

POLLUTION

As nuclear waste piles up, scientists seek the best long-term storage solutions

Researchers study and model corrosion in the materials proposed for locking away the hazardous waste

by **Mitch Jacoby**

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IN BRIEF

More than a quarter million metric tons of highly radioactive waste sits in storage near nuclear power plants and weapons production facilities worldwide, with over 90,000 metric tons in the US alone, emitting radiation that can pose serious risks to human health and the

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As nuclear waste piles up, scientists seek the best long-term storage solutions

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us from our parents' generation," says Frankel, who is a materials scientist at the Ohio State University. "And we are—more or less—handing it to our children."

Like other specialists studying the root causes of corrosion and degradation in nuclear waste storage materials, Frankel isn't content to kick the can down the road. Instead of waiting for the leaks to get worse or for governments to finally decide to permanently store the waste, these scientists are investigating how to recognize and predict damage to nuclear waste storage

environment, the waste, much of it decades old, awaits permanent disposal in geological repositories, but none are operational. With nowhere to go for now, the hazardous materials and their containers continue to age. That unsustainable situation is driving corrosion experts to better understand how steel, glass,

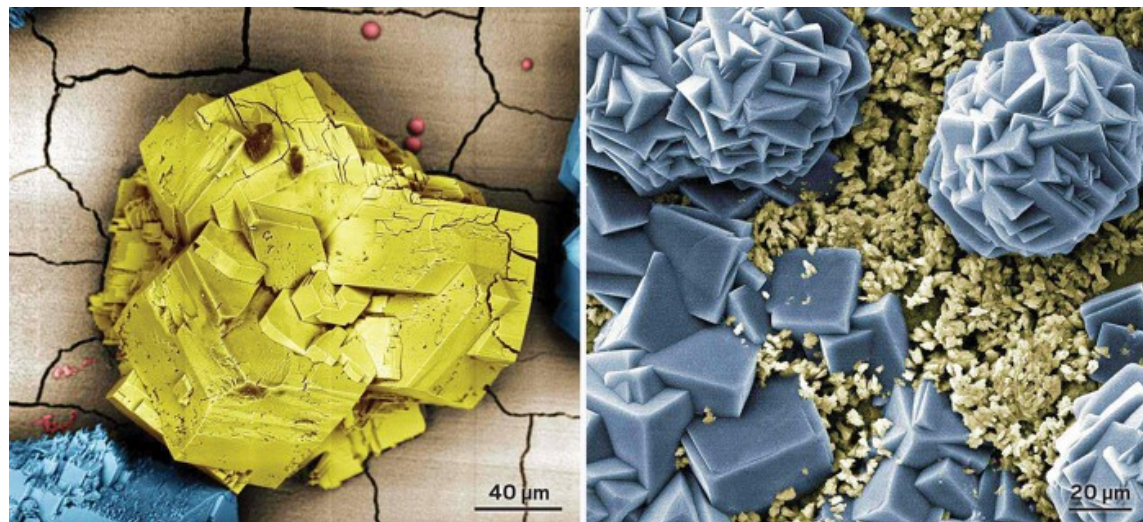
containers and how to control it to safeguard people and the environment.

WHERE DID IT COME FROM?

Highly radioactive waste, often called high-level waste, comes mainly in two forms. One is leftover fuels that were used in **nuclear power plants** to generate electricity. The other is the waste made by facilities

and other materials proposed for long-term nuclear waste storage containers might degrade. Read on to learn how these researchers' findings might help protect people and the environment from waste leakages.

making reaction. Chemists want to change that



Credit: John Vienna/Pacific Northwest National Laboratory

Scientists are studying glass samples to understand long-term corrosion of vitrified nuclear waste. These micrographs show the results of accelerated aging tests on two types of aluminosilicate glasses. Ions have leached from the glasses and crystallized on their surfaces: the largest of these false-color crystals (yellow on left, pale blue on right) are sodium aluminum silicate hydrates of various composition and structure.

involved in nuclear weapons production or by facilities that reprocess and recycle used power plant fuel.

All these wastes can remain dangerously radioactive for many thousands of years. For that reason, they must be disposed of permanently, experts say. About a dozen countries, including Finland, Switzerland, and other European nations, are planning deep geological repositories for their nuclear waste. In the US, government officials have proposed storing the country's waste in a repository beneath Yucca Mountain in Nevada. The site lies about 300 m below ground level and 300 m above the water table. But the Yucca Mountain site has gone **in and out of favor** with changes in the US's leadership. For now, waste accumulates mainly where it's generated—at the power plants and processing facilities. Some of it has been sitting in interim storage since the 1940s.

“ It’s a societal problem that has been handed down to us from our parents’ generation. And we are—more or less—handing it to our children.

— *Gerald S. Frankel, materials scientist, Ohio State University*

During World War II and throughout the Cold War era, the US generated millions of liters of radioactive waste—a mix of liquid, sediment, and sludge—in the name of national defense. The toxic waste, a by-product of creating plutonium for nuclear bombs, was collected for 45 years in underground storage tanks mainly in Hanford, Washington, and the Savannah River Site in South Carolina.

In Hanford alone, more than 200 million L of this waste still sits after many decades in underground tanks waiting to be processed, according to Thomas M. Brouns, who leads the environmental management sector at nearby Pacific Northwest National Laboratory (PNNL). About one-third of the nearly 180 storage tanks, many of which long ago outlived their design lives, are known to be leaking, contaminating the subsurface and threatening the nearby Columbia River. Another 136 million L of the stuff awaits processing at the Savannah River Site.

Related: Proposed nuclear waste storage materials may have a corrosion problem

Today, no nation in the world would consider storing high-level liquid waste indefinitely like this, says PNNL materials scientist John D. Vienna. “That’s a thing of the past,” he adds.

THE WONDERS AND WORRIES OF GLASS



Credit: US Department of Energy

These underground tanks in Hanford, Washington, were built in the 1940s to store liquid radioactive waste from plutonium production. Today, the contents have been transferred to newer tanks in preparation for vitrification.

One way that scientists have come up with to store liquid nuclear waste more permanently is to vitrify it. In this process, the hazardous material is converted to a more easily managed immobile solid—glass. Not only does glass prevent toxic species from leaking into the environment, but it also provides some shielding against radioactivity leakage and is highly durable.

NUCLEAR WASTE BY THE NUMBERS

~442

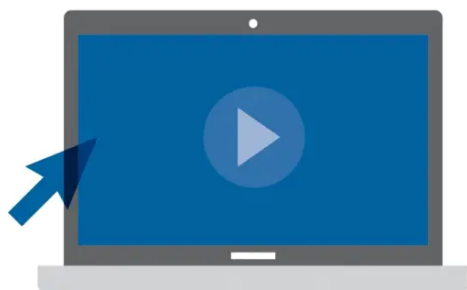
For years, India, France, the UK, and other countries have carried out vitrification of liquid waste from weapons production and fuel recycling—and still do. In the US, operators at Savannah River have been vitrifying weapons-related waste for about 20 years. And although the US does not currently recycle fuel, it did so in the 1960s and '70s near Buffalo, New York, in a program known as the **West Valley Demonstration Project**. The operation generated 275 canisters of glass from vitrifying high-level waste; the canisters are stored there and await permanent disposal in a repository.

At Hanford, the site is gearing up to vitrify its waste. The work will be done on-site at the Hanford Tank Waste Treatment and Immobilization Plant. Also known as the **Hanford Vit Plant**, the multibillion-dollar Department of Energy (DOE) facility has been under construction since 2002. According to the DOE, some of the waste treatment operations are scheduled to begin by 2023, but the massive project has already seen several delays and may see more.

Stabilizing nuclear waste via vitrification isn't a new idea. The process involves blending waste materials with glass precursors, heating the mixture to above 1,000 °C to melt the components, pouring the molten glass into a storage container, and letting it cool and solidify, locking the harmful constituents in the glass matrix.

“Vitrification of nuclear waste seems to be **well established** by now, but actually it still faces complex problems,” says Ashutosh Goel, a materials scientist at Rutgers University. The plan at Hanford, for example, calls for entombing nuclear waste in borosilicate glass and encasing the glass in stainless-steel canisters. Yet the exact formulation of the glass, or glasses, is still under investigation.

Open questions include the following: What glass compositions will lead to the highest uptake of nuclear waste? How suited are those glasses to vitrification? And how well will they resist corrosion after being interned for eons in a repository environment?



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After 1,000 years or so, Goel says, the steel canister surrounding the glass will likely corrode, and groundwater may seep in and interact directly with the glass, degrading it. “The stability of the glass in the presence of groundwater represents the last line of defense against release of radionuclides” into our environment, he adds.

Number of nuclear power reactors operating worldwide

96

Number of commercial nuclear power reactors currently operating in the US

>90,000 metric tons

Total mass of highly radioactive nuclear waste in the US, including spent fuel and other material

212 million L

Volume of radioactive waste in underground storage tanks in Hanford, Washington

Sources: US Energy Information Administration, US Government Accountability Office, World Nuclear Association, International Energy Agency, Hanford Vit Plant.

So scientists would like to better understand how and if glass might leach any radioactive materials locked inside. Whether groundwater degrades the glass enough to cause it to release its radioactive cargo depends on several processes, experiments have shown. For alkali-borosilicate glasses, a well-studied family, the degradation steps would include ion exchange between ionic species in the water and alkali ions in the glass; hydrolysis of silica, boron, and other chemical groups that compose the glass network; and dissolution and release of glass components into solution or onto the surface of the reacting glass.

Goel and colleagues in the US and the Czech Republic tackled the complex relationship between these and other processes and ways to model them in a recently published study (*J. Non-Cryst. Solids: X* 2019, DOI: **10.1016/j.nocx.2019.100033**). The authors recommend that to better understand the long-term fate of vitrified materials, researchers in this field should focus on experiments that help determine the rate-limiting mechanisms of glass change over time—especially during long periods of very slow change—and help explain how composition affects the rate at which glass changes.

Related: Radioactive Waste Safety

Some models for predicting how vitrified waste will corrode over millennia in a geological repository assume that the stainless-steel canister eventually disintegrates, leaving groundwater, if present, to react with the glass. Others consider water's reactions with steel and glass independently.

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A new study, however, shows that water-based solutions can trigger **unexpected corrosion chemistry** that occurs only at the wet steel-glass interface (*Nat. Mater.* 2020, DOI: **10.1038/s41563-019-0579-x**). The idea for the study came from Xiaolei Guo and coworkers, who reasoned that if water seeps through cracks in the steel canister, penetrating the microscopic gap between the glass and steel, it could set off reactions in the confined space that would not occur in a more open setting. So Guo, a corrosion researcher at Ohio State who works with Frankel, teamed up with PNNL's Vienna, Stéphane Gin of the French Alternative Energies and Atomic Energy Commission (CEA), and others to test the hypothesis.

To simulate damp repository conditions, the researchers pressed glass and steel together tightly and set it in a salt solution held at 90 °C. The temperature was chosen to accelerate the normally slow corrosion process and to mimic conditions caused by ongoing radioactivity. After a month, they found accelerated pitting and corrosion of the glass and steel compared with control samples in which glass and steel were not held in intimate contact. Analyses showed that reactions between metal ions and the water acidified the solution. The acidity corroded the steel and glass, releasing additional ions, thereby accelerating the corrosion process.

The study uncovered a previously unknown corrosion mechanism involving dissimilar materials in close contact; some researchers say this process could decrease the durability of glassy nuclear waste. Vienna cautions against drawing that conclusion. This ongoing study turned up “an interesting finding that can be used to improve our models,” he says. He adds that the accelerated process is limited to roughly a 10 μm thick surface region on a standard vitrified waste sample. According to Vienna, the results suggest that less than 1 in 10,000 of the waste packages proposed for storage at Yucca Mountain would fail in 150,000 years.

Widespread storage

Tens of thousands of metric tons of radioactive spent nuclear fuel sit in steel-and-concrete storage casks (cutaway) at nuclear power plants across the US (map) as they await permanent disposal.



Credit: Pacific Northwest National Laboratory

At Pacific Northwest National Laboratory, Jodi L. Meline pours a sample of molten glass to study corrosion in vitrified nuclear waste.

Related: Nuclear Efficiency

The radioactive material in the casks is a controlled solid, not an unruly liquid-sludge combo that's leaking into the ground and warrants vitrification, as is the case for the material in the Hanford tanks. Experts consider dry-cask storage **safe in the short term**. But because the spent-fuel containers sit in limbo, many of them will remain where they are for decades longer than originally intended. That leaves Eric J. Schindelholz wondering whether environmental factors will eventually take a toll on the stainless-steel canisters' integrity. Schindelholz, who recently joined Ohio State's team of corrosion experts, studies a type of chemically induced damage known as stress corrosion cracking, which can occur in metals at stress points such as weld joints—like the ones used to seal the stainless-steel canisters of spent fuel.

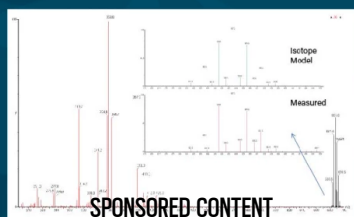
“Vitrification of nuclear waste seems to be well established by now, but actually it still faces complex problems.

— *Ashutosh Goel, materials scientist, Rutgers University*

He explains that during manufacturing, stress develops at weld seams as they cool and contract. If corrosion sets in at those spots, then some materials can start to crack and fail. The iron-chrome-nickel-based stainless steel used in dry casks is a material prone to fail when corrosion kicks in.

What might cause the corrosion on these concrete-covered casks? Many nuclear power plants in the US were built along coastlines for convenient access to cooling water. Proximity to the coast means exposure to sea-salt aerosol. Because of the cask design, which blocks radiation but allows air flow—for cooling—between the steel cylinders and concrete silos, aerosols can reach the cylinder

surfaces. Salt particles, which are hygroscopic and deliquescent, can settle on canister welds and other stress joints, take up atmospheric water, dissolve, and form chloride-rich corrosive brines. Those conditions could lead to small cracks that breach a cylinder and release harmful material and radiation.



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Schindelholz, together with researchers at Ohio State and Sandia National Laboratories, is developing a model that can be used to predict when, how, and where dry-cask cylinders might crack. The model includes input derived from the team's electrochemical and microscopy studies that analyze pitting, crack initiation, and the effects of relative humidity on the morphology and distribution of pits formed in stainless steel exposed to marine atmospheres (*J. Electrochem. Soc.* 2019, DOI: **10.1149/2.0551911jes**). These researchers are hoping their efforts will protect the temporarily stored waste until a more permanent solution can be agreed upon in the US.

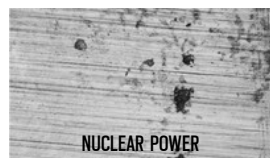
The tens of thousands of metric tons of radioactive waste that accumulated from commercial power plants and years of national defense operations continue to age at sites around the globe. As the hazardous material and the containers it sits in await permanent disposal, the stockpile keeps growing. Corrosion experts are doing their part to safeguard people and the environment from this danger, but it's still there. "It's a difficult problem, but we need to deal with it now," Frankel says. "Putting it off any longer isn't good for anyone."

Chemical & Engineering News

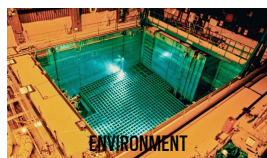
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COMMENTS

Purniah

(March 30, 2020 6:33 AM)

Its well known that the spent fuel taken out of a reactor contains a lot of useful material. Unused fuel, Plutonium and radioactive fission fragments which can be used for medical treatment etc. If these are removed then the remaining waste is very less. So it makes sense to reprocess the spent fuel

Reply »

Mike Keller

(March 30, 2020 3:02 PM)

Reprocessing is an ideal that more or less guarantees no solution will emerge. Get rid of the stuff, as in deep underground.

Reply »

Phil

(March 30, 2020 10:37 AM)

Partially spent nuclear fuel will be used as a fuel source for Generation 4 reactors being developed now by Bill Gates' TerraPower and others. It will supply electricity for decades without mining any more uranium. Dry Cask

storage is safe and adequate for the near future. In my opinion and that of many others permanent disposal is costly and not necessary.

Reply »

Tony

(April 19, 2020 2:10 PM)

Yes, TerraPower is by far the best way to use spent nuclear materials, e.g. fuel. However, I think its development is stuck in the midst of US-China trade disputes as well as internal NRC-DOE issues here in the US.

Reply »

Mike Keller

(March 30, 2020 2:59 PM)

Yes, I suppose we can have scientist continue to "study" the issue and never actually solve the problem.

Or we could have engineers manage the effort and actually get something accomplished. The solution does not have to be perfect, it only has to be good enough for a couple of hundred years. At that point the radiation levels are reasonably low. Worrying about the disposition of plutonium in the distant future is a classroom exercise.

Reply »

TptDac

(April 1, 2020 1:30 AM)

I have given thought to the issue of what kind of people could run a successful nuclear waste disposal project. I spent a fair part of my career as a scientist working on such projects. When the scientists were in charge, the funding tended to be a feeding trough for people who did what they wanted to do anyway. When the engineers were in charge, things were more focused on the end result. but not as good as I would have hoped. There was always some component of basic science that was actually needed to attain the end result. What could be done was always limited by either "dilution of needed science by unneeded science, like by a factor of 100:1", when the scientists were running things, or schedule/funding limitations, when the engineers were running the show. Some kind of hybrid scientist/engineer is required to manage things, but there is also the problem of political interference (e.g., certain national labs and academics MUST be funded - I won't say who here, but you know who you are). I am not impressed with this article. It presents a happy picture (Wow, now we know what to do!), when the reality is quite different. Look up the history of project failures, going back about four decades or so (e.g., Yucca Mountain, the Salt Repository Project, the Basalt Waste Isolation Project, and many smaller efforts). The article should have included something about the long history of failures, especially those related to vitrification. Only WIPP (TRU waste in bedded salt) ever got anything done in terms of actual disposal.

Reply »

Tom

(March 30, 2020 4:21 PM)

This industry has never known what to do with the waste. They are idiots for ever making any of it. Nature out of place.

Reply »

TptDac

(April 1, 2020 1:32 AM)

Don't blame the industry. The federal government promised to figure out the waste disposal issue.

Reply »

James E Hopf

(March 30, 2020 5:17 PM)

These researchers complain about kicking the waste "problem" down the road. The truth is that their own remarks, and articles like this, make it more likely that it will continue to be kicked down the road. Hying small to non-existent risks/hazards associated with nuclear waste makes it all the more likely that no communities will step forward to accept a repository.

The nuclear waste "problem" is purely political. It has been technically solved for a long time. The fact is that any risks (long-term as well as shorter term) associated with nuclear waste are tiny compared to those associated with other industries' and energy sources' pollution and waste streams. Even with all the (supposedly significant) issues these researches go on about, the long-term risks of other waste streams are orders of magnitude larger.

Nuclear has done the most, and has been the *most* conscientious, with respect to not leaving problems and posing risk to future generations. It is the only industry that is containing all its wastes and is ensuring that they remain contained for as long as they remain hazardous. NRC has concluded that Yucca Mountain would meet that impeccable, unprecedented requirement (that no other waste streams come close to meeting). Other industries just release their wastes and toxins directly into the air, simply heap them into piles (like coal ash) or carelessly shallow-bury them. Depleting earth's reserves of valuable hydrocarbons, destabilizing the planet's climate, and lacing soil and water all over the world with toxins like mercury and arsenic; now THAT's a gift to future generations!

If one is concerned about overall public health and safety, as well as the climate, the way to help is not to nitpick about tiny nuclear-power-related risks or try to make tiny nuclear-related risks even smaller. The way to reduce overall public health risks is to maximize the *deployment* of nuclear power, since its risks are thousands of times lower than those associated with the (mainly fossil) alternatives. (Even solar and wind power pose larger risks than the ones these researchers seem to be so concerned about.) The only real issue nuclear power has is cost, and almost all research efforts should be directed at bringing nuclear power costs down. THAT is how you reduce public health risks.

Reply »

Dennis Huber

(March 31, 2020 5:43 PM)

It is really straightforward to resolve the spent fuel issue. Reprocess the spent fuel into four product streams - transuranics that go to a burner or breeder reactor, fission products that are further separated into short lived

(less than 33 years) that can be vitrified and stored for 1000 years or so at Yucca Mountain, and the seven bad actor fission products with long half lives that need to be sent to the burner reactor. The fourth stream - the rest of the "waste" - is Uranium dioxide, and the deficit mass from the fission products and transuranics can be filled with weapons grade U or Pu from US or former Soviet Union weapons such that the resulting average enrichment is sufficient to use the entire lot to power another nuclear reactor without having to mine additional uranium for an extended time. We should eliminate our wasteful once-through practice and deal with the problem we have created, not pass it onto the next generation. Certainly I have simplified this: there are small issues with this approach (few technical, mostly regulatory), but it is much better than the alternative - which is continue to do nothing.

Reply »

Noel Wauchope

(March 30, 2020 6:45 PM)

Look, this is a really informative and interesting article. But, I continue to be amazed that none of these experts have thought of the idea of STOPPING MAKING RADIOACTIVE TRASH.

Reply »

Steven Curtis

(March 30, 2020 7:47 PM)

Great article, however, recycling should be explored more in-depth. There is a process called pyroprocessing when used with fast reactor technology, can extract the remaining 95% of the energy from the existing used fuel material (or any source of depleted uranium, such as the "tailings" from the uranium enrichment process). Purniah is right about recycling commercial used nuclear fuel, however, taking out medical radioisotopes must be done quickly for them to be useful. No process plans to do so yet, but it would be great if it could happen. Nevertheless, getting the remaining power from material currently considered waste should not be ignored.

Reply »

Shane Broussard

(March 31, 2020 7:40 AM)

We should be recycling the fuel as much as possible. Continuing to study the problems and doing nothing is what has been done for decades.

There is no way to guarantee any storage solution for millenia. Get off the pot and put this stuff in Yucca mountain.

Or why not just drop these storage containers into the ocean above the Mariana' Trench? 35,000 feet of water, it can't go anywhere and when the containers begin to degrade there is a vast ocean that will disperse and neutralize it.

Reply »

G.R.L. Cowan

(March 31, 2020 11:58 AM)

"As nuclear waste piles up" – not a good beginning.

For ~60 years, nuclear waste – especially nuclear *power* waste, which the article painstakingly fails to distinguish from the bomb production waste, liquid, at Hanford – has been piling up nowhere in the world and injuring nobody a year. The Vermont Yankee casks in the page-top photo could I suppose be considered a one-deep pile.

Each of them contains nuclear fuel that has raised ~50 million tonnes of steam and is now a little tuckered out. Each represents, at today's prices, about \$35 million in natural gas royalties and tax revenues that government didn't get. At today's prices, uranium miners would have got only \$10 million. Years ago, both gas and uranium prices were much higher, and government's loss was accordingly greater.

"... reactor engineers submerge the assemblies in on-site pools for a few years until the fuel cools and the radioactivity starts to fall ..."

An assembly that has been shut down only four days can do real damage, at least to itself and other equipment (as shown at Fukushima). Supposing it goes from pool to cask after five *years* of rest, it then has lost 97.5 percent of the four-day radioactivity. Otherwise said, it's down 40-fold.

Rather than *starting* to fall at that time, the radioactivity has already fallen greatly and quickly, but the reduction will from then on be slower.

Reply »

Dallas

(March 31, 2020 12:04 PM)

Or we could consume all that waste in a next gen reactor and provide 100 years of safe, carbon free electricity and process heat. Bet you didn't know we could do that.

<https://youtu.be/aHsljVnY6ol>

Reply »

Dr. Jacob D. Paz

(March 31, 2020 6:29 PM)

Both the scientific community and the state of the Nevada have challenged the proposed high nuclear waste repository at Yucca Mountain, Nevada based on scientific and legal grounds. On the other hand, the Department of Energy (DOE) maintains that the Yucca Mountain Project (YMP) is a suitable place to bury high nuclear waste. There is uncertainty as to whether the engineering barrier system will be corroded. There are two major corrosion concerns: electrochemical corrosion and microbial induced corrosion. All the corrosion studies at Yucca Mountain were conducted in laboratories due to the chemical and geological complexity of YMP, which raises serious questions. In order to evaluate properly how the repository will comply with regulatory requirements, the DOE

should have conducted long-term studies in real-world conditions prior to the approval of YMP. One DOE assumption that the tuff will absorb all contaminants might be inaccurate since the Nuclear Waste Technical Review Board stated that the tuff is highly fractured, about 85%. In addition, the DOE did not incorporate into their computer model deliquescence corrosion. The 2016 Nuclear Regulatory Commission (NRC) Environmental Supplement stated that both radionuclide and non-radionuclide contamination would be "small." This raises scientific questions: Did DOE comply with the NRC and submit a complete and accurate license application? Have the DOE and/or NRC approached the Environmental Protection Agency for assistance in the development of risk assessment for the mixture of radionuclides and heavy metals? Why were no studies of the coefficient of distribution of radionuclides and heavy metals submitted

Reply »

Imfene Endala

(April 1, 2020 7:37 PM)

Defense waste like the vitrified glass is destined for WIPP a salt mine in New Mexico. the facility exists and has been in use for quite some time. The salt is waterproof and plastic. So the containers will eventually be completely encapsulated and water ingress will not be a worry.

Spent fuel in Finland will be entombed in a hardrock mine. In France they want to bury it in clay. Water doesn't move in clay.

While these materials will be radioactive for a very long time, within a 1000 years they will have decayed to a level where they are not all that dangerous any more. Of course they will still be toxic, but Lead, Arsenic, Copper and Mercury and even some man made materials will also be toxic forever.

I think the US should bury the spent fuel either at WIPP or in a similar salt deposit.

We dig up metal tools and ornaments that are lots older than a 1000 years so I can not see any reason why we can not make containers that will outlast this period.

Reply »

AD Rice

(April 1, 2020 7:44 PM)

This problem was solved in the late 50's. I saw the research. Bury in a salt dome, of which the U.S. has many. It is entombed and gone forever. All other "solutions" are just jobs programs.

Reply »

Joe Atkinson

(April 19, 2020 9:11 PM)

One solution is to load the waste in a rocket and shoot it into the sun. This is currently unacceptable because of the fear that a rocket malfunction and drop a load of radioactive waste back on earth.

A scientific solution is to find a controlled way of doing nuclear reactions on a kilogram scale and so convert radioactive waste into non-radioactive elements. I am an organic chemist and have no idea of even where to start.

Reply »

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