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Shifting Radioactivity Risks

A Case Study of Waste Management at the Fernald Nuclear Weapons Site

BY ANNIE MAKHIJANI
AND ARJUN MAKHIJANI[1]

Since the late 1990s, the U.S. Department of Energy has produced several versions of "accelerated" cleanup schemes of its nuclear weapons sites. This has led to more rapid decommissioning of two large weapons plants – notably the Rocky Flats Plant in Colorado, where plutonium pits for nuclear weapons were mass produced, and the Fernald Plant near Cincinnati, Ohio, where uranium metal was produced, mainly for plutonium production reactors. These sites are considered flagship sites in the federal government's effort to "clean up" and "close" some of the most contaminated sites in the country.

"Accelerated" has not necessarily meant better or lower-risk. To examine the effects of hasty cleanup, where timetables were tied to bonuses without reference to radiation doses to future generations, we did a case study of some radioactive waste at the Fernald plant.

The specific wastes we studied derived from the extraction of high-grade uranium ores. Some of these ores were processed during the World War II Manhattan Project and in the immediate post-war years at the Mallinckrodt Chemical Works in St. Louis. Others were processed at Fernald. Besides uranium itself, uranium ore contains thorium-230 and radium-226, which are decay products of uranium-238, the main isotope of natural uranium. (See uranium decay chain on page 6.) Since the ores in question were high-grade, they were also concentrated in radium-226 and thorium-230, which are long-lived, bone-seeking radionuclides. The highest grade ores, up to two-thirds uranium oxide content, were known as "pitchblende" and came from the "Belgian Congo," so called because the region was then a Belgian colony.

PHOTO COURTESY FERNALD AND DOE OHIO FIELD OFFICE

A Fernald worker packages thorium-containing radioactive waste for shipment to the Envirocare facility in Utah (now called EnergySolutions). Disposal at this facility is problematic due to a loophole in nuclear waste regulations.

The wastes from the processing of uranium ores and uranium concentrates (yellow cake) were stored in three large concrete structures called silos. Silos 1 and 2, which were emptied and demolished last year, stored radium-containing wastes from the processing of the "Belgian Congo" pitchblende. Silo 1 contained only waste produced at Mallinckrodt. Silo 2 contained waste produced at Mallinckrodt and Fernald. Silos 1 and 2 were also known as the "K-65" silos after the name of the process used to extract the uranium from the ore.

Silo 3, which also has been emptied and demolished, contained radioactive waste from the processing of uranium concentrates. Silo 3 wastes are less radioactive than the K-65 silo waste and have far more thorium-230 than radium-226.

This article examines the silo cleanup decisions made by the U.S. Department of Energy (DOE), how they changed over time, how they were implemented, and what their long-term radiological implications are likely to be. It summarizes our August 2006 report, *Shifting Radioactivity Risks: A Case Study of the K-65 Silos and Silo 3 Remediation and Waste Management at the Fernald Nuclear Weapons Site*. The full report, including references for

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this article, can be found online at www.ieer.org/reports/fernald/.

We have chosen to study the significant problems and failures associated with the management of these wastes because they illustrate problems in remediation and long-term stewardship that hold lessons for other sites.

The Fernald site

The Feed Materials Production Center (FMPC), now called the Fernald Closure Project Site, covered 1,050 acres and operated from 1952 to 1989. During these years it produced about 1.3 billion pounds of uranium metal to support the U.S. nuclear weapons program. Its main function was to provide reactor fuel and target rods for plutonium production at the Hanford Plant in Washington state and the Savannah River Plant in South Carolina. Fernald operations resulted in the contamination onsite and offsite of air, soil and water.

The radium and thorium waste that was stored in the three silos posed the most immediate risk to the workers and the offsite residents in two ways: (1) radon emissions, created by the decay of radium-226 into radon-222, and (2) the potential for the roofs of Silos 1 and 2 to collapse and release radioactive waste into the environment. Emptying the silos and putting the waste into a form that would not be prone to dispersal in the short term and that would be stable in the long term was recognized to be a challenging and critical task in the decommissioning of Fernald.

The silo waste

Table 1 on page 4 shows the concentrations of the various radionuclides in Fernald silo waste. Table 2 shows the silos' radioactivity content from some radionuclides. The contents of Silos 1 and 2 were similar because they were generated from high-grade ores. It is important to note that while the concentration of radium-226 is far higher than that of thorium-230, the latter has a much longer half-life — 1,600 years and about 75,000 years, respectively.

Moreover, thorium-230 decays into radium-226, so over a period of a few thousand years their concentrations become approximately equal. They are approximately in equilibrium at times far longer than the half-life of radium-226 but far shorter than that of thorium-230. Hence the half-life of thorium-230 controls the rate of decay of the waste over the very long term. Moreover, when inhaled, the radiation doses to most organs are considerably larger per unit of radioactivity for thorium-230 than for radium-226.

"Accelerated" has not necessarily meant better or lower-risk.

The main radionuclide of concern for Silo 3 waste is thorium-230. However, radium-226 will build up over the millennia to approximately equal the concentration of thorium-230 for a time. Silo 3 also contains arsenic, cadmium, chromium, and selenium. It is common for ore processing wastes to contain significant heavy metal contamination. Lead-210 and polonium-210 are decay products of radon-222, which is a decay product of radium-226.

Silo waste classification and disposal

The waste in all three silos is classified under the Atomic Energy Act as byproduct material resulting from the processing of uranium or

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6935 Laurel Avenue, Suite 201
Takoma Park, MD 20912, USA
Phone: (301) 270-5500
FAX: (301) 270-3029
E-mail: ieer@ieer.org
Web address: www.ieer.org

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Principles for Safeguarding Nuclear Waste at Reactors

The following principles are based on the urgent need to protect the public from the threats posed by the current vulnerable storage of commercial irradiated fuel. The United States does not have a near-term solution for the permanent storage of high-level nuclear waste. The proposed Yucca Mountain site is unsafe for geologic storage of nuclear waste and the program remains mired in bad science, mismanagement, and yet another design overhaul. Even if licensed, Yucca Mountain could not legally contain all of the waste produced by existing reactors. Under the U.S. Department of Energy's unrealistically optimistic scenario, Yucca Mountain is not predicted to begin receiving waste until at least 2017 and transporting waste to the site would take more than 30 years. Meanwhile, irradiated fuel at reactor sites remains vulnerable to accidents and attacks.

The Institute for Energy and Environmental Research, along with a number of other national and grassroots groups, has endorsed this set of basic principles on how to deal with spent nuclear fuel in the near term. The statement was coordinated by Michele Boyd of Public Citizen with input from many groups and individuals. To sign on your organization or your local or state government, contact her at mboyd@citizen.org. The full list of signatories is available at www.citizen.org/documents/PrinciplesSafeguardingIrradiatedFuel.pdf

The undersigned organizations' support for improving the protection of radioactive waste stored at reactor sites is a matter of security and is in no way an indication that we support nuclear power and the generation of more nuclear waste.

- **Require a low-density, open-frame layout for fuel pools:** Fuel pools were originally designed for temporary storage of a limited number of irradiated fuel assemblies in a low density, open frame configuration. As the amount of waste generated has increased beyond the designed capacity, the pools have been reorganized so that the concentration of fuel in the pools is nearly the same as that in operating reactor cores. If water is lost from a densely packed pool as the result of an attack or an accident, cooling by ambient air would likely be insufficient to prevent a fire, resulting in the release of large quantities of radioactivity to the environment. A low-density, open-frame arrangement within fuel pools could allow enough air circulation to keep the fuel from catching fire. In order to achieve and maintain this arrangement within the pools, irradiated fuel must be transferred from the pools to dry storage within five years of being discharged from the reactor.
- **Establish hardened on-site storage (HOSS):** Irradiated fuel must be stored as safely as possible as close to the site of generation as possible. Waste moved from fuel pools must be safeguarded in hardened, on-site storage (HOSS) facilities. Transporting waste to interim away-from-reactor storage should not be done unless the reactor site is unsuitable for a HOSS facility and the move increases the safety and security of the waste. HOSS facilities must not be regarded as a permanent waste solution, and thus should not be constructed deep underground. The waste must be retrievable, and real-time radiation and heat monitoring at the HOSS facility must be implemented for early detection of radiation releases and overheating. The overall objective of HOSS should be that the amount of releases projected in even severe attacks should be low enough that the storage system would be unattractive as a terrorist target. Design criteria that would correspond to the overall objective must include:
  - Resistance to severe attacks, such as a direct hit by high-explosive or deeply penetrating weapons and munitions or a direct hit by a large aircraft loaded with fuel or a small aircraft loaded with fuel and/or explosives, without major releases.
  - Placement of individual canisters that makes detection difficult from outside the site boundary.
- **Protect fuel pools:** Irradiated fuel must be kept in pools for several years before it can be stored in a dry facility. The pools must be protected to withstand an attack by air, land, or water from a force at least equal in size and coordination to the 9/11 attacks. The security improvements must be approved by a panel of experts independent of the nuclear industry and the Nuclear Regulatory Commission.
- **Require periodic review of HOSS facilities and fuel pools:** An annual report consisting of the review of each HOSS facility and fuel pool should be prepared with meaningful participation from public stakeholders, regulators, and utility managers at each site. The report must be made publicly available and may include recommendations for actions to be taken.
- **Dedicate funding to local and state governments to independently monitor the sites:** Funding for monitoring the HOSS facilities at each site must be provided to affected local and state governments. The affected public must have the right to fully participate.
- **Prohibit reprocessing:** The reprocessing of irradiated fuel has not solved the nuclear waste problem in any country, and actually exacerbates it by creating numerous additional waste streams that must be managed. In addition to being expensive and polluting, reprocessing also increases nuclear weapons proliferation threats.

thorium ores. Generally this means the tailings or waste produced by the extraction of uranium or thorium from their ores. Byproduct waste is exempt from RCRA (Resource Conservation and Recovery Act) regulation even though all three silos contain toxic metals at concentrations exceeding the RCRA limits. However, the fact that the tailings are exempt from RCRA does not mean that these toxic metals are not going to adversely affect the health of future generations. On the contrary they are more likely to do so since the exemption makes lax waste disposal more likely.

Mill tailings from U.S. ores generally contain relatively low concentrations of radioactivity compared to other radioactive wastes, but represent the largest volume of such wastes (with the possible exception of uranium mining wastes). That is because U.S. uranium ores have typically been of rather low uranium content — less than one percent. By contrast, the high-grade ores — including pitchblende, from which the Silos 1 and 2 waste came – have up to about 65 percent uranium oxide content resulting in relatively small volumes of waste with high concentrations of thorium-230, radium-226 and the solid decay products of radium-226. Table 3 compares the concentration of radioactivity in tailings from 0.2% ore to the waste in the silos.

Fernald operations resulted in the contamination onsite and offsite of air, soil and water.

Mill tailings have the most lax procedures for disposal of any radioactive wastes largely because they are typically very voluminous and have low specific activity (radioactivity per unit weight). They are required to be put in lined tailings ponds that have a cover of water to reduce radon air emissions.

The management of typical mill tailings is not protective of public health in the long term due to the very long half-life of thorium-230. However, it is broadly comparable to the category of radioactive waste called Class A low-level waste so long as the *combined* concentration of uranium, thorium-230, and radium-226 (the long-lived, alpha-emitting materials in tailings) remains below 10 nanocuries per gram.

As can be seen from Table 3, the concentration of typical U.S. mill tailings is comparable to that of Class A waste, which restricts plutonium and other transuranic radionuclides to under 10 nanocuries per gram. That is not the case for the waste in the three Fernald silos. The waste in Silos 1 and 2 even exceeds 100 nanocuries per gram of alpha-emitting, long-lived radionuclides, though they are not transuranic radionuclides. Rather, they are decay products of uranium-238 and uranium-235. One hundred nanocuries per gram is the upper bound for the content of long-lived, alpha-emitting transuranic radionuclides

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TABLE 1: ESTIMATED RADIONUCLIDE CONTENT OF SILOS 1, 2, AND 3 WASTE

Radionuclide	Mean Concentration, picocuries per gram		
	Silo 1 (V=3,240 m^3)	Silo 2 (V=2,845 m^3)	Silo 3 (V=3,890 m^3)
Actinium-227	5,960	5,100	618
Lead-210	165,000	145,000	2,620
Polonium-210	242,000	139,000	Not given
Protactinium-231	Not given	2,350	487
Radium-226	391,000	195,000	2,970
Thorium-230	60,000	48,400	51,200
Uranium-234	800	961	1,480
Uranium-238	642	912	1,500

SOURCE: Adapted from the 1997 IEER report, *Containing the Cold War Mess*, on the Web at www.ieer.org/reports/cleanup/, citing D. Paine (Silos Project Manager), *Operable Unit 4: Project History and Status Presentation,* Fernald, OH: Meeting of Independent Review Team, November 14, 1996, pages 8 and 11.

NOTES: Volumes (V) for Silos 1 and 2 do not include 357 and 314 cubic meters (m^3), respectively, of bentonite clay added in the 1990s to reduce radon emissions. Bentonite clay was not added to Silo 3. There is a slight discrepancy between the volumes cited in Paine for Silos 1 and 2 (3,240 + 2,845 = 6,085 cubic meters) and the total volume listed in the Record of Decision (6,120 cubic meters).

TABLE 2: RADIOACTIVITY IN SILOS 1, 2, AND 3 WASTE

Radionuclide	Silos 1 and 2 combined	Silo 3
Lead-210	1,800 curies	4 curies
Radium-226	3,700 curies	26 curies
Thorium-230	600 curies	450 curies
Uranium	28 metric tons (See note)	about 20 curies

SOURCE: 1994 Record of Decision for OU4, and *Safety of the High-Level Uranium Ore Residues at the Niagara Falls Storage Site, Lewiston, New York* (Washington, DC: National Academy Press, 1995).

NOTE: 28 metric tons of natural uranium correspond to less than 20 curies.

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in Class C low-level waste. Waste with concentrations of long-lived, alpha-emitting transuranic radionuclides greater than 100 nanocuries per gram is called "Greater-than-Class-C" waste, which cannot be disposed via shallow land burial as a general matter. Rather, it must be put in a deep geologic repository, unless a special exemption from such disposal is granted by the Nuclear Regulatory Commission. (Further information about radioactive waste classifications is provided in the box below.)

The National Academy of Sciences is in general accord with our analysis. Referring to Silos 1 and 2 waste, it states that:

> Although radium is not a transuranic, the K-65 wastes produce a substantial external dose due to gamma-ray emission and the risks they pose may even exceed those posed by some transuranic wastes and are at least similar based on the intrinsic toxicity of the isotopes involved.[2]

According to U.S. regulations, transuranic wastes with a specific activity above 100 nanocuries per gram and half lives of more than 20 years must be disposed in a deep geologic repository.[3] The concentrations of radium-226 in Silos 1 and 2 waste are well above this. Moreover, the radiological properties of radium-226 (and also thorium-230) are similar to those of transuranic elements. Disposal in shallow land burial is therefore not in accord with the spirit of the regulations as they exist, even though there is no explicit classification for Silos 1 and 2 waste that would require deep geologic disposal.

Silo 3 waste has a concentration of thorium-230 that is

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TABLE 3: RADIOACTIVITY OF SILO WASTE COMPARED TO TYPICAL U.S. URANIUM MILL TAILINGS

Material	Concentration in nanocuries per gram	Ratio of silo waste/tailings
Mill tailings arising from 0.2% uranium ore	2 to 3.4	—
Silo 1 waste	452	133
Silo 2 waste	245	72
Silo 3 waste	54	16

NOTES: Mill tailings arising from 0.2% uranium ore are typical in the United States. Values for silo waste include only long-lived alpha-emitting radionuclides – U-238, U-234, Th-230, Ra-226, and Pa-231. For Silo 1, no Pa-231 value was available so the value for Silo 2 was used. U-235 was omitted because it does not play a significant radiological role in this context.

SOME CLASSIFICATIONS OF RADIOACTIVE WASTE

TRANSURANIC WASTE, also known as TRU waste, contains elements with atomic numbers greater than 92, the atomic number of uranium. TRU waste contains alpha-emitting transuranic radionuclides with half-lives greater than 20 years and total concentration greater than 100 nanocuries per gram. This is the U.S. Environmental Protection Agency definition. The U.S. Nuclear Regulatory Commission definition is slightly different and is part of a broader category called Greater-than-Class-C waste.

LOW-LEVEL RADIOACTIVE WASTE (LLW) is defined by what it is not. According to U.S. Nuclear Regulatory Commission (NRC) regulations, low-level waste is "radioactive waste not classified as high-level radioactive waste, transuranic waste, spent nuclear fuel, or by-product material [i.e., uranium or thorium mill tailings]…"

The "low-level" radioactive waste category thus includes everything from slightly radioactive trash (such as mops, gloves, and booties) to highly radioactive activated metals from inside nuclear reactors. It includes both short-lived and long-lived radionuclides.

NRC regulations sub-divide commercial low-level waste into four classes which are determined by the types of radionuclides and their concentrations which make up the waste. These classes are labeled Class A, Class B, Class C, and Greater Than Class C.

CLASS A waste is the least radioactive on average, and is contaminated primarily by what the NRC terms "short-lived" radionuclides.

CLASSES B AND C wastes are more radioactive: Class B may be contaminated with greater amounts of "short lived" radionuclides than Class A, and Class C with greater amounts of long-lived and short-lived radionuclides than Class A or B.

GREATER-THAN-CLASS-C waste is typically much more radioactive than the other classes, and generally is considered unacceptable for near-surface disposal, which is how Classes A, B, and C are generally disposed of in the U.S. Shallow-land disposal used to be simple dumps mainly, but the concept now also includes more elaborate structures.

Sources: Makhijani and Saleska, *High-Level Dollars, Low-Level Sense,* IEER (New York: Apex Press, 1992), and *Classifications of Radioactive Waste*, IEER On-Line Classroom, last updated April 29, 1996, www.ieer.org/clssroom/r-waste.html.

far larger than the 10 nanocuries per gram limit for Class A low-level waste and therefore should not be disposed of without the precautions for Class B low-level waste or in a facility that is not licensed for Class B waste.

> Mill tailings have the most lax procedures for disposal of any radioactive wastes

The management of the silo wastes at Fernald has been an abject failure when these considerations are taken into account. This conclusion is borne out by the detailed radiological assessment that we carried out, described starting on page 7. First we will outline a brief history of the silo cleanup program and its current status.

The silo remediation program: promises and reality

In 1994, DOE issued a Record of Decision (ROD) stating that the best option for treating the silo wastes was to put them into a melter similar to a glass melter, heat them, and vitrify them (convert them into a glassy material). The glassy blocks of silo waste would be disposed of at the Nevada Test Site. Another option considered but rejected was to mix the silo wastes with cement.

In the ROD, the comparison of the vitrification and cementation options for Silos 1 and 2 waste was discussed separately from that for Silo 3 waste because of differences in chemical composition. Vitrification of the waste from all three silos was selected as the best option because laboratory tests indicated that this would be the best way to prevent the migration of radionuclides into the environment. Other benefits cited in the ROD were waste reduction and the fact that cementation (mixing the waste with cement) did not reduce radon gas emissions as efficiently as vitrification.

At that time the cost for cementation for Silos 1 and 2 waste was assessed to be $73.1 million versus $43.7 million for vitrification. For Silo 3 it was $36.8 million versus $28 million. The lower cost for vitrification derived in large measure from the estimated savings in transportation cost due to the volume reduction produced by vitrification of the waste.

URANIUM DECAY CHAIN
(principal branch only)

Uranium-238
(half-life: 4.46 billion years)
alpha decay
⇩
Thorium-234
(half-life: 24.1 days)
beta decay
⇩
Protactinium-234m
(half-life: 1.17 minutes)
beta decay
⇩
Uranium-234
(half-life: 245,000 years)
alpha decay
⇩
Thorium-230
(half-life: 75,400 years)
alpha decay
⇩
Radium-226
(half-life: 1,600 years)
alpha decay
⇩
Radon-222
(half-life: 3.82 days)
alpha decay
⇩
Polonium-218
(half-life: 3.11 minutes)
alpha decay
⇩
Lead-214
(half-life: 26.8 minutes)
beta decay
⇩
Bismuth-214
(half-life: 19.9 minutes)
beta decay
⇩
Polonium-214
(half-life:
163 microseconds)
alpha decay
⇩
Lead-210
(half-life: 22.3 years)
beta decay
⇩
Bismuth-210
(half-life: 5.01 days)
beta decay
⇩
Polonium-210
(half-life: 138 days)
alpha decay
⇩
Lead-206
(stable)

But this scheme was not implemented due to a series of management and operations failures and bad decisions. The actual program that was implemented was substantially inferior to vitrification.

Whereas the vitrification of fission products from the reprocessing of spent fuel is performed on a routine basis, the vitrification of the type of waste in the silos was a more challenging problem because it had never been done before on a large scale. Fluor Fernald, the DOE contractor, decided to do laboratory experiments and then build a pilot plant to test the concept prior to large scale construction. However, poor design of the pilot plant and the failure to take into account the specific characteristics of Silo 3 waste, despite prior indications of possible problems, led to the failure of the melter before any radioactive waste was put into it.[4]

After the failure of the pilot plant, Fluor Fernald proceeded with the vitrification plan but decided, as it should have from the start, that the waste from Silos 1 and 2 should be vitrified separately from the waste from Silo 3. Thereafter, however, there was a steady deterioration in the silo remediation program.

The deterioration was intimately linked to DOE's "Accelerated Cleanup" program in the late 1990s, followed by its Top-to-Bottom Review in 2002, which moved the closure date of the Fernald site up to 2006 from its previous, less ambitious deadline of between 2006 and 2010. DOE's reasons for speeding things up were the need to address cost escalation and to remove the contamination in a timely manner for protecting human health and the environment. An additional reason proffered was that it would save money.

However, at the same time the schedule was accelerating, the site's long-term cleanup performance goals were essentially being abandoned. The bonus promised to Fluor was geared solely to the deadline for finishing the job. There were penalties for being late, but there was no bonus for better environmental protection nor any penalty for inferior long-term disposal performance.

The result was predictable. In stages, the vitrification program was abandoned for all three silos. Silos 1 and 2 wastes were cemented. The cementation program for the

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Silo 3 waste was abandoned and that waste was packaged — with minimal treatment for reducing dispersal potential — in large plastic bags (see cover photo).

The State of Nevada differed with DOE on whether the Silo 3 waste could be safely disposed at the Nevada Test Site and threatened to sue DOE if the waste was shipped there. Left without other disposal options, DOE shipped the Silo 3 waste to the Envirocare facility in Utah,[5] a low-level radioactive waste facility that is licensed to accept Class A low-level radioactive waste and byproduct material. Envirocare is allowed to accept byproduct material if the average concentration of the waste in the incoming trucks is below 4,000 picocuries per gram for radium-226, and below 60,000 picocuries per gram for thorium-230.

There is, to date, no final disposal solution for the waste from Silos 1 and 2.

Silo 3 waste meets Envirocare disposal criteria today for these two radionuclides. However, the license does not take into account that thorium-230 decays into radium-226. The waste will exceed the limit of 4,000 picocuries per gram radium-226 in about 50 years.[6] If it were shipped to the site at that time, the waste would no longer meet the disposal criteria.

Moreover, for most organs in the human body, the dose delivered per picocurie of thorium-230 ingested or inhaled is far greater than that of radium-226. For instance, the inhalation dose per picocurie to the bone surface or to the liver is about 300 times bigger for thorium-230 than for radium-226.

The Division of Radiation Control (DRC) in Utah, which granted Envirocare's license, has not carefully done its homework in creating a regulatory framework for Class A low-level waste that protects the health of future generations. The DRC has tried to make such problems go away by simply assuming that no one will ever go to the site, even thousands of years from now.

Estimated doses to future generations

We have done RESRAD calculations to verify whether shallow land burial of the silo waste would meet criteria set forth in federal rules that limit exposure to 25 millirem per year per person in the general population.[7] RESRAD is a government-approved computer program that allows the estimation of long-term health impacts of radiologically contaminated soil.

Silos 1 and 2

There is, to date, no final disposal solution for the waste from Silos 1 and 2. Because the waste far exceeds the 4,000 picocuries per gram limit for radium-226 for the Envirocare site, it could not be shipped to Utah. The cemented (grouted) wastes have been shipped for temporary storage to a facility in Texas owned and operated by Waste Control Specialists, which does not as yet have a license to dispose of radioactive waste.

In fact, Waste Control Specialists' ability to handle waste is in question because of egregious errors in its license application.[8] For instance, Waste Control Specialists (WCS), using DOE data, claims that Oak Ridge will ship it over 12,000 metric tons of uranium-235 as waste for disposal. This amount is more than the entire inventory of U-235 that has ever been mined worldwide. If WCS cannot detect obvious errors in waste data (and this was not the only absurd value in its license application), how can it be expected to, say, prevent illegal disposal of prohibited radioactive materials during operations?

We performed an assessment of radiation doses far into the future should the WCS site be inadvertently occupied or even used occasionally for such purposes as grazing or hunting. These are uses to which the site has been put in the past. Table 4 shows the estimated peak doses and when they will occur based on the erosion rate of the waste cover. This is for a "rancher scenario," which assumes a future person will spend a limited amount of time on the site and will not build or grow food on the site.

Fernald silo wastes should be disposed of in a deep geologic repository.

Depending on the erosion rate, the projected external dose to the maximally exposed person in the future is 346 to 800 times higher than the present regulatory limit of 25 millirem per year. Note that these peak doses are from the radium-226 originating from the decay of thorium-230. The original radium-226 in the waste will be almost completely gone after about 9,150 years and totally gone after about 91,500 years.

Silo 3

Silo 3 waste is slated to be disposed of at the Envirocare site in Utah. Table 5 on page 10 shows the doses estimated by RESRAD modeling of that waste. Kd is called the partition coefficient and refers to how tightly the contaminant is bound to the soil. As described in the

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TABLE 4: PEAK DOSES FROM GROUTED K-65 WASTE DISPOSAL – RANCHER SCENARIO

Erosion rate (cm per year)	Peak external dose (rem per year)	Peak inhalation dose (rem per year)	Years until peak dose
0.1	20.1	0.078	9,150
0.01	8.7	0.037	91,500

The Environmental Transport of Radium and Plutonium

BY BRICE SMITH

Understanding the mobility of radium and plutonium in the environment, especially through soil and into groundwater, is very important. Radium is one of the principal contaminants associated with uranium mining and milling and affects a large number of sites. It also affects thousands of secondary oil recovery sites, where radium pollution is quite common. Plutonium is one of the most long-lived and dangerous radionuclides in the nuclear weapons production process. Plutonium-contaminated wastes have been dumped into unlined disposal areas at several sites around the United States near critically important bodies of water.

Both radium and plutonium mobility vary widely depending on the circumstances. Estimates of radiation dose to future generations are highly dependent upon the assumptions about how contamination in the soil actually affects groundwater. IEER undertook a review of the environmental transport of radium and plutonium because of the scientific difficulty of the question as well as the widespread importance of these two radionuclides in cleaning up the messes from decades of uranium and plutonium processing. This piece summarizes that review.[1]

—Arjun Makhijani

Given the complex chemical, biological, and physical properties of soil, predicting the mobility of radionuclides is far from simple. There are a number of examples where models relied upon by the Atomic Energy Commission and later the Department of Energy failed to accurately predict contaminant transport. After the discovery of radionuclides spreading further and more rapidly into the environment, these models had to be fundamentally revisited. These failures were due in large part to the failure to adequately characterize the systems.

In real systems there may be chemical or biological processes that affect the mobility of contaminants that may vary over space and time. There may also be more pathways for radionuclides to move than originally expected. Finally, the transport model itself might be adequate, but information on what parameters to use may not be available.

This article provides a brief review of the transport of two specific radionuclides, radium and plutonium. Radium is a naturally occurring radionuclide that is part of the uranium and thorium decay series (see uranium decay chain on page 6). In the body, radium is a calcium analog, and goes primarily to the bone. Plutonium is a human-made radionuclide produced in nuclear reactors. Inside the body, plutonium also preferentially goes to the bone.

A number of important site-specific properties can either enhance or retard the mobility of both radium and plutonium. As a result, detailed, site-specific analyses are essential. Performance assessments which are predicated on simplified models or default values should not be accepted for demonstrating compliance with regulatory limits. Transport modelers should seek to learn from past surprises. This is particularly true for radium, plutonium, and many of the other transuranic elements (elements with an atomic number greater than uranium), given the long half lives of many of these radionuclides and their parents.

The Kd approach

Many contaminants, including radium and plutonium, can adsorb onto soil through ion exchange. The strength of this interaction is quantified by the partition coefficient (Kd). The partition coefficient relates the concentration of a contaminant adsorbed onto the soil to that dissolved in the water after the system has reached equilibrium.

$$Kd = \frac{\text{concentration of contaminant in soil (pCi/kg)}}{\text{concentration of contaminant in water (pCi/L)}}$$

The partition coefficient therefore has the somewhat unusual units of liters per kilogram (L/kg) or, equivalently, milliliters per gram (mL/g). A large value for the partition coefficient implies that the contaminant is tightly bound to the soil and will therefore migrate slowly. A small value implies the opposite. Due to its relative simplicity, the constant Kd approach is the most widely used transport model today. It is called the "constant" Kd approach because the model assumes that an entire site can be characterized by one value of Kd that does not change over time.

Limitations of the Kd model

Despite its widespread use, there are a number of important limitations of the constant Kd approach. Most importantly, the value of Kd is strongly dependent on local chemical and physical conditions and can thus vary greatly between sites and even across a single site. The values of Kd for plutonium measured at the Hanford site, for example, vary by more than a factor of 1,000.

This variation is because the adsorption of contaminants, like radium or plutonium, changes depending on various factors. These factors include the action of plants and bacteria; the oxidation state of the radionuclides, which can change over time (a particular concern for plutonium, which can exist in four different states); and the amount of clay, sand and organic matter in the soil. Specifically, the adsorption of contaminants decreases as the acidity of the soil increases and as the concentration of chemically similar ions in the system increases.

Even the U.S. Environmental Protection Agency (EPA) explicitly acknowledges that default or generic Kd values can result in "significant error" when used to predict contaminant migration.[2]

An additional limitation is that there are five methods for measuring Kd. As noted by EPA, "it is not only common, but expected that Kd values measured by different methods will produce different values."[3] For radium, additional uncertainties arise when other alkaline earth metals are present during measurement because co-precipitation can lead to erroneously large estimates of Kd.

More sophisticated models have been developed that overcome some of the limitations of the constant Kd approach. These models have more successfully described migration involving lead contamination and sulfate contamination at uranium mines. The challenge of these newer models is that they require significantly more information about the site.

Other transport pathways

In some cases, other pathways and natural phenomena can be important. Sulfate-reducing bacteria can cause radium precipitates to dissolve, thereby enhancing mobility, while the bacteria *Bacillus subtilis* has been found to impact the mobility of plutonium. In addition, animals cause soil mixing, which can enhance near-surface contaminant migration. Finally, the similarity of radium to calcium may cause it to bioconcentrate as it is transferred up the food chain.

One of the important processes that can affect the transport of plutonium and other transuranic elements is adsorption on colloidal particles. Colloidal particles are small particles that occur naturally and are easily suspended in ground and surface water. The potential for colloidal transport to affect the mobility of contaminants like plutonium was recognized more than 50 years ago, but the interest in this phenomenon has grown since the discovery of rapid colloid transport of plutonium at the Nevada Test Site in the late 1990s. The impact of colloid transport is highly site-specific, and there is evidence both supporting and questioning the importance of colloid-mediated transport at different DOE facilities.

In addition, plutonium adsorbed onto sediment may become mobilized by resuspension. Likewise, contamination bound to surface soil can become mobilized through erosion. The transport of plutonium via surface water and erosion is known to be particularly important at Los Alamos National Laboratory. The significance of this pathway has increased since the May 2000 Cerro Grande fire that burned approximately 43,000 acres in and around the lab. The loss of vegetation has led to increased erosion, particularly during the flooding that follows storm events.

Transport modelers should seek to learn from past surprises.

Finally, at sites contaminated with transuranics, it is important to consider decay and ingrowth of elements which may have significantly different mobility or half lives than their parents.

Conclusions and recommendations

One of the major recommendations by the EPA in a 2004 review of radium's mobility was that "for site-specific calculations, partition coefficient values measured at site-specific conditions are absolutely essential."[4] (Emphasis in the original.) Similar conclusions are reasonable for plutonium. Assessments that are predicated on default values of the partition coefficient should not be accepted for regulatory purposes. At a minimum, an effort should be undertaken to determine a well-founded, site-specific value for Kd.

While in many cases a suitable Kd value can be determined, consideration should be given to the use of more complex transport models in light of the well-known limitations of the Kd approach, especially at sites with highly concentrated and reactive wastes. In cases where erosion and surface water transport or colloid-mediated transport are potentially important, a model capable of including these pathways should be used, particularly in light of the relatively rapid migration of plutonium already observed at some sites.

1 Brice Smith and Alexandra Amonette, *The Environmental Transport of Radium and Plutonium: A Review,* Takoma Park, MD: Institute for Energy and Environmental Research, June 23, 2006. Available on the Web at www.ieer.org/reports/envtransport/.

2 *Understanding Variation in Partition Coefficient, Kd, Values: Volume I: The Kd Model of Measurement, and Application of Chemical Reaction Codes.* Office of Radiation and Indoor Air, Office of Solid Waste and Emergency Response, U.S. Environmental Protection Agency [and] Office of Environmental Restoration, U.S. Department of Energy, Washington, DC, August 1999. (EPA402-R-99-004A). Page 1.1.

3 ibid., p. 3.1.

4 *Understanding Variation in Partition Coefficient, Kd, Values. Volume III: Review of Geochemistry and Available Kd Values for Americium, Arsenic, Curium, Iodine, Neptunium, Radium, and Technetium.* Office of Air and Radiation. July 2004 (EPA402-R-04-002C), p. 5.67.

centerfold on pp. 8-9, the value for Kd can vary dramatically among sites and even within a site.

The more rapid approach of peak doses for Silo 3 waste is due to a less thick cover over the waste than at the WCS site. The external dose from radium-226 dominates in this case as well.

The estimated external doses from the Silo 3 waste, in the 250 rem range, are 10,000 times bigger than the dose limit of 25 millirem. Hence, if a hunter or recreational vehicle user or other visitor in the far future spends just one hour at the site, he or she will have exceeded the allowable dose limit.

These two sets of dose estimates illustrate why the Fernald silo wastes should be disposed of in a deep geologic repository. While there would still be some risks with a deep geologic repository, the risks of exceedingly high doses from shallow land burial would not occur.

PHOTO COURTESY FERNALD AND DOE OHIO FIELD OFFICE

The Fernald site from above. *The very large rectangular area at the top right is the On-Site Disposal Facility, which contains about 2.5 million cubic yards of radioactively contaminated soil and demolition debris.*

IEER's proposal for Silos 1 and 2 waste

Our RESRAD calculations indicate that WCS is a poor site for burial of the wastes from Silos 1 and 2. It is highly unlikely that any shallow land burial site would be able to meet the dose limits set by the regulations. Wet sites would deliver high doses mainly from the water pathway, while dry sites deliver high doses mainly due to the uncovering of waste by erosion. In the latter case, the principal component is external dose from radium-226.

The estimated external doses from the Silo 3 waste are 10,000 times bigger than the dose limit.

The question of the disposal of the K-65 silos waste bears a lot of similarities to the question of the disposal of depleted uranium that IEER has extensively addressed.[9] It is IEER's scientific conclusion that both should be buried in a deep geologic repository since shallow land burial is highly unlikely to meet compliance dose criteria.

In the interim, until a repository disposal option is developed, the grouted K-65 waste should be put into monitored storage. After that, it should be disposed of in a deep geologic repository with depleted uranium from past U.S. uranium enrichment activities. There is at present no deep geologic repository for the vast amounts of DU from uranium enrichment plants.

The volume of the DU would be several hundred thousand cubic meters. This is much larger than the present grouted volume of waste from Silos 1 and 2. Even with additional packaging to make the cement blocks compatible with future repository waste acceptance criteria, the Silos 1 and 2 waste would remain small compared to the DU waste. As a result, the marginal cost would not be high, though the average disposal cost will likely be many times the shallow land burial cost.

Other challenges at Fernald

The last two issues we will discuss are important because parts of the Fernald site will remain contaminated for tens of thousands of years to come. The issues are (a) residual radioactivity at the On-Site Disposal Facility and (b) permanent monitoring and community education.

The On-Site Disposal Facility (OSDF), or "waste cell," is

TABLE 5: PEAK DOSES FOR SILO 3 WASTE DISPOSAL – RANCHER SCENARIO

Kd	Peak external dose (rem per year)	Peak inhalation dose (rem per year)	Years until peak dose
Low Kd, HIR, HC	245	0.698	6,800
High Kd, LIR, LC	273	0.756	9,000

NOTES: HIR stands for high infiltration rate, HC for high conductivity, LIR for low infiltration rate, and LC for low conductivity. We have chosen to pair the low Kd with the high infiltration rate and the high conductivity, and the high Kd with the low infiltration rate and the low conductivity in order to obtain the lowest and the highest leaching of radionuclides from the disposal cell. The results were the same for both high and low erosion rates.

SEE **FERNALD** ON PAGE 11, ENDNOTES, PAGE 12

MAIN FINDINGS

1. Mismanagement and design flaws led to the failure of the vitrification program for silo wastes at Fernald. Instead of fixing the management and design, the U.S. Department of Energy (DOE) decided to change the waste forms, thereby significantly degrading the expected long-term performance.
2. The changes in waste form and expected performance have resulted in an uncertain future for the processed wastes from Silos 1 and 2. If the grouted waste is disposed of by shallow land burial, the long-term estimated doses will be far in excess of the allowable regulatory limit of 25 millirem per year.
3. Silo 3 waste is estimated to produce radiation doses in the long term that would be far in excess of legally allowable limits, even though the waste is far less radioactive than Silos 1 and 2 waste. The long-term build-up of radium-226 to high levels (far higher than allowed in waste at the time of disposal at Envirocare) is a significant part of the problem.
4. The large increases in the cost of disposal, despite the significant degradation of performance, do not appear to have any readily identifiable engineering basis. The cost changes made by the DOE and its contractor were not transparent in their technical justifications.
5. Expediency and short-term gain have driven the process of decision-making about the waste form, resulting in the sacrifice of long-term performance. The DOE's failure to include long-term waste form performance in its decision-making for bonuses created a perverse incentive to finish rapidly at the expense of long-term health and environmental protection.
6. The DOE altered and loosened the remediation goal for uranium contamination of groundwater at Fernald after remediation commenced because the U.S. Environmental Protection Agency promulgated a more lax standard in the year 2000 than the remediation goal that had been set earlier.
7. The Department of Energy has abandoned its commitment to the community to provide guaranteed funding for an education program for the community as part of its Natural Resource Restoration Plan. This has been regarded as essential, since the community accepted that a very large volume of low level radioactive waste could be disposed of in an on-site waste cell. This backtracking on long-term stewardship parallels the degradation of waste form choice for the silo wastes.
8. The Waste Acceptance Criteria for the On-Site Disposal Facility at Fernald were not subjected to adequate quality assurance.

RECOMMENDATIONS

1. Contractor bonuses should not be tied to schedule alone. Long-term radiation dose consequences for all communities affected by the waste, including those to which waste is shipped, should be central to the contracting process.
2. State and local governments and communities should have stronger legal leverage to prevent the U.S. Department of Energy (DOE) from degrading performance or cleanup goals once a Record of Decision (ROD) is issued.
3. Shallow land burial of Fernald's Silos 1 and 2 waste should not be permitted. This waste should be disposed of in a deep geological repository. It can be co-disposed of with the depleted uranium (DU) waste resulting from historical uranium enrichment operations, which also need repository disposal. (This DU waste, currently 740,000 tons in unstable hexafluoride form, are stockpiled at three DOE sites in Kentucky, Ohio and Tennessee awaiting a disposal plan. See SDA vol. 13 no. 2, June 2005, for details.) Because the volume of the Silos 1 and 2 waste is far smaller than the depleted uranium waste inventory and because the specific activities of each waste type are comparable, the silo waste is unlikely to significantly add to radiation doses resulting from DU disposal.
4. The DOE should be required to create a permanent fund for monitoring and education large enough for the interest to be sufficient to cover annual costs for the programs. The federal government must disburse the principal amount to state and local governments up front, with provisions for openness and community participation built into the fund.
5. The federal government should be liable for the costs of legal proceedings, including litigation, arising from a demonstrated failure of the DOE to live up to its cleanup or long-term stewardship commitments.
6. An independent quality assurance program for computer programs and for the calculations done using those programs, including input parameters and software logic, is needed to ensure that cleanup decisions are technically sound. These programs include those used to estimate the total burden and the maximum residual radionuclide concentrations of radionuclides that can be put in waste cells. Specifically, the computer program and the input parameters need intensive verification. There is very likely to be at least one major error in the software that needs correction. A new performance assessment for the Fernald On-Site Disposal Facility should be performed once this independent quality assurance is done.
7. The state of Utah should tighten its rules for waste acceptance so that the total amount of radium disposed of in shallow land at any time remains less than 4,000 picocuries per gram after its in-growth from thorium-230 has been taken into account.

FERNALD
FROM PAGE 10

a lined, landfill-like structure located permanently on the Fernald site. (See photo.) It contains about 2.5 million cubic yards of soil and demolition debris contaminated with low-level radioactive waste. The main problem is that, in estimating allowable residual radioactivity in the waste cell, DOE used a computer program that appears to have a serious systematic error. Specifically, we found that DOE estimated that *four grams* of neptunium could be left as residual radioactivity *per gram* of soil (total weight). This is of course physically impossible. These calculations must be checked and redone after the computer program has been debugged. The publication of an absurd residual radioactivity value indicates a lack of quality control that raises questions about the rest of the effort.

Further, the DOE went back on its promise to provide obligatory funding for an education program that would keep the public informed about the waste cell and residual radioactivity far into the future. The State of Ohio is a natural resources trustee of the site under the law. Funding to the site for damage assessment is required under the Superfund law under such circumstances, but the DOE is not providing it. The Ohio Attorney general has protested and the matter was under negotiation at the time this issue went to print.

Our main findings and recommendations for Fernald silo waste management, waste cell problems, and monitoring and education funding can be found on the opposite page.

1 Based on Annie Makhijani and Arjun Makhijani, *Shifting Radioactivity Risks: A Case Study of the K-65 Silos and Silo 3 Remediation and Waste Management at the Fernald Nuclear Weapons Site* (IEER: Takoma Park, MD, August 2006). Detailed references are to be found in the full report, which is on the Web at www.ieer.org/reports/fernald/.
2 National Research Council, *Risk and Decisions About Disposition of Transuranic and High-Level Radioactive Waste,* Committee on Risk-Based Approaches for Disposition of Transuranic and High-Level Radioactive Waste, Board on Radioactive Waste Management, Division on Earth and Life Studies, National Research Council of the National Academies. Washington, DC: National Academies Press, 2005. On the Web at www.nap.edu/books/0309095492/html.
3 40 CFR 191.02(i).
4 For a detailed review of the failure of the melter, see Chapter 4 of IEER's report, *Containing the Cold War Mess,* on the Web at www.ieer.org/reports/cleanup/ccwm.pdf.
5 The company is now called "EnergySolutions."
6 It will reach about 50,000 picocuries per gram in about 8,000 years.
7 10 CFR 61 subpart C.
8 These errors are documented in the IEER report, *Costs and Risks of Management and Disposal of Depleted Uranium from the National Enrichment Facility Proposed to be Built in Lea County New Mexico by LES,* online at www.ieer.org/reports/du/LESrptfeb05.pdf.
9 ibid., section II.A.

Sharpen your technical skills with Dr. Egghead's

Atomic Puzzler

Calculating CO_2 emissions from a coal-fired power plant

Dr. Egghead's dog, Gamma, is back from a long hiatus studying at obedience school. While there, he heard a lot about global warming and is quite concerned about it. Gamma needs your help figuring out how much carbon dioxide, a major contributor to global warming, is emitted by nuclear power plants compared to fossil fuel fired power plants.

You and Gamma will tackle this problem over the course of several Atomic Puzzlers. In this one, you will calculate the carbon dioxide (CO_2) emissions from a pulverized coal-fired plant. In the next one, you will calculate CO_2 emissions from a natural gas fired power plant. In the Atomic Puzzler after that, you will at last calculate CO_2 emissions from a nuclear power plant — well, actually from the enrichment of the uranium fuel that is used in that plant. (Whew! This global warming stuff is a lot of work!)

In this problem, we will be calculating the CO_2 emitted directly from the coal-fired power plant — that is, the amount of CO_2 released as a result of burning the coal. There are additional indirect emissions associated with the mining and transportation of the fuel and the construction of the power plant. For fossil fuels, the direct CO_2 emissions are dominant. Good luck!

1. The complete combustion of one metric ton of coal releases 22.88 million British thermal units (MMBtu) of energy. One Btu is equal to 1,055 joules. How many joules of energy would be released by the complete combustion of one kilogram of coal? (Hint: One metric ton is equal to 1,000 kilograms.)

2. The watt is defined at one joule per second. How many joules are there in one kilowatt-hour? (Hint: There are 1,000 watts in one kilowatt)

3. How many kilowatt-hours (thermal) would be released by the complete combustion of one kilogram of coal? (Hint: You will need to use the answers to (1) and (2).)

4. The efficiency of a modern pulverized coal fired plant is typically about 34 percent. That is, 34 percent of the energy released by burning the coal is ultimately converted into electrical energy while the remaining 66 percent is wasted as heat and other types of energy losses. How many kilograms of coal would need to be burned in such a plant to produce one kilowatt-hour of electricity? (Hint: For a plant with this efficiency, how many kilowatt-hours of thermal energy would have to be released by the burning coal in order to generate one kilowatt-hour of electricity?)

5. Coal in the United States is 61 percent carbon (by mass) on average. How many kilograms of carbon would be released by such a plant in producing one kilowatt-hour of electricity?

6. When the coal is burned, the additional weight of the oxygen means that every kilogram of carbon emitted is equivalent to 3.67 kilograms of carbon dioxide (CO_2). How many grams of CO_2 are emitted by a coal fired plant per kilowatt-hour of electricity generated?

Send us your answers via e-mail (ieer@ieer.org), fax (1-301-270-3029), or snail mail (IEER, 6935 Laurel Ave., Suite 201, Takoma Park, Maryland, 20912, USA), postmarked by November 30, 2006. IEER will award a maximum of 25 prizes of $10 each to people who send in a completed puzzler, by the deadline, right or wrong. One $25 prize will be awarded for a correct entry, to be drawn at random if more than one correct answer is submitted. International readers submitting answers will, in lieu of a cash prize (due to exchange rates), receive a copy of IEER's newest and hottest book, *Insurmountable Risks: The Dangers of Using Nuclear Power to Combat Global Climate Change* (IEER Press and RDR Books, 2006).

It pays to increase your jargon power with Dr. Egghead

Actinides

a. Also "Act in Ides." In Roman times, a mandatory service a subject had to give the emperor on the 15th of every month.

b. Children who frequently throw temper tantrums and otherwise act up.

c. Elements with atomic numbers between 89 and 103, e.g. uranium, thorium and plutonium, neptunium, americium, etc.

Radioactive decay

a. Also known as "Active decay on the radio," the increasing use of foul language by some disk jockeys.

b. The aging of the world's cadre of nuclear physicists.

c. The spontaneous transformation of an unstable atom into one or more different nuclides accompanied by either the emission of energy and/or particles from the nucleus, nuclear capture or ejection of orbital electrons, or fission. Unstable atoms decay into a more stable state, eventually reaching a form that does not decay further or has a very long half-life.*

Decay products

a. In medieval times, the Lords' rotten leftovers given to his subjects.

b. The stuff at the bottom of a compost heap.

c. A series of radionuclides formed by the nuclear transformation of another radionuclide which, in this context, is referred to as the parent.*

Decay chain

a. Necklace made of very ripe fruit.

b. Bindings made with biodegradable materials used in prisons long ago. Once degraded, the prisoner was free. The time the chain took to decay matched the sentence of the prisoner.

c. A serial decay relationship where a parent radionuclide decays to one or more radioactive progeny, which in turn decay to form a third, fourth, or more generation of radioactive progeny. The final decay product in the series will be a stable element or an element with an extremely long half-life.*

Precipitation

a. Very wet sweat.

b. What happens before cipitation.

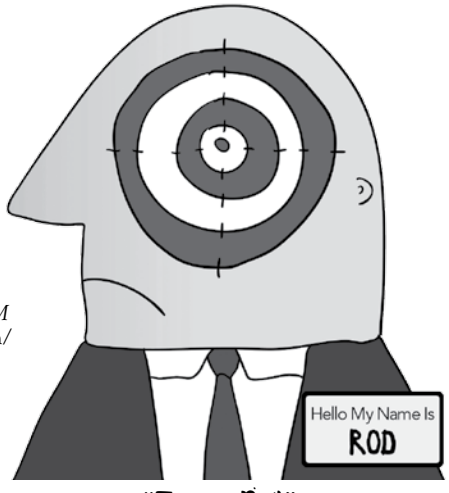
c. The production of an insoluble solid from a solution during a chemical reaction.

Target rods

a. In the animal kingdom, the male rep[Censored]ans.

b. Souped-up vehicles that are prone to being crashed into by other vehicles.

c. Specialized rods that are bombarded with neutrons. For instance, uranium-238 is converted into plutonium for use in nuclear weapons. Uranium-238 target rods were used for this purpose in some military production reactors, like at the Savannah River Site in South Carolina.



"Target Rod"

*Source for three definitions: Argonne National Laboratory, *MARSSIM Glossary/Acronyms,* on the Web at http://web.ead.anl.gov/marssim/acrogloss/

Answers: c,c,c,c,c,c

Thank you very much.

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