Lead Containing mainly isotope Pb-208: New Reflector for Improving Safety of Fast Neutron Reactors

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Berta Oates
Thank you, again, for joining today's Gen IV International Forum webinar presentation on Lead Containing Mainly Isotope Lead-208: New Reflector for Improving Safety of Fast Nuclear Reactors. The presenter is Dr. Evgeny Kulikov.

Doing today's introduction is Dr. Patricia Paviet. Patricia is the technical group manager of the Radiological Materials Group with the Nuclear Sciences Division at Pacific Northwest National Laboratory. She is also the chair of the Gen IV International Forum Education and Training Task Force. I give the floor to you, Patricia.

Patricia Paviet
Thank you so much, Berta. Good morning, everyone.

It's a pleasure to have Dr. Evgeny Kulikov with us today. He earned his Ph.D. at the National Research Nuclear University MEPhi in Moscow in 2010 and is currently the associate professor at the Institute of Nuclear Physics and Engineering. His areas of professional interest include improving fuel burn-up, nuclear fuel cycle, non-proliferation, and fast reactor safety. Currently, his scientific research is supported by the Russian Science Foundation. He lectures on theoretical aspects of nuclear reactors and conducts laboratory works on experimental reactor physics. He is serving on the Gen IV International Forum Education and Training Task Force.

Thank you so much again, Evgeny, to volunteer to give this webinar today and I give you the floor. Thank you.

Evgeny Kulikov
Thank you a lot for a kind introduction and hello, everyone, and I would like to thank you for joining us this day.

Now, I have a presentation for you. You saw the title, which is about lead, which contains mainly isotope lead-208. Such lead is proposed as a new reflector to improve safety of prospective fast reactors.

As you know, safety is very important and actually the number one priority in nuclear field. And if we look at the respective types of Generation IV reactors, then 3 out of 6, that is half, are fast. This idea that I will be speaking about is applicable for any type of fast reactor, sodium cooled, lead cooled, or gas cooled. That is why this topic is very important and urgent.

At this slide, you may see the outline of the speech. We'll start with a review of how to slow down chain reaction. After it, I'll show you how slowing down chain reaction could improve safety, first, in a simple case when there are no feedbacks, and then we will continue with a more real case when there are feedbacks in the reactor. And, at last, we will look from where we can take these sources of lead-208.

Let us start with a review of how to slow down chain reaction. To start with, I would like to remind you that, as a result of nuclear reaction, we get neutrons that are divided into prompt and delayed. Prompt neutrons are emitted by the fragments immediately. The fraction of them is over 99%. And delayed are emitted a bit after it, sometime after it in some seconds. That fraction is less 1%. Even if we have only a bit of them, thanks to their essential delay time, they help us to control a reactor. And comparing to spread of thermal reactors, respective fast reactors are more difficult to control, that is because a lifetime of prompt neutrons here is 1000 times shorter as you see it here. It's not 1 millisecond as in thermal, but 1 microsecond. And the fraction of them is twice smaller as well.

The idea of this speech is safety improvement by slowing down chain reaction. And here you may see how we can do it. Fast neutrons from the core should go deeply into reflector, you may see it here, and then they should have high probability to return to the core as a result of diffusion. In some way, we may call them delayed because they will go back with a delayed time.

Let us formalize these requirements to slow down chain reaction. Requirement number one is to penetrate deeply into reflector. It means that neutron age should be as large as possible. Neutron age shows us how far fast neutron travels from where it is born to where…

This distance is migration of neutrons slowing down from fast to slow, while this one is migration at diffusion from thermal to absorption. To fulfill the second requirement to have high probability to return to the core, this value should be larger than this value. In other words, diffusion length should be larger than square root of neutron age. You see the idea to slow down chain reaction is that fast neutrons go deeply into reflector and return to the core with essential time delay.

Here, you may see characteristics of chain reaction rate. To have a large neutron age, we need to have a reflector with large atomic mass. So something heavy. To have a large diffusion length here, we need a reflector with small absorption. And for a lifetime of a neutron, we also need a reflector with small absorption.

Looking at these requirements, we may conclude that lead-208 is a good choice for a material of reflector because it is heavy, 208, and it absorbs very small, as you may see it at this slide. Here, you may see a capture cross-section of lead isotopes and some other elements. Natural lead can consist of four isotopes, lead-204 here in black color, 206, 207, and 208. We can see that lead-208 captures less than any other lead isotope and even less than deuterium and graphite at thermal energy range. So it captures extremely small actually.

It leads to the following. First, neutron age and diffusion length are very large, as you may see it here. So it means that fast neutrons from the core will go deeply into reflector, this value is large, and it will go back to the core because this value is also very large and bigger than this one.

If we compare it with natural lead, then we may see that also a neutron from the core would also go same deep, it would not go back to the core. Other elements here are the same. There's natural lead and graphite, but for it, a neutron cannot go very deep into reflector.

Second, slowing down probability is high. It means that a neutron would go into reflector and it will be slowed down there to thermal energy and it will be safe. It will not be absorbed. For a natural lead it is not the same because it captures higher. And third, as you may see it here, a lifetime of thermal neutron is long, which is very important for safety as we see it soon. Lead-208 thermal neutrons live longer than in any other material of a reflector.

This slide shows you albedo. Albedo is an important characteristic of a reflector. Albedo is defined as ratio of returning to living neutrons. The maximum value of albedo is unity. It means that all neutrons leaving the core go back thanks to a reflector.

This slide shows thermal neutron albedo on thickness of reflector. We may see that the largest thickness of reflector the large albedo. But at a certain thickness, there is a saturation, a limit. For natural lead and light water, albedo is low. That is because to large capture cross section of these elements. Heavy water is considered as best moderator thanks to very low capture and due to this fact it has high albedo. However, at thickness over 70 centimeters, lead-208 has even higher albedo, which is really impressive. It is even better than heavy water. So this slide proves that lead-208 is really a very effective reflector.

Now, let us consider properties of lead-208 as a moderator. A good choice would be something that has large scattering cross-section and low absorption cross-section. Here at the top of this slide, you may see a composition of natural lead. Roughly, half of it is lead-208, one-quarter is lead-207, a quarter is lead-206, and just a bit of lead-204. Lead-208 is double magic nucleus with closed proton and neutron shells. And therefore its excitation levels are higher than for other elements and for other isotopes of lead. They are higher than 2 mega electron volt while for other lead isotopes, they are pretty low, below 1 mega electron volt. That is why the lead-208 has a high threshold of inelastic scattering comparing to other lead isotopes. As you know, the average energy of neutron that results from a nuclear chain reaction is 2 mega electron volts. It means that there is no inelastic scattering of such neutron from lead-208. Therefore, a neutron that leaves the core almost won't lose its energy by inelastic scattering on lead-208 and would go deeply into a reflector.

At this slide, you may see comparison of moderator properties of different materials. First property here is a logarithmic energy decrement. It describes average energy loss at collision. It is not dependent on energy; it depends only on atomic mass. This value is small for heavy elements such as lead. It means that many, many X of collisions are needed for a neutron to slow it down on lead. Because of the low logarithmic energy decrement, moderating ability for lead is also low here. But integral characteristic is moderating ratio that takes into account absorption. And as you remember, absorption for lead-208 is very low. That is why it results in a high value. We may conclude here that lead-208 is a better moderator than light water or beryllium oxide or graphite and is inferior only to heavy water.

Now, let us see how these good properties of lead-208 help us to improve safety.

This slide shows contribution of neutrons with different lifetimes into reactor criticality. As already said, in fast reactor, prompt neutron lifetime is very short. It's about 1 microsecond here as you see it on this axis. It is very short comparing to that in thermal reactors where it is about 1 millisecond for PWR or CANDU type of reactors. Control of reactor is possible thanks to delayed neutrons. Their lifetime is very long. It's 1 second or even tens of seconds, but its fraction is very small. And reflector from natural lead almost does not increase neutron lifetime as you see it in brown curve. But if we have a reflector from lead-208, here in red, we can increase prompt neutron lifetime up to a delayed neutron lifetime. And fraction of these neutrons is considerable. It's 34 beta where beta is a fraction of delayed neutrons. And if we add a bit more of a reflector, it would add us a longer time.

This slide shows you peculiarities of reflector neutrons. These are neutrons that go back from a reflector. Their origin is prompt and delayed neutrons that leaves the core, spend a relatively long time in reflector, and then go back to the core. Their fraction is much less than prompt but much more than of delayed neutrons. Their lifetime is between prompt and delayed, which is very important for improving safety. These neutrons spend a relatively long time in reflector and return to the core at lower energy comparing to a typical value of prompt and delayed neutrons. It’s important to say that reflector neutrons have inputs into nuclear chain reaction only after returning to the core, which means that there is always some dead time when they can’t affect reactivity. This is also important for improving safety. As these neutrons come from a reflector, their inputs into nuclear chain reaction are mainly at the edge of the core where effects of reactivity are lower comparing to center of the core. Also, we may change the amount and energy by changing reflector geometry and material. Finally, reflector neutrons play the role of additional delayed neutrons.

This slide shows you importance of reflector neutrons considering reactor runaway. Here you may see three options. Option number one, on the left, is critical reactor with a reflector. It is critical here. Multiplication factor is equal to a unity and its components are multiplication factor on prompt neutrons, delayed neutron fraction, here in red, and input of reflector neutrons here shown in green.

Second option is runaway on delayed and reflector neutrons. If there would be no reflector neutrons, then a reactor would be supercritical on prompt neutrons, which would mean a rapid runaway. But, thanks to reflector neutrons, even insertion of a reactivity that exceeds delayed neutron fraction does not lead to a rapid runaway as you see it here, which is still subcritical on prompt neutrons.

The third option, on your right, is runaway on prompt neutrons. Only if there is insertion of a very large reactivity that exceeds delayed neutron fraction plus the input of reflector neutrons, then, yes, there is super criticality on prompt neutrons.

Here you may see an example to understand how neutron lifetime is important for safety. Let us consider a fast reactor runaway without feedbacks induced by step insertion of reactivity for two cases, reflector from a natural lead here in red color, and reflector from lead-208, here in blue. If reactivity does not exceed delayed neutron fraction, on your left here, then there is only a little advantage of lead-208. But if reactivity does exceed delayed neutron fraction, then in this case we may see that if we have natural lead, reactor power increases by 500 times just in 3 milliseconds. It is almost an explosion. While in the case of lead-208, much, much slower. That is a big difference and lead-208 as a reflector in fast reactor could slow down chain reaction and improve safety.

Now, let us continue with more real case where there are feedbacks. According to the model of neutron flash, if inserted reactivity exceeds delayed neutron fraction, beta, then this is a state of prompt super-criticality. Heat does not have enough time to reach the coolant and that's why only Doppler effect has enough time to act. And duration of neutron flash is proportional to a neutron lifetime while energy yield is independent on neutron lifetime.

All this is correct if we have a reflector of natural lead. But when we have lead-208, neutron lifetime increases by three orders of magnitude, not 1 microsecond but 1 millisecond. And duration of neutron flash would increase proportionally, it's about 1 second. And 1 second is actually even more than time constant of fuel element as a parameter that indicates how fast heat is transferred from fuel to coolant. And so in this case, part of the heat has enough time to reach the coolant and negative coolant feedback has enough time to act in addition to a Doppler effect. So we may conclude that if we have lead-208 as a reflector, there would be not one but two feedbacks, Doppler effect and negative coolant feedback.

Here, you may see fast reactor power at the neutron flash. In fact reactivity is twice as much as delayed neutron fraction. In the case of natural lead, a neutron lifetime is about 1 microsecond while in the case of lead-208, about 1 millisecond. That is why processes are 1000 times slower here. And peak power is 1000 times slower.

When processes are slow, there is enough time for the heat to be transferred from fuel to coolant, and about a quarter of the heat would be transferred to the coolant. It leads to the fact that temperature of fuel increases not that quick, 1000 times slower, and reaches a lower level because part of the heat is transferred to coolant.

Finally, here you may see one more comparison. These are dependences of relative power growth rate on reactivity. If we consider fast reactor with natural lead as a reflector, then insertion of reactivity about delayed neutron fraction would mean almost explosion. You may see it here, very, very strong increase of power. While lead-208 makes the reactor much safer, runaway would happen as slow as in thermal CANDU type of reactor.

Finally, we consider from where we can lead-208. Actually, lead-208 is a final product of a radioactive decay chain of thorium, while lead-207 and 206 are accumulated due to decay chains of uranium. Lead that is produced in radioactive decay chains of thorium and uranium isotopes is called a radiogenic lead and a relative content of lead isotopes in radiogenic lead depends on age of ore and admixture of natural lead. So we can get radiogenic lead from thorium and thorium-uranium ores.

This slide shows ore deposits containing radiogenic lead. Here, we can see the ores that have high quantity of thorium. Because lead-208 is accumulated due to decay chain of thorium, you may see that there are ores in Brazil, in Australia, in Ukraine that contained radiogenic lead with about 93% of lead-208. Let's see if such a composition is enough to give good properties of lead-208 or would a small presence of other lead isotopes ruin them. In other words, do we need to enrich lead or can we just get it from the earth?

To make a decision, let's consider dependence of required reactivity on asymptotic period. This value is period of time during which reactive power changes approximately 2.7 times. Long asymptotic period means that tension processes are slow and reactor is easy to control. So this area is easy to control area. While short asymptotic period means that processes are rapid and reactor is difficult to control. Red curve number 1 indicates a case of a bare core. Insertion of reactivity that even slightly exceeds delayed neutron fraction leads to a very short asymptotic period as you may see it here. And it means that power increases rapidly. Curves number 2 and 3 correspond to adding a reflector from a natural lead. And as you see, the situation is almost the same assertion of reactivity that even slightly exceeds delayed fraction leads to a nuclear accident.

Green curve number 4 indicates a case of reflector from a radiogenic lead that is extracted from thorium ores. So that is more or less a real case. Asymptotic period is dozens of times longer but still it's rather short. Pink curve number 5 indicates a case of reflector from lead-208. In this case, as you see, asymptotic period is almost 1 second; that is slow enough for emergency systems to react. That is good. Curve number 6 is an imaginary case when capture is equal to zero. You may see that there is almost no further improvement comparing to curve number 5 and it means that lead-208 has small enough to capture and there is no need to look for other materials that exceed it in properties.

Enrichment plants in Russia are able to enrich lead up to the level of 99.8% of lead-208. It means that enriched lead would still contain 0.2% of lead-207. This isotope has a rather large capture cross-section and even its very small content has a noticeable effect. As you see it from curve number 7, you see we added just 0.2% of lead-207. And comparing to curve number 5, it looks more difficult to control. The conclusion here is that it seems that we should enrich lead because what we have from the earth, curve number 4, is not enough for systems to react.

One more issue to solve is how thick the reflector should be. This slide shows a neutron lifetime in fast reactor with different lead reflectors.

Parameters are fraction of lead-208 on this axis and thickness of reflector which is presented by different curves. You may see that a half meter reflector is not able to prolong a neutron lifetime even if we have high value of lead-208. One meter reflector is able to prolong a neutron lifetime by at least an order of magnitude. A combination of 2 or 3 meters of reflector and high quantity of lead-208, which is here, is able to prolong a neutron lifetime by several orders of magnitude approaching 1 millisecond, this value. There is no sense to make the reflector thicker because, as you see, after 3 meters all is the same. So, here, we may conclude that it's reasonable to use enriched lead of thickness of about 2-3 meters.

The conclusion is that a new approach is proposed to improve nuclear safety of fast reactors thanks to slowing down chain reaction. Lead-208 as reflector in fast reactor can considerably prolong lifetime of prompt neutrons by three orders of magnitude but it would be about the same as in thermal type of reactors. Long neutron lifetime and short-time constant of fuel elements substantially improve safety even if we insert reactivity that exceeds delayed neutron fraction.

I would like to thank you for joining us and if you have something to ask, please do it now. Thank you.

Berta Oates
Thank you very much, Evgeny. While questions are coming in, we'll take a look at the upcoming webinar presentations. In September, a presentation on the Gen IV Coolants Quality Control by Dr. Christian Latge out of CEA in France. In October, Passive Decay Heat Removal System by Dr. Farmer with Argonne National Lab in the USA. And, in November, the Czech Experimental Program on MSR Technology Developments by Dr. Uhlir with the Research Center in the Czech Republic.

As a reminder, the presentation today is being recorded and, yes, it will be available to watch again or to share with others. The link will be provided on the Gen IV International Forum webpage.

The link will be posted in the e-mails that distribute after today's presentation.

If you have questions, please go ahead and type those in now.

There's a question, how does lead-208 influence neutron spectrum.

Evgeny Kulikov
How does lead influence neutron spectrum, right?

Berta Oates

Yes.

Evgeny Kulikov
Well, yeah, I don’t think that I have a slide to show a comparison of neutron spectrum that is formed in bare core and in the case of the core that is reflected by natural lead and by the lead-208. Well, yeah, sure, it would make it a bit more thermal but not that much because lead is a heavy elements, though a neutron, it would not be slowed down to thermal and the reactor would still be a fast type of reactor.

Berta Oates
Thank you. How expensive is it to enrich lead-208 to the desired level including removing lead-207?

Evgeny Kulikov
Yeah. It’s a good question. This information about enrichment of lead in Russia and up to what level they can do it, we asked it and they answered us only up to what level they can do it. But when we also inquired about the price, there was no answer from them. So I just think that maybe its industrial secret or something else. So we don’t have any info on that, sorry.

Berta Oates
In your opinion, which kind of Gen IV system is most suitable for these reflectors?

Evgeny Kulikov
Well, as I said at the beginning of the presentation, it should be applied to any type of fast reactor. There are three of them among Generation IV systems. It’s lead-cooled, gas-cooled, and sodium-cooled. In all these three types, it is applicable.

Berta Oates
Thank you. Did you assess the impact of slowed down neutrons returning from the reflector on power peaking at the edge of the fast core?

Evgeny Kulikov
Sorry – and power peaking?

Berta Oates
Correct. The question is did you assess the impact of slowed down neutrons returning from the reflector on power peaking at the edge of the fast core?

Evgeny Kulikov
No, I don’t think so. No. Power peaking. I would write it down on power peaking at the edge. Could you tell it one more time please, Berta, on –?

Berta Oates
You bet. And also hopefully you'll be able to see it.

Evgeny Kulikov
Can I see it?

Berta Oates
Yeah. If you look in your control panel, there should be a pod that you allow you to see the questions and then I've also shared it on the chat pod with all. The question is did you assess the impact –

Evgeny Kulikov
Yes, I can see it now.

Berta Oates
You see it. Okay. Great.

Evgeny Kulikov
Did you assess the impact of slowed down neutrons returning from the reflector on power peaking at the edge of the fast core? No. The answer is no. Unfortunately, we did not do it yet. Perhaps we should, yeah. Thank you for this.

Berta Oates
Have you considered reactor control from the reflector?

Evgeny Kulikov
Have you considered reactor control from the reflector? Could you specify what do you mean reactor control from the reflector?

Berta Oates
Craig Smith, if you're still listening, can you please clarify your question? In the meantime, there's a question, in an MSR reactor which is using thorium as fuel, is there a possible interaction between the fuel and the in situ production of lead-208 as self-moderator.

Evgeny Kulikov
Interaction between fuel and lead-208?

Berta Oates
Correct. The in situ production of lead-208 as a self-moderator.

Evgeny Kulikov
You mean a molten salt reactor. Okay. In a molten salt reactor which uses thorium as a fuel is there a possible interaction between the fuel and the in situ production of lead-208?

Actually, I don't think so because it takes time for accumulation of lead-208. It accumulates in nature due to long decay chain. And some reactions that are part of this chain, they happen during a very long time. So I don't actually see how we can do it in the core while it has this type of fuel. Anyway, there is enough of lead now in the earth, so there is no issue if we have it enough for now. Yes, we have it enough. The issue is only how cheap or not cheap it is to extract it to enrich it.

Berta Oates
Thank you. Let's see. The thickness of reflector would result in a non-negligible effect on neutron lifetime is significant, larger than 1 meter. Have you done any evaluation or optimization that accounts also for the impact on the size of reactor?

Evgeny Kulikov
I'm just looking for this question. Yeah, I see it. The thickness of reflector that would result in a non-negligible effect on neutron lifetime is significant. Yes, right, more than 1 meter. Any evaluation position that also accounts for the impact on the size of the reactor?

No. This research is a fundamental research that was done during about one year and we did not go yet that far. So, here is just a demonstration how to do it. And, of course, many aspects should be considered in future. That's of course for sure.

Berta Oates
There's another question on cost but I believe the answer to that is probably the same as before. And it is unknown. The cost to enrich lead-208 at 98% from natural lead. But before, you indicated that you are not sure of that.

Evgeny Kulikov
Yeah. Well, enrichment level is 99.8. It's 99.8. Yes. But we don't know the price because we did not get any reply from the company that does it.

Berta Oates
Could the control rods be located in the reflector instead of the core?

Evgeny Kulikov
Well, in principle, yes, but their impact would be not that effective, not that strong, because as you know, the number of neutrons and reactions is much lower there. Normally, they are located in the core. Well, you may do it in the reflector but effectiveness of such an idea I don't think would be very high.

Berta Oates

Thank you. I think that's all the questions. We've managed to go through the whole list. There's quite a few today. Thanks to everyone for your participation. It's always such a great indicator of interest in the topic. We appreciate your participation.

Evgeny Kulikov
I also have some that we should think about. I will write them down. Thank you.

Patricia Paviet
I am planning to do that. We will talk about that with the education and training task force in a few days. I wanted to thank you again, Evgeny, and thank you to the audience for all these very good questions.

Evgeny Kulikov
Thank you.

Patricia Paviet
Okay. Goodbye.

Berta Oates
Thank you, everyone.

Evgeny Kulikov
Thank you. Good day.

Berta Oates
Bye bye.

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