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NUCLEAR WASTE TECHNICAL REVIEW BOARD

ENGINEERED BARRIER SYSTEM PANEL MEETING

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I N D E X

SPEAKERS: PAGE NO.

P R O C E E D I N G S

8:05 a.m.

DR. ELLIS VERINK: Good morning. We're ready to reconvene the activities today and we'll be starting with spent fuel characterization overview. Ted's going to start off. Okay.

MR. TED PETRIE: I'm Ted Petrie, again. I just wanted to point out where we are. We've gone through the waste package strategy and implementation plan, the waste acceptance process, high level waste glass, and we're about to talk about spent fuel and then go into the waste data base development.

Again, as in yesterday, there will be quite a few speakers coming through here to talk with us today. Ray Stout is the first one and then he will introduce the next one and the next speaker will introduce the next one as we go along trying to keep it a little bit organized here.

So, again without further ado, here's Ray.

MR. RAY STOUT: Good morning, my name is Ray Stout. This talk is on spent fuel. It's an overview. I have broken it down into three parts. There's about five or six viewgraphs on an introduction. I'll talk about the distributional aspects of properties of spent fuel. Primary variables which we're looking at to select spent fuel and to correlate initially is burnup and fission gas release on the fuel pellet. I'll spend some time talking about conceptual models that are under development. These models are in a preliminary state. We are using them to plan tests and eventually we'll use them to describe the response of the spent fuel for performance assessment and waste package assessment.

Why perform spent fuel characterization? This is the objectives of the activities which we have in our technical area. We want to provide data, testing, and models that describe the degradation and radioactive release responses of the spent fuel. This information is used for waste package and system performance assessments for the Yucca Mountain Project.

What does this mean in a practical sense? You'll see this viewgraph several times this morning. What I wanted to indicate is that in the event that a container does fail, there is then a potential event sequence in time which you can expect to see for the spent fuel in a container. First off, there will be cladding exposed to the environment. From that cladding, there will be gaseous release response which is the carbon-14 release. If we're going to provide data, we will need to know the initial inventory of that cladding. We'd like to know where it comes from. We think it comes from the oxide film, the early release. Testing will provide that kind of information.

Model development will tell you how to extrapolate in time once you have some testing information for other pieces of spent fuel and which you haven't tested. The next event that will follow will be degradation; in fact, this is going on at all times. The primary driving force is the gas pressure inside or one other driving force is inside the cladding. Gas pressure is due to initial gas that's put into fuel pins when they're manufactured and it's also augmented by fission gas released which occurs during reactor operation. We'll spend more time on this topic later.

If the cladding fails, then there will be spent fuel exposed. The amount of spent fuel exposed depends on the amount of cladding that's failed. So, we would like to understand this better. This would give you a feel for how much inventory will be subjected to oxidation. The oxidation response of spent fuel occurs in the higher temperature, higher oxidation rate. The oxidation response is important because it influences the dissolution response of spent fuel. It turns out that the oxidation of UO_2 goes up to different states, U_4O_9, U_3O_7, U_3O_8. We need to study each of these states and see the dissolution response. Once water contacts spent fuel, there will be radionuclides in the water. This will bring in the geochemistry part of the problem and we will have a talk on geochemistry modeling.

We have a milestone. We hope to bring all this information in its preliminary state together in a characteristics report. That report is due in March of '91. Briefly, the contents of that report will have the physical property data of both existing and projected inventories. We'll talk about the radionuclide data for these inventories and what models and test data that are available will go into that report at that time.

I thought I'd like to give you a feel of what spent fuel is like. This is a viewgraph courtesy of PNL. This is the size of your hand, a fuel pellet in your hand, or about a half a centimeter by a centimeter or a couple centimeters long. The 200 or 300 of these are stacked in a fuel pin. A fuel pin is about 160 or 170 inches long. Fuel pins are put together in assembly. This is a typical BWR assembly, that's a boiling water reactor assembly. They come out usually in a 7 by 7 array in the assembly for BWR. The typical PWR is very similar. These are usually now 17 by 17 arrays of rods so the assemblies go into a core. This is a typical core. The point here is that across a core, the radial direction, you will have a distributional, or a non-uniformity, and the axial direction will be non-uniformity. So, even if you had all the same cores and all the same operating conditions for all the reactors, all your spent fuel would still not be absolutely uniform.

DR. PARRY: Excuse me, the fuel is moved throughout the operation of the reactor and there is every attempt to homogenize the fuel, but you're quite right, there is a flux distribution across the core.

MR. STOUT: Yes. Flux heat gradient, yes. And, that fuel management cycle is for economic reasons, as well. Thank you.

This brings us into talking about the properties of spent fuel, the distributional aspects which is the second topic. This is important to us because when you start looking at what's out there, you find it perhaps not described in a way that you typically do experiments or modeling. Typically, we look at what's going to be the response of a rod, and when you look at the data, it's assemblies, tons, things like this. So, one of the things that we're looking at is how do we get back to rod information. I'll show you how we're going to do that.

As I mentioned, there is a large distributional aspect. Part of it comes about because there are many classes of reactors, many classes of assemblies. This is a typical -- this has more information I'm going to cover, but I just want to show that BWR is clumped together. This is the GE rods. This is Westinghouse reactor fuel here. This is metric ton of heavy metal. What we know is for each assembly the number of rods in assembly. If we take that, we go to a viewgraph which you've seen before which is burnup per 1000 metric tons. We can get a relationship between the coordinates of this graph and put these two distributions together; in other words, add the history and the projection and come out with information which gives us information like this which is the number of rods per unit burnup and plotted against burnup. This is like a distribution function, it's not a probability density because we're probably not going to normalize it to one, but from this then, for any increment of burnup, we can tell you the number of rods that have that burnup.

Now, why is this important? We're going to be getting into more detail when we do analysis. One of the things that I mentioned already is oxide film. Its thickness is an important attribute. Here, it shows, a courtesy of MCC -- that's the Material Characterization Center at PNL -- burnup versus oxide film thickness. We can get an upper bound line. If you have an upper bound line, you have a linear or perhaps a transformation function which you can go from previous viewgraphs. You can now talk about rods per unit oxide film thickness versus oxide film thickness. This is a way of getting to rod data with its associated attribute.

We can do that for a variety of, what I call, attributes. These are just the characteristics of the spent fuel. For example, rods per unit carbon-14, this is something we'll need to know. Rods per unit hydrogen, this gets into the hydriding problem in cladding. Rods per unit heat decay, we will need to know that for thermal analysis. Rods per unit activity species, this is a release kind of problem where you need to know the inventory in the spent fuel to study release.

The other variable besides burnup, a primary variable -- there are secondary variables -- is percent fission gas release. This is an important variable and we are looking for data which we can get the rods per unit fission gas release versus percent fission gas release. The reason it's important, the fission gas release augments the initial gas pressure in the rod and this has been, as I've mentioned before, a force driving degradation or failure of fuel pins.

Fission gas release also tells us something about redistribution of inventory in the spent fuel. This was mentioned, I believe, briefly by Mike yesterday. It turns out that fuel pins with high fission gas release redistribute what's called the volatiles. These are cesium, iodine, technetium, strontium. These also are highly soluble species. If they get out in the gap, rapidly when water is contacted, it contacts the spent fuel. So, where they are is very important for that initial rapid pulse of release.

We see here that fission gas release can be correlated with the percent inventory that's out there. We can use that information, again make a linear transformation, get a plot which says the number of rods per unit inventoried in the gap and grain boundaries. What we see is that like fission gas release, we don't expect a large number of rods to have high gap inventory or high pressures; however, those that do have those high pressure will probably be the first to fail. These will also be the ones with the highest inventory and so we can keep track of these in terms of a correlation between those two attributes. That's all I wanted to say about the characteristics.

This part of the talk will be the conceptual models that, as I mentioned, are under development. These are not in any final state. We are using them to plan tests. It's an energy process where you look at test data and you look at what you're doing in your model, you refine both. And, as I said, we will eventually use these then to describe response of spent fuel.

Now, in going through these, the subsequent talks will amplify on these topics. The gas release response, that's work being done at PNL and Harry Smith will talk about that. Cladding degradation response, again Harry Smith will talk about that. UO_2 oxidation response, that's Robert Einziger from PNL at Pacific Northwest. Water contact with spent fuel and the dissolution response, really we only talk about dissolution response, this is Charles Wilson of PNL and Herman Leider. They will talk about modeling and work that's being planned at Livermore. And, finally, Carol Bruton will talk about a geochemistry solution response and that's the model EQ 3/6 and solubility of a species. So, this will be quite rapid.

Gaseous release, this was found to come out quite quickly from cladding. What we're doing is performing tests on it. What we're looking at is a diffusion model. We think that the rapid release resides in the oxide film primarily and Harry will talk about this more in detail.

Zircaloy cladding degradation, we're looking at three modes of degradation. What we now feel is the one analysis path is to consider the oxide film failure. Oxide film forms on zircaloy in reactor. The film forms with a significant volume increase. That means the film itself will be in a compressive state of stress. As long as it remains in a compressive state of stress, you can't drive a crack across it and this will be a failure/no failure kind of criteria. We're looking at modeling a deformation then due to the gas pressure during the rods' history and a repository considering elastic, plastic kind of creep response, thermal, and any strains due to hydride precipitation. So, that's a preliminary model.

I mentioned hydrides. Hydrides, hydrogen goes into the cladding during reactor operation. It is in solution of high temperatures. When temperature comes down, the hydrides precipitate out as platelets. The platelets shown here, I call circumferential. In this alignment, they would not degradate the cladding and you would expect no failure. However, if the stress is high, they do have a tendency to reorientate. That's a hoop stress in the cladding. If they reorientate, then you can have a crack path across the cladding. These would be the radial hydrides, and in that case, you would predict a high probability of failure.

The third topic or mechanism we're looking at is fluoride attack on the zircaloy. Fluoride, ions are known to corrode zircaloy. This appears as a pitting corrosion mode. We're worried about pin hole pathways through the cladding. This was an electro-chemical corrosion response. The degradation of cladding will be discussed by Harry Smith, as

I mentioned earlier.

If cladding fails, then you can expect oxygen. In the event that the containers also fail to contact the spent fuel fragments, the fragments occur because when you go up in the power cycle, thermal strains crack up the fuel pellets. This exposes more area. So, oxidation response is higher in this case than it would be if you had whole pellets. What we're seeing is that there is a propogation of oxidation front into a fragment. You have U_4O_9 crystalline structure behind the oxidation front. So, this is a phase change. You have UO_2 in the center. We're looking at modeling which we can talk about representing these fragments and following the propagation of this boundary into the grain or into the fragment. That way, we can develop a model which certainly is related back to test data to describe oxidation.

The results indicate that we have to worry then about fragment distribution sizes. We have to worry about different temperatures. What's happening is that the grain boundaries oxidize first. They open up and then expose individual grains. And, this problem will be discussed further by Bob Einziger when he talks.

This brings us to spent fuel radionuclide release which right now we're considering aqueous release. In other words, fuel is exposed to water. We have to worry about what's in the gap. This is rapid release. We have to worry about grain boundary response because the grain boundaries load up if you have high fission gas release. They also load up with the soluble species just normally because there's migrations out of the grain. And, finally, there's the grains itself which contain most of the actinides. The dependence is quite complicated. We do not have at this moment what I would call a detailed working model. We're still looking at experimental data/approaches trying to isolate various effects. And, what we're looking at is major solubilities of the soluble species, ways to get release rates, and also study the release of the solubility limited species. These turn out to be the actinides. This will be talked about by Chuck Wilson who has done hot cell work and Herman Leider from Livermore.

Okay. Well, I guess I had one more viewgraph on this. The two kinds of experiments which we are going to study radionuclide release are flowrate control experiments and semi-static experiments. The semi-static experiments are those of Chuck Wilson in the hot cell and he's also done some flowrate on UO_2. This data, so far, provides us input and ideas to initiate modeling and, of course, to couple to the EQ 3/6 geochemical simulation code that is at Livermore. The simulation work will be discussed by Carol Bruton.

This is sort of a summary viewgraph, the philosophy which we have in terms of testing. Because of the long times, we are looking at what is called mechanistic model development. You have to do this based on short term tests so we look closely at what goes on at a microscopic domain a lot of times. We always try to perform testing over a range of experimental variables which exceeds what we call repository conditions. This is not always possible, particularly because at low temperatures, nothing can be measured, the response is so slow. But, if we can do this, this means that we will be able to interpolate rather than extrapolate on function variables and data that we need for model development.

As I said, you'll see this viewgraph many times. The first two topics will be talked about by Harry Smith of PNL and the topic will be called the spent fuel cladding degradation.

Any questions?

DR. DEERE: Yes. In your Viewgraph 21 when you note that the oxide film will generate large compressive stresses as it's being formed.

MR. STOUT: Right.

DR. DEERE: Can they get to the point where they will actually lead to either a shear failure or a splitting tendency parallel to the maximum principal stress?

MR. STOUT: That's never been observed as far as I know in reactor operation. Oxide film does have a porosity and there have been attempts to analyze that porosity as perhaps a buckling or some kind of a failure to a wavelength of buckling around the clad. But, that's never worked out well, either. Work at Bettis indicates, you know, it may have 400,000/psi compressive stress in the oxide film. Some work we sponsored doing elastic analysis sets in this same kind of ballpark, 200,000/psi, 400,000.

DR. DEERE: How does that compare with the unconfined compressive strengths?

MR. STOUT: I have no feel for that. You see, this is like a pre-surface on the film. It's tied down to this boundary. When you say compressive strength, you're talking about confined? I don't know --

DR. DEERE: Unconfined. The reality being a biaxial stress state. It probably will fail before it gets up to its maximum shear strength.

MR. STOUT: Yeah.

DR. DEERE: And, you'll get a premature splitting failure.

MR. STOUT: Yes. That hasn't been seen. What seems to have happened is that if you distribute the volume change in all three directions of strain, you would have even higher stress. There is some work out there which says that when this form is on the surface that it is anisotropic and most of the growth grows normal to the surface. And, so that keeps the enormous stresses from building up. Otherwise, you would predict millions of psi compressive stress in the hoop direction. So, it's not a straight forward problem. We hope to do tests to see what the failure strain is in the cladding oxide.

DR. PARRY: Ray, on your Slide 9, you list some quantities by various reactor type and fuel type.

MR. STOUT: Yes.

DR. PARRY: I'm assuming that those quantities are based on a reactor running for 20 years and then shutting down and not having an extension of its operating life by licensing its extension. Is that correct?

MR. STOUT: I really don't know. This work, as I didn't mention, I should give credit to the Oakridge people. Karl Notz, who I think is in the audience, could answer that.

MR. KARL NOTZ: Well, the answer is yes.

MR. STOUT: The answer is yes.

DR. PARRY: Thank you.

MR. STOUT: Thank you, Karl.

Okay. Harry Smith will talk next.

MR. HARRY SMITH: I'm Harry Smith. I will discuss with you briefly the work we've done in spent fuel cladding degradation. I'd like to cover these topics. First, the carbon-14 release from zircaloy cladding. Then, we will look at what we've done with C-ring failure testing from scoping studies and then some pressurized tube testing which is planned. It's just really begun, barely. And then, some zircaloy/fluoride corrosion testing which we've done only scoping studies on, but enough to get some indication.

Why zircaloy cladding degradation studies? Well, first, we need to get carbon-14 release data so that its release can be properly assessed in post and pre-emplacement conditions. And then for the cladding degradation, it is an initial barrier to release of any gap inventory radionuclides and so forth. And, if you can show what its barrier characteristics or its resistance to degradation is, you can consider it as a barrier and then you will not maybe need another duplicate barrier to perform its duty. But, you have to be able to show what it does do.

Just to give you a quick picture again and remind you of what cladding is, I'll show a few spent fuel rods in cross section as they might be in a consolidated container in repository setting. This is just one of the possible geometries. But, the cladding is a metal sheath around the spent fuel. It has an oxide film over the top. Crud can deposit on top of that, and then in the various repository type scenarios, you may have water that does contact the surface of the film even before a container breaches because of water-logged rods loaded into the container, or after container breach, water that comes in from a repository setting.

The zircaloy cladding, the oxide film, and even the crud contained in carbon-14 that I will discuss this morning; water, like J-13 does contain some fluoride, corrosion models could be some kind of a uniform corrosion -- unlikely because of the oxide film -- but pitting, crevice corrosion perhaps could occur and then the cladding itself can degrade also by cracking like in stress corrosion or delayed hydrogen cracking mechanism.

Just for your information, the experimental material we are using for experimental work, essentially work we're reporting on this morning comes from PWR reactors, H.B. Robinson and Turkey Point. They're a medium or a slightly low, I guess, burnup material. And, we've looked at thick oxides which we interpret to be 12 to 20 microns thick film of oxide or thin, the dark, 3 to 6 microns thick oxide film.

Discussing first the thermal release of carbon-14 from the cladding, we've developed and demonstrated, I believe, a technique that will quantitatively measure the release and we will apply or would like to apply this technique to spent fuel, entire population or good samples from the entire population, and reactor hardware.

Our apparatus shown schematically consists of a furnace with a homogeneous temperature zone, a gas flow path that brings its own atmosphere across the cladding surface as it resides in the temperature zone, brings it out to a series of traps which allows us to trap tritium that's released and carbon-14, to separate those two. And then, by taking it through a copper oxide furnace, determine what proportions of the release may be in some less oxidized state that weren't caught in the caustic trap or initial tritiated water trap. This is then brought back into the hot cell.

Modeling for release, two really simple and really almost the same models are the semi-infinite plane or semi-infinite fitness plane, simply a plane for which the diffusional release from that plane does not see the back surface. So, it looks semi-infinite. Or if it does see, if the source is thin enough so that the diffusional release profile sees the back wall of the reservoir, then you get a finite thickness plane model. And, what happens is at some particular point, depending on the geometry and release rates and so forth, you would see a divergence between these two, something such as I've shown here.

First, I'd like to point out from an initial carbon-14 concentration profile we made through the upper surface of a piece cladding shown here, I would like to point out that there is a slight scaling error. So, you should make that correction in your viewgraph. So, if you have a question about it, that's the reason it looks strange. The oxide film depth on this piece of cladding was about 10 microns. It looks like there's a higher concentration of carbon-14 present in that oxide film and then drops off to some lower uniform value in the middle. Though we did not penetrate those usually, those were -- it's a very arduous, tedious experimental measurement to make because to get the good uniform stripping, we had to etch at a slow, very low temperature, very slow in the system, and each of the points on there represented several hours of etching time with also the trafficking of the released carbon-14. So, it was a rather delicate measurement to make.

Okay, back to the model. Here, we did show some data plotted in the same way of cumulative release against the square root of time. We see for a high temperature, the scale on the left, versus low temperature, the scale on the right, we can see that for the highest temperature, we seem to see a falling off such as you might expect from a finite plane diffusion model release. The others seem to be pretty straight like an infinite (sic), semi-infinite plane, simply indicating they have not seen the back of the reservoir yet.

A summary of our data to date is shown in this plot, plotted a little differently than the first one, but shows two things. One, there's quite a temperature dependence on the carbon-14 release from the cladding, all the way, almost four orders of magnitude there over a temperature range of 100 to 350 C. And then, there is a significant atmosphere effect. All the rest of these were done in air. Three Argon tests shown here at 350 are considerably below similar tests done in air.

Okay. A summary then of our work and some conclusions, we have developed a system that will allow us to look at the temperature, atmosphere, and other environmental factors as release as a function of those things. And then, our initial data indicates that -- now, I'm saying that 10% of the carbon-14 cladding inventory. This is in no way different than what Ray said earlier. It's just related to the cladding -- specifically and this is at eight hours, 350 in air. We notice that argon gives about an order of magnitude or more lower release than in air for the same time period and that the carbon-14 appears to follow some kind of a finite plane source model suggesting that the carbon-14 might be there in more than one species or there's more than one reservoir; maybe the oxide film, as Ray suggested earlier, and the cladding itself, the metal, the carbon in maybe solution in the zircaloy and other reservoirs.

Moving on to the cladding degradation work, looking at tracking of zircaloy-4, we've done some C-ring experiments that were done to determine whether or not we'd see cracking at all. Just really to tell us if this was something to expect and then to see if we could determine some kind of relationship between time to failure and the stress that we placed the zircaloy under.

And then, the pressurized tube testing, the design is a followup to the C-ring experiments to look at what kind of strain is necessary to actually rupture the oxide film to potentially initiate cracking, and then to try to also take those measurements to lower stress levels by some other experimental tricks, in a sense, of maybe mechanically defecting the cladding and so forth and certain ways to get cracks initiated and then watch them actually move rather than just an all or none as it was for the C-ring experiment. And then, to look at the question of hydride reorientation. This is a possibility for some repository type temperature profiles, and if it does occur, it can degrade the mechanical properties of the zircaloy.

Okay. Looking at the C-ring cracking experimental apparatus we used, we took an actual piece of cladding, made a C-ring out of it simply by cutting off an arced segment, placing it under compression between two anvils. The relationships here, if this is one, this is five, and this side here to the LVDT engagement screw is a factor of two. So, you can scale all these things and measure a deflection of the C-ring as a function of time under an environment that you establish in this little pan. And, you can either put water in here or, as we did in some cases, also put air, run it with air by cranking up the oil bath a little bit. The thermocouple is in place to tell you that indeed you've achieved the right temperature. We ran these experiments at 90 degrees.

Just to give you an idea of what you see when you get in this high strained area of the cladding C-ring, when those drain, you see some porosity here in this rather thick oxide film, but you don't see radial cracks. In the high strain area, you can clearly see a set of radial cracks develop that penetrates the oxide film.

Some results look like this. They all look like this with various lengths of time to failure. Basically, you had initial relaxation time, a probably creep in the zircaloy because these are all done at quite high stresses compared to the actual yield stress of the material tied to some large percentages of the yield stress. You saw then a slow deformation. In some experiments, it was almost imperceptible. A period of increasing deflection and then you would go into a period of consistently increasing deflection until you actually approach a failure such as you see here. When we observed failure in one when it was unexpected, we usually had it break on us, shocking the whole system, and we deflected then the neighboring specimens. And, this is the specimen shown here that was a neighbor to this. We saw this deflection, interesting enough. The recovery appeared to go back out to what you might have expected to have been the original failure profile suggesting that it was an environmental effect maybe at the cracked pit, something going on there such as the gathering of hydrogen to lower maybe the crank, or it may be a fresh corrosion phenomena going on. Something going on at the crack that needed to be re-established when you saw failure at the original rate.

If we take the data for all of the C-rings run in water and plot the percentage of yield stress -- and this could have just been stress, but I chose to use percent of yield stress because we also determined that that is an initial step in doing our experiment -- you see this kind of a relationship between log of time to failure and that stress, a logarithmic relationship.

DR. PARRY: Is that the stress of the irradiated zircaloy?

MR. SMITH: That's correct. This is the calculated stress for the outer fiber of the C-ring.

DR. VERINK: As installed?

MR. SMITH: As installed, yes. Now, we did preliminary experiments to determine how accurate we had to be on where you installed it and so forth. And, we can be quite accurate, it looks like.

DR. VERINK: But, with time, you're changing the thickness of the film and so on, aren't you?

MR. SMITH: You don't see any hint of that, though you don't have before and after obviously. But, I had done other experiments in which I had just immersed zircaloy cladding material in J-13 for up to periods of like a year and could detect absolutely no change. Now, there's a lot of discussion on how accurate I could do that and it certainly wasn't that fine. But, I would say that the change in thickness of the film during these experiments was negligible because of the fact -- another thing, because I had both thick and thin film specimens represented here and I haven't distinguished them for you. There may be a difference between them, there may not.

DR. LANGMUIR: Harry, to what extent does it matter which kind of water you've got there, J-13 or something else, since the unsaturated zone water is about three times the saline and you also might have condensation water which would be distilled?

MR. SMITH: I would say good question and that's something we would address, you know, if we use one kind of water, J-13, for the test or just plain hot cell air for the air test that we run.

DR. DEERE: What is the approximate yield stress?

MR. SMITH: Well, the yield stress that I measured based on looking for a proportional limit -- I mean, waiting until I got a deflection from the proportional limit -- were numbers like 150,000 psi which is high for irradiated cladding. But, you have to remember that in this outer film area, we've got an oxide film there. We have got an oxygen saturated zircaloy, an irradiated zircaloy right beneath it. Now, a typical number, you might have expected 120 maybe or even 130 maybe. So, these seem to be high, but I've also done the experiment using an HT-9 stainless steel for which we had all the properties and I was able to predict those from my measurements to get similar values that were measured by other methods, such as the yield stress, doing the same kind of experiment.

DR. DEERE: Is it fairly brittle material or does it have a yield before you get to ultimate?

MR. SMITH: It yields. Now, there's some discussion on this. Other people told me it was going to be very yielding. I expected it to be like glass with what some people have told me. Though we did on a couple of experiments drop the weight, as you saw, crushed it and it didn't break. If you take a look at the surface oxide film at the failure time, you will see areas where you get very sharp crack that has penetrated down and is propagating down through the cladding. And, the surface of that crack looks like a transgranular fracture. If then you also look at areas that failed in this catastrophic failure where it broke, you see surfaces that are dimpled, but look kind of like a ductal yield. And, you find places on the surface where the oxide film has been cracked and separated and you get just a sharp, you know, rather V-notch crack. So, it will look like under a rapid deformation, it was acting at least partially ductal, but these things did break in a rather catastrophic manner.

DR. VERINK: What is the ratio between the specific volume of the oxide that's there and the metal from which it was formed?

MR. SMITH: You're asking me how much volume increase there was in forming the oxide film?

DR. VERINK: Yes. So, it's only a very small one?

MR. SMITH: Yes, it's a small amount. For me, we have not made measurements. We know what the oxide film thickness was and predict from other measurements --

REPORTER: I can't hear you.

MR. SMITH: Ten to fifteen percent.

DR. VERINK: So, it's not a question of over-straining the adhesion of the oxide versus the cohesion of the oxide?

MR. SMITH: I think that's true. The fracture that propagated into the cladding showed absolutely no deflection at magnifications as high as 20,000x on the SEM. I've never seen such a tight adhering film. The porosity does sometimes project pretty close down to the boundary and they -- porous film and that's where I've concluded that a lot of the excess stress was being taken up.

DR. VERINK: Um-hum. Well, it sounds like there's greater adhesion to the substrength than there is cohesion within the oxide?

MR. SMITH: Oh, yes. Another observation to support that is that these fractures, we took a number out before we failed so we could see the fracture patterns that formed on the surface. We had inactual on cracks. The shear zones between those cracks that formed, you would see the top surface of the oxide film fall off, but there was still an adhering, a tightly adherant layer, maybe highly fractured below, but it was still adhering.

DR. VERINK: That would only be on very thick films or would it be?

MR. SMITH: Well, it was on all the films.

In summary, we've kind of discussed some of this. We would just propose that perhaps delayed hydrogen cracking is maybe the principal mechanism involved here. Based on the kinds of fracture patterns we have observed and in comparison with other experimental work, we've observed that the time to failure, log time to failure, was proportionate to yield stress. And, we observed that time to failure under air was about twice as long as it was in the water when we used J-13. And, just for your information, if you extrapolated that stress to maybe some conservative upper limits for the repository situation, you would expect failure to occur in hundreds to thousands of years. We're not claiming that that's a good extrapolation or at all accurate, but that's what you have.

Okay, planned pressurized tube testing. First, we were going to determine the strain to produce cracks through the oxide film. We'd like to then extend firm results from C-ring testing. Then, we'd like to look at the question of hydride reorientation under possible repository conditions and then determine the impact of any reorientation we might see on the mechanical properties of that cladding.

To get an idea of the level at which the oxide film cracked, our stress level, we've started performing experiments such as the one you see here shown schematically. We put pressure fittings on a piece of cladding, attached a strain gauge, and actually even considered acoustic emission. This is tassivated except for the oxide area of the cladding section and we've simply measured the resistance through conductive water which could be J-13 to another electrode using a short duration pulse resistance meter.

There's also such measurements shown in this next figure. What you see is that at a strain of about 23%, resistance begins to drop very rapidly to a rather low value, a relatively low value, out at about .56% stress. During that same test, the acoustic emissions began at less than .1% strain. Of course, that can be due to any kind of fracture in the cracking, anything going on in the oxide film or the oxide film and substrate. And, we see a relatively uniform increase of the number of events with increasing strength. Though we have not analyzed that in great detail, it appears that the resistance method is probably the best technique to use for determining when we're getting through the oxide film cracking.

DR. VERINK: Will there be complimentary experiments in the presence of irradiation?

MR. SMITH: Well, we're doing this first with unirradiated cladding just to get the method down and then we would go in the hot cell for the irradiated cycle. We have actually got some potential --

DR. VERINK: Okay.

MR. SMITH: Now, just the whole general test setup for pressurized tube testing looks like this. Basically, we have a pressurization system in which we have some data readout to follow the actual pressure that's maintained in the cladding section itself shown schematically here. It will be filled with some kind of inert material if we take out the spent fuel. If we leave it in, we'll let the spent fuel take up the volume. And then, this piece of cladding can be put in some kind of an environmental chamber here, small stainless steel pressure vessel of some kind that we can then put water or various concentrations or various chemical concentrations in. We can put different atmospheres in there. This item then fits in an isothermal block, say like aluminum, that we can then impose various -- on the system and we imagine that maybe the test will be done like this. You see, this is the part of the system that sits in the hot cell, the rest of it is outside the hot cell.

Now, we've talked about this hydride reorientation. Hydride reorientation may occur because when you put the spent fuel in a repository, the temperature raises for a period of time. This would cause hydrogen that was exolved as hydrides to re-dissolve into the zircaloy and then during the slow cool-down can re-precipitate. Now, when this happens, the hydride may reorient due to that tensil stress that is in the cladding itself due to the internal gas pressure, both because of initial fill pressure and fission gas release.

DR. VERINK: There doesn't seem to be any orientational thing that I can tell with cracks through the oxide though, correct?

MR. SMITH: No. In this particular picture, I wasn't trying to illustrate anything like that. What we would expect to do is first to see if we can reorient the hydrides, at all. If we can't reorient them, there's nothing to worry about. If we can reorient them, then we have to see what happens to the mechanical property and then it becomes another type of specimen we put into our standard pressurized tube testing maybe with some modifica-tions that become obvious at the time. That's what we plan to do.

Just some data that suggests that we need to consider this, in a normal reactor shutdown, you come down from a temperature like 350 and you have a fairly rapid cool-down rate shown here and they observed no radial hydrides. And, if you do the same thing with a similar rod, but not in the reactor, but without any internal pressure, you see at a slow cool-down rate, you see no reorientation of the hydrides. Now, this pressure shown here is equivalent to something you might expect at one, it may be slightly extreme, but not unreasonable for some of the spent fuel rods. You take it for 323, cool it at 5 degrees an hour and they do see radial hydride reorientation. And, that would be enough for significant degradation of the mechanical properties. So, this needs to be checked out.

Just to show you what we mean by reorientation, this is normally the way that hydride looked in a piece of spent fuel cladding that you get out of the reactor that, in essence, stayed that way.

DR. VERINK: Now, that's internal pressure in the tube or --

MR. SMITH: Yes.

DR. VERINK: All right.

MR. SMITH: Okay. When it cools down in a reactor, they've pressurized the cladding to keep it from collapsing because of the reactor pressure, itself. So, they kind of balance off. So, there's not a big stress gradient in the cladding itself at that time. But, when you get outside, the external pressure is now removed and you have additional fission gas release. Now, you have the potential for a significant pressure inside that can cause the hydrides to reorient when it cools down at the repository rate, whatever that is. Okay. So, this is what they look like in a reoriented state.

Now, I'd like to switch to the zircaloy/fluoride corrosion testing. It has been recognized for many years that the fluoride is very detrimental to zircaloy and great efforts are made to make sure that you don't have it when you're completing your fuel rods. Fluoride is present in the groundwater, such as J-13 at 2.2ppm. And, we really have no quantitative studies that exist under the conditions of interest. Now, it should be pointed out also that the fact you have fluoride in the groundwater is important, too, because it could be concentrated by refluxing and so forth during the repository lifetime. So, you may be considering something up to even 100ppm or greater in the reactor water or at least that's what I am told. And, so we have done studies down into that range.

Now, what we tried to do in the zircaloy/fluoride interaction study was first to develop and demonstrate the techniques that would allow us to look at corrosion rates at the kind of levels you might see in a repository situation. Then, we applied this technique to a series of zircaloy-4 specimens and then we would like to apply, eventually, this to irradiated spent fuel cladding to determine if our unirradiated cladding results are the same as you would get with irradiated cladding.

Okay. What we found with the zircaloy/fluoride interaction studies was that a pH stat system was a good way of making measurements and we believe we could do it very nicely on irradiated fuel samples. The pH stat either was a pH stat or a fluoride stat technique that you use for high pH areas. Zircaloy-4 corrosion rate is a function of pH and fluoride content and temperature very clearly. And, we did see pitting all over the place. However, the way we were doing the experiment, you could not demonstrate exactly what condition, which condition was the cause primarily because we were using a particular specimen at a number of different pH's and we would then at the end examination observe this pitting that had taken place.

DR. VERINK: What you're doing is controlling the pH. That's what you're doing, right?

MR. SMITH: Well, yes. What happens is that as the corrosion reaction occurs hydrogen ion is consumed in the reaction. And so what you do is by statting it, you're adding a little hydrogen ion and maintaining a constant pH. And, from that information, you deduce a corrosion rate.

DR. VERINK: Um-hum. And, you're assuming that the pH, wherever you're sensing it, is where the action is. Is that right?

MR. SMITH: Yes, you do assume that. However, we use a small reaction vessel and we do vigorous stirring while the test is going on. So that you can judge how accurate that is, but that's what we're trying to do. We do see consistent results, I can tell you that. We do see hystoresis results which does indicate some interesting mechanisms going on and so forth, but we do see consistency.

Some more conclusions, we do see a scale-forming reaction which appears to be the standard reaction for zircaloy reacting with a water producing its trassivating film. The film that's going reactive seems to be probably fluoride complexing with the zirconium oxide film and producing a soluble species. That's the second slide, I think. And, these seem to be consistent with other literature work done on this using other methods for measuring corrosion. Now, we need to do this on an irradiated cladding. We have not done that and that does need to be done to confirm that it will work.

Summarizing my work, first with carbon-14, we've got a functional apparatus to do the work and we have seen that the release rates are a function of both temperature and atmosphere. And, the zircaloy cladding cracking studies, we've got evidence of cracking at high stress levels. If you extrapolate that to repository levels, you'd expect failures in hundreds to thousands of years. Pressurized tube results will extend this hopefully to lower pressures and look at the potential for hydride reorientation and the impact that it will make.

The fluoride/zircaloy corrosion is observed. Corrosion down at 100ppm, measurable for 100ppm fluoride, and it's strongly dependent on pH fluoride, as well as I say, fluoride concentrations.

DR. VERINK: And, that was pitting, is that right?

MR. SMITH: Well, it's pitting, but you know, if you do some calculations, and we also do weight loss studies to kind of give a separate confirmation of our measurements, it's not clear whether the calculated weight loss implied pitting. Pitting may be another mechanism that's active, but not the total mechanism.

DR. VERINK: So, there's also a surface film?

MR. SMITH: Yes, a reaction type of thing going on, right. Pitting was definitely there, though.

DR. LANGMUIR: Will you extend the work you've done in the aqueous phase with respect to the cladding cracking and fluoride induced corrosion to steam and high temperature effects which is what's really going to be going on for a long time?

MR. SMITH: Well, this is the reason for the little environmental chambers that we were going to produce. As I said, we can go into high temperature water, 170 degrees, for instance, and have actually water in there or steam or go to a higher temperature and do the same thing in those chambers. That pressure is not significant enough to really modify the pressure we put in the tubing. It would be a much higher stress level. And, all those things could be done, I believe, with the system. That is the plan.

DR. VERINK: Have you had a chance or is it part of your normal scheme to see whether there's any knowledge to be gained from checking these kinds of results with the appropriate Pourbaix diagram for these?

MR. SMITH: We have looked -- it seems to parallel the species predicted by Pourbaix and levels -- we really haven't looked at it a lot because we've been worrying about actually getting good numbers first. I felt that we needed good numbers before we worried about comparing those others.

DR. VERINK: Um-hum.

DR. DEERE: Yes. Could you go back, please, to Slides

24 and 25? These are the hydride reorientations.

MR. SMITH: Sure.

DR. DEERE: I guess really it's the diagram 25.

MR. SMITH: 25.

DR. DEERE: Now, under normal, do you have this orientation of the hydrides before you begin to cool down? I mean, during the operational phase?

MR. SMITH: Well, this is in the normal phase you would expect to see from cladding that has been removed from a reactor, after it comes out of the reactor. This is what you would see. This is what I see all the time.

DR. DEERE: Yes.

MR. SMITH: Exactly like this.

DR. DEERE: Do you think this also looks like this when it's in the reactor?

MR. SMITH: Well, in the reactor, you're at a considerably higher temperature. I would guess a lot of these have come out perhaps during cool-down. Of course, you realize that manufacturers of cladding are very particular about getting a proper structure in the cladding itself so that the hydride will form in this orientation in reactor service. Now, there's some hydrogen probably as hydrides, but most of it is in solution at 350. As you come down, a lot of the thing barges those hydrides.

DR. DEERE: I would think this orientation is the one that might want to exist during operation because you have the high compressive stress or circumferential stress in the cladding.

MR. SMITH: Um-hum.

DR. DEERE: And, free surface for expansion is out into the container. So, you would have a tendency for anything that's going to form or any reorientation to have exactly that. But, when you come over here in the reoriented specimen, even though you state that this is in a compressive field because the container is pressurized --

MR. SMITH: Well, it's more neutral. Let's put it this way, it's more of a neutral. It's not a high stress state at that time. It's more neutral. The cladding has simply been fabricated so that the hydrides will tend to come out and this is the manufacturers' knowledge that goes into producing cladding in this form. So, this will have it because they realize degradation from mechanical properties if you've got radial hydrides. And, so what happens in the repository or what could happen -- this is what we would want to look at -- is that in that situation now you have no constraining outer pressure. As the temperature goes up, these hydrides then redissolve into the matrix. Then, as you'd reach peak temperature, and start back down again, they start coming out. But, now the pressure field stress they feel they see in the cladding is not a low stress field. It's a much higher stress field and it's tensile.

DR. DEERE: That's what I was going to say. That's what it looks to me like, that you might have external pressure, but if you're cooling down, you're developing tensile strains or extension strains or tension in that and you get the new reorientation.

MR. SMITH: Yes.

DR. VERINK: Are those distributions, say, so-called normal and basically intergranular?

MR. SMITH: Yes.

DR. VERINK: Are they sometimes transgranular? Well, I'm intrigued by the distribution to the other orientation. It seems like it would be easier to do that intergranularly than it would be transgranularly.

MR. SMITH: The other orientation usually is on the grain surfaces when you reorient.

DR. VERINK: Yeah.

MR. SMITH: And, it's a stress driving --

DR. VERINK: Yes, but you'd think that faster diffusion pass would be intergranular rather than through the bulk of the grain.

MR. SMITH: Yes.

DR. VERINK: It's interesting.

DR. DEERE: Another way I'd like to look at this, our cracks did form in the orientation that I thought it might, but they immediately became filled with the oxide in here. The cracks that formed in the other direction and then were -- well, they had to be filled because it moved from one position to the other. I don't know what was the mechanism of the transfer.

MR. SMITH: Now, I'd like to introduce to you Robert Einziger who will talk about spent fuel oxidation.

MR. ROBERT EINZIGER: The only thing anybody remembers from my hydride experiment is the mistake I made. I'm Bob Einziger. I'm with the Pacific Northwest Laboratory. I'm going to spend a few minutes talking about spent fuel oxidation.

I'd like to briefly review the information we know about spent fuel oxidation response, then go into two of the sets of experiments that we've been doing. One is the thermogravimetric analysis and the other is dry bath testing.

Somewhere in the neighborhood of approximately less than .1% of the rods are going to enter the repository in a breached condition and you can define breach any way you want. But, the cladding will have been compromised. And, at the time the canister is compromised, oxygen will be available to the pellets for oxidation to take place. In addition, while in the repository, additional rods may breach due to the corrosion that Harry just talked about. If you looked at the phase diagram for oxygen uranium, you'll find out below 150 degrees C that UO_3, not UO_2, is the stable oxidation state.

Now, if you get oxidation, a number of things are going to happen. First, you're going to change the phase of the fuel. You can have UO_2, U_4O_9, U_3O_7, U_3O_8. You will open additional internal fuel surface so that if a leachant contacts the fuel, there will be additional surface area to release radionuclides. You'll release some of the trapped fission gas both from the surface of the grains and also internally to the grains as oxidation proceeds. And, if you have sufficient oxidation, eventually you'll split the cladding and change the path that the leachant has to take.

The bottom underlying question is what is the change of O to M as a function of time? If it's very slow, oxidation may be taking place, but it doesn't take place in a time frame that we're interested in. If it's rapid, then we may see any or all of these effects.

This is some pictures of what happens as the oxidation process takes place. This was from an experiment where we took a rod segment and we drove a small hole in it.

We oxidized it for a fairly short time. In this case, it was only for about 10 hours and it was 360 degrees C. And, you'll notice that we split the cladding open. It was about 7% deformation of the cladding. The split was quite long. In this case, it went pretty much straight up and down the cladding. We've seen it go around the cladding. We've seen less deformation in cases. We've seen it happen at lower temperatures, but the bottom line is if you get sufficient oxidation, you'll split the cladding open.

If you look at individual fragments, this is a picture of fragments of fuel as they've been taken out of a fuel rod. As you oxidize to U_4O_9, you'll see a reflective area around the grain boundaries. These reflective areas are the U_4O_9 that's forming on the grain boundaries. As you go through the U_3O_7 state on to the U_3O_8 state, you have a reduction in density which expands the fuel and eventually it will tear the fuel apart. So, this fuel becomes this fuel. If you continue further into the UO_3 state and even further beyond that into the hydrated UO_3 state, you'll start to tear the grains themselves apart. This experiment was done at a fairly high temperature, approximately 360 degrees C, but the same effect has been seen at lower temperatures to a lesser degree.

Looking at the information that was available on oxidation before we started this experiment and continued to develop in other experiments as we were going on, we formed some basis for establishing the oxidation work. I think it's very important to realize that the oxidation behavior of irradiated fuel could not be inferred from the behavior of unirradiated fuel. There was a significant amount of work that was done on unirradiated fuel. None of it agreed with what we're finding with irradiated fuel. Although UO_2 is the basis for spent fuel, it's not the thing that seems to be governing it.

We also know from the literature that temperature is a very important variable. One of the surprises that was obtained early in other programs, the oxidation rate tended to be lower with increasing burnup. Now, as we get to higher burnups into the 30 and beyond range, we're finding not much of a burnup effect, but we oxidized UO_2 on rating UO_2 much faster than we oxidized spent fuel. You look at the literature, the effect of moisture was unknown. There were studies that said that the moisture in the atmosphere significantly increased the rate of oxidation. There was other studies that indicated that it significantly decreased the rate of oxidation.

DR. PARRY: Well, you do have one important data point. You have failed fuel in the reactor and you don't get oxidation --

MR. EINZIGER: Well, you're dealing with a different situation in the reactor and the water state, the same as one of the mistakes that people tend to make is using very high temperature steam data to infer what happens with the oxidation of a fuel. So, we try to stay away from using that data. But, in a reactor, there is not any extensive oxidation.

Another thing that was apparent from the literature is that the low temperature oxidation data was not available. There were a few tests done on unirradiated fuel under conditions that really weren't applicable to the tests we were looking for.

What were the purposes of the spent fuel oxidation program? First was to determine the mechanisms and rate of oxidation. We wanted to find the effects of temperature, burnup, and moisture on this rate of oxidation. In addition, we wanted to get input for modeling of the oxidation and lastly provide samples for further leach testing.

We did three types of things in the program. One was the thermogravimetric analysis testing. This was done on single samples. It was used to provide mechanistic data. There were short-term tests, approximately 3,000 hours or less and basically what they did was continuously weigh the sample as it oxidized. Concurrent with that, we did dry bath testing. These were for longer terms, usually two years or greater. There were many samples so that we could determine repeatability of data. We provided extrapolation data so that we could take what we found in the short-term TGA testing and fit it to some lower temperature information. We went down to lower temperatures and it was done by interim weighings. Approximately every six weeks, we weighed the sample. Additionally, some samples were removed from both of these tests for examination; ceramography, transmission electron microscopy, x-ray diffraction, ion microprobe in some cases, and fission gas analysis.

Let's look for a moment at the TGA tests. We've used two types of fuel, PWR fuel at about 27 GWD/MTU. This was the Turkey Point fuel and we've also done some work with the ATM-101 which is H.B. Robinson fuel. These should be sister fuels. The only difference is the grain size, approximately 25 microns in the Turkey Point fuel and about 5 microns in the ATM-101. We've also studied some BWR fuel, about 32 GWD/MTU. This was Cooper fuel, ATM-105, and for the information of the committee what the ATM's refer to are the fuel designations of the approved test material provided by the Materials Characterization Center.

The TGA tests ran temperatures between 140 and 225 degrees C. At 140, insufficient oxidation occurred for us to measure it in a significant time period. At 225 degrees C, the reaction went too fast for us to also measure it in the TGA system.

We used dew points mainly of 14.5 degrees C, although some tests were done at -70 degrees C. Just to give you an indication, -70 degrees C has about 5ppm or less moisture in the atmosphere, and at 14.5 degrees C, it's orders of magnitude higher. These tests ran anywhere from approximately 300 hours to 2600 hours. In the course of the experiments, we obtained final O/M's ranging from 2.02 to about 2.4.

We tried to analyze some of this data and basically we used a model which assumed that the grains were spherical; not true, they're not spherical. That the grains oxidized independently; not true, they don't oxidize independently. That there was a planar oxidation front; well, for a short time, that's true. And, if you use those assumptions, you come up with a description that predicts the O/M as a function of time and gives you a rate constant. So, if you plot this side of the equation versus $t^{1/2}$, you should come up to determine a rate constant.

In fact, this is what we obtained. We have a period where we're increasing and this is what we think is basically due to the fact that the grains are not independent. That we don't satisfy the assumption that they're all independently oxidizing. We think at this point that the oxygen is migrating down the grain boundary very rapidly. Eventually, we have a time when the assumptions are pretty well satisfied. The grains are independent. We haven't had sufficient oxidations of the grain themselves. So, we're still having a planar front. If you continue the data longer than this, then eventually it turns up and you don't have the linearity anymore.

DR. PARRY: You mean off the scale or off the --

MR. EINZIGER: Excuse me?

DR. PARRY: You mean off this -- out past 28 hours?

MR. EINZIGER: Well, eventually, what will happen is that you will get sufficient oxidation. So, you go above .33 and the model blows up.

Basically, the model that gets put together is that there's oxidation in the drain boundaries and this is a similar picture that you saw before. The individual grains have oxygen around them. There's been a rapid transport path of the oxygen to the grains. That gives them their individuality. And, the oxygen starts to penetrate into the grains. Here's a grain boundary and this is a TEM photograph and here you see a layer of U_4O_9 and here is the boundary.

It's a very sharp boundary between the U_4O_9 and the UO_2. If we had changed the lighting a little bit, you would have seen the same thing on the other side of the photograph.

There are complicating factors though. This is a fragment of sample that was oxidized in the TGA. From the curvature of the surface we can tell that that was an outer surface of the pellet as it was originally fabricated and these are cracked surfaces that happened in reactor.

DR. VERINK: We're looking down on the outer surface?

MR. EINZIGER: You're looking down at a cross section.

DR. VERINK: This is a cross section?

MR. EINZIGER: That's right. You'll notice that the oxidation front is significantly further into the pellet as it comes in from the outer surface than it does from a fractured surface. We think this may deduce some rim effects. We haven't explored it in sufficient detail to find out what's going on there. It does create a complicating factor in analyzing the data, though.

You can summarize the TGA data by saying that spent fuel oxidation is a two step process. It occurs by oxygen penetrating the grain boundaries and followed by oxidation of both grains. We also know that there's a strong temperature dependence and it does affect essentially an arrhenius type dependence. The activation energy though seems a little bit strange in that it's independent in this stage of the oxidation process. The tests did not give us any indication of strong moisture effects and still essentially left the question up in the air. We know that oxidation occurs more rapidly at the pellet surface. The majority of the information in these tests does not come from the weight gain data. Mechanistic data comes from the microstructural examination of the oxidized fuel. There are two TGA apparatus at PNL, currently both of them are idle and examination of the samples and data to determine the effects of grain size, fuel type, and gas release to date have been deferred.

Let's move on for a second to the dry bath tests. As I mentioned before, the lowest temperature that we could get in the TGA tests was about 140 degrees C. So, we're obtaining some high temperature data. A thousand years into the repository lifetime, temperature is not going to be 140 degrees C, it's going to be somewhere down around 95 degrees C. So, we'd like to know that if we extrapolate the high temperature data obtained from the TGA down into temperatures that might occur in a repository that we're still sitting with the same mechanisms and our projections are correct.

DR. VERINK: What do you mean by shortest measurable oxidation rate?

MR. EINZIGER: Actually that should mean for the TGA we could obtain data in a certain amount of time. We ran for about 3,000 hours, and if we ran at a lower temperature for that amount of time, we would not find anything there. It would just --

DR. VERINK: Immeasurable?

MR. EINZIGER: Immeasurable.

DR. VERINK: Um-hum.

MR. EINZIGER: The TGA is a one sample device. We can put one sample in and run it and we opted for about four samples a year so we could get just more than one data point. And, based on that, we're talking about 3,000 hours. There's nothing in the experimental setup that precludes us from putting a sample in at a lower temperature and running it for a year, 10 years. Beyond that, we have institutional problems.

The dry bath test used initially Turkey Point PWR fuel, 27 GWD/MTU, and we used 50 samples. We followed that up with Cooper BWR fuel, approximately 30 samples. Currently, we're putting in Calvert Cliff fuel of two different types. Both of them are high burnup fuel. One has a low fission gas release, less than a percent, and the other one is a high fission gas release of 18%. We're running at three temperatures, 175 degrees C, so that we can overlap with the TGA data, and down as low as 110 degrees C. Once again, a temperature that for any substantial length of time, if we went lower, we couldn't get good data.

There's two different dew points, -55 degrees C and +80 degrees C. Once again, -55 degrees C, you're down in about the 10ppm moisture range. At 80 degrees C, you're up at about 200,000ppm.

Current duration of the tests, some of them have run as high as 25,000 hours and are continuing. The highest O/M we've gotten is 2.38 and, by the way, that sample has been removed and it's being examined at the current time.

A quick view of the setup in the dry bath test, basically we have a dry bath with just a heating mantle. In it are three aluminum plates, anodized (phonetic) aluminum, with holes drilled in them. Inside each hole is a nickel chromium crucible with about 10 grams of spent fuel. They're covered with a protector of nichrome so that in case we drop one of these samples -- you have to realize each of these are being done in a hot cell with two fingers. Imagine all the tests, take two fingers, and do it. So, that's one of our limitations. Each of the baths has a blank crucible in it so we can tell what weight gain is occurring from the crucible itself and generally it doesn't vary by more than +/-.3mg. Each of the blocks also has a thermocouple.

To date, we've run these tests for up to 25,000 hours, the O/M essentially change as high as .4. Interestingly enough, the oxidation is reproducible to about 10%. That means that if we take a series of samples and oxidize them under the same conditions and spread in the observation data, it's about 10%. We feel it's pretty good.

There is reasonable agreement between the rate constants obtained in the short-term TGA tests and in these dry bath tests. As we're getting to the lower temperatures, we find that the predictions from the TGA tests tend to form an upper bound, that we're getting slightly lower oxidation rate constants from the dry beds.

The finer grain BWR fuel tends to oxidize faster than the PWR fuel under all temperature and atmospheric moisture conditions. It appears, though, that this isn't a matter of PWR versus BWR, but a matter of grain size effect.

To date, we're still not sure on the affect of atmospheric moisture. We're starting to see a slight effect of moisture, but it is slight. It's barely coming out of the data after 25,000 hours.

Current experiments in fuel, we are using tailored samples, using only exterior fuel fragments or interior fuel fragments in order to accommodate modeling needs. Currently, only the long-term dry bath tests are being conducted with some limited sample examination and analysis being done.

DR. VERINK: Is it reasonable to conjecture that the temperature dependency on your uranium spots corresponds to something like diffusion of oxygen in uranium or --

MR. EINZIGER: Within the error in the data in -- you have to be careful when looking at diffusion of oxygen in UO_2. It can range over six orders of magnitude because it has a strong O/M effect in it. But, in general, when you look at -- it's more believable that it's in the right range, yeah. It's got about the right activation energy.

DR. VERINK: That's fairly consistent between the oxides, though?

MR. EINZIGER: Yeah.

DR. LANGMUIR: The oxidation rate is not a function of P_{H2O}, apparently. Would you think that's --

MR. EINZIGER: Excuse me?

DR. LANGMUIR: The rate is not a function of the availability of moisture, as you say. Might that suggest that the rate is -- moisture availability is perfectly adequate at all the levels you looked at; it's not limiting? That's the inference I would have.

MR. EINZIGER: Yes.

DR. VERINK: We're due for a break now and this is just about right on time. So, let's take a break until 10 minutes of 10:00.

(Whereupon, a brief recess was taken.)

DR. CHARLES WILSON: I'm Chuck Wilson and I'm with PNL and I do spent fuel dissolution studies on actual fuel specimens in the hot cell. I'm going to talk about some of the properties that influence the radionuclide release behavior from spent fuel and Ray already this morning gave a little background on that -- and briefly describe a couple of methods we used in the laboratory to do spent fuel dissolution tests and then go into some highlights of results from the tests that we've conducted to date.

Relative to release, the radionuclides in the spent fuel fall under two general categories; those that are very low solubility and the more soluble radionuclides. The soluble radionuclides can be in either true solution or can be low solubility species or adsorbed onto particulate matter that are in suspension. Carol later will talk about modeling. You can with thermodynamic models calculate solubilities. Colloids are a little more difficult to deal with with models.

One of the properties of the low solubility nuclides which are mainly the actinides is that these reach their maximum steady states and concentration in the solutions fairly rapidly in the tests which would indicate that the release will not be particularly dependent on the characteristics of the spent fuel waste form itself. Now, the more soluble species, it's a different story. In fact, the tests we've conducted to date would indicate that to meet the regulations you can't depend on the durability of the waste form itself. You're going to have to look at other waste package factors and environmental factors such as time distribution of cladding container failures and low probability water contact scenarios and other site-specific criteria.

We'll go on to talking a little more about soluble radionuclides in general. The spent fuel, as mentioned in Ray's talk, is a non-homogeneous material. When water would first contact the fuel, we'd have radionuclides that may be present in the gap, fuel cladding gap, on surfaces that have migrated out of the fuel during radiation and the soluble species and particularly iodine and cesium dissolve almost immediately on contact. So, this gap release occurs like in a matter of days from first contact. After that, we'll have a preferential component where we're preferentially leaching soluble radionuclides from areas like grain boundaries or other secondary phases of the fuel where they may concentrate during irradiation. With time, assuming that the fuel doesn't become substantially degraded by some environmental process such as oxidation before water contact, the current contention is that the dissolution rate of the UO_2 matrix where most radionuclides reside will be the rate controlling factor.

Okay. Our first tests were conducted in a recent procedure we developed back in the 1983-84 time period and we called them semi-static tests. And, here, we submerged the specimens in the water and take periodic solution samples until we reach some kind of steady state condition. Say, after six months, we'll change the water and restart the specimens for a second test and subsequent test cycles. These tests give data on the steady state concentrations you reach with the actinides and you can find secondary phases forming and so it provides information on what secondary phases may be controlling actinide concentrations. Also, we can measure the rate of dissolution, both the instantaneous gap dissolution component and the preferential and continuous release components for the soluble radionuclide dissolution rates.

One thing we can't measure in the static test because uranium in the actinides saturate in solution, and don't remain in solution, are the matrix dissolution rates. We've completed three series of tests in the program using ATM and Turkey Point spent fuels. More recently, we've been working on developing flow-through techniques so we can try to measure the matrix dissolution component of the fuel dissolution and that, so far, is concentrated mostly on the technique developments using UO_2, although the UO_2 reviews have been on actual production pre-irradiated fuel pellets.

This diagram just kind of schematically shows what one of the semi-static test systems look like. This happens to be the system we used for what was called the Series 3 tests and these tests were conducted with two types of configurations. One was the bare fuel configuration where we split the cladding open and let the fuel just fall into the bottom of the basket that contains, you know, the fuel where everything is submerged in the groundwater. These tests used J-13 water. And, another configuration where we would put water-tight end fittings on clad segments and artificially introduce a defect. Some of the defects were like a 200 micron diameter laser drilled holes or slits and then we would periodically sample the solutions until we reached a steady state and then terminate and go on to sequential cycles with the same fuel specimen.

DR. PARRY: Excuse me, your J-13 water, is that synthetic?

DR. WILSON: No, on these tests we used the water that came from the actual J-13 well, you know, and we have --

Barrel G was how we, you know -- and then we have a Barrel F and we characterized this water. Every time we use it, we analyze it to kind of follow its chemistry.

In the flow-through apparatus we're developing -- and we want this to be fairly simple if we're going to apply it to hot cell. Imagine the hot cell wall being right here, we have a feed water container and this could be a sealed vessel with a known cover gas to control the chemistry of the water and we have a control flow pump and in the hot cell we have a specimen column pumping this solution through and collecting samples. The objective here is to measure the dissolution rate for uranium and soluble radionuclides in a test where all the uranium remains in solution so that we know that we're measuring matrix to solution. And, it turns out not to be a real easy thing to do because the uranium solubility is below or fairly low -- in fact, the concentrations which you get played out in adsorption effects are even much below the solubility as we measure in a static test. So, we have to have flow rates that are sufficiently high that the uranium isn't played out, but when we raise the flow rates the concentrations of water may be only in contact with the fuel for a few minutes and those dissolution rates to the concentrations of the soluble rate you do apply are often too low to measure chemically.

DR. LANGMUIR: Chuck, how do you control the oxidation state of the system which is --

DR. WILSON: Of the system?

DR. LANGMUIR: Which is so critical in terms of release.

DR. WILSON: Okay. In tests we've done on the bench, this will be a sealed vessel or bubble. We'll equilibrate it with a gas and then we'll have an oxygen probe that will go down into the water to measure the -- the probe reads out in like ppm or actually it reads out in an EMF and you use a chart, a conversion chart, to convert to dissolved oxygen. That's where we're at now in the developmental tests. We haven't really done controlled oxygen potential tests in the hot cell with real fuel yet.

DR. LANGMUIR: But, you're not scrubbing oxygen out of the system then?

DR. WILSON: By scrubbing it, we're doing gas purging. I'm --

DR. LANGMUIR: But, you're going to have finite amounts of oxygen within your hot cell during the reaction?

DR. WILSON: Oh, yes, but this is a closed -- all this tubing here is a closed system until you get to here, but we run this for months. You know, we're measuring concentrations over a long period of time with a controlled oxygen potential solution going through there and it takes a period of time to reach some steady state conditions.

DR. LANGMUIR: Yes, but with any oxygen at all, you're oxidizing the U(IV) to U(VI) so you're going to have higher solubilities?

DR. WILSON: Yes, this isn't an equilibrium type of measurement. So, I don't think we get down to the oxygen concentrations that would keep the system reducing to UO_2, but we measure kinetic factors.

I'm going to just review some of the data from tests and this is just a key to understand, I'm talking about five air/fuel test specimens here, H.B. Robinson Series 2, Series 3 at 25 degrees and 85 degrees C. I'm going to just summarize some results on the actinides from these tests. The result suggests that the actinide release will be solubility limited and that's because we reached steady state concentrations relatively rapidly in the system. However, we haven't really completely assessed the affects of colloids and, you know, actinides may adsorb onto suspended particles in the system, whatever it be, in the repository or in the tests.

Sample filtration results indicate that the majority of the actinides, particularly americium and curium measured in samples and particularly in lower temperature tests were removed by filtration around suspended particles.

As we raised the temperature, the kinetic factors appear to favor precipitation of stable secondary phases over the colloid formation and the filtration effects tend to be reduced. If you look at the effects of temperature and filtration of a function of the actinide, we saw the greatest effects with the americium and curium and next the plutonium, uranium and neptunium are fairly similar, although uranium had a little more temperature effect than plutonium on the results.

Just really two points I want to make with this chart. First, that the steady state concentrations are reached relatively rapidly. In the first cycle, we have some super saturation effects when the water first contacts fuel that's been in the air for some time and the concentrations decrease and then we relatively rapidly reach steady state conditions in the tests. The second thing is that these three tests here, the upper tests, are the 25 degrees C test and we see significantly lower, like two orders of magnitude lower in this case, plutonium activities in solution when we raise the temperature to 85 degrees C. This is a .4 micronfilter. If we filter these samples, the activity drops down not quite to the 85 degree C level, but it does drop because of the colloid content.

Looking at filtration effects, this happens to be a test where we're looking at americium in a 25 degree C test. We see that the unfiltered and the 0.4 micronfilter data, there's little difference, but there's about a two order of a magnitude drop in the measured activity. In the membrane are two nanometer filtered sample activities and we see as we go off on to Cycle 2 that a two order of magnitude drop persists. At the higher temperature, we see the recycle, one, that we're rapidly decreasing in concentration or activity with time and that the effect of the membrane filtration at the higher temperature is much less indicating that the colloids are probably less of a factor at the higher temperature.

DR. PARRY: I think we're missing Page 11 in some of these handouts. We would appreciate getting a copy.

DR. WILSON: Which one is Page 11?

DR. VERINK: The one you just used.

DR. WILSON: This one is not in the handout? Okay.

If we look at the 25 degree C results from these tests which would be the more conservative results in .4 micronfilter samples which should include both colloidal and true collusion fractions and we look at the steady state concentrations that we measured and assume we have a flow rate of 20 liters per year per waste package, we can calculate some release rates assuming that quantity of water saturates at these actinide concentrations and using those parameters we calculate release rates on the order of 10^{-9} of the inventory of each actinide per year which is much below the 10^{-5} 10 CFR 60 limit.

DR. LANGMUIR: Isn't this also a strong function of whatever oxygen content you had in your solution since it's incongruent dissolution?

DR. WILSON: Yeah, if this was a -- well, I don't think it has much to do with congruent dissolution. It has to do --

DR. LANGMUIR: Well, no, it isn't. But, the rate of dissolution of the UO_2 which is the key to all of this is function of the oxygen.

DR. WILSON: I don't think the rate of dissolution of the UO_2 has much to do with what these concentrations are. Carol will be talking about this more later. This is determined really by chemistry in the repository.

DR. LANGMUIR: I don't see how. I mean, if you're talking about -- you're putting water through the system.

DR. WILSON: This isn't a flow-through. This is static test.

DR. LANGMUIR: That's static, but the release rate will be a function of a reaction which involves oxygen and UO_2.

DR. WILSON: Well, the release rate is a function of how much water is becoming saturated at these concentrations and it has nothing to do with the dissolution rate of the fuel.

DR. LANGMUIR: But, your product is a U(VI) product.

DR. WILSON: Right.

DR. LANGMUIR: You're taking it from U(IV) to U(VI) which is an oxidation process.

DR. WILSON: Yeah, I'm assuming that there's oxygen in the repository and at that rate the oxidation and dissolution of the uranium and the actinides into the solution occurs rapidly compared to the time that it takes to transport the water. I can talk --

DR. LANGMUIR: We can talk about it later.

DR. WILSON: Okay. We're going to talk about it later because I'm running out of time. This figure just shows a particle out of -- this is a fuel particle that's been in tests in J-13 water for about a year and a half and we can see the grains of the fuel and grain boundary grooving and dissolution along here and then we see all these cycular crystal phases which turn out to be uranophane which is a calcium/uranium silicate that's forming on the surface of the fuel. As the fuel dissolves, the uranium isn't going very far and it's these crystals that are determining at the uranium concentration, presumably some other phase will be determining plutonium and americium concentrations. And, so it's not really dependent on how fast this fuel is dissolving. It's really more related to these other phases.

DR. LANGMUIR: Well, where does the silica come from?

DR. WILSON: The silica is in the J-13 water and so is the calcium. And, the rock in the repository has a lot of silica in it. So, we presume it's going to be in the water.

Okay. This is kind of a summary of some of the soluble radionuclide release data. Technetium, cesium, strontium, and iodine in terms of inventory fraction per year released in these tests were on the order of like 10^{-4} per year at 25 degrees C and went up by a factor of about 600% in the 85 degrees C experiments in the range close to like 10^{-3} per year or slightly less or in the range shown, 3 times 10^{-4} to 1.2 times 10^{-3} were measured.

Carbon-14 tended to be preferentially released even farther into the test than just a gap inventory measurement and, in fact, 1% of the specimen inventory in the test conducted in sealed vessels were measured as being released in the first year of the test. And, this release was primarily from gap and grain boundaries, but not much of it appeared to be from cladding exterior. And, it wasn't from fission gas either. This was dissolved out of the fuel, this carbon-14, and if we conducted the tests in unsealed vessels, we really didn't measure it because it was lost as CO_2.

This figure kind of shows the relative activities of all the different radionuclides we measured in solutions during the test. You can see it spanned a range on this plot of 11 orders of magnitude. The most active radioactive species in the young fuel, of course, are the shorter half-life cesium-137, strontium-90, which in a couple hundred years have decayed out, but the longer half life soluble species like carbon-14, technetium-99, cesium-137, iodine-129, you know, these all have half lives on the order of thousands of years or greater. In Cycle 3, I've shown the solubility limited activities of plutonium and americium isotopes that we also measured in solution just for a comparison.

If we look at this data in terms of fraction of inventory released over given periods of time -- and here I'm plotting inventory fraction measured in solution versus time -- we see the gap inventory release of cesium-137, a fairly strong preferential release of Cycle 1 of carbon-14 and iodine-129. The cesium and technetium and iodine are much lower in the sequential cycles, but the carbon-14 is still being released at a substantial preferential component of release.

DR. PARRY: When you speak of inventory, you mean inventory within the --

DR. WILSON: In the fuel. This is inventory in the fuel plus the cladding in the case of carbon-14, but it's the inventory that was in the sample that was in the test.

DR. PARRY: How did you measure that inventory? By burnup?

DR. WILSON: Well, carbon-14 was measured radio-chemically in the same lab we did the measurements on the leach solution samples. And, the cesium and iodine and other radionuclide activities, we used ORIGEN code output for the fuel that we used.

This is pretty much just a repeat of the previous slide. It happens to be a different test, but it just magnified the scale of Cycle 2 and 3 that show that these soluble radionuclides are being continuously released with time. And, in this slide, it looks like it's close to a congruent release.

Here, I want to show the effects of temperature and oxidation to key variables in the release. We've got four identical Turkey Point fuels except for two of them were oxidized to both O/M's of 2.21 and 2.33. We've got one of the unoxidized samples, at 25 degrees C test and one at an 85 degree C test, and the two oxidized samples, both at 25 degrees C, and we can see that the oxidation caused substantial increase in the false release or gap release component of technetium. And, the temperature effect again was about a factor of six and that preferential release as a result of oxidation for technetium carried on into the second test cycle and these tests are still going. We're at about, oh, 1300 days on these two tests now.

DR. LANGMUIR: Chuck, the O/M ratio is the --

DR. WILSON: Oxygen to metal ratio.

REPORTER: Could you repeat the question? I couldn't hear you?

DR. WILSON: The O/M ratio of the fuel is the oxygen to metal ratio, the bulk oxygen to metal ratio in the fuel.

DR. LANGMUIR: So, basically, it's UO_2 at the low end and $UO_{2.33}$ at the top end of that?

DR. WILSON: Yes. Right. And, that's a bulk, that's not a particular phase.

DR. PARRY: Chuck, I'm sorry to trouble you, but would you go back to #16, please?

DR. WILSON: Let's see, okay.

DR. PARRY: I just want to check the dimensions there on the left hand side. Is that 1% or --

DR. WILSON: This is 1% release or close to 1% of the cesium-137 in the tests was released almost immediately or .9%, as soon as the water contacted it in this particular fuel. Another fuel that maybe have a different gas release for the fuel, like the H.B. Robinson fuel -- this is the H.B. Robinson fuel. Turkey Point fuel release a half of 1% of the cesium on initial contact.

DR. PARRY: Thank you, I misunderstood you.

DR. WILSON: Okay. Every time I try to explain this slide, I get tied up. So, I just want to make a couple points here. This kind of shows some of the effects in a flow-through test that you can measure of water composition and temperature and I'd like to use this for a bit of also an introduction of the next topic by Herman Leider because he's going to talk about doing these types of tests in more detail. But, what we're seeing here is we're starting in J-13 water. We change to a dilute bicarbonate, the same bicarbonate concentration in J-13. We increase by a couple of orders of magnitude and we started putting things back into it like the silicon and the calcium. The dissolution rate drops, just a constant flow. So, this is also a proportional dissolution rate. And then, we see an irreversible effect when we raise temperature which led us to the conclusion that something has to be precipitating out or happening. We may not even be measuring dissolution rate. We started examining after that the surfaces of the particles, the UO_2 particles, first on an electron microscope. We couldn't see anything, but when we started looking at it in the Auger microprobe we were able to detect that we formed very thin, like 50 angstrom layers of a calcium silicate containing layer on the surface that was apparently formed and acted as a kinetic barrier and made these tests irreversible. But, we were able to partially dissolve that layer off by going back to deionized water or more rapidly we dissolved it off by going back to bicarbonate.

This is just an example of another test which this data has been run out somewhat further and we've done some different oxygen potentials after this data was taken. But, it just shows that there is an effect of reducing oxygen potential in the solution. This is air-saturated deionized water, about 8ppm dissolved oxygen. We've Argon sparged it and the oxygen potential with time went down to about -- or I shouldn't say potential, concentration when down to about 10ppm of dissolved oxygen, a substantial decrease in the rate of dissolution. We went back to the air sparged system. We immediately increased and then we just run the air cover.

DR. LANGMUIR: In your previous experiments, did you have well-defined control on this option content? Did you know what it was?

DR. WILSON: We didn't measure it, per se, like we did with the probe here. It just was air equilibrated solutions.

DR. LANGMUIR: In all cases it was air?

DR. WILSON: In all the semi-static tests, it was pretty much air equilibrated solutions, air cover in the experiments.

Okay. It looks like I'm about out of time here, but kind of in summary, the actinides in the tests appear to reach solubility of limited concentrations fairly rapidly. So that would indicate the release should be solubility limited. Soluble nuclides, though, will be somewhat more complicated to model because the fuel is a non-homogeneous material. We have the gap and grain boundary, matrix release components, plus the fuel is likely to change with time as a result of oxidation or degradation of the fuel in the repository and oxidations, grain boundary dissolution can cause an increased surface area over time. But, for the samples we did run, we found that there are gap releases of cesium and carbon-14 on the order of 1% of the inventory in the first year. And, for other soluble radionuclides in later test cycles, technetium-99, cesium, strontium, and iodine we measured release rates generally in the range of 10^{-4} to 10^{-3} of the inventory per year in later test cycles.

There's a lot of things that we really haven't figured out yet about what's going on. And, some of the things that would be good additional information needs would be radionuclide distributions in the spent fuel. This is something that has been done through MCC and currently there isn't a lot of activity there, but in particular, it would be nice to know a little more about the carbon-14 distribution in the fuel itself. The dissolution behavior of oxidized fuel and other fuel types such as high gas release fuels, we really haven't done that yet. The effects of colloids, the actinides that sorb out onto colloids are already in the groundwaters and how do colloids come about and what are they and what are their effects? That will particularly have an effect on this conclusion up here. The effects of water conditions on matrix dissolution rates. Herman is going to talk about this a little more in the next paper, but it's something that we've just scratched the surface on.

In the models that are available to date for modeling spent fuel radionuclide dissolution behavior are similar to the types used on glass where you have a known surface area and you don't have that in spent fuel. It's going to change with time and it's difficult to measure, and to use these types of models, we really need some kind of time dependent model for the surface area of the exposed surface area of the fuel on how it evolves with time.

Everything I've presented has actually been published in different documents and I gave Helen a copy of four references which describe most of this data in more detail and she'll make those available to the panel members.

The next talk, Herman Leider will be talking about dissolution experiments on UO_2 and experiments to make some of the measurements I was talking about in the last conclusion.

DR. HERMAN LEIDER: I'm going to describe to you how good it's going to be. These are experiments we're about to begin at Lawrence Livermore; in fact, they're imminent. They may even have begun today, but I'm not quite sure. This will be on undersaturated flowing dynamic dissolution tests on uranium dioxide.

In anticipation of possibly your first question, why study UO_2? Basically this is a study in chase of a mechanistic model for dissolution of spent fuel and what we have to do is be able to describe the basic maker's dissolution first. Irradiated fuel is a rather complicated chemical mixture and so we feel it is essential to be able to describe the dissolution of uranium dioxide, per se, at least uranium dioxide in contact with a number of chemical conditions. Once we do this, we can make a direct comparison with spent fuel and, in principle, learn a lot of things about the effect of the presence fission products in the matrix which are to several percent usually in an irradiated fuel. We can examine the suspected, at least postulated, chemical effects of the irradiation field in changing the chemistry of the water because of the high activity near the fuel and we can possibly have a good deal to say about the contribution of grain boundary to solution since they are a source of the some of the fission products which are exolved from the irradiated fuel.

Your second question would be my second question, too. Do we need any more experiments? It's a big literature. And, the answer is yes because there's lot of data and it's not self-consistent nor is it useful for our purposes. I'll show you some of this.

For instance, these are published dissolution rates for uranium dioxide, initially uranium dioxide, under various conditions. Some spent fuel is included. It doesn't take too much of a careful look to see there are orders of magnitude differences as a function of pH is the way this is plotted.

DR. LANGMUIR: Is it largely because of the oxygen concentrations not being --

DR. LEIDER: I believe that's partly because of inadvertent lack of control on important things like oxygen potential. It's also due to deliberate differences which people didn't recognize as important, say, in water composition.

DR. LANGMUIR: Complexation.

DR. LEIDER: Yes. There's lots of things that were never done carefully and well-defined. That's why we have to do this, I believe.

It's not just the rates of the dissolution that are scattered. The solubilities as reported in the literature are pretty bad, especially for tetravalent uranium which is less important for our case because we expect oxidizing conditions. For uranium-6, there's a big scatter, less than in 4, but a big scatter. So, we don't even know very well the equilibrium concentrations of saturated solutions, saturated in the sense of the solute being dissolved. And, to some extent, I'm guilty of this. Some of the old data was mine.

DR. LANGMUIR: Do you have the data from Parks & Pohl in here?

DR. LEIDER: I do.

DR. LANGMUIR: That's after '88, isn't it?

DR. LEIDER: That's in there, too. This came from a recent SKB report and I believe that's included. They referenced all their points, but I took it off because it complicated the viewgraph. However, this particular report listed on the bottom does have all the sources for this.

Static or semi-static tests do have this limitation. These are the kind of things that Chuck Wilson just reported to you being done on spent fuel and they're essential on spent fuel. It's very hard to do much with extremely hot material. But, the static or semi-static tests can't give you rates except for the very soluble materials which you can see build up in time. They will give you solubility levels at steady state. So, we want to do dynamic tests to be able to determine rates, as well as saturated conditions.

Now, in the unsaturated flow tests for many important species in spent fuel, but certainly for your uranium which is what we're going to be studying, per se, you can produce measurable concentrations. So, you can measure what you get out of a product in the flowing system. This is also true for things like cesium and strontium. Some of the others may be below detection limits and they'll have to be looked at in detail. Analytical techniques have limitations. I don't know exactly how many fission products you can accurately determine with this method, but you can certainly do uranium.

In the tests we're proposing, we will be far below the saturation level of a solute and we will be using what amounts to simplified water. We will not use J-13 well water which contains many components. We will use certain simplifications so that we don't have the complication of secondary failures precipitating.

The philosophy of these measurements is precisely the same really as what you heard described for the glass dissolution. In fact, the apparatus and the techniques are very similar to what Kevin and Bill Bourcier talked about yesterday. We expect in a flow-through test -- and you will know this expectation is right since Chuck showed you a slide already -- that we will plot the inverse flow rate versus concentration you achieve and it will be linear. And, the slope of the plot will give you a dissolution rate and he already showed you that's largely so, at least in the preliminary tests. We can measure solubility directly. Basically, you get to slow enough flow rates and you've got a static condition. And, so if you have this slope which goes up and then bends over, or you plain stop and measure when it gets to be static, you can get solubility in these experiments, as well.

The last is a point for people who are interested in a lot of details about dissolution properties. I'm sure it's a case if you see a change in slope while in the unsaturated condition, you're looking at a change in mechanism. We don't anticipate this, but at least we'll be able to see it if it happens. So, surprises can be dealt with in this kind of a measurement. Surprises can be recognized, I cannot say dealt with.

Here is something you saw shortly a little time ago by Chuck Wilson. This is his flow-through dynamic tests on UO_2 unirradiated fuel in this case. He did it in deionized water and in synthetic J-13 water. In both cases, you've got a pretty good linear relationship between the reciprocal of the flow rate and the uranium concentration. And, the slope is the rate. So, we know this is going to work pretty well, and once we start, it is going to be wonderful.

We will use simplified solutions. We are going to limit the variables in the water to pH, that is hydrogen/ion concentration, to carbonate/bicarbonate concentrations which are the complexing agents that will enhance solubility of uranium, hexavalent uranium, of course, and the oxygen activity or concentration, if you like, because the oxygen will be crucial here in getting to that hexavalent state which is the soluble or much more soluble version. And, in principle, you can see it, but the limiting material we'll be looking at is something like schoepite which is uranium trioxide all the way up.

Under repository-like conditions, you know, the other constituents of the water you expect, such as the J-13 water, which has silica and calcium will definitely produce a lot of insoluble secondary phases which will be solubility limiting. Interpretation of this complex system is probably beyond our means and so that's why we're simplifying it. And, in any event, these will produce lower levels of concentrations than it will without them. So, we're looking at the worst case which is what you want to do for modeling purposes. Because if you don't look at the worst case, you're not predicting safely.

Now, if you'll look at irradiated fuel, you're going to have radionuclides that are not uranium from three sources. These have been alluded to a number of times. I'm not going to bore you with this. The gap which is volatiles released external to the fuel and is immediately available for dissolution. Stuff that's gone out of the grain because of insolubilities into the grain boundaries to form other phases which are going to behave differently kinetically because they're more accessible in principle than in the stuff that dissolved in the matrix. And, the matrix itself, the flow-through tests are going to attack mainly an exploration of the bottom one. We hope to be able to, by inference, say something about the others.

A schematic of this, it's somewhat different than ones you've seen before, but I like it. I just like it.

But, it simply shows the total release rate, not release, and it's plotted on a log/log. Seal (phonetic) as a function of time with the total being the upper envelope that covers everything. We have a constant contribution presumably from matrix dissolution. You have a very fast and quickly decaying contribution from the gap, the stuff that's free immediately. It goes in and then it's gone. And, the grain boundaries are faster than the matrix in principle and will -- you can see stuff nearer the surface in the grain boundary will go pretty fast. And, if the water has to penetrate deeper and deeper, it will slow down. Eventually, everything will asymptotically approach the matrix given enough time, whatever that happens to be. We hope to say something about that with our experiments.

Now, this is not to say people don't understand anything about dissolution, they certainly do. Going back to a very elementary text, you say, hey, people have decent models that describe dissolution. This is back in the early parts of this century. An equation like this which describes the rate of dissolution of a non-dissociating material. It's a function of how quickly the solution becomes concentrated in the solute. This describes lots of things very well. However, we're dealing with ionic materials. So, even starting with such a simple model -- and this isn't so awful as you might think because it's so easy to look at -- complicates things because once you start getting ions you start complicating the chemistry of it.

Now, this is a very overly simple equation for dissolution of UO_2 and it's just a pedantic tool here. So, disregard the formation of +4 ion which people in solution chemistry know is not something you're going to find. But, in this case, it's simply illustrative that when you dissolve an ionic material, you make ions. Other things strongly influence your dissolution properties. For instance, in this case, you can see that it's very strongly dependent on the 4th power, in fact, of the hydroxyl ion concentration and thus on the pH, the 4th power of pH which is -- water constant related directly. And, so already we've come to our first complication.

Now, a schematic of what happens when you immerse UO_2 or spent fuel into water of some sort and we'll say whatever repository water is, this water is going to have some oxygen dissolved in it. There's going to be some oxidation potential. It's going to oxidize to a certain degree, the surface of the UO_2. What's more, the water is going to be contain things other than water. The L written in this equation are some undefined cation. In our case, we could say that's bicarbonate. These things will form a stable complex with uranyl ion, for instance, which dissolves from the oxidized fuel and make it more soluble than it would otherwise be. All this has got to be considered. As I say, this is in simply for schematic purposes.

When we look at what happens in the UO_2, there's a lot of things that may determine the rate of dissolution. We know it's going to oxidize. We don't know for sure how it's going to oxidize, whether it's going to be molecular oxygen dissolved in the water making the uranium oxide go to a higher state or whether the oxygen is going to have to adsorb on the surface of the solid and dissociate into ions or whether it has to dissolve in the solid. All these things are possible.

The point I'm making here is, for instance, the dependence on external parameters for these processes are understood. We don't know which ones are important, but we can describe them sort of in first principles. And, once you make it oxidized and you have it in the water, especially in our case when we're talking about carbonate/bicarbonate, you have all these processes that go on in solution as part of the dissolution process. These aren't exhaustive, but it's simply an exposition that there are a lot of things we understand might happen, probably do happen, and we can describe their dependencies on basic properties of the system accurately. We just don't know which ones are important yet without doing the tests, but we know which ones are probably involved. Some of these are going to be involved and unimportant. You can throw them out in the model.

We decided because of the variables even in the simplified solutions that the only sensible way to deal with this in an economical fashion, and I mean economical in the real sense, is to do a statistical approach. We have designed an experimental matrix using a commercially available program, for instance, a well-defined mathematical technique for doing statistics on an array of variables in experiment which will allow us in a set of reasonable tests, which may be done simultaneously in part, to determine the interaction between these various parameters, say, the bicarbonate, the temperature, the oxygen, et cetera, and determine the relationships between them in a quadratic fashion. That is take an equation and give it to you in terms of a polynomial, $a + bx + cx^2$. Now, these aren't first principle relationships, but we do know the first principle format of a relationship and can decide which ones fit a quadratic or a linear one, at best. This will not only give us an actual empirical relationship for the conditions we look at, but will allow us to decide which of the theoretical forms fit it best. Anyway, this allows us to get a lot of information that one by one variation of parameters will take years to do. Hopefully, this will not take years.

Here's an example, a little bit hokey in terms of the top scale, but this is what we're going to do is describe the matrix of 19 experiments which include replication in the alkaline region, that is, you can see the pH in this case is going to range from 8 to 10. At least, that's written down. We're going to look at temperature between 25 and 75 or it could be any range we chose, you know, zero and 100. We're going to look at the activity of -- in this case it should be the carbonate/bicarbonate concentrations. In the acid case, it will be the gaseous activity which determines the solubility of CO^2 and the oxygen activity. This matrix is easily achieved. It's not too horrible in these flow-through tests to do. That's only 19 tests. We may be running six at a time. And, we can get a lot of information out of it and I'm convinced that the only way to look at a system this complicated is from a statistical point of view.

Now, if we look at what we're going to do with spent fuel, we will do the same thing ideally. I don't know if Chuck is smiling or groaning back there, but at least within the limitations of what you can do in hot cells, we're going to try and repeat as many of the elements in that matrix or in such a matrix as we can with spent fuel so we have a direct comparison. And, this will, in principle, allow us to extract the differences in behavior due to the differences in composition because of the fissioning that went on or the irradiation that went on.

We're also looking at some interesting, new, and novel techniques that are very sensitive for looking at the process, one of which has been explored a bit for us by Rick Russo at LBL in Berkeley and this is a technique which is called spectro-electrochemistry. But, basically, let me just tell you what it is. They do the electrochemistry using a uranium oxide probe and they establish an oxidation potential electrically and they use a parallel beam of a laser which is deflected and it's deflected because of gradients in the refractive index due to both concentrations of chemical species or thermal gradients that are produced during the reaction. And, in principle, can also do spectroscopy simultaneously because if you use a laser and you're watching it deflect, you can also tune it on a transition you want and see something come in or go away.

The other thing which is a little easier for me to visualize is using scanning atomic scale microscopy. This is new stuff. This includes scanning tunneling microscopy and atomic force microscopy. This is stuff that's available at the laboratory right now at our laboratory. Siekhaus and

Balooch are operating these now. Both of them are available and will operate under water. They'll operate under real conditions and you have in principle the capability of seeing on a given flat surface, pretty flat surface, an atomic layer disappearing and the rate at which is disappears. This is a direct measurement of rate without having to fool around with anything else. You just watch it go away. This has been done. They've done it recently on oxidation of pyrolytic graphite. This is not a related thing. But, they've watched it oxidize layer by layer and they can measure how far, atom by atom, how fast it goes away. You're seeing things that under conventional techniques are 12 orders of magnitude too slow to measure. That's nice. We hope this will pan out for us. That's a very good check, by the way, on any model we have because if you don't fit that observation, you're in trouble.

So, in summary, we think we have found a way to do tests in a satisfactory fashion which are statistically satisfying. We have an environmental system capable of the fine control necessary to eliminate the variables that cause -- scatter in data. We're about to start and it's going to be great.

Thank you.

DR. VERINK: At this stage, if there are no questions, I will introduce someone who probably knows more and is prettier than I am. Carol Bruton will come up and tell you all about the geochemical implications of all this and the value of EQ 3/6 code.

MS. CAROL BRUTON: What I'd like to do today, is to discuss some geochemical modeling work we've been doing. The modeling of dissolution of spent fuel. So, the planned outline for today is to first give you an idea of what geochemical modeling is, what the tools we are using are, and then to give you a conceptual idea of how the models work. And then I'm going to show you some sample simulation results and compare the simulation results to the results that Chuck Wilson has got in his hot cell test. Then we want to investigate implication of changing water chemistry on our simulation results. And, in conclusion to give you an indication of what important variables that we must consider in trying to predict radionuclide concentrations in solution.

Why do we use geochemical modeling? Because, we are being asked to evaluate nuclear waste repository performances over time periods as great as ten thousand years in response to a wide number of variables, such as temperature, fluid flow and other conditions. Now the traditional approach to this type of problem is to set up an experimental program which we have done. But obviously in any experimental program, there are a number of limitations, such as we are restricted to a laboratory time scale of a number of years, or if we have to make predictions, we are up to ten thousand years.

In addition, for a given experiment, you can only set a given number of variables in their combination. So when we are asked to evaluate performance under a different set of conditions, it can be difficult. And that leaves the last one, it's very difficult to extrapolate the results of one experiment to multiple repository scenarios.

That's why we are suggesting that the most powerful combination to attack these problems is to combine the experiments with geochemical modeling. The combination of them is more powerful than experiments alone or geochemical modeling alone. By using this combination, we hope to develop quantitative process oriented models of repository response. And yesterday you heard one of the best examples of that in Kevin Knauss and Bill Bourcier's work in their developing a kinetic model for glass waste form dissolution.

Once we have these process oriented models, we can simulate complex interplay among the proxies that control rates of waste form degradation. And ultimately to predict the changes in the chemical environment that will occur throughout the post-closure period.

The geochemical tools that I am using for the simulations you'll see is the EQ3/6 software package. Now it's composed of two codes. EQ3 which is an aqueous species distribution code and EQ6 which is the more interesting one, because that actually carries out the dynamic simulation of interaction among host rocks, repository components and the fluids. Now both of these codes are based on fundamental thermodynamic principles. And thus, they both call upon common thermodynamic data bases with data for almost 2,000 solids, gases, and both inorganic and organic aqueous species.

Now since the simulation was made using EQ6, what I'd like to give you now is a conceptual overview of how EQ6 works and then you'll have a better understanding of how I got the results.

What I've got now is a series of three color overlays which are compressed into one black and white one in your handout. Okay, how does EQ6 work? How do I model in the interaction of a solid phase in the presence of a fluid? Well the first thing you do is you give it the composition of your starting phase whether that be a spent fuel pellet or glass as Phil was doing yesterday. And then you specify the rate at which the components of this spent fuel pellet in this case are released into the solution. Now if you have a rate equation such as you have for glass, then use that absolute rate equation. If you have no rate equation, you can assume the dissolution is congruent. Or for spent fuel, we may want to assume that the grain and gap boundary components are at least a little bit faster.

Now since this simulation was done for spent fuel before we had a kinetic rate equation, I assume that the components of the spent fuel were released congruently into the adjacent solution. And by congruently, I mean that they were released in proportion to their quantity in the initial spent fuel pellet.

Well the next step in the code is to calculate the species distribution of these elements released in solution. Carbon gets released in solution and it speciates as a carbonate and a bicarbonate species. You'll see that when uranium goes into solution it forms a uranyl ion. And then the uranyl ion sees this carbonate in the solution and combines to form a uranyl carbonate. That's a function of EQ3.

Now the powers of geochemical modeling approach is in the next step. When you look at the composition of the solution and check to see whether you are saturated or super-saturated with respect to any solid phases, for example, this spent fuel, are we saturated with respect to the calcium uranium silicate haiweeite. If the code finds for super-saturated with respect to any solids, it will simulate the precipitation of these phases until we produce a secondary phase assemblage. And you see, for each thing it precipitates it has to revise the fluid chemistry. So once the solution has come back down to equilibrium for the secondary set of phases, you go right back to the beginning, dissolve a little bit more of the spent fuel, change the solution composition, check for precipitates and so on. So through the simulation, you can see we will predict the sequence and the identity of the solid precipitates that form as we dissolve the spent fuel, and also how the fluid chemistry changes as it dissolves.

Okay, now we'll look at some simulation results. What I did was to take a representative PWR spent fuel composition, which included both fuel and non-fuel components, that is sub-components from the zircaloy cladding and hardware and I dissolve it in J-13 water at 25 degrees centigrade. What I predicted was the solid base that precipitated and the changing composition aqueous phase concentrating on the radionuclide concentrations in and solution.

Okay, this is the set of solid that precipitated as I dissolved that spent fuel. The horizontal bars here refer to the stability ranges of each solid phase. And for your information, on the next page of your handout, I won't show it, it's got the names of these minerals and their compositions in case you are curious.

So you will notice that I've expressed the sequence of solid phases in terms of the amount of spent fuel that got dissolved. The log of the grams of dissolved spent fuel per kilogram water. This axis is our proxy for time. When we get a kinetic rate equation, you will see on this axis time, but, for the time being now, we're just calculating the extent of reactions in terms of the grams of spent fuel dissolution.

Now there are about 25 components in this spent fuel. And you see we get a wide range of solid precipitates. Those in the beginning ranging to those that are more representative of J-13 water and those at the end more representative of the components in spent fuel. But rather than going through this whole thing, what I'd like to do is concentrate on the results for three radionuclides, uranium, neptunium and plutonium, look at the solid phases and then see how the solid phases control the composition of the radionuclides in solution.

Okay, let's look at the results for uranium first. On the top part of the diagram, what I've done is to excerpt from that paragenesis plot the stability phase of minerals; the three phases that precipitated to sequester uranium. Haiweeite, which is a calcium uranium silicate, soddyite a uranium silicate and schoepite which is a hydrated uranium oxide.

Now look at how the precipitation of these phases were affected by the composition of the solution. On the bottom part of the diagram here, I plotted the elemental concentrations in milligrams per kilogram of silica and uranium. Again as a function of the amount of spent fuel dissolution. As long as the silica in solution was high and it is high in J-13 water, uranium concentrations were maintained at the .03 milligram per kilogram level. But as soon as I depleted the solution in silica, uranium increased in the concentration to over 10ppm.

This suggests that as long as we have sufficient quantities of silica in J-13 water to support the precipitation of uranium silicates we'll keep uranium low in solution. Now I should mention here Chuck Wilson did observe the precipitation of haiweeite and soddyite in his experiments. He also observed precipitation of uranophane, which is another calcium uranium silicate. But unfortunately the data in the data base for uranophane was actually estimated by Don Langmuir and is not precipitating in the runs. I think we have to do more calorimetric studies on uranophane.

DR. LANGMUIR: Just an estimate.

MS. BRUTON: Okay, let's compare it with the results of Chuck Wilson and his studies of Series 3 at 25 degrees centigrade with H. B. Robinson fuel. This is the concentration of uranium he measured, about .3 milligrams per kilogram in solution. A little bit higher than our predicted .03, but we'll see a little bit later there are other probable explanations for that.

But it does suggest there are sufficient silica in Wilson's water to maintain relatively low concentrations of uranium in solution.

Let's turn to neptunium. Look at the same diagram here. These are simulation results at 25 degrees centigrade. Now neptunium is a little bit different. There was only one phase that precipitated to sequester neptunium. That was neptunium oxide. And I made the initial simulations assuming atmospheric conditions, a fergacity (phonetic) of oxygen in equilibrium with the atmosphere.

And my neptunium precipitated late in the sequence. When I look at the log of my neptunium concentration in molality it increased in solution until I started precipitating neptunium oxide. When you look at the results of Wilson, and he measured neptunium concentrations not at 10^{-6}, but 10^{-9}. But we know that the neptunium concentration in solution is very dependent on the redox potential. And so when I artificially went in and made another simulation assuming that the fergacity of oxygen of 10^{-12}, we obtained a much better match to Wilson's experimental results.

Now Chuck has just mentioned that he did not control or he did not measure the redox state in his vessel. And he did have air cap in it. These results may suggest that we would have to consider gradations in the redox potential in these reaction vessels.

The next diagram we see, actually I tried to explicitly show the dependence of neptunium solubilities on Eh and pH. This is called a Pourbaix or Eh and pH diagram and I've tried to simplify it a bit. On the Y axis, the Eh in volts, on the X axis, the pH. And what I have contoured here are the molalities of neptunium in equilibrium and neptunium oxide as a function of Eh and pH. And what you can see is that we started our run at about a pH of 8 and under very oxidizing conditions you can see how a very small change in Eh can drastically change neptunium concentrations in solution. That suggests in a repository scenario that we are going to have to have some idea of what controls the redox state to be able to predict what neptunium concentration would be in solution.

Now for plutonium, is that also dependent on Eh or pH or on something else? When we take the same type of diagram, Eh and pH, and these contours are now for the allowed molality of plutonium in equilibrium of PuO_2. And again you can see that under pH of about 8, as in many of these experiments, that the concentration plutonium is highly dependent on the Eh. There's another factor that we have to consider in plutonium. And that is it's not only dependent on Eh and pH, but it is also dependent on the nature of the phase of the solid precipitate that first forms from solution.

Here I have superimposed a set of different lines on this diagram, so let's go through them. While my initial simulation--if you remember that first diagram under oxidizing conditions, I precipitated a plutonium oxide. Now that plutonium oxide constrained plutonium concentrations in solution to about 12^{-14}, depending on the fergacity of oxygen. A very low level. But the experiments of Rai and Ryan, et al, suggests that the first plutonium precipitate may not be this crystalline form, but a more amorphous hydroxide phase. So when we go make the simulation constraining the precipitate form to be plutonium hydroxide, we get much higher concentrations of plutonium in solution, about 10^{-4} and 10^{-6} versus 10^{-12} and 10^{-14}.

So looking at the results for uranium, neptunium and plutonium, we can see that while uranium concentration was dependent on the overall chemistry of J-13 water, neptunium was mainly dependent on the Eh of the solution. But, plutonium tells us that not only do we have to consider those variables, but you also have to consider what phase would be the most likely one to precipitate kinetically.

All right, there's always a question that comes up in any estimates of radionuclide concentrations in water--I've been using these simulations assuming it's J-13 water. There's a question, is J-13 water representative of the composition of water at the proposed repositories sites? In fact, is J-13 water sampled from the saturated zone representative of water from the unsaturated zone? And finally, what if the composition of this water varies areally over the site. How will that affect our predictions of radionuclide concentrations?

The next diagram you have is just a table showing you what ranges and compositions have been measured for J-13 water. And things in small concentration 10ppb, but we can get up to a few milligrams per kilogram change in solution of radionuclides. In fact when you look at the compositions of extracted pore waters which come from a non-welded unit of unsaturated paintbrush tuff, we see higher concentrations of these elements in solutions. How will these changes in solution composition affect our predictions of radionuclide concentrations in solution?

An additional thing we have to consider is the water that compacts the waste package will probably not be the composition of the water that's out on the site now, because it's going to undergo a lot of interactions on its way into the package and also on the way out. For example, would it be heating up the host rock. What happens when we heat up the water and it interacts with the host rock? We are going to have metal corrosion going on. It may interact with cement or any organics that are left in the bore hole; interactions with any manmade components and you have a radiation field. All of these parameters can have an affect on the pH and Eh of the solution and also an affect on the major and minor components of the water, through processes such as precipitation of phases, sorption and in the case of organics, complexation.

All these things may have to be taken into account. I did some sample calculations to show that yes, you do have to take them into account because they can affect the radionuclide concentration.

On this diagram what I've plotted is the uranium concentration in milligrams per kilogram as a function of pH. And these are minimum solubility envelopes for uranium and, for the solid line, for J-13 water. That is at a given pH the uranium concentration can increase but it reaches a maximum when these labeled phases tend to precipitate. So this gives you an idea of what the concentrations in uranium in solution are as a function of pH.

Now if we do this calculation assuming standard J-13 water, we get the solid curve here. Now my question is, what if the calcium content dropped by order of magnitude from 12.5 milligrams per kilogram to 1.25 milligrams per kilogram? That is shown by this dash line here. Note that the concentrations in uranium in solution can just about double.

Now if we take it one step further, let's not only decrease the calcium by an order of magnitude, but let's change the silica to an order of magnitude dropping it from cristobalite saturation to quartz saturation. Look at the drastic effect it has here. Bring it up to about three orders of magnitude change of potential uranium concentrations in solution because we've changed the major element chemistry of J-13 water. So I think we do have to take these interactions with all the components of the repository into account before we can reliably predict radionuclide concentration in solution.

DR. LANGMUIR: Carol, is it going to be a single number anyway when you get to a near field situation of the thermal gradient and all these variations probably will occur in a few inches, plus a few more.

MS. BRUTON: We hope to get ranges. We hope to make conservative estimates. But we have to make the simulations under the different conditions to see what our ranges are, and then to compare all of the different ranges so that you can get a coherent range that will take into account all possible scenarios. That's how we are looking at it now.

So to summarize, I think we found, which is no surprise that we would be able to confirm it is that actinide concentrations in solution can vary significantly with changes in Eh, pH, solution composition, and the nature of the actinide-bearing precipitate. And here I show you a little chart that sort of summarizes the results from this talk and from some other things I didn't include.

But what I'd like to emphasize is that they have an impact, but the solution chemistry must change by orders of magnitude to impact actinide behavior.

And if you remember the chart I showed earlier of compositions of J-13 water, it doesn't appear that the observed variations in the chemistry of J-13 water and the extracted pore waters from the unsaturated zone do not seem large enough to affect actinide concentrations.

And this is what I want to emphasize, it's the interactions among the waste form, the repository components and the host rock that can result in such order-of-magnitude changes in solution chemistry. And that we much consider these interactions in order to reliably predict radionuclide concentrations through time.

Thank you.

Questions?

DR. LANGMUIR: I know EQ3/6 has a kinetic subroutine which you haven't used here and I suspect there simply isn't data for the uranium minerals at this point to put that subroutine into action.

MS. BRUTON: Right. You mean for spent fuel dissolution?

DR. LANGMUIR: Well for example you are suggesting that uranium is limited by some secondary uranyl phases, but you're just strictly using thermodynamic arguments to determine what the levels might be and to compare those to some experiments in which rates presumably were involved.

MS. BRUTON: That was really curious because that is one of the reasons for making the comparison. Okay, we're making the comparison, there are simulation based on purely equilibrium considerations. When comparing it to Chuck's experiments which have both equilibrium and kinetics, how well are we doing?

And the other thing was I didn't mention, but looking for results in like neptunium and plutonium, now Chuck measured these concentrations to reach steady state levels, but he didn't observe any solid precipitates that sequester these phases. So one of the things was, okay, can we use the modeling to give us an idea of what phases might be precipitating in his experiments because we'll never be able to see them. They are just going to occur in such small quantities.

DR. LANGMUIR: So you really can't validate what you've got at this point. It's inferential.

MS. BRUTON: We don't want to--yeah, we weren't doing this to say, okay, we want to. Our results should match Chuck's. It was basically, okay, can we use the results to give Chuck a better indication of what might be happening in the system and take it a step further. How sensitive are Chuck's results to changing different parameters like solution chemistry or something else.

DR. VERINK: You'll note by the schedule that we are right on schedule again. Because of the change in the general thrust of the activities in the next section, it seems appropriate to take lunch now and try to be back here so we can kick it off at 12:10.

(Whereupon, a lunch recess was taken off the record.)

AFTERNOON SESSION

DR. VERINK: Let's reconvene.

I call your attention to the fact that at the close of the formal program, there will be time for some questions from the audience as well as from the panel.

MR. PETRIE: We are about to start the final session. This afternoon we are going to talk a little bit more about spent fuel and the characteristics and the data base. Your first speaker this afternoon will be Alan Brownstein from the Department of Energy. I'll turn it over to him.

MR. ALAN BROWNSTEIN: Thank you.

This is going to be a little bit of change of pace from discussions, technical discussions that we have heard in the last day and a half. You've heard mentioned reference to data bases, characteristics data base and spent fuel data base. What we want to do today is to show you what it is we collect, how we collect it and where we collect it and what we do with it, what stage we are in in the process of putting together what I would call a reference set of information that the researchers, the participants that you've heard from, use as a base of information and where we are going to go from here. That's the objective.

The data bases, there is really two data bases that the OCRWM program uses. I've listed up here three. The integrated data base is really executive or summary level data that represents a waste from a broader scale low level waste we'll get into from the lead sites. But it's really a document for the public. It's not the detailed data bases that are contained in the reactor-specific spent fuel data base and characteristics data base.

In the presentation today, we are going to have Rick Walling from PNL talk about the reactor-specific spent fuel data base and Karl Notz on the characteristics data base.

The spent fuel data base, it comes from the utilities, the owners and generators of the waste. And what we get from them is the inventories and projected spent fuel discharges. They provide us quite a wealth of information, the key fuel definition operational parameters, the type of fuel, burn-up, enrichment and items like that that Rick will get into in detail. And we get from them their projections which we dovetail with EIA projections so that both the utilities and we in projecting out the end of life all are singing off the same song sheet. Both the spent fuel data base and the characteristic data base are used by all the OCRWM and extended family of participants in the program.

Now the characteristics data base, what that does is really go the next step from the spent fuel data base. It provides the technical details of the waste that either will or may require geologic disposal. And that includes more than just the spent fuel. It includes the high level waste and miscellaneous waste. The data--and I'll show you a flow chart later, is compiled principally for the spent fuel from the spent fuel data base, but they compile information from fuel vendors, NRC, waste generators and lead sites. And they get into a great deal of detail as Karl Notz will describe of the physical descriptions, chemical compositions, radiological properties. Again these are the two data bases that form the reference set for the OCRWM family.

For completeness there is another data base that is widely used for the public. This is summary or executive level data from the integrated data base. And again, principally it's from the CDB, the characteristics data base, and the 859, but they, too, get some information, some summary level data from the lead sites. It serves a very important purpose. It makes sure and gives the opportunity for the DOE family including EM and NE to make sure that we are all talking about the same thing and to use for a planning base that we all know what each other is dealing with.

Unlike the characteristics data base and the spent fuel data base this is not OCRWM's alone. We are supported in this roughly half and half with EM. And there is a little piece of NE another part of DOE in there. So this is not our data base. It's something that we work with, we contribute to and it's again more for providing baseline summary data to Congress and the public when they ask questions as opposed to the details.

In terms of what we use the data for, all the program elements used the characteristics data base and the spent fuel data base for their planning decisions. And obviously in the waste acceptance process, both from the waste acceptance process you've heard for the last day and a half, the waste acceptance process, described standards at close of contract that we have with utilities, transportation, all the elements up to and including our fee adequacy analysis. So we do have this reference system where everybody is integrated and coordinated.

This gives you a real quick synopsis of how the data bases are put together. We get the data from the utilities for the spent fuel. This is the commercial spent fuel. We go through, do QA, reconcile information. This is a continuing process. In some of these double lines, there is analysis done at several levels and there is a double feedback to indicate that there are a number of checks and balances so if something looks anomalous they'll come back and understand what's wrong. That whole process Karl and Rick will get into in detail.

The input from the CDB, we've talked about the fuel vendors and waste generators, there is some additional, especially from the lead sites of other information from the TRU waste and low level waste that the characteristics data base really pulls from the integrated data base. Since the CDB gets into a little more detail or much more detail than the IDB, they will use that as a starting point and then actually go back to get some additional specific information.

In terms of answering questions, in terms of planning, we really use these two in order to answer the questions that come up. It's not a question of taking one or the other. There is a great deal of integration and coordination and communication between these two and really the questions that have come up, in order to answer them, you can't use one alone and we rely heavily on both to answer the questions in there.

In terms of where we are and where we are going, the reactor-specific data base is essentially complete for inventories. It's updated annually as new fuel is discharged. History is, it's about in a cooperative way since the mid-70's, but in 1983 after the Nuclear Waste Policy Act was passed we signed, I think you've heard reference before, this standard disposal contract with utilities, and one of the requirements in that contract with the utilities provide us information on something called a RW 859 form.

So the project that was started and evolved through a cooperative nature is now mandatory. We have held a number of meetings and continue to hold meetings with the utilities and discuss whether the data is--it started out as quite an imposition on the utilities certainly getting the inventories lined up on all the information we were requiring. We hold regular meetings with them to see if there is data that we don't really need, that we can stop; if there is more data that we need to add. We are in a process now of revising the RW 859 which is the principal input to the reactor-specific data base. We anticipate revising it by May, so, we are beginning to discuss things now to examine, since the last update three years ago, has the program determined that they need more information than is there. The utilities have been very cooperative in discussing this as long as we can make a strong case and understand the rationale, and to make sure that there are no frivolous data demands. And I think that's reasonable on their part and it's worked out pretty well.

We know already that at this next update that we are going to be concentrating on getting information on defective fuel and non-fuel components. That's something that we talked about the last time in the last revision. It's difficult to understand how best to put what we need and what questions we should ask in a way that all the utilities can answer them. And we have been working rather intensely for the last three or four months with them in trying to frame those in the best way that we possibly can. Those negotiations if you recall, were going out to our entire family now to ask about new data requirements that the program needs, especially in these areas we will get together with utilities in the next few months and hopefully propose a revision of RW 859 in May. That's a process that has to go through OMB to get the federal data form approved.

Now the characteristics data base that began in '83, understanding that we needed more information on the characteristics of the fuel than the utilities can provide us. It uses literature searches, input directly from fuel vendors and utilities. The data base unlike the spent fuel data base is incomplete in certain areas. There is a concern from fuel vendors and certain fuel vendors on proprietary issues. It's a concern that we are trying to get a handle on so we can acquire the information that the program needs. Karl will be talking about that, but it's a hurdle that's out there. And this is updated periodically and I believe there is another update coming very shortly.

So now what we do is go back and hear from Rick Walling who is the key person involved in putting together the reactor-specific data base development and then Karl Notz.

If there are any questions, I'll be happy to answer them.

MR. RICK WALLING: As Alan said, my name is Rick Walling. I work for Pacific Northwest Laboratory, and I've been involved with the reactor spent fuel data base for a number of years.

This data base is actually sort of a co-effort between the Energy Information Administration and PNL. They generally do the interface with the utilities and put out the data form to the utilities and do a lot of collecting, and we are involved in reviewing the data after they have gotten in and also help put it to use quite a bit with the various programs.

I think actually we've been hearing for the past couple of days why the spent fuel data base is important. It's the reason why we are here. We have this fuel to dispose of and there's a need to know how much of it there is and how much of it there's going to be.

The reactor-specific spent fuel data base is waste generator oriented. Primarily the commercial reactors contains inventories projections and associated site information and it's maintained and updated annually for the user base.

The primary source of data for the spent fuel data base is the nuclear fuel data form RW-859. This is, as Alan mentioned, a mandatory survey of the utilities conducted by the Energy Information Administration. The data submission is actually a report by exception. The utilities do not have to update the entire data base each year. They are asked to update existing submittals and supply new information. And the data is generally a snapshot in time.

As of generally the end of fiscal year--end of calendar year. What is the status, what fuel have you discharged and what have you put in the reactor, et cetera. In some cases it's necessary to get a little bit of data from other sources and alternate data sources include direct contact with the utility. The nuclear fuel data form does not collect every nuance of possibility that we might want to capture, and sometimes it's necessary to contact the utility especially if something they've submitted looks funny. And there's other data that's collected just from other available sources as needed.

Alan got into the evolution of the 859 a little bit. Prior to the Nuclear Waste Policy Act and the development of the 859, there were surveys that were done, but these were all voluntary. And most of the utilities were helpful and did respond, but it was not mandatory. Generally the data level requested was simplified to the extent since it was voluntary to keep it simple, and all that was collected was batch level data on the spent fuel, and there was really no cycle history. There were 20 assemblies discharged as of that date, but there was no detail back on how many cycles that had been irradiated beyond the final discharge.

DR. PARRY: When you say batch oriented, what do you mean?

MR. WALLING: In this case batch was just--I think a batch at that time was defined as a group of fuel that essentially went through and basically achieved the same level of burn-up. I think it was plus or minus say 5,000 megawatt days per metric ton.

Currently batches, at least as we have it in the system, as defined by several other parameters being the same fuel type and having the same irradiation history and quite a few other parameters. But at that time, for a given discharge jig you might end up with two or maybe three batches by this burn-up criteria.

MR. BROWNSTEIN: Let me add, where we are now, one of the complaints that the utilities were making is that we are asking for quite an amount of information for each specific assembly. And they were saying, why can't we handle similar assemblies that have gone through--you know have the same enrichment and have gone through the same cycle, why can't we just handle them in a batch. So we met and got together and defined those parameters such that every batch has the exact, identical characteristics and they can report once on this batch. The only thing individual you will get is assembly ID's. But they now no longer have to report thirty times for the one discharge. So that's how we have sort of evolved and that's what a batch is now. We'll have specific assembly ID's but they will have the same exact characteristics.

MR. WALLING: Okay, come calendar year 1984 the original

RW-859 went out and this first 859 was an annual update by all of the utilities. As of the end the calendar year, the utility got the whole data set that they had submitted the previous year back to them and were asked to verify all the data that was there. Again it was set up in a computerized form so they didn't have to re-enter all the data. They could just verify it was there and input new data. But, they were essentially asked to sign off on all of it.

They submitted assembly-level detail on the fuel. There was a cumulative burn-up reported at the end of each operating cycle on each assembly of fuel, although this was a non-mandatory request, but probably 90 percent of the reactors did comply with that.

Future discharges in this initial 859 were predicted all the way to the end of the reactor life as projected by the utility. And after three years of using this form it was felt that the historical and kind of static information, things like crane capacities and dates of when the reactor started to operate or whatever some of this information didn't have to keep being re-submitted each year and was well-established.

So then we got to the current form as it exists today. There is now an option for the utilities to either submit this data as an annual update or a couple of months following their restart after refueling they are to submit the data. Approximately two-thirds of the reactors use the annual update option and the other third submit following the recharge.

The information collected on the 859 has now been really reduced down to the set of data that is needed to keep the data base up-to-date. And that is primarily information on new fuel that has gone into the reactor, fuel that is being re-inserted into the reactor and an update on their--they are now projecting their future discharges only for the next five operating cycles so there is an update of that. And there is an update of any fuel discharge from the reactor at that time.

The new data submitted is merged with the existing data base by EIA to create a complete data set. And as Alan mentioned we went from early submittal to batch-level data to assembly-by-assembly, very detailed submittals. And now it's gone back to a little more of a batch-level orientation. Although an assembly-level, a data record for each assembly is maintained in the EIA, but it contains kind of a batch average data for the assemblies that that pertains to.

Also the burn-up on the fuel is only collected at the time of discharge of the fuel from the reactor. If the fuel is reinserted and comes out a second time we would have a second burn-up point. But, we would not have--if a spent fuel assembly was burned in cycles one, two, three, taken out of the reactor and then reinserted for cycles five, six and seven, we would not have burn-ups for one and two, we would have it for three; we would not have it for six, and we would have it again for seven. But if a similar one burned in cycle one was taken out and only burned in cycle five and then taken out for good, we would have a full burn-up history.

Okay, this is a brief overview on what kinds of data we have in the data base. Generally there are four areas; reactor characteristics, some information on spent fuel storage, there's characteristics on the spent fuel and then there are some subsets of data on the operating characteristics of the reactor.

On the reactor characteristics, this is fairly--this is kind of some of that static information that I was talking about earlier. Things like the name, kind of an ID that we put on the reactor to keep track of things. What is the reactor type? Who was the reactor vendor? What utility are we talking about? Where is the reactor located? What are the power ratings? Dates related to when the reactor started up, when they entered low power testing. When are they planning on shutting down and retiring? When does their license expire? And, some data regarding fuel handling capabilities such as crane capacities and a few other things.

DR. PARRY: Do you also indicate whether access is by water or by train or truck?

MR. WALLING: Yes, there is some of that. That is information that was collected with the original 859 and that is not currently being updated via the 859, but there are other studies going on within OCRWM to continue assessing that data and actually add to that data base.

On spent fuel storage, in order to kind of have an idea of how much the reactors are getting into trouble with time, as they continue discharging fuel, we need to know pool capacity and things. You know, when they will probably have to start using dry storage if they haven't already. So we need things like pool capacities and pool configuration data. There's a little bit of information on dry storage capacities or what dry storage they are using. And also, the utilities each time they report now do report an accumulated inventory by contributor. There are a few reactors that have transhipped fuel back and forth and we get kind of a baseline number for a reactor for how much fuel it is holding from another site.

On spent fuel characteristics, on every assembly that is out there, there is an assembly identification and a batch identification. The cycles irradiated, which cycles the fuel was in the reactor is maintained. There is a fuel vendor and assembly type ID in there. That's a key element because that is a key field to interface with the characteristics data base since it maintains data on the physical characteristics of various fuel assemblies. This is the key to get us in to merge the inventory data with the characteristics data.

Also we pick up the initial enrichment and the heavy metal content of the fuel, the initial pre-irradiated heavy metal content and the burn-up.

In addition to the data I just said which is in for all assemblies, special characteristics that are picked up as they are reported, the fuel is identified if known by the utility as they report whether it has a defect or not. Any modifications that have been done to the fuel as if it's been reconstituted or reprocessed or something like that. Also if the fuel has been--there's a few assemblies or a few reactors out there that do shuffle fuel around between their reactors, so if the fuel has been irradiated in more than one reactor, then there is a field to pick that up. And also fuel that's been sent to an offsite location, then we also have identified where the fuel is at and when it was shipped and how it got there. And in some cases we know the cask type and how it was shipped.

Okay in operating characteristics, this is a fairly small set. We have a cycle ID and the shutdown date for the cycle, and the effective full power days during that operating cycle. The cycle shutdown date is the date that is used to help assign a date of "discharge" to the fuel for purposes of waste acceptance or age of fuel that it is in the repository or whatever.

Well, the data that is collected on inventories is good, but to date that represents only about 19,000 metric tons that we anticipate is going to be somewhere around 80,000 metric tons or so through the end of all the existing reactors' lives. But as I mentioned before, the RW-859, the utilities only project five cycles out in the future and that only gives us data for the next five to ten years, depending upon what cycle length they are operating under.

So there are also some projections that are done. EIA takes the first step and does some aggregate spent fuel projections just on general energy projections and percent of nuclear growth and percent that nuclear is helping generate electricity in the U.S. and such, and this eventually ends up creating a projection for the amount of spent fuel that will be produced in total. That is merged with the utilities projections of what they will do over the next few cycles and also merging that with a few other pieces of information like an assumed capacity factor which is somewhat based upon historical data. We end up with a data base that gives us a data base for reactor by reactor guesstimate of what fuel will be out there in the future.

This is not so much important for this group, but it is important for the people doing systems engineering that like to have a semi-realistic data base for predicting things like cask fleets and logistics and such.

The no new orders case with increased burn-up is used as a planning base, but EIA projects a number of different cases and there are high and low energy growth and burn-up variations that are used to create sensitivity data bases which are also used.

These forecasting assumptions are reviewed annually and there are new data bases that are created annually, so we are in constant move.

And then this is a list very similar to what Alan showed near the end of his talk. The reactor-specific data is used by a lot of people for a lot of different purposes. We've heard about ATM's that the Material Characterization Center has been providing and collecting. They've done some work with the reactor-specific data to--well they are now looking at whether those ATM's are representative within the population. In the past there's been some looking at items like what is the population of say the stainless steel clad fuel and can we tell if that is a significant enough piece to need to acquire an ATM for that purpose. So there is some use by the people that we've been hearing from here for this data base, but it's also used for a lot of other purposes like systems engineering and such.

That's the end. Are there any questions?

DR. PRICE: Do you have in your data base on fuel handling, you indicated crane capacities, other limiting features like infrastructure features, clearances, roadbed limitations and these kinds of things in here?

MR. WALLING: Not in this data base. But there are other--there have been some programs at Oak Ridge that are more tasked to do that. There are also some ongoing studies to go to each reactor and try to figure out what capabilities they have or may be able to have if they or somebody else, if they were somehow upgraded. Say, how far are they away from being able to accept a rail cask as opposed to having to use a truck cask.

MR. BROWNSTEIN: We've gone out and we have something called the FICA study, the Facility Interface Capability Assessment and that is sort of within the fence assessment of not only what is there but with some minor modifications of what could be there. And there is another study that looks at the same type of considerations outside the fence, roads and so forth, clearances to get there, to see how much flexibility we have.

DR. PRICE: I think we are aware of both of those studies. But these haven't been integrated into your data base?

MR. WALLING: No, not at this time.

DR. PARRY: On your last two slides, you did not indicate the possibility of license extension. Have you considered that yet?

MR. WALLING: Oh, thank you. The standard--somehow it got missed. I'm sure it was on there once upon a time. The standard assumption that was made with the projection data is that each reactor with few exceptions will operate for forty years from the start of their operating life.

The few exceptions are the reactors that have shut down or have indicated very strongly that they will shut down in the near future. Ranch Seco, Ft. Saint Vrain, those are recent candidates.

DR. PARRY: Peach Bottom-1.

MR. WALLING: Peach Bottom-1. But like I said, the standard assumption is that they will operate forty years. I'm not aware--currently there is an extended life case beyond that point, but there have been in the past. There has been sensitivity cases at some of these points.

DR. PARRY: It certainly might have a real impact on the capability of any site to handle all the fuel.

MR. WALLING: That's very true. Any other questions?

All right I think our final speaker for the day is Karl Notz and he will be talking about the characteristics data base.

MR. KARL NOTZ: Good afternoon. My name is Karl Notz. I'm the task manager for the waste characteristics data base. And, I appreciate this chance to tell a little bit about the data base.

The physical evidence of the characteristic data base are these eight volumes here which were printed in two printings, six volumes in December of '87 and two volumes in June of '88. These were backed up by five PC data bases. There's a demo disk inside the first volume to give the users an idea of what's in the data bases. Those data are collected on 48 of these 5 1/4 inch floppy disks. There's roughly five million pieces of information in there. And the eight volumes contain maybe two percent of that.

In addition we use special studies and technical studies to provide additional backup information and set the stage for improvements and those have been published as ORNL reports.

The objectives of the characteristic data base are to provide a single unified source of data for use for the federal waste management system, including all those parts of it that have been mentioned by both Alan and by Rick. And to provide the detailed characteristics of those wastes that will, or may require geologic disposal.

The scope is to include those things that may require geologic disposal and that is obviously LWR spent fuel, immobilized high level waste, non-LWR spent fuel and miscellaneous waste which is largely greater than class C low level waste, which is not acceptable for shallow burial.

We provide data in these categories, the physical descriptions, the dimensions, the weights, the method of assembly; the chemical compositions both in terms of both total elemental content, and where we have the information the chemical composition of it because those are important in determining the behavior. The radiological properties represent probably 80 percent of the data that we have and that includes of course the thermal power, the gamma, and the neutron radiation. We have it here for the individual nuclides as well as collectively and the integral heats. And of course inventories and projected quantities.

Just to give you a rough idea of about how much we are dealing with and what kind of peak load we have, this is of course a very summary type of presentation. It involves a lot of assumptions, but it indicates clearly that you do have largely--and this has been reduced to canister to have something we can count. So this is the largest amount, but the immobilized high level waste is also a significant quantity. The non-LWR and special case fuel, although they are a smaller quantity because of special problems with them, and then the greater than class C waste has a potential to be a large contributor. The LWR spent fuel is going to be the source of most of the heat, but the others are not insignificant. And you'll notice also there's a rather wide range, wider ranges of uncertainties in these heat loads here.

Spent fuel of course had a wide range of heat load also depending on the burn-up, but they are going to package those in such a way to help levelize that out. But, that's a separate topic altogether. That's just not our task to deal with that.

DR. PARRY: Excuse me, your watts per canister, does that take into account decay?

MR. NOTZ: This is--of course decay is an important part of all of this, and this represents a nominal level at the time of implacement so you recognize is it five years old, is it 15 years old? This is kind of a levelized number if you would take everything you have, you would average out to something like that. Some canisters will have more than that and some will have less. And of course, when it decays, that will drop since the bulk of that heat is going to be from your short-lived fission products. After 30 years of age this number will drop down almost in half.

DR. PARRY: How about on the immobilized high level waste, does that include the strontium and cesium capsules?

MR. NOTZ: Yes and no. I'm not quite sure how they are going to fold those in or what they are going to do with it. If they leave them as a separate entity and create a special package for them, they are going to be limited because those generate so much heat that you cannot fill the canister all the way up with them. You'll have too much heat.

There is some consideration that they may open those up, recombine that material with other high level waste at

Hanford and put it into the glass wall (sic). The benefit is the glass wall is a little character out of waste form. Whereas the capsules, well we know some of them are leaky right now, we are going to have to invent a special waste package and it may not be economical to do that. It might be cheaper just to recombine them.

In summary then, we have extensive properties, the inventories and projected quantities. We have the intensive properties, the physical descriptions, the chemical compositions, the radiological properties, and then we also have what I call exception properties. These of course apply to everything (indicating). These exception properties only apply to some things. And defective fuel is obviously one such category. The most obvious special fuel is TMI rated fuel which doesn't even look like LWR fuel anymore.

And then of course, the non-LWR spent fuels are such a wide range and types of fuels out there.

For the LWR spent fuel, this is my flow chart of the data flow. We receive data from the EIA and PNL on inventories and on projections. And this also includes the data that we are supplied by the utilities, which we have to have to do our work. We have to have the initial enrichment. We need to know the burn-up. We need to know the heavy metal content. We need to know the assembly type and how many them there are. We also need to have detail data on those assemblies which we have obtained from the vendors with the exception of GE and we do it ourselves in that case. With this information we now can provide physical and chemical descriptions and inventory projections.

To get the radiological properties, we take the appropriate data and run it through the ORIGEN2 code which draws upon PWR and BWR models and nuclide libraries, and do that calculation and see that it's available to all those users out there.

We printed a thousand copies of this and we are down to 200. It went out on category distribution; it went out to special distributions and we answered requests for it and we probably have 150 copies left right now that we will probably send off to various universities and educational institutions because we are now in the process of doing a revision on this. And I will close my talk by talking about some of the improvements that the revision is going to have in it.

When we did the first set we had 78 assembly types out there. That was confusing. As we have continued our work over the years, we are now up to 126 assembly types and there has to be a systematic way of categorizing these descriptions, otherwise, it is too difficult to deal with. So the basis that we chose to simplify all those assembly detailed descriptions was what we call classes, which are based on reactor core configuration.

Once the reactor has been built and the core designed, then that fixes the length, cross-section and in the case of BWR, the array size. The BWR's have cruciform so you can use different array sizes. And there are seven BWR classes and 16 PWR classes. In each case we have the generics, in otherwords there are several and in some cases many reactors of a certain type. And then again for both of them we have a number of one-of-a-kind. And these are the early reactors of which there was only one built and then the design was changed.

Now within these classes, you can break it down further by the model type, each has it's own design. And for a given vendor they change their design as time goes by. They find ways to minimize defective fuel and improve neutron utilization and things of that sort.

This is just one example and you've seen this before. Ray Stout showed you this graph earlier. These are those 23 classes. There is only 22 on here. Pathfinder is not shown. The heavy bar is what we had of December 31, 1987. The open bar is what's projected out to 2020. And Rick gave you what the ground rules were for that. So, these are all BWR's, the bottom part is all PWR's and you can see the generic classes have by far the lion's share of the spent fuel that's out there.

We calculate the radiological properties using

ORIGEN2, a code that's been around for a long time. And in order to do that for spent fuel, we must know the initial enrichment, the burn-up and the discharge date. And we do that for each batch. Cycle data, we have used generic cycle data for BWR's and generic cycle data for PWR's, but this is based on data supplied by utilities and accumulating and analyzed by EPRI.

For immobilized high level waste, we need to know the composition and the canister fill date. We just arbitrarily said we are going to put the high level waste data out on the current projected date of filling the canister, which may or may not be realized, but still of in the future a little bit. But in the case the spent fuel went to discharge, that's the fixed point and we go forward from there.

One of the important things that we've learned in the last year is that the enrichment is a very important parameter in doing the ORIGEN2 modeling and it must be handled correctly. So we do have the data available batch-by-batch. We know what the enrichment was. We know what the burn-up was. And this is a graph of those data. The solid circles are 80 or more assemblies in the batch. The open circles were less than 80 assemblies per batch. So you can see it does group. We did the regression analysis and got the relationship. So when we do a calculation--now, we did it at 10, 20, 30, 40, 50, and 60 up to 50 for BWR's and up to 60 gigawatt days for metric ton for PWR's.

We used this statistical relationship between the enrichment and the corresponding burn-up. As you can see though, a lot of fuel isn't on the line exactly. And we determined that by going plus or minus 7/10ths of a percent we included virtually all the fuel. So in our data base we have the calculation not only for the center line, but also for the two extremes and then we have built in an interpolation function which allows you to interpolate on enrichment or also on the burn-up, if you wish, to pick up any point in there.

This is a similar curve for PWR's and you can tell that it's not quite the same as the other. It doesn't have as much curvature and doesn't flatten out quite as much. That's one reason why we have to have separate models for PWR's and BWR's.

So these radiological properties then include the individual nuclides. The code actually works on the gram-atoms of individual nuclides. That's the basic unit that it works with. And it groups them into fission product, activation produce and actinides. And the data output includes the alpha emissions, the beta-gamma emissions, and these are broken down to 18 energy groups. The neutron emissions, and it breaks it down by spontaneous fission and alpha-N neutrons, and of course the thermal output and the integral heat.

Here it gives one example. The amount of radiological detail of course is very extensive, but I know there is interest in C-14 for Yucca mountain, so we pulled together this particular view graph. This is on the basis of a metric ton of fuel whether it's a PWR or BWR. And it shows you that the Carbon-14 is found in the UO_2 fuel. It's found in the cladding. It's found in the assembly hardware that holds the pins together. In the case of BWR it's found in the channel. There is also a little bit in that plenum space and there's a little bit in the crevice on the outside.

The important thing in this chart, I think, is the fact that the Carbon-14 doesn't come from the fuel. It comes from the nitrogen impurity, by and large. In the case of the crud, we are assuming it comes from the activation of the oxygen that's in those oxide forms that are on there. But we've had to make some assumptions as to the nitrogen content of the fuel and that's probably a pretty good number. And we've used some pre-existing data here for the nitrogen content of the zircaloy and the inconel and stainless, and these maybe--probably are on the high side. And that's an area where we need to do some more work.

Although these numbers here for the plenum and the crud look small, these are very accessible fractions. As soon as you penetrate the container, the crud is on the outside of the container and it's available. Or, as soon as you penetrate the cladding then what is in the plenum space is available.

Okay, referring back to our characteristics report, it's a lot of material here, but the first two volumes include a summary plus those four major waste form types that we talked about. Everything else that is in there is to back that up. Some of this is additional detail and some of it is user's guide for the PC data bases. These are all supporting appendices of one form or another.

We have five PC data bases for spent fuel and one for high level waste. The ones for spent fuel, one deals with the quantities that have been discharged, and that also includes the projections and it also includes detailed descriptions of the hardware that hold those assemblies together and that go with it.

That is just the quantities area and it includes data on defective fuel, for example. It also includes the identifiers that we have used collectively between PNL and EIA and Oak Ridge. We use a common set of identifiers for assemblies. That's identified in here also, so you know not only how much it is and when it got discharged, but what kind of assembly it's in.

The assembly data base gives a detailed description of individual assemblies; these 126 types I mentioned including the hardware that hold them together. And this also includes the radiological properties of that hardware. Because, in the hardware you have only a few activation species which appear.

The spent fuel however has hundreds of the radioactive species. The radiological data bases for the spent fuel itself is on the basis of metric tons of contained heavy metal.

The non-fuel assemblies hardware data base--there's two kinds of hardware out there. It's what we call spent fuel disassembly hardware. If you leave the assembly intact, then the assembly hardware stays right with the assembly. But if you consolidate, then that hardware is separated and that's why we call spent fuel disassembly hardware.

On the other hand, the non-fuel assembly hardware which is largely PWR control assemblies and BWR control blades, that is a separate item even if you do not consolidate. So we have a separate data base for this which includes both the quantities and radiologic properties. And we have a new one which is not in these documents but it's in the revision we are now working on and that is the serial numbers.

Looking ahead to the day when a accountability is going to be a requirement, the serial number data base will allow you to go by individual assembly serial number and go back to how much it represents, what its properties are, its radiological properties are, so that everything can be tracked back in a unique manner.

For a high level waste, we have data on both the interim form because after all that's going to be the parent of those canisters and then on individual canister basis.

All of these data bases are menu-driven. They are user-oriented. They have gotten a good reception from the people who have used them. Most people don't even have to refer to the user's guide in order to get them up and running.

Here are just a couple of very simplistic type examples of the data that we've extracted on an assembly class basis and I've picked on fuel rod diameter here and to show you some of it. And there's some interesting things to show up here. For example, within a given vendor, as he goes to a later design, the diameter gets smaller. It's happened here with B&W and it's happened here for Western Engineering. It's happened here for Westinghouse within theirs. The newer designs are going to smaller diameters.

And the same thing is evident here in the GE fuels. As you go to newer designs you go to smaller diameters. In the case of GE, the same basic fuel design can be used in both of these classes. The primary difference is that Class 4, 5 and 6 is about six or eight inches longer than Class 2 and 3.

We were unable to acquire details on GE assembly from them. They treat that all as proprietary. So we did a study referenced here at the bottom in which we got NRC docket type information and literature publication to get the descriptions for the GE fuels. One thing we do not have in detail though are their burnable poisons and their enrichment patterns. BWR fuels in particular run through a more complex loading pattern within an assembly than do the PWR's.

There was an earlier question about how much variation there is across the core. And it's true within the core they will rotate those assemblies and move them to different positions to balance out the burn-up and to keep them from overheating on the first cycle. But within a given assembly and this is especially true for BWR's, there is a wide range in the BWR where you have the fissile form coming in. You have those four corners which is different from the outside corners. And in this one example, the enrichment can vary for a given assembly between 1.3 and 3.3 percent. As the enrichment varies you also vary the burnable poison content and you will vary the burn-up within those individual rods.

The data we have are assembly average burn-ups. We do not have individual rod data. We do have individual assembly data, but not individual rod data. I hope we don't ever need it again.

DR. PARRY: Are there variations longitudinally?

MR. NOTZ: Oh, of course there are. If you take a look at the burn-up curve, it bellys out and both the top and bottom ends slack off considerably.

DR. PARRY: In loading or just in the way it burns?

MR. NOTZ: In the way it burns, because the neutrons fall off.

DR. PARRY: No. You indicated that you had a variation in enrichment across an assembly. Do they do any adjustments longitudinally?

MR. NOTZ: I think the BWR's do. To my knowledge PWR's don't do it yet. But it is probably a future refinement that may be coming, I don't know. But again the burn-ups that we have are assembly averages and the ORIGEN2 code is designed to give you an assembly average. It recognizes that there is a distribution of neutron flux within the core both radially and longitudinally and that's been accounted for in constructing the model.

DR. PARRY: Such variations will play hell when it comes time to reprocess if you do.

MR. NOTZ: They could.

Okay, the hardware is becoming increasingly visible as something to be concerned about. A report on the hardware has been put out by PNL and the concern of course is if you consolidate then you've got all this spent fuel assembly hardware to dispose of. And some of it is going to be greater than class C. And it's not real clear how much of it might be. So he took measurements on three different assemblies and took samples from three different assemblies, measured 12 different areas, divided it up into zones and also did ORIGEN2 calculations for comparison. And just in summary, we have divided the total assembly into the top end fitting region and the bottom end fitting region and then the glass plenum region which is unfueled and then the core region which is the fueled part. And taking samples from all these areas and looking at a dozen or more different nuclides, the factors if you take the core region as being 100 percent activation, this is on a per gram basis of whatever it is you are looking out, whether it is zirconium or niobium or whatever, then in the plenum region is 20 percent of that and the top end is 10 percent of that and then the bottom end is 20 percent for PWR and 15 percent for BWR. It would take a fair amount of experimental work to refine those numbers much better than that.

The non-LWR spent fuel is a concern for a number of reasons. And again we have published a report that is referenced here on this. The major contributors here are HTGR reactors, Ft. St. Vrain and Peach Bottom-1, and also a special fuel is a degraded LWR fuel from TMI-2. And you can see that here we have represented most of the contributors. Some of these are unique in the things they--I'll get that in a minute, about your special properties.

The other contributors here are the LWBR, the FERMI-2 Blanket (sic), the TRIGA research reactor and then collectively a lot of research and development type reactors.

Now these are of interesting concern because they are different from LWR fuel. Their chemical composition is different. They might be carbides or they might be metals. Their cladding is different. There might be silicon carbide or it might include sodium bond. Their physical condition in some cases is quite different.

Going back to this chemical composition for those HTGR's, they are embedded in a graphite matrix. Many of them because of their nature are high enrichment and low burn-up. This causes us to be concerned about the criticality. So we did address the criticality issue in this study.

And then finally some of them contain thorium and/or U_{233} so you are going to wind up with some U_{232} which has some rather nasty radiation properties of its own.

Because the ORIGEN2 code is such an important part of our work and the radiological data are so crucial to all this work, we did a sensitivity study, and we did use a new cross-section set that had been developed and published. We wanted to find out the relative importance of some of the variables that you can deal with in setting up the models. It turned out that enrichment was by far the most important one and so in our re-issue here, we have dealt with enrichment explicitly, as I just pointed out. It had a major affect on the actinides. And the actinides in turn are the source of your neutron emission. They are what contribute to criticality and it also has in fact on some activation products.

Things which had a minor impact, fission products are dependent almost entirely just on the burn-up, so they were not affected by enrichment. We tried running at different power levels and that changed the number of days it took to get the burn-up, but it didn't have much affect on the output, particularly when you are looking at fuels of more than five years old.

Also the decay time does not influence our interpretation of these results, again when you are looking at fuels over five years old.

Okay, we just completed a draft of the first revision of five volumes of our data base. The other three volumes will come out next fiscal year. The major improvements that we have in this revision is that we have improved the classification scheme. I've told you about that. We have additional data on LWR assemblies, especially the GE and I've told you about that. We have revised and improved our radiological data and we've just talked about that. I've mentioned the interpolation functions. The integral heat allows you to pick any starting time and any stopping time and to calculate what the integral heat is over that total time period.

We've added the serial number data base. We have these new improved factors for activation of the hardware. We have added to the assembly data base a fuel pin data base which is a sub-set. And if you do consolidate, just allows you to subtract the hardware away from the pin.

We have an improved way of doing the neutron source strength for high level waste data. Some of the old numbers were too low. And then in all of the PC data bases, we have taken advantage of comments from users and upgrading of software to improve the interfaces of those data bases for the users.

That concludes what I have to say. I'd be happy to donate these eight volumes to anybody who wants to take them back with them or I would be happy to mail them to you if you would rather get them that way.

DR. DEERE: No, we'll take them now. We'll have them boxed and we'll ship them back.

MR. CLONINGER: For the record, would you note that Mr. Notz gave the board a whole set, volumes 1 through 8 of DOE/RW-01.

DR. PETRIE: Thank you. That concludes the prepared presentation. We have time now for some closing questions. If there are any additional comments or questions we can take those now. Or in fact I guess that concludes at this point.

DR. VERINK: I think the time would be particularly appropriate for any questions from the panel that might cover any aspect of what we've talked about in the last two days. And when we get through with the panel's questions, we'll open this up for questions from the audience.

Does anyone from the panel have any particular point to raise?

One question that has kind of come up is considering the budgetary constraints and one thing and another that crunch on everyone in various ways. What priority do you feel is or should be given to the container development kinds of things that have been discussed?

DR. PETRIE: Well, I don't believe there is a specific priority given to it. I can just state that it is lower than the site suitability issues.

DR. VERINK: Does that mean that it will be worked or does that mean that it probably won't be worked?

DR. PETRIE: In the present budget--where we are today with the present budgets, I would say there will be some modest amount of money provided or resource provided for those activities. It will very likely be less than last year.

DR. VERINK: Will that option be exercised by your organization or will it come from Washington, or what will that be? Where's the pinch from?

DR. PETRIE: The general frame work is laid out by Washington. Within that we'd have some flexibility, but basically I suspect this is a policy decision made at headquarters level.

DR. VERINK: So if there were to be any alteration that would be the place to talk to?

DR. PETRIE: I would think so, yes.

DR. DEERE: This might be a right topic to bring up at the strategy planning meeting in a few weeks to be held in San Francisco.

DR. PETRIE: Let me just ask Max, are you familiar with a strategy planning meeting that's coming up?

MR. BLANCHARD: No, I have not heard about it.

DR. PETRIE: Okay, so I hadn't either so--

DR. DEERE: I won't say anything more about that.

DR. PETRIE: So, you've probably hit the right place.

DR. DEERE: Well, I think one of the reasons that Ellis brought up the question is that we see the workload that you have laid out and particularly in many of the different presentations, but specifically in the first two that we had, where you are doing the flow down and traceability for the source of requirements for the waste package design alternatives. And obviously there has got to be a great deal of work done on that. There hasn't been a lot done to date. At least we haven't seen a lot presented as yet. And we just are wanting to know what the time frame that this kind of assessment is going to be able to be brought forth.

DR. PETRIE: Can you give us a minute to caucus?

DR. DEERE: Sure.

DR. PETRIE: I would like to repeat something which Max said yesterday maybe in response to that to put it in perspective. And I think the numbers that he gave you yesterday were we were planning on getting something like $275 million for the project next year. And our best guess--you didn't say that? I thought you did.

MR. BLANCHARD: If I did it was a mistake. To the best of my recollection which may not be all that accurate the most optimum case for FY91 is $172 million. And the current spend rate for the project is $194 million. That is not doing any new site characterization work.

DR. DEERE: On page three of the transcript--

MR. BLANCHARD: That's only doing the monitoring work, like seismic monitoring and meteorological stuff that you've seen when you've been out to the site. So there are no new starts there.

So, the expectation was if the permits were lifted or we got clearances and we could start, then we would do everything we could to focus our money in initiating new site characterization work so that we could focus on early identification of potential disqualifying conditions, should they be at the site.

DR. PETRIE: The $275 million I mentioned was what we would expect to have if we were going to maintain a schedule like '92 for start of ACD, which is what we are talking about here. That's my recollection and I'm sure you will quote me, but nonetheless it's only a recollection. It's not a guaranteed number. Our best expectations, most optimistic expectations for next fiscal year are less than what we would hope to get to carry on our program with that--some program.

If that helps you at all, and I hope it does, that's about the best I can give you at this point.

DR. VERINK: One of the thoughts that's been going through our mind as a panel member, is that--well, we were very pleased and impressed with the kinds of discussions that were made about how to make the canisters or containers more robust and so on in the improving climate with regard to receptiveness to such an idea on the part of some of the regulatory people.

And so we were thinking it might be very timely to have a workshop which would deal with the development of the robustness of the container and fleshing out in some more detailed terms how the combination of environmental control and materials could work together to accomplish this. And we were thinking that if we could shoot for something like in the middle to late January time frame, that might be a kind of thing to shoot for, which could keep the momentum going in this very important area. And perhaps focus some plans which could be very helpful.

DR. PETRIE: In response to that I would think that we would like to confirm that with you at some later date. I don't think we could commit to that today.

DR. VERINK: I understand. But I thought you should know the direction we are thinking and get your reaction to it.

DR. PETRIE: Well, let us consider it and get back with you.

DR. DEERE: Because we do know that one of your early parts of work on this program is to set up the requirements for these alternative systems, because the requirement for one system might be somewhat different than the requirement for another. And we felt that during a workshop, these are the types of things that could be discussed in an informal sort of way.

DR. PETRIE: Oh, yes. I think a workshop would be in order. The question is the timing of it.

DR. DEERE: Yes.

DR. PETRIE: When would be an appropriate time to do that.

DR. DEERE: Yes.

DR. NORTH: I'd like to reiterate the question that I raised yesterday about the relative lack of emphasis on performance assessment in the first phase. The phase that you are really in now. This is on your flow diagram which is page

12 in I believe it was the first presentation that Les Jardine made.

And my reaction from the point of view, the risk and performance analysis panel which I chair is we've heard a lot of very interesting science and engineering in the last two days, but with the exception of Dr. Jardine's second presentation, there was very little attempt to focus and prioritize what it is that you are really going to need for performance assessment. And it seems to me that one can focus that at the level of what is most critical for the design concept that is your reference case right now, but that the same issues apply perhaps with different priorities to the alternative of an extended life canister which many of us on the board are very interested in, seeing further worked out the ideas for the alternative that Les Jardine presented. And it seems to me that a workshop might well take as part of its emphasis, trying to get a first cut at some of these prioritization issues.

Now you said in your waste package plan, page 317, that you are going to do it, and you even quoted the language from that paragraph. And I think we'd all be very interested in seeing just how it is you propose to do it, to set the priorities and how you might do this against various levels of your budget that you might have in the coming year.

DR. PETRIE: I'm not sure I know how to answer that.

DR. NORTH: I didn't expect that you would.

DR. PETRIE: I do see Les taking notes galore here.

DR. VERINK: Are there any questions from the audience? Would you please come to the microphone and give your name for the record.

MR. PHIL NIEDZIELSKI-EICHNER: Good afternoon. My name is Phil Niedzielski-Eichner. I'm here representing Nye County, Nevada. Just a couple of questions. One I'm afraid is a follow-up on the theme that the board members were just asking about. And that's related to process on the alternative design approach for waste package and engineered barrier. The question would be how and when would the programmatic policy requirements be identified and be integrated with the technical requirements. Dr. Jardine made a point of distinguishing between the two and my question is how will those two be integrated and when?

The second question related to that is the methodology for the selection of ranking for the acceptable design solutions, how will they be developed. And what is the likelihood that decision ranking methodology will be tool for that purpose?

The third question is related to the greater than class C in a waste package plan. I think also on page 317 it's indicated that no efforts will be expended in pre-ACD for developing detailed plans for accommodating lower than class C waste. And I'd just like to have some elaboration on the reason for that.

Thank you.

DR. PETRIE: Did anybody get that? I'm afraid there were a lot of questions there.

DR. VERINK: Maybe if we could have them one at a time.

MR. NIEDZIELSKI-EICHNER: Okay. I just wanted to get them on the record for you.

DR. PETRIE: They will be on the record, I suspect. Is that what you were asking? Would you like to have a response at some later date? Is that what you are looking for?

MR. NIEDZIELSKI-EICHNER: Let's just take it one step at a time. In terms of Dr. Jardine's distinction between programmatic policy and technical requirements, there was a lot of discussion on how the technical requirements might come into play, but what is the process for how the programmatic policy requirements will be integrated with the technical requirements.

MR. CLONINGER: Well I don't know if I can totally answer your question. Regarding the when, we have addressed that. It's a function of priorities and budget. The non-technical aspects of requirements flow down will have to be addressed through some decisionating methodology yet to be identified for this part of the program.

Does that cover your first question and it does cover your second question.

MR. NIEDZIELSKI-EICHNER: From a time standpoint. So you are suggesting it's really a budget-driven issue.

MR. CLONINGER: Partly, yes. Max?

DR. PETRIE: It's budget to address the timeliness but not whether or not it gets done.

MR. BLANCHARD: This is Max Blanchard with DOE. Although it's clear, like Mike mentions that timeliness and budget factors enter into this, I think when you look at the regulations that govern the program that had been in existence over the last ten years, it's clear that there is some very high level policy issues that were established in the process of originating those regulations. And those, although may have encompassed some technical aspects, they were basically strategies.

For instance an example of a strategy is primary reliance on a natural system and not allowing an engineered barrier to compensate for deficiencies in natural barriers. That's a judgment call on the part of those who are involved in creating the regulations. And there is no way for the Department to do anything except to try to build a program that demonstrates compliance with that strategy.

In a similar fashion, the length of time the repository is to remain open for retrieval, that also is a policy call on the part of those who are creating the regulations. They are asking for comments from other people as of the time they create these regulations, but someone has to take the first step. And at some point in time the first step was taken back in the early '80's when NRC started after EPA was involved in setting up 40 CFR 191.

I think one of the reasons why this board was formed was to look not only at the DOE technical program but also to attest to the viability or the reasonableness of some of those policy calls that were made in the early regulations.

To that extent, I think the topic of a long life waste package is right for discussion. And I would suggest that the NRC would be just as interested as the DOE is in looking at that aspect, even though the current program is geared towards emphasis of the natural barriers.

MR. NIEDZIELSKI-EICHNER: My question is down to one of process and sequence. When you look at the discussion of taking the systems approach to looking at alternatives and you have recognized the programmatic policy of requirements that come into play there, can you enter into a systems approach without--if you readdress policy issues, can you enter into this systems approach without first looking at those or somehow setting up a mechanism for getting those integrated with your process? That's really where my question comes in.

MR. BLANCHARD: I think the systems approach accommodates that quite well. And as far as we've been able to deal with that in our existing decision analysis methods that are applied to the ESF alternative, Calico Hills risk benefit and the surface base testing prioritization activities this year, we have tried to encompass an aspect of this policy judgment, which in some ways is geared towards trying to build confidence in the other parties that are involved in this program including the public that aren't up to the technical aspects.

MR. NIEDZIELSKI-EICHNER: Nye County has just submitted a letter to Carl on the question of outside or external or independent involvement in that type of a process because of the public confidence issue.

The other question that I had was this one of the waste package plan and the greater than Class C consideration.

MR. CLONINGER: Okay. Would you like to repeat that question for me.

MR. NIEDZIELSKI-EICHNER: Sure.

MR. CLONINGER: Thank you.

MR. NIEDZIELSKI-EICHNER: I was just looking for an elaboration on why that waste element that 10,000 canisters that Dr. Notz would indicate was not considered--will not be considered during pre-ACD?

MR. CLONINGER: Oh right. The main reason we haven't considered or planned for disposal for greater than Class C waste is we don't know what that encompasses at this point.

The waste stream is not well defined at all in terms of amounts or radioactive content or chemical form. As I mentioned in my second talk yesterday, the Environmental Restoration and Waste Management office is going to issue this calendar year, a scope of greater than Class C report. At that point we'll have a planning basis or an initial planning basis.

MR. NIEDZIELSKI-EICHNER: Okay. That's helpful. Thank you.

MR. MIKE LEE: My name is Mike Lee. I'm from NRC. I have a couple of questions.

In today's presentation there was reference to this data base. Is this the same data base that's going to be used to compile the waste form characterization report described by Dr. Jardine yesterday?

MR. CLONINGER: That data base will be part of the input into that report. However, the entire last several years of testing and analysis and calculational efforts at the project as well as throughout the DOE program that are referenceable and are available will be included in that report.

MR. LEE: Okay. With reference to the waste package plan that Dr. Jardine also described yesterday, has that been sent to NRC for its information?

MR. CLONINGER: Not at this point. It was just issued I believe--is Dick Morissette here? I believe he has the issue date. We provided drafts to the NRC and the panel in the March meeting. We have given final uncontrolled copies of the issue document to the board here, but it was only issued I believe either last Friday or this Monday. So, no we haven't yet to the NRC.

MR. LEE: Just from a planning standpoint, we are interested in studying these documents so we know what we can do and what we might have to support in the out years.

MR. CLONINGER: Just for the record, it was issued as a controlled document, August 17th.

MR. LEE: Okay. Thank you.

DR. VERINK: Any other questions? Any other questions from the panel? Any comments from you all?

MR. CLONINGER: No. I would just like to thank the board very much for their attention and time.

DR. VERINK: Well, I know I'm speaking for the board when I say we greatly appreciate the time and the effort and enthusiasm that has gone into this presentation.

Thank you very much.

(Whereupon, the meeting was concluded.)