density and these targeted fuel alloy additions.

Decreasing the fuel smeared density. As I said before, metallic fuel swells very rapidly within about the first 2% burnup, up to 30%, but once fuel swelling hits 30%, what happens is the fission gases, bubbles, begin to interconnect, and they begin releasing their fission gas at a very high rate, probably 80% of the fission gas beyond that point is released, and that dramatically reduces the driving force for additional swelling. Now you still have the accumulation of solid fission products that does continue to drive fuel swelling but at a much slower rate, as you can see here, than the gas-driven swelling.

So the reduced smear density, 75%, allows you to accommodate that 30% swelling, but as the swelling continues on, eventually you won't have enough void space to accommodate even solid fuel swelling.

Our estimations are that if you wanted to go to 40% burnup in metallic fuel, you need to take the fuel smeared density down to about 55%. And so we've been doing some testing with 55, 65, 75% smeared density metallic fuel. And one thing that we've noticed is a 55% smeared density metallic fuel with a solid cylindrical fuel slug, there are some questions on how geometrically stable that will be under irradiation. It may be subject to some strange effects. Some people are concerned about slumping that you certainly wouldn't want to happen. And so to address that issue, we've been looking at annular fuel, which may be a better way, a better geometrical form for these low smeared density fuels.

So what you're looking at here is not a piece of cladding. This is an annular metallic fuel slug, 55% smeared density. That would be put right into a cladding tube with essentially contact with the cladding right from fabrication. And an additional feature of an approach like this is, if you don't have a large fuel cladding gap, you essentially have no fuel cladding gap, you put all your void space into this annular, now you don’t have a need for a sodium bond, and in fact, most of our annual fuels that we're testing have no sodium bond.

Now the real question is, what's the swelling behavior of this annular metallic fuel going to be? If it swells outward and begins mechanical interaction with the cladding right away and strains the cladding, that probably will be a feature that will limit burnup. What we were hoping was that the swelling would go inward, into this central void, this annulus, and then it would be easily accommodated. But that had to be demonstrated by testing.

The other thing that we've been looking at out of what we call these targeted fuel alloy additions. Those who are familiar with metallic fuel performance know that one of the issues, one of the limiting issues that we have is fuel-cladding chemical interaction. So this is a piece of cladding here. Fuel would be down below it. And not the fuel alloy itself but specifically the rare earth fission products, which are created by fission, tend to migrate to the cladding inner surface and react with the cladding inner surface, and they produce this brittle interaction layer that can't support any load, and so as this reaction continues, you're basically reducing the load-bearing capacity of your cladding. But even worse than that, the composition of this reaction product is very much eutectic-like. It's like a eutectic between rare earth elements and iron, and it melts at a relatively low temperature, and so it doesn't tend to be a problem under normal operating conditions, but under sort of transient over-power condition, what you can get is melting at that reaction product, which will then propagate into the fuel.

So this is a limiting performance feature of metallic fuel. One way we're attempting to address it is by developing cladding coating and liners to act as a diffusion barrier. These tend to have a difficult fabrication process.

What we've been looking at recently are, well, we've been asking ourselves the question, is there an alloy addition I can make to the fuel, a minor alloy addition, that would react with the rare earth fission products when they're created, chemically bind with them in the fuel matrix, immobilize them, and if I can prevent them from ever transporting to the classing surface, then I've essentially eliminated the FCCI problem at the source.

So we're looking at a variety of elements that might be capable of doing that. One of the early ones we looked at was palladium. So here you see not irradiated fuel but fuel being examined after casting study. So we cast metallic fuels with the palladium added and with lanthanides or rare earths added as well, and then characterized the macrostructure of what's happening to the lanthanides?

In casting studies we have found that the palladium and the lanthanides do in fact precipitate in phases together, so the palladium is reacting with the lanthanides and hopefully keep them from transporting.

We've also done a lot of out of pile diffusion couple work to look at this issue, so for instance, here's a diffusion couple of iron versus neodymium, a pretty simple diffusion couple, at 700°C, and we held it I believe for 100 hours. And the interface of the couple is here, and what you see is not only extensive interaction between iron and neodymium, but at 700°C there was significant liquefaction of that interface. This is not unexpected. This is what would be expected. This is what you get as a result of FCCI.

But what we did is we looked at a couple where iron was put against neodymium with the addition of palladium in the one-to-one ratio, and not only was there no interaction at 700°C, not only was there no liquefaction, there's essentially no interaction under the same condition, 700°C and 100 hours. So that gave us a lot of hope that a palladium addition could be beneficial for this problem under irradiation.

So those are just two things we're looking at, alloy addition, as well as the geometry of the fuel slug, but we're looking at other things, like tweaking the alloy, the fuel alloy, looking at a variety of smeared densities, looking at different burnup ranges. We are looking at cladding, coatings and liners, and so what you see in the ATR test program is a series of irradiations to explore these various design features.

We designed some of our tests to be very short-term irradiations, get them into PIE early so we can see, is the proposed innovation looking to be feasible, but we're also always interested in hearing some of these tests onto much higher burnup because ultimately we do want to demonstrate high burnup potential.

Okay, that's most of what I have to say. One simple slide to summarize the major things that I'd like to emphasize.

In the area of fabrication, we've done a lot of work on casting especially and our approach to casting to address issues of waste generation, as well as americium loss during volatility. We started that out in a surrogate approach, but we've moved on to validating what we've learned using surrogates in real systems, and looking good. Specifically, the issue associated with americium volatility during casting operation has for all practical purposes been entirely resolved.

Under irradiation performance, I showed really just a huge spectrum of fuel alloys that have been tested in our ATR test program, both with and without minor actinides. Performance tends to be consistently good and consistently typical of stored metal fuel behaviors. So we're confident we can rely on what we've learned over decade and know that most of those behaviors will be borne out as we change the fuels as well.

Now it's definitely the case that we've doing most of our testing in the ATR, which is not an entirely prototypic environment, but what we've been doing is a lot of analysis and collecting data toward publishing what we call a comparison report, and it's going to come out later this year, where we are comparing fuels that we have tested in our ATR cadmium shrouded concept directly within cases where possible identical or very similar fuels that were irradiated in EBR-II, FFTF, and Phénix, genuine fast reactors, under similar condition, to validate that the results that we're getting in the ATR tests do represent what would be expected in a true fast reactor.

And then I ended with where we're going on some future directions related to the Advanced Metal Fuel Concept, additives to control lanthanide fission product and hopefully mitigate fuel cladding-chemical interaction, low smeared density fuels that hopefully will be capable of much higher burnup performance. Our ultimate goal is to demonstrate reliable performance in metallic fuels to ultra-high burnups, 30, 40%.

And I think that concludes my presentation.

Oates: Thank you, Dr. Hayes. If you have questions regarding today's presentation, feel free to type those into the chat box and we will go through and take as many of those questions as we have time for.

A couple of things that we didn't know when we started, I apologize for not taking care of this earlier, the presentation slides, the slide deck as a PDF is available for download in the Files 2 pod. Click that title of the file name and it should download directly to your computer.

Today's presentation is archived and has been recorded and will be achieved and available with that slide deck from the GIF website as well.

And last and certainly not least, we do appreciate your feedback and invite you to participate in an online survey, and the link to that is posted in the Notes pod there for your use.

While questions are coming in, let's just take a quick look at the upcoming webinar presentations in September: a presentation from Dr. Richard Stainsby on Energy Conversion, in October, we have schedule Dr. Geoffrey Rothwell with a presentation on Economics of the Nuclear Fuel Cycle, and in November, we look forward to hearing from Dr. Joel Guidez on the Feedback from Phénix and Superphénix.

And with that, let's turn our attention to the questions.

Hayes: So Berta, you may need to help me.

Oates: There's a question from Ken Karcher. "What is the best current report summarizing metallic fuel irradiation performance and how can one obtain a copy?"

Hayes: Oh, okay, I just found out how to…

Oates: Okay. I was going to say, I knew you would. It just takes a minute.

Hayes: I just can't see the whole question. Thank you. "What is the best current report summarizing metallic fuel irradiation performance?"

So probably, there's a series of journal papers we've been producing on specific things. One that we published seven or eight years ago is Crawford, Porter and Hayes. I thought I had… Ah, "Fuels for Sodium-Cooled Fast Reactors," Crawford, Porter and Hayes. That's published in *Journal of Nuclear Materials*, 2007, Volume 371, starts at page 202. That's probably the most comprehensive summary of metallic fuel performance up to that time.

After that, we've been publishing papers primarily in *Journal of Nuclear Materials*, so look for them there on specific experiments, but we're probably overdue for another review article summarizing what we know since 2007.

Oates: Then there's a very long question.

Hayes: I'm looking at a very long question here from India talking about experimental simulation of sodium infiltration using surrogate materials.

We've been looking at a little of that too, probably not as much as you. There's the problem I deal with is about the initial swelling of metallic fuel at low burnup followed by interconnection. Yes, the problem of fuel swelling is solved by providing a large gap between fuel and cladding.

Yes, that's all true. Liquid sodium in the fuel infiltrates into the fuel and does, I agree, lead to some recovery of fuel thermal connectivity which is degraded by the swelling. He says, please let me know about numerical validation for variation in thermal connectivity before and after sodium infiltration.

Okay, so there's not... We have some... It's more than anecdotal evidence but probably stops short of being very quantitative data supporting this idea that once metal fuel swells and the porosity interconnects, that interconnected porosity certainly allows for the escape of fission gases out of the fuel, but it also creates this pathway for sodium, the bond sodium, to in effect wick into some of that porosity.

A lot of what we think we know about that has come out of TREAT or Transient Reactor tests that were done back in the '80s, and the references for papers published on that are not immediately in front of me, but I think those papers were mostly published in *Nuclear Technology*, and the principal author there is going to be Ted Bauer, B-A-U-E-R. If you look in *Nuclear Technology* for Ted Bauer.

I think he found that maybe about 30% of your open porosity could be infiltrated by the bond sodium and that it did allow for some recovery of the effect of thermal connectivity. He found that in the first percent or two, when you're just generating the fission gas porosity, the thermal connectivity can basically be cut in half prior to the sodium infiltration occurring, and then after the sodium infiltration occurs, you have a recovery of fuel thermal connectivity up to, maybe up to about 70% of its original value. So it doesn't ever go back to its as-fabricated level, but it improves over the minimum condition.

Okay, a question here from Alexandre, "Do you integrate in your approach ways of recovery for valuable elements in spent metallic fuels?"

Like, perhaps you're thinking of the rare earth. I've heard people bring that up before, but, no, I'm not aware of anyone who has tried to pursue that.

More questions?

Oates: I don't see more questions. Thoughts of appreciation and accolades on your presentation. There's one come in.

Hayes: Okay, so from Derek Bass, "What lengths are you currently casting in bottom casting?"

So we haven't quite gotten up to the lengths that we've historically done in counter-gravity injection casting. I apologize. The figures are most readily in my mind in English units, which won't be helpful to a lot of people, but I'll give you the normalization factor.

So in EBR-II, the metallic fuel slugs were cast at, they were finished at 13.5 inches, so they were typically cast 14, 15, 16 inches long. In our bottom pour castings, we're not quite that high. We're probably about 10 inches currently. That's not to say we couldn't get longer with some work, but that's where we are now.

Ken Karcher asks, "What do you consider to be the lower cut-off temperature for eutectic formation?"

So that's a hard question to answer with great precision. It's a function of your fuel composition. It's definitely a function of burnup. A minimum, it's a little bit different whether there's plutonium present or not as well, even though plutonium tends not to be incorporated into that reaction product, but there does tend to be a difference.

Okay, so all that to say, the minimum temperature is probably somewhere around 700°C, maybe 725. It's certainly well above 650. Under certain conditions you might see it as low as 675, but more traditionally, 700, 725 is kind of the minimum temperature for that eutectic.

A question: "Can you tell me a little bit about pyroprocessing?"

I could tell you very little about pyroprocessing. I don't know, maybe Patricia wants to say something about it. I'm not a recycle guy, but it's a molten salt process. It's an electrochemical process where the spent fuel is chopped up and put in a basket that basically serves as an anode, and then transported uranium and in some cases the plutonium minor actinides can be transported through the molten salt to the cathode and recovered there.

It's not like PUREX, which can be made to have very high decontamination factors. Electrochemical recycle tends to bring with it different things, depending on how you operate the electrorefiner, but certainly the rare earths can be brought along to some level.

Did you have anything more to add on that, Patricia?

Paviet: No, I think you did a good summary.

Hayes: Okay. A question: "What about the cladding material for the Advanced Metallic Fuel Concept?"

So the US has been... There's no specific cladding although cladding development is an activity that's being pursued as part of the Advanced Fuels Program by DOE. We have typically migrated to ferritic/martensitic steels, which are fairly resistant to swelling, at least relative to austenitic steels. They are low swelling, and so, because of the reduced swelling they are capable of higher burnup, but they tend to have less strength than the old austenitics. So most of the cladding development is staying within the generic ferritic/martensitic class of alloys but looking at ways to improve the strength, typically by the addition of oxide particles.

So I would say HT-9-like ferritic/martensitic steels with ODS enhancement. That's sort of what's being worked on in US programming.

Let's see. Sal has a question. "Can you discuss a bit more about the motivations of eliminating sodium bonding and what the benefits and drivers are?"

So I'm torn a little bit on this issue. As a fuel performance guy, the sodium bond brings in some very positive benefits. It just gives extremely high thermal margin to the metallic fuel element that you can use in a lot of different ways. There's also some indication in transient performance that it provides a little bit of lubrication between cladding and fuel that allows for rapid axial expansion of the metal fuel in over-power situations, which can be a really nice negative feedback mechanism.

But the negative parts of it, it's an added step in fabrication that costs you a little bit. But the bigger issue is generally perceived to be on the back-end disposal of sodium bonded metallic fuel. Here in the US – I'm sure everyone, every nation's legal requirements are probably a little bit different – but in the US, the legal disposal laws basically don't allow for direct disposal of sodium bonded metallic fuels. Yucca Mountain as a geological repository when it was under development would not have accepted sodium bonded metallic fuel for direct disposal.

And so that means you're faced with having to treat spent sodium bonded metallic fuel prior to sending it for geological disposal, even if you're not doing something about actinide transmutation. So that's a significant added waste disposal cost that would be associated with sodium bonded metal fuel. If you had a metal fuel concept that didn't have sodium, then the potential for direct disposal presumably could be back on the table if you wanted to do that.

So there's a recommendation for the book *Plentiful Energy: The Story of the Integral Fast Reactor* as an intro to pyroprocessing for the one who had the question on pyroprocessing.

There's a comment really about accountability, so IAEA requirements relative to accountability issues with electrochemical recycle. That's certainly not an area I can talk about.

Michelle Bales has a question: "I've heard that metallic fuel's swelling behavior varies by manufacturing method. Is that because manufacturing methods produce different smeared density?"

So yes and no on what you've heard. Generally, metallic fuel behavior can be affected by fabrication method if that method introduces severe microstructural orientations, if a pronounced texture. So for instance, extrusion. Extrusion or rolling of metallic fuels, which can create a highly textured metallic fuel microstructure, that can change some of the swelling characteristics. Michelle, you're right. That was observed long ago, decades ago.

However, most approaches to metallic fuel fabrication today, if you're using a fabrication process, say, like extrusion that would produce a textured microstructure, it's typically always the case that you would then have some post extrusion annealing process to homogenize the alloy. So as long as you homogenize the alloy prior to irradiation, metallic fuel performance seems to be fairly consistent.

I just lost my connection, Berta. So if there's another question...

Oates: "From your experience kindly tell is mechanically bonded fuel zirconium liner between fuel and clad better or sodium bonded is better for metal fuels?"

Hayes: Well, so again, I would say it's a trade-off. Okay, I'm back now and I can see the question. I'd say it's a tradeoff. Sodium bonding brings some performance benefits. It brings some cost and disposal challenges. Oh, sorry, I was jumping to the next question.

In your experience kindly tell me...

So I don't know which is better. Like I said, there are some definite benefits to sodium bonding for performance, basically, the very large thermal margin it gives you. Zirconium liners are a great idea if you can get them on. We definitely know if you can get a zirconium liner onto the inside diameter of the cladding, it will be an extremely effective diffusion barrier that will essentially eliminate your fuel-cladding chemical interaction issues.

Our problems have been in purely manufacturing, coming up with a manufacturing process to apply that liner on a very small diameter, a very long cladding tube in a cost effective way and in a way that doesn't destroy the heat treatment on your base cladding material that you like. So we're still working on that as are others. If the manufacturing issues can be resolved, I think that's a great way to go.

Derek Bass has a question. "Do you know if there were any cost-benefit discussions of challenging the law restricting direct disposal of sodium bearing fuel?"

I might defer to Patricia if she has anything to say. I can't speak to the issues, to that issue myself.

Oates: I think Patricia has had to step away.

Hayes: Okay. There was a lot of discussion in the early days, in the heated days of Yucca Mountain work leading up to the license application related to metal fuel disposal, and the decision was reached that it needed to be treated prior to disposal in Yucca Mountain. I don't know how aggressively that was challenged or if it was. It's just not my area.

A question from Ken Karcher: "Have you tried the same zirconium-line fabrication method as is done for LWR fuels?"

No. For a couple of reasons. The liner thickness that we would need tends to be a bit thicker, and our tubes are just much, much smaller, which complicates the approach. That's really the complication; the small diameter and very long tubes make application of a liner difficult.

Oates: Thanks, everyone, for making this presentation such a benefit to others by asking such good questions. And thank you, Dr. Hayes, for having spent so much time developing your presentation and then your graciousness for answering all of these questions. It is really most appreciated.

Hayes: It's been a pleasure. Thank you.

Oates: I don't see more questions coming in. This webinar has been quite lengthy and I think that's a tribute to how good the information was presented and of what interest it is to folks.

Thank you again, Dr. Hayes. I think that the questions have dropped off, and so I think I will give you the rest of your day back. And thank you again.

Hayes: Okay. I appreciate the invitation.

Oates: Bye-bye.

Hayes: Bye-bye.