The RadioActivist Campaign

Comment: to the Blue Ribbon Commission (BRC) of the DOE

Title: **Concern for unassessed, local failure of a repository in salt**

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Summary

What if WIPP's first 11 years of operational experience might reveal prospects for long-term integrity of a repository in geologically stable salt beds? Here are scenarios of ***local*** **failure** of containment, based on an example of K Basin sludge from Hanford. *Local* scenarios alter the risks enough to warrant performance assessment.

This conclusion implies that adequate placement of wastes in geologically stable salt beds is unexpectedly complicated, along with concern that the envelope of performance assessments for WIPP is insufficiently comprehensive. Finding an adequate plan for the back end of America's nuclear fuel cycle will, thus, be even more challenging than it has appeared.

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Problem Statement

While the front end of the nuclear fuel cycle moves ahead, the back end of the cycle has been fraught with delays, technical dead ends, and loss of public confidence. The Blue Ribbon Commission is charged with mapping a new policy direction.

The complicated history of repository siting in the U.S., in its context of competing interests, is an obstacle course for the BRC. While nuclear wastes have accumulated, the whole subject has itself become quite the mess.

Experience with geologic repositories over several decades has indicated that "plastic" containment by salt (the Waste Isolation Pilot Plant — WIPP) or clay is probably superior to "solid" containment by rock (the Basalt Waste Isolation Project and Yucca Mountain).

Nuclear waste repositories emplaced in salt deposits that have been stable for millions of years are exceptionally attractive, based on WIPP and international experience. So the BRC will examine WIPP closely as an important part of its mission.

With this in mind in June 2010, I began, for the first time, to consider scenarios for possible failure of long-term integrity of WIPP: After 11 years of experience gained at the pilot plant, *How robust does WIPP seem nowadays?*

This is a fresh, independent, *pro bono* evaluation of WIPP's long-term integrity with an eye to implications for next-generation waste repositories.

Introduction

The layout of the Department of Energy's WIPP facility is sketched, below:

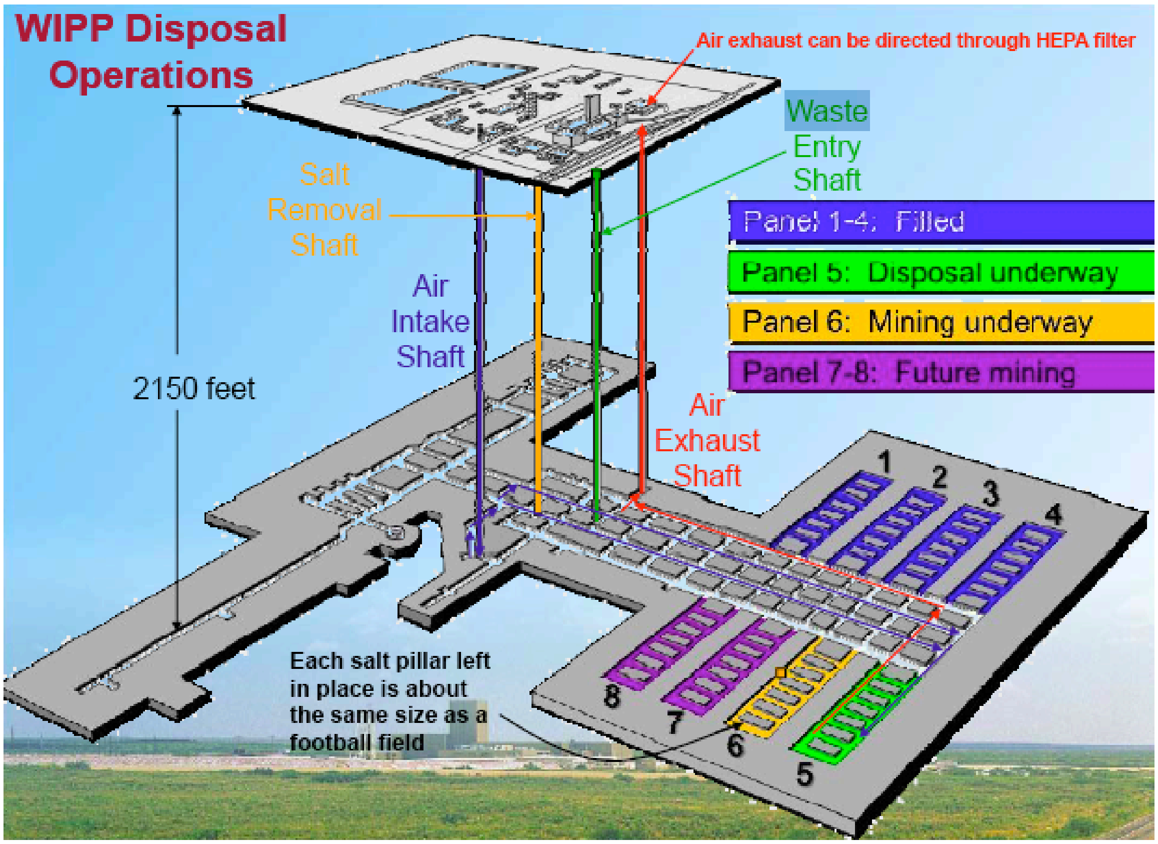

Fig. 1. Three-dimensional sketch of WIPP, including surface support buildings, four shafts, and the mined underground operations areas. The repository is situated about 660 m below the surface, within the Salado Formation, a 250+ million year old sequence of bedded evaporite salt. Eight storage panels are planned, each consisting of 7 rectangular rooms, 10 m wide and 91 m long [sketch from Nelson, p. 11].

Waste emplacement into a room in the WIPP repository looks like this:

Fig. 2. Boxes of magnesium oxide desiccant are placed on top of stacks of waste containers to limit solubility of radionuclides [from Nelson, p. 9].

The general path by which containers from source sites move wastes to WIPP is sketched in (P1), below:

(P1) Passage of wastes into WIPP:

characterized waste
⇓
generic allowance determination: regulations + authorities
⇓
candidate waste container
⇓
screening: handling and transportation criteria
⇓
screening: WIPP waste acceptance criteria*
⇓
WIPP

* maximum acceptance criteria per container [SEIS-II, Chap. 2]:

- chemical composition
- production of flammable gas
- liquid content
- plutonium-239 equivalent activity to avoid criticality
- thermal power generation

After WIPP is filled and closed, late this century, long-term integrity will become a main concern. For the near term of the next century, WIPP will be monitored. After that, the general schema to adequately maintain and assure post-closure integrity for hundreds to thousands of years is passive, and comparatively simple and robust:

(P2) Bases for WIPP repository long-term integrity

- warning signs to discourage future human intrusions
- containment within a geologically stable salt bed
- natural salt creep having sealed / healed of the repository
- provision of a continually reducing environment in the repository
- restrictive waste acceptance criteria
- introduction of an adequate desiccant

—This comment addresses the question: How sound are these bases for long-term integrity?

Method

I have considered a modest range of risks to WIPP integrity informally, by identifying, exploring, and refining scenarios that have come to my mind. The next day, I would check out a different scenario. This process continued until I found a scenario that was sufficiently interesting that checking it out did not come to a satisfactory conclusion.

I checked out several scenarios involving seepage of pore brines. I concluded that bore hole data and the variability of brine chemistry do preclude any important pathway.

After two weeks, "bronze disease" came to mind.

"Bronze disease" refers to corrosion of old bronze coins that have survived hundreds or thousands of years, in diverse conditions in the seabed, swamps, dry ground, indoors, etcetera. Then the coins are moved to seemingly better conditions and usually thoroughly cleaned. On some ancient bronze coins, a spot of unstable cuprous chloride (CuCl) forms, becoming a catalyst at that point on the surface of the bronze coin, in a reaction that is difficult to stop. The coin may be consumed by this *disease*. Even bronze coins touching it might be *infected* and destroyed. An example of an old coin with "bronze disease" is shown in Fig. 3.

Fig. 3. The bronze coin (top) has a tiny pit (arrow head, below) that is the site of "bronze disease" corrosion that will consume the coin, unless there is skilled intervention [Jason Sanchez and Ken Harl, "Bronze Disease: Understanding, Curing, and Preventative Treatment, <http://www.crescentcitycoinclub.org/seminars_and_programs/Bronze%20Disease.pdf> p. 12].

By analogy, I wondered whether there might be some prospect for a local condition in a room in WIPP that might grow upward or laterally and then upward through 400 m of halite above the repository, thus destroying the integrity of the salt bed that has been stable for hundreds of millions of years, under varying environmental conditions. I wondered whether the introduction wastes into the stable salt bed might create a spot of instability, with some reaction that would be difficult to stop.

The length scale for the initiation of such a scenario in WIPP, analogous to "bronze disease" in a coin might be in the range of

local length scale for this analog scenario: 1 - 10 meters

This range is about the width of one waste container to the width of a room at WIPP. Thus, the 400 m of halite overlying WIPP would be 40 to 400 local length scales deep.

As this comment, I summarize my informal exploration of this scenario and its seeming implications, in a way that I hope might be helpful for the BRC's mission.

A Local Scenario

Historically, "bronze disease" was thought to have a biological cause, because of the way it spread and was transmitted. Hence the terminology as a *disease.*

The process is actually a catalytic chemical reaction. In respect for the history, while avoiding the false connotation, I dub the catalytic process *BronzeDisease,* for the purpose of this comment. The two-step process, simplified as a single step, can be written as:

(P3) BronzeDisease:

$2Cu + H_2O + 2CuCl \rightarrow CuO_2 + H_2 + 2CuCl$

--with H_2O from moisture in air

--with HCl intermediary

So I searched for an analogy at WIPP for this BronzeDisease-in-coins by Googling "wipp" and various terms including "reaction" and "water". One result was "Mitigation of hydrogen gas generation from the reaction of water with uranium metal in K Basin sludge [PNNL]."

The reaction of uranium metal with water in K Basin sludge is catalytic. By analogy with the BronzeDisease catalysis, I dub this catalytic reaction *UraniumSludgeDisease.* The two-step equation can be simplified to one step, written as:

(P4) UraniumSludgeDisease:

$U + 2H_20 \rightarrow UO_2 + 2H_2$

--with H_2O either a condensed film or liquid

As with BronzeDisease, UraniumSludgeDisease is difficult to stop. Uranium metal scavenges water, as does cuprous chloride, to continue catalytic oxidation and generation of free hydrogen.

The total uranium content of K Basin sludge is about 2.7 metric tons [calculated from EPA, p.9]. (*I wonder what fraction of this uranium is as metal.*)

Figure 4 is a picture of K Basin sludge, before removal.

Fig. 4. Sludge in K-East Basin at Hanford [M.W. Peres, "A Comparison of Challenges Associated with Sludge Removal, Treatment and Disposal at Several Spent Fuel Storage Locations, WM'07 Conference, <http://www.wmsym.org/archives/2007/pdfs/7287.pdf> (25 February - 1 March 2007), from Fig. 1].

Generation of a flammable gas like hydrogen is of concern for safety during transportation of wastes and during operation of WIPP. After WIPP is sealed and closed, there are other concerns for gas generation. Gas Generation is an important input for several repository Performance Assessments, as I'll come to shortly.

WIPP offers a default solution for K Basin sludge; namely, that the waste destined for WIPP be "diluted (by using partially full drums) until it does meet the gas generation limits [SEIS-II, Comment Response Document, Sec. 02.07(04)]." This default solution equates the gas generation by this oxidation of uranium, with gas generated by radiolysis. Radiolysis is part of a "Feature, Event, and Process" (FEP) for which performance assessments have been accepted for certification of WIPP under Title 40 CFR Part 191.13.

If the K Basin sludge were sufficiently diluted volumetrically, "the number of **drums for certain streams must increase by factors in excess of 100** to accommodate the contained uranium metal, and its associated H_2 generation [emphasis added, PNNL, p.1.1]..."

The volume of K Basin sludge (as-settled) is about 45 m^3 [calculated from EPA, p. 9]. That would fill about 215 drums of 55-gallon capacity. That is a tiny fraction (0.026%) of the 170,000 m^3 total waste inventory projected for WIPP through 2033 [SEIS-II, Table 2-2]. If the volume of sludge waste were expanded hundreds of times by fractionally filling waste drums, K Basin sludge would become a substantial occupant of WIPP. —As PNNL notes, there are compelling economic reasons to diminish or eliminate hydrogen gas release from K Basin sludge.

PNNL has been trying to solve the problem of hydrogen generation in K Basin sludge for about a decade. The introduction of ~0.5 molar nitrate salt into the waste drums presently seems the most promising of several possible fixes. The added nitrate effectively scavenges hydrogen that is produced in the sludge, rather than abating the uranium metal corrosion that generates the hydrogen [PNNL, p. iii].

Meanwhile, WIPP authorities have good reasons to be wary of accepting wastes that have been altered in order to *work-around* a waste acceptance criterion. In the case of K Basin sludge, about a half gallon of sodium nitrate added to a drum of sludge would practically eliminate free hydrogen generated in the drum, ***now***. But as time would pass, this nitrate would be used up, and gas generation in the waste would resume. That is to say, a drum of waste that might nominally be acceptable for WIPP storage this year could become seriously unacceptable after years of storage, by reason of future gas generation.

If more nitrate salt is added to the sludge to provide longer duration of gas elimination, the contents of the drums would be made more oxidizing, possibly locally overcoming reducing conditions in the repository. Thus, there is an interesting question of WIPP's acceptance of altered wastes having time-dependent properties. As PNNL puts it:

> **The acceptability of any waste form to be disposed at WIPP must be determined, including the potential introduction of nitrate, nitrite, or U(VI) to the sludge** [PNNL, p.4.5].
>
> **However, objection to the use of nitrate, nitrite, or U(VI) is unlikely given the existence of these constituents in prior wastes disposed to WIPP from Rocky Flats and Hanford** [PNNL, p.3.37].

(P5) This scenario makes the following points:

- ***Local* FEPs are of concern.**
- **Time-dependent, physical and chemical processes might well be important for post-closure repository performance.**
- **The real example of K Basin sludge approaches the envelope of WIPP performance assessments.**
- **Some isolated mistakes in almost one million waste containers will very probably make it into storage at WIPP.**

—The fourth point can be seen from the fortunate circumstance that there had been no reason for Hanford Site management to limit hydrogen production in the K Basins. Had there been reason, the operators might have put something like enough nitrate into the basins to eliminate hydrogen generation in the sludge for a few decades. Then the nitrated sludge would probably have been characterized as acceptable for storage at WIPP. After several more decades, hydrogen gas would then have begun to be generated in the nitrated sludge in the repository ... without having been assessed.

Local Assessment Considerations

WIPP's Waste Acceptance Criteria (WACs) are updated as more experience is gained. The criterion for chemical compatibility of acceptable waste, effective 30 June 2010 is:

Chemical Compatibility [emphasis added, WAC, Sec. 3.5.3]:

> **TRU waste containing** incompatible materials or **materials incompatible with** payload container and packaging materials, shipping container materials, **other wastes, repository backfill, or seal and panel closure materials are not acceptable** for transport in the TRUPACT-II and HalfPACT or **for disposal at the WIPP**.

—The highlighted parts of this criterion focus on the chemistry of post-closure WIPP. The essence of this actual WAC is a kind of catch-all and end-all: "**TRU waste containing materials incompatible with other wastes ... are not acceptable for disposal at the WIPP.**"

This actual WAC is akin to an illustrative, tautological, over-arching WAC for WIPP waste (**not actual!**): *TRU waste containing materials incompatible with repository integrity for >10,000 years are not acceptable for disposal at the WIPP.* Such an illustrative, tautological WAC might seem to solve every failure possibility without the bother of any performance assessments at all! The rub with both the actual and illustrative WACs is their failure to provide practical guidance for what should not be accepted into the repository.

So we find ourselves yet with not too much experience and guidance regarding time-dependent chemical compatibility between nearby creep-crushed containers of waste, in the post-closure era of WIPP. It is not clear whether such a change in waste behavior over time is tolerable for long-term repository integrity.

Thus does the basis for repository integrity provided by waste acceptance criteria translate, in practice, into a set of formal (albeit changing) guidelines, with final determinations made by WIPP authorities, with regulatory acceptance, on a somewhat case-by-case basis. (*—What a long way this seems from the starting notion: Geologically stable salt beds make excellent repositories; no worries!)*

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The 24 Performance Assessments (PAs) that have been run for certification of WIPP were independently peer reviewed in 1996 [PEER 1]. Of these, the Peer Review Panel judged 9 to be Not Adequate, with consequences. Two of the **Not Adequate** PAs were [p. ES-2]:

Gas Generation
Chemical Conditions

The Panel was generally concerned with both these PAs because their outputs are inputs to other PAs. So their inadequacy might have affected the overall adequacy of the risk predictions generated by the PAs (see Fig. 5).

The Peer Review Panel concluded that four issues had not been resolved adequately in the Gas Generation PA, the first of which was [PEER 1, p. 3-150]:

- **Analysis of hydrogen generation by corrosion of metals other than steel in the waste is inadequate. Ignoring gases generated by corrosion of other metals could result in underestimating gas pressure in the repository.**

The Gas Generation PA corresponds to the concern expressed in the previous section of this comment. The oxidation of uranium in K Basin sludge is an example of another metal that has, indeed, turned out to be just such a problem. Thus, the work-arounds that PNNL is exploring are seen to be outside the envelope of the PAs that assure WIPP's post-closure integrity.

The Peer Review Panel judged the Chemical Conditions PA to be Not Adequate because of an assumption in the PA: "Actinide and ligand **inventories are fixed** [emphasis added, PEER 1, p. 3-154]." That is, the Panel believed that some of the inventory of the repository might be time-dependent in important ways.

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In accord with the requirements of Title 40 CFR Part 194, the purpose of the peer review for WIPP's PAs was [PEER 1, Plan, p. 1]:

> **Specifically, a [peer review] PR will be conducted to determine whether the conceptual models developed and selected by DOE reasonably represent future states of the disposal system.**

Sandia National Laboratories "determined which processes are significant," and "developed conceptual models which represent possible future states of the disposal system and subsystems." The conceptual models had been translated into the 24 PAs, along with supporting documentation. The Peer Review Panel reviewed the 24 PAs, one by one, and their inter-connections.

DOE responded to the Peer Review Panel, and an increasingly complicated back-and-forth ensued, until the Panel finally accepted DOE's PAs in April 1997 [PEER 2, Appendix 1.15]. (—*Shame on DOE!*)

The Peer Review Panel was ***not*** asked to consider overall adequacy: ***Is the set of 24 PAs adequately comprehensive to include all processes that might reasonably be significant to determine future states of the disposal system?*** It must have seemed obviously, usefully inviting for DOE, at the end of that peer review process to have asked its Peer Review Panel: ***Has DOE omitted any scenarios/assessments that the Panel thinks might be important for WIPP?***

The logic of the risk analysis for long-term WIPP performance involves "the formation of scenarios for consequence analysis from combinations of FEPs that remain after screening [DOE/CAO, p. 6-61]." —This is certainly feasible and conceptually valid, but is the envelope of PAs (in contrast to FEPs) comprehensive?

There is a certain impression that the overall ***envelope of PAs*** is comprehensive because ***all*** significant Features, Events, and Processes (FEPs) that might affect the disposal system for 10,000 years were systemmatically identified and thoroughly evaluated [DOE/CAO, Sec. 6.2]. But comprehensiveness of the FEPs does not logically assure comprehensiveness of the PAs. The question of the overall adequacy of the envelope of PAs seems not yet to have been addressed. Instead, it has been dismissed in terms of a logic diagram [DOE/CAO, Fig. 6-7].

The fundamental issue of comprehensiveness of the set of PAs for WIPP might seem academic. DOE has modified and updated the PAs over the years, for its own purposes and in response to concerns by regulatory agencies. That might be good enough. On the other hand, the concern of this comment for local failure of containment strongly suggests that a sensitivity analysis of the comprehensiveness of the current set of PAs for WIPP should be performed. Then DOE should ask one of its independent peer review panels to evaluate the adequacy of the envelope of PAs and to recommend appropriate additions, if any.

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Risk models for radioactive releases from WIPP, thousands of years into the future, have significant outcomes deriving from human intrusions (Disturbed Performance). The modeled, total risk is shown in Fig. 5:

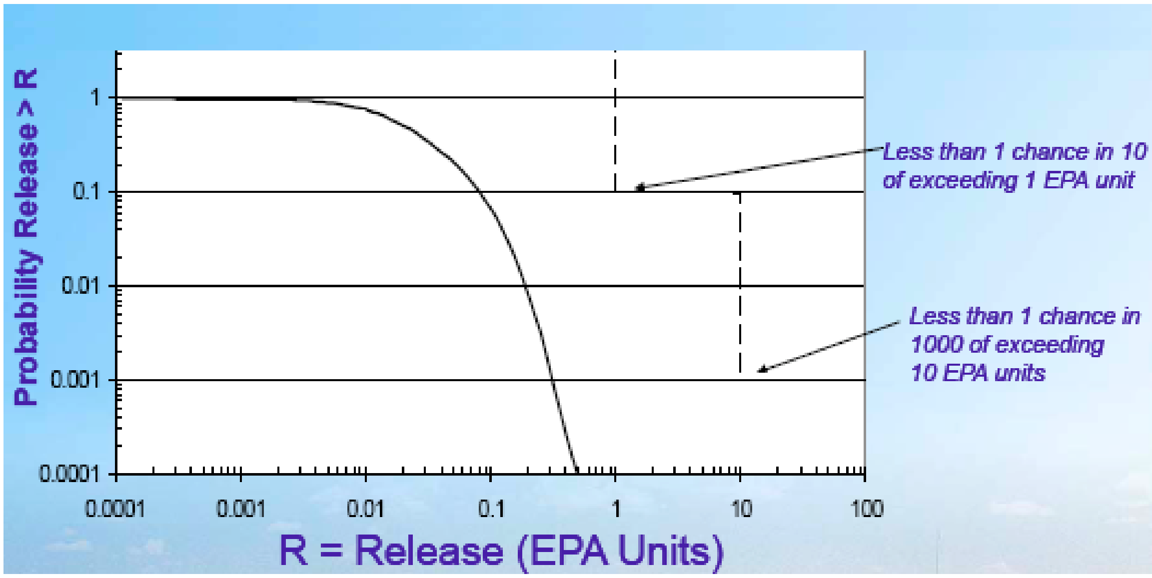

Fig. 5. Mean total risk probability of release R [from Nelson, p. 18].

The risk models show that probabilities of releases from Undisturbed Performance (no human intrusion) of the repository are below concern; that is, total risks for all undisturbed scenarios are to the left and below the area shown in Fig. 5. Thus, WIPP claims "no release" from any and all scenarios not involving human intrusion [Nelson, p. 22].

Notice the steep slope of the total risk probability curve, close to the upper step of the EPA Containment Requirements, on the right of Fig. 5. This steep slope means that adequacy of containment is much more sensitive to the magnitude of modeled release than to the modeled probability of a release occurring.

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In 2003, Lawrence Allen and James Channell analyzed placement of actual wastes in Panel 1 at WIPP and implications for risks predicted by PAs for WIPP. They found that wastes from the priority shipping campaign of residues from Rocky Flats had not been not stacked randomly [PEER 2, App. 4.4.10, p. 18]. They concluded that the consequences are great enough that it "may invalidate the premise of the performance assessment bounding analysis for spallings."

Allen's and Channell's analysis was itself bounded by a minimum of 810 drums of waste. That would be at about the upper limit of my concern with *local* scenarios. Allen and Channell noted that the 810 drums might be too large to "represent an adequate bounding case for non-random emplacement of waste [p. 16]." So, wastes already emplaced in Panel 1 at WIPP might already exceed bounding cases for the risk analyses that demonstrate WIPP's compliance, Fig. 5. —*How can this have happened?* ...***There are no FEPs, no PAs, and no WACs for a Priority Shipping Campaign.***

...So, let's make up a scenario with K Basin sludge: Suppose that WIPP authorities decide to accept K Basin sludge, with just enough nitrate added to meet the hydrogen gas generation criterion for the next several years. For this scenario, further suppose that the roughly 215 drums of nitrated sludge will be shipped in groups and emplaced in groups in storage rooms in WIPP.

After creep closure of the waste rooms, this scenario supposes the nitrate will be consumed as it mitigates hydrogen gas generated by continuing uranium metal oxidation with sufficient moisture provided by local seepage of pore water into the locales. —*Would gas pressure rise sufficiently, locally, around some of these drums or groups of drums to exceed the tolerable input bounds of any of the PAs for WIPP? Or would compensatory processes suffice to localize any such excesses?*

This is where the analogy of "bronze disease" comes into the repository picture: *With generic Gas Generation and changing Chemical Conditions in mind, are there post-closure scenarios beginning in one or several locales that might spread enough to compromise the integrity of waste containment in the Salado Formation?*

A set of performance assessments (PAs) of local scenarios would answer such questions, at least in general. Such PAs would show how much concern repository managers and regulators should have for both allowance and placement of wastes that approach the limits of acceptance criteria: *How robust is repository integrity relative to local variations? Would the risks and consequences of assessed, local failures of containment over the next hundreds or thousands of years be permissible without additional safeguards?*

From the standpoint of numerical modeling, consideration of local scenarios would entail reducing the grid scale for the repository (Region 23), the disturbed rock zone (Region 22), and the surrounding, impure halite (Region 19), illustrated in Fig. 6.

Such small-scale modeling would distinguish the locations where the desiccant boxes are placed on the waste stacks and how the desiccant might spill as in-creep of the halite breaks the boxes. Likewise, various heights of pore brine seepage into the creep-collapsed repository might be included in the small scale of such a PA model.

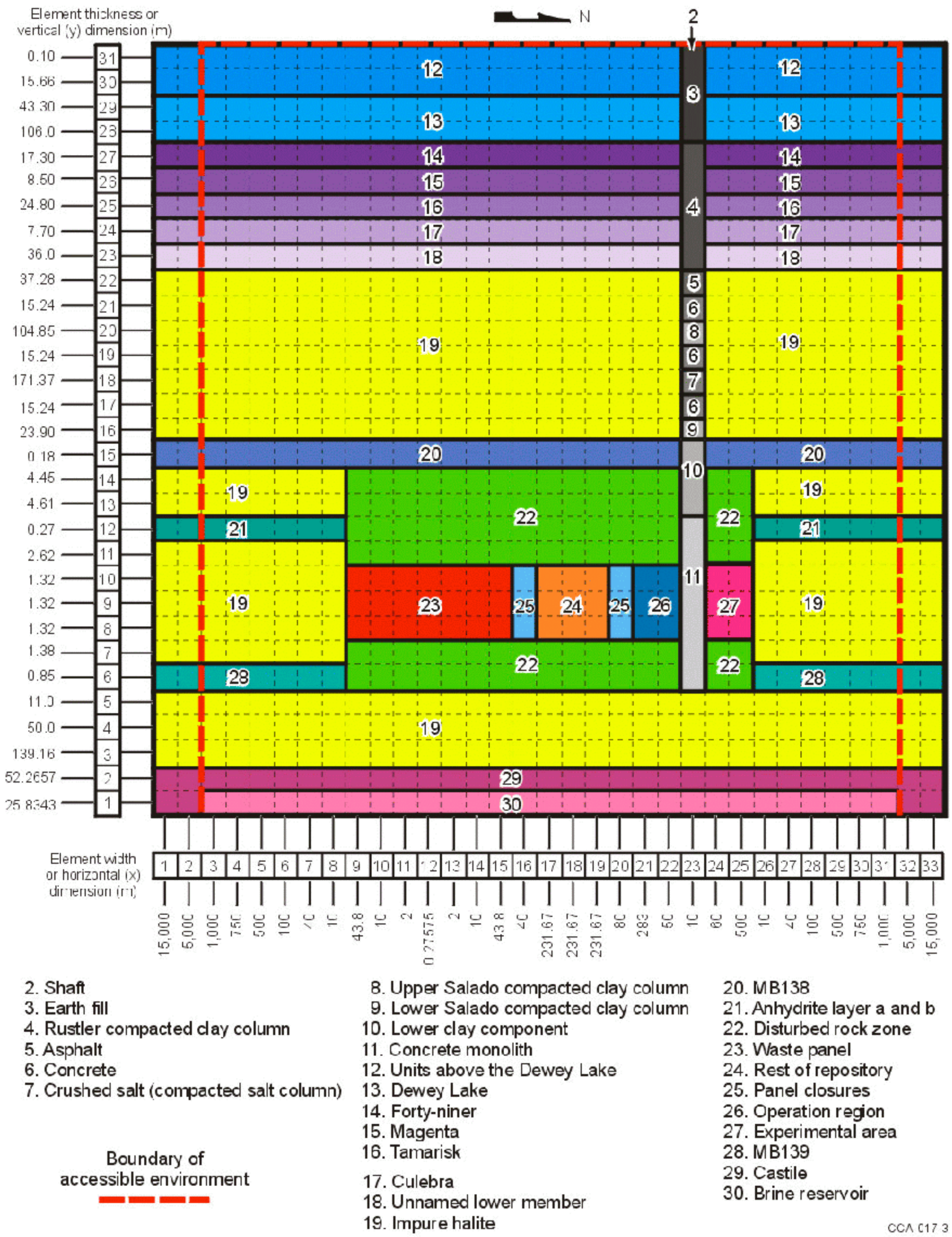

Fig. 6. Side View of the Elements and Material Regions for Modeling Undisturbed Performance [DOE/CCA, Fig. 6-13].

Based on my limited consideration of scenarios, here, I believe that such a local-source PA should be run, along with a sensitivity analysis of it.

I believe that DOE should perform a sensitivity analysis of the comprehensiveness of it current PAs for WIPP. Than ask one of the independent peer review panels to evaluate the adequacy of the envelope and to recommend appropriate additions, if any.

Conclusions

1. **Detailed technical requirements for long-term integrity of a repository in bedded salt are more complicated and challenging than was evident at the outset.** The natural robustness of such waste disposal does not invite adequate technological resources that are expensive. A useful feature for a candidate repository is an *appearance of less natural integrity than exists*, so that extra effort goes into solving each and every problem. **An apparently safe solution does not invite good risk management. —**This conclusion is counter political.

2. **WIPP will probably not be *inherently* sealed by the Salado Formation**, beyond a few hundred years. Assuming that operational and closure plans are carried out well and that there is no future human intrusion; then containment will likely come to depend on details of the emplacement of particular wastes and their local surroundings. Whether containment will be breached locally or not, will depend on the detailed development of local processes within the, then, long-closed repository. These details will be more or less unique in the thousands to hundreds of thousands of waste *locales* within the sealed repository. —*What is the chance that the particular properties in **none** of these locales will lead to a local breach of containment in hundreds or thousands of years?*— This questioning of long-term integrity of containment at a multitude of locales would be a different kind of performance assessment.

3. **K Basin sludge is an informative, real-case of time-dependent hydrogen gas generation that is outside the envelope of Performance Assessments for WIPP.** If one-percent of nitrate had been added to the sludge in K Basins, this TRU waste would likely have been accepted into WIPP, without consideration of or regard for future gas generation after the nitrate would have been consumed by the generated gas.

Recommendations to the BRC
<u>regarding repositories in stable salt beds</u>

1. **Consider how much more complex and expensive waste storage in seemingly robust, geologically stable media will turn out in practice than is apparent at the outset.** "Bronze disease" of old coins illustrates the illusion that undisturbed stability implies continuing stability after a novel perturbation.

2. **Assess whether the *envelope* of Performance Assessments for WIPP is adequately comprehensive?** If not, what lessons are there for future repository siting?

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