A Practical Solution to Hanford's Tank Waste Problem

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A “Troubled” Project

- “Since 1988, roughly $2 billion per year has been spent on Hanford cleanup.”
- “Despite the big money and big employment figures (typ. 10,000)..., many feel that cleanup is off-track. Very little of the radionuclide and chemical inventory has been stabilized after thirty years of effort.”
- “GAO estimates that the final bill may be as much as $120 billion and may take another 50 to 60 years to complete.”

http://www.hanfordchallenge.org/the-big-issues/how-hanford-works/

A FEW EXAMPLES

http://www.power-eng.com/articles/2013/02/hanford-nuclear-waste-storage-site-panned-in-gao-report.html (Feb2013)

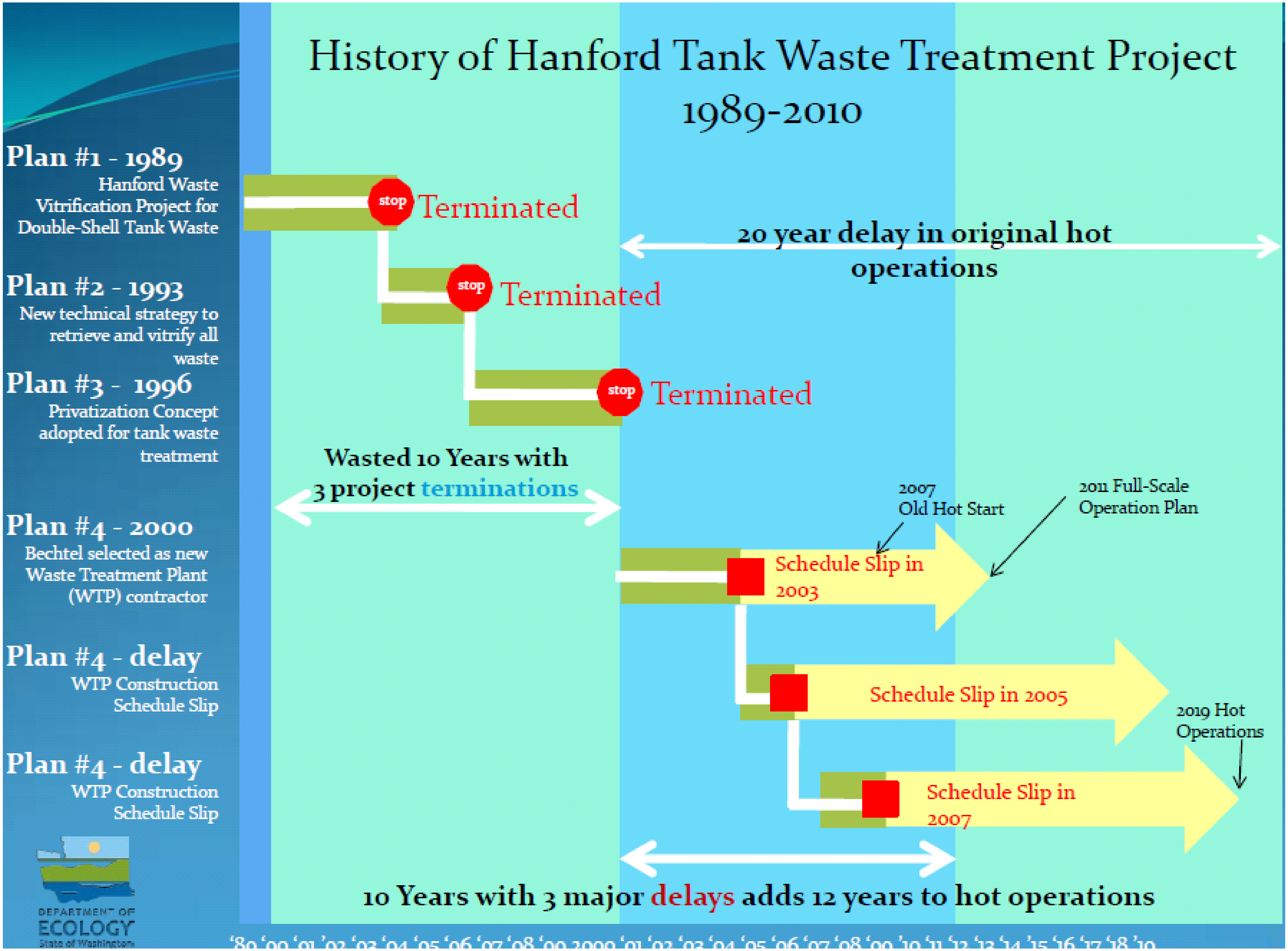
http://www.nwnewsnetwork.org/post/federal-report-blasts-hanfords-waste-treatment-plant-project (Jan2013)

http://www.forbes.com/sites/jeffmcmahon/2012/08/29/bechtel-incompetent-to-complete-hanford-nuclear-waste-cleanup-doe-memo/ (Aug2012)

http://ehstoday.com/safety/news/safety-board-hanford-site-atmoshpere-adverse-safety-0707 (Jul2012)

...etc., etc. back to the early-90’s

The National Academies identified the cultural “symptoms” responsible for Hanford’s (& INL’s) EM woes in 1996: *see “Barriers to Science*: Technical Management of the Department of Energy Environmental Remediation Program”, *www.nap.edu/catalog/10229.html*



Slide 7, **"Tank Waste Final Waste Form Perspective", Suzanne Dahl, WA dept of ECOLOGY, Jan. 13, 2010**

The Galvin Commission's Conclusions (1995)

"One of the consequences of the troubles has been the enhancement of a syndrome common to large bureaucracies: risk aversion. It has a name: "the Hanford Syndrome." It has become widespread and severe in the EM program. Its symptoms are an unwillingness to alter familiar behavior patterns, to stick with unproductive or failing procedures, to enhance tendencies for excessive resource allocation and regulation, and to oppose innovation. It is an important element in sustaining unproductive patterns of work."

"The Tri-Party Agreement at Hanford, and similar ones elsewhere, have proven to constitute major constraints on remediation progress because, in many instances, they are unrealistic, not having had proper input from those experienced in actual cleanup. The milestones they incorporate, along with penalties for noncompliance, force continued activities, some of which are make-work and should be abandoned

www.lbl.gov/LBL-PID/Galvin-Report/Galvin-Report.html

Root Causes

The reason why Hanford's WTP project has morphed into a huge boondoggle* is that it was based upon two technically unrealistic assumptions/promises: 1) its radwaste will/must be separated into "high" & "low" fractions so that the former can be dumped into someone else's "back yard"[note1], and 2) both fractions will/must eventually be turned into borosilicate glass[note2]

**www.gao.gov/products/GAO-13-38*

Recommended paradigm shift

- Homogenize (not separate) Hanford's tank wastes: i.e., simultaneously retrieve from multiple depths from multiple tanks
- Pug-mill mix with crude phosphoric acid[note] & powdered iron ore, vitrify with a stirrer-equipped melter, and make "aggregate"* of the glass
- Slurry this aggregate with a MgO/KH_2PO_4 ("Ceramicrete") - type grout & pump it back into Hanford's best-condition waste tanks**

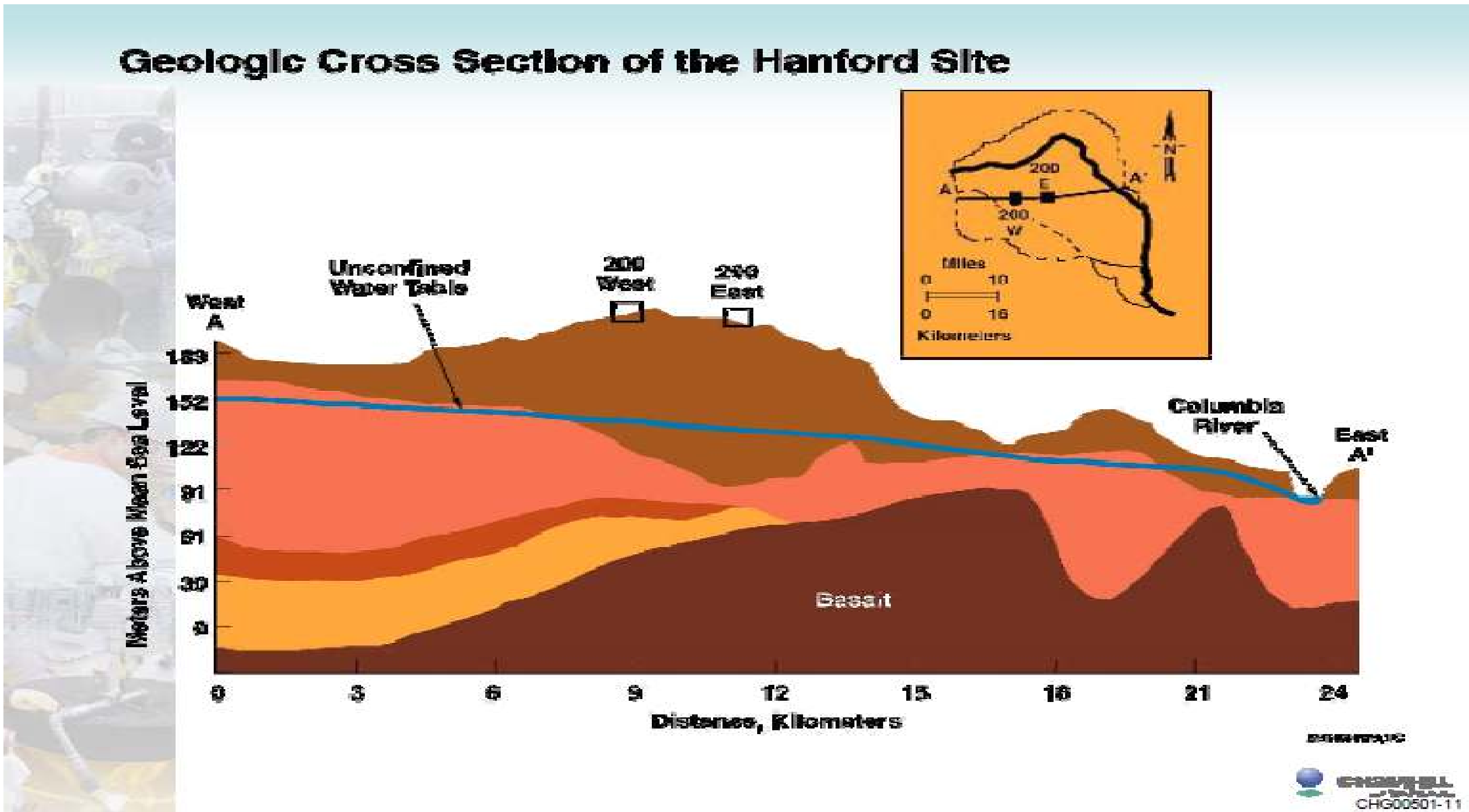
* Either glass marbles, "gems" or cullet

** this grout would simultaneously serve to seal any leaks in those tanks .

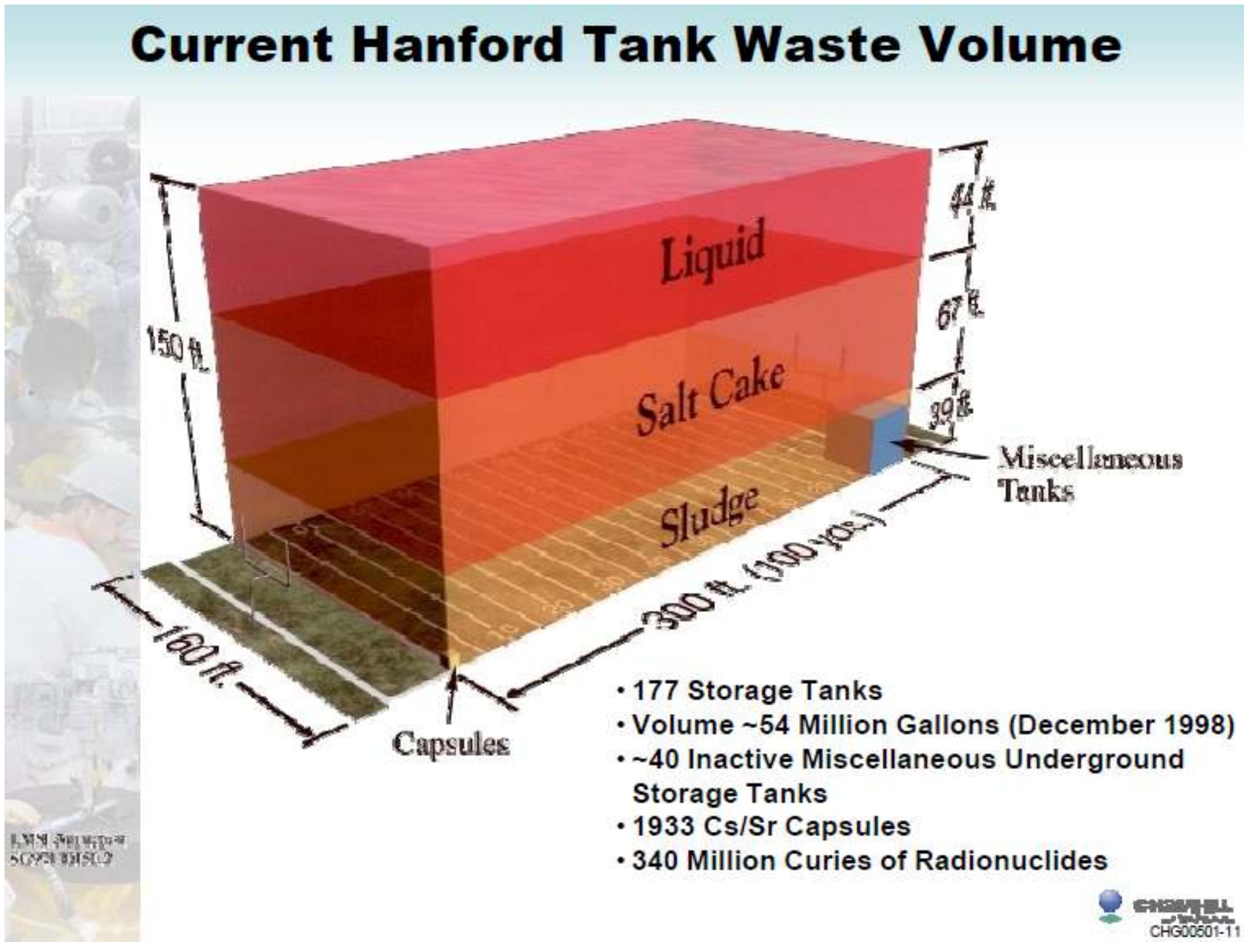
Background Slides*

**srnl.*doe*.gov/emsp/day1_overv/*hanford*-gaspl.pdf* (CH2MHill, 2005)

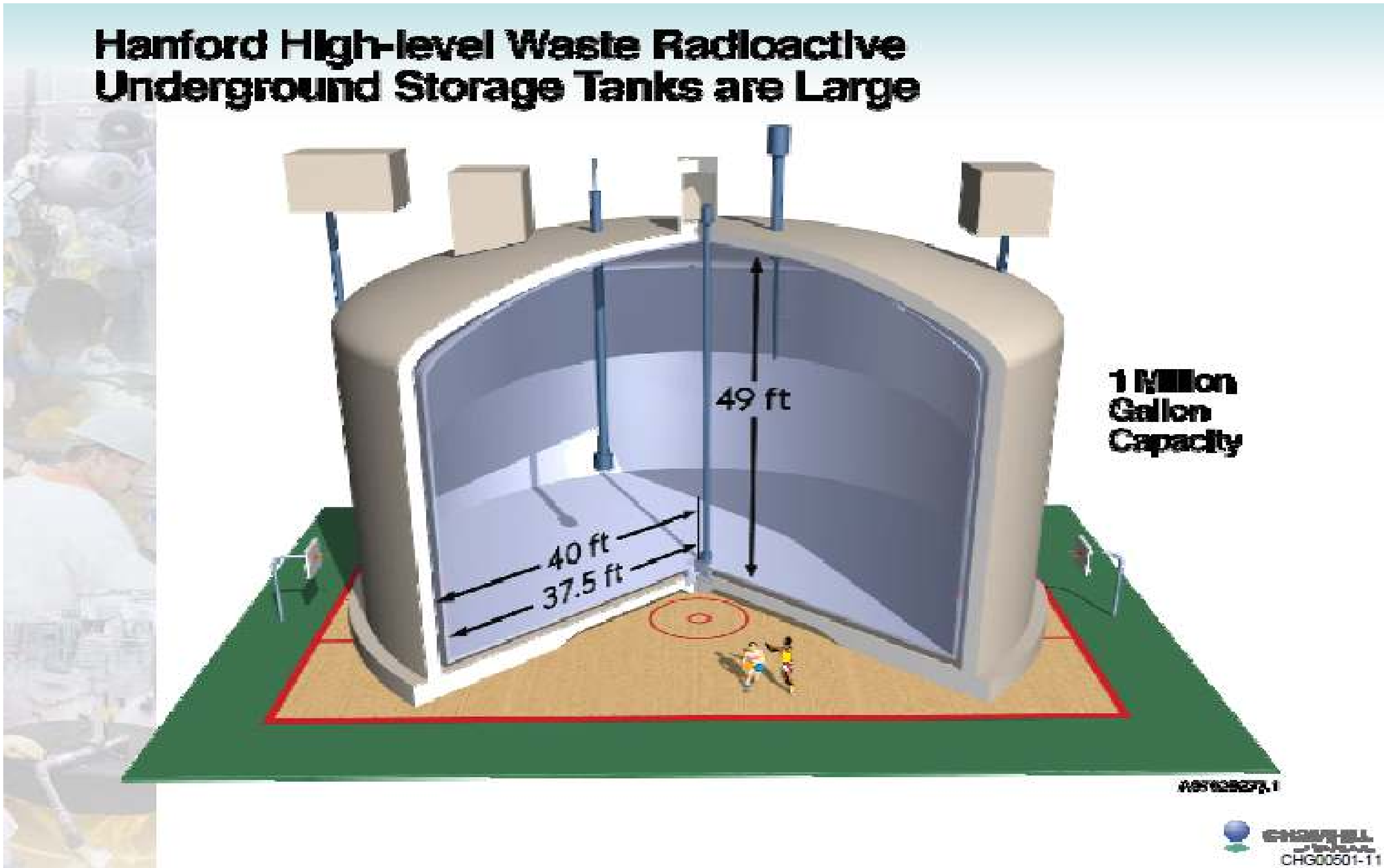
More background...note



more...note

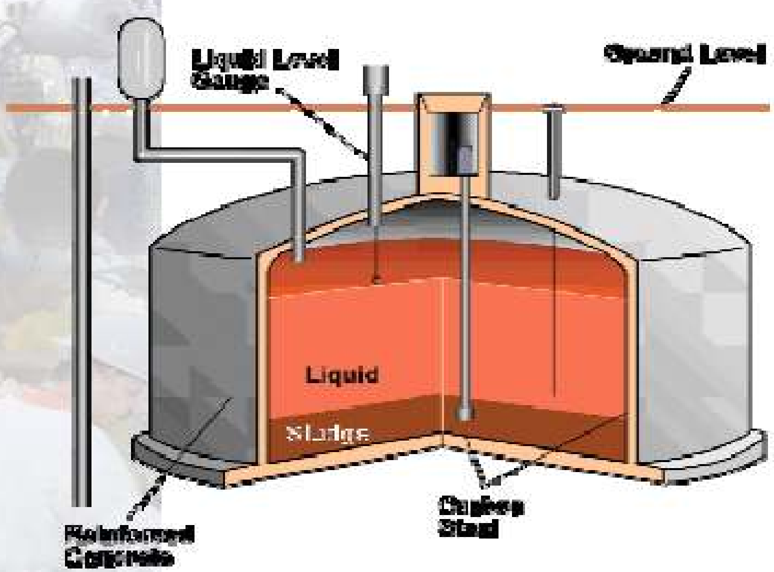

Current Hanford Tank Waste Volume
Liquid
Salt Cake
Sludge
150 ft.
44 ft.
67 ft.
39 ft.
Miscellaneous Tanks
160 ft.
300 ft. (100 yds.)
Capsules
• 177 Storage Tanks
• Volume ~54 Million Gallons (December 1998)
• ~40 Inactive Miscellaneous Underground Storage Tanks
• 1933 Cs/Sr Capsules
• 340 Million Curies of Radionuclides
CHG00501-11

more...


Hanford High-level Waste Radioactive
Underground Storage Tanks are Large
49 ft
40 ft
37.5 ft
1 Million
Gallon
Capacity
CHG00501-11

more...

Single-Shell Tanks



- 149 Tanks Constructed 1943-64
- ~210 m^3 to 3,800 m^3 Capacity (55 kgal to 1 Mgal)
- Bottom of Tanks at Least 50 m (150 Feet) Above Groundwater
- No Waste Added to Tanks Since 1980
- Tanks Currently Contain:

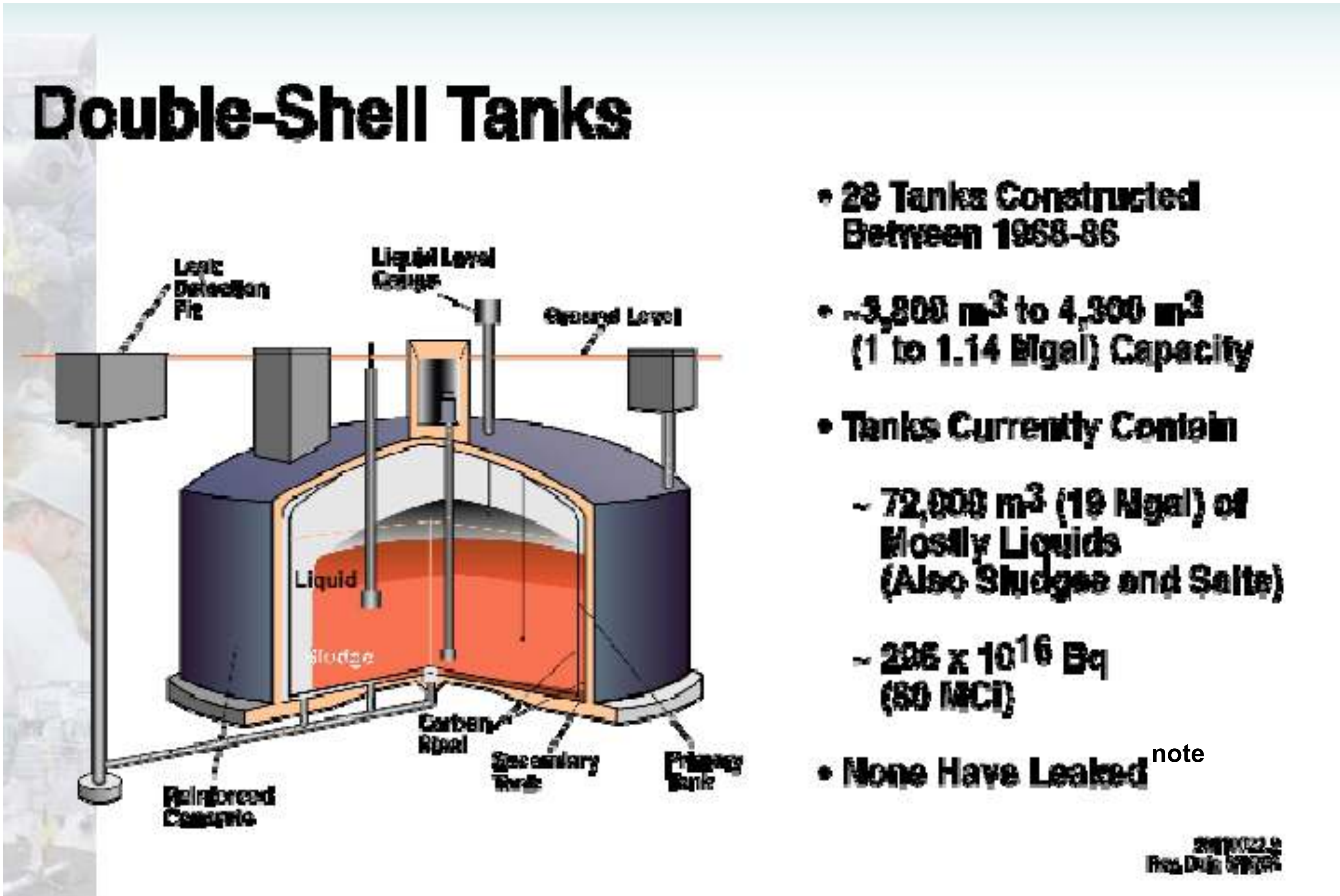
  ~132,500 m^3 (35 Mgal) of Salt Cake, Sludge, and Liquid

  ~407 x 10^{16} Bq (110 MCi)
- 67 Are Assumed to Have Leaked ~ 3,800 m^3 (~1 Mgal)

more...

Inside Tank SX-109

more...

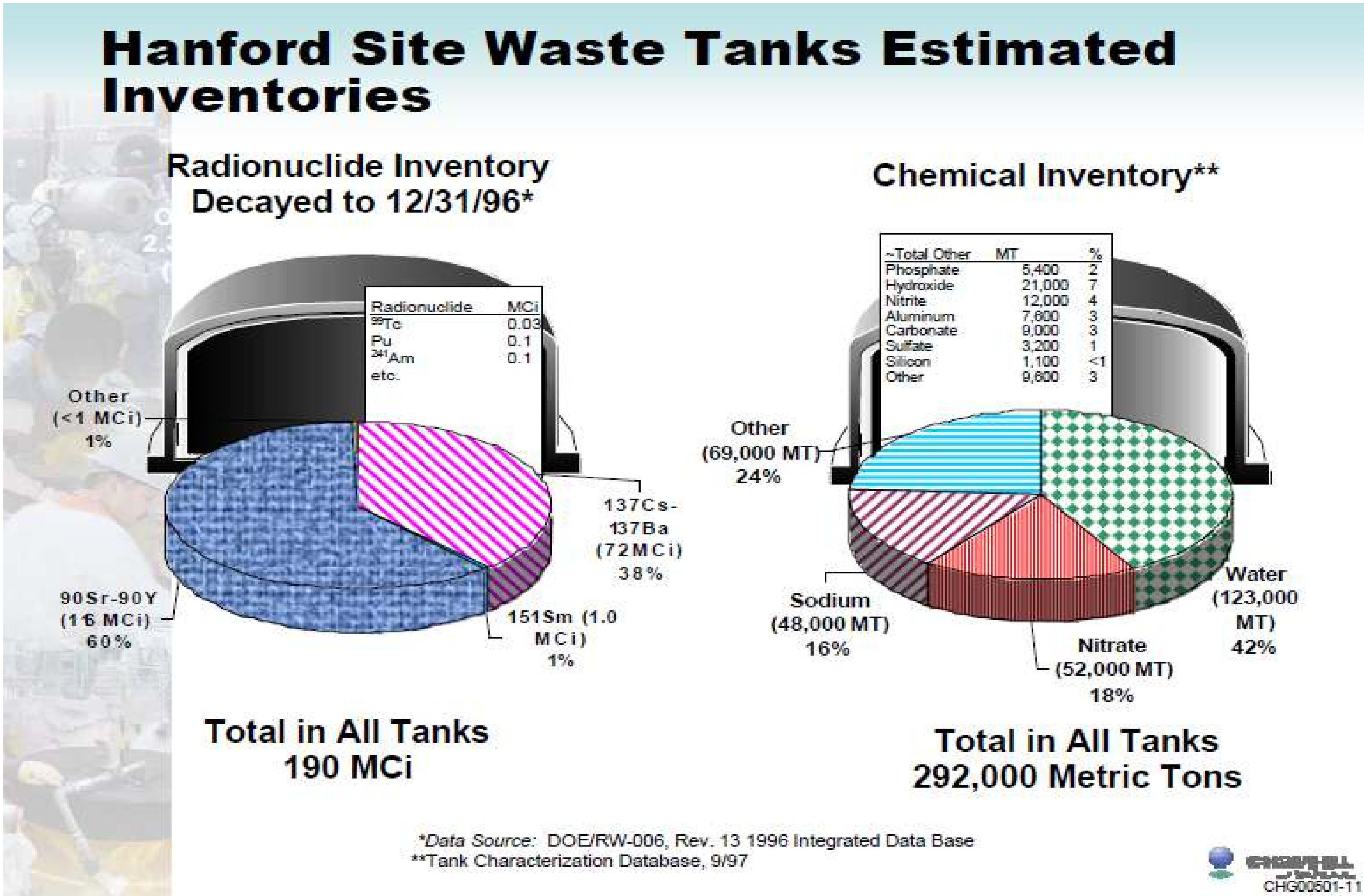


Double-Shell Tanks
Liquid Level Gauge
Ground Level
Liquid
Sludge
Carbon Steel
Secondary Tank
Primary Tank
Reinforced Concrete
• 28 Tanks Constructed Between 1968-86
• ~3,800 m3 to 4,300 m3 (1 to 1.14 Mgal) Capacity
• Tanks Currently Contain
~ 72,000 m3 (19 Mgal) of Mostly Liquids (Also Sludges and Salts)
~ 296 x 1016 Bq (80 MCi)
• None Have Leaked
note

more...

AP Tank Farm

more...


Hanford Site Waste Tanks Estimated Inventories
Radionuclide Inventory Decayed to 12/31/96*
Radionuclide MCi
99Tc 0.03
Pu 0.1
241Am 0.1
etc.
Other (<1 MCi) 1%
137Cs-137Ba (72MCi) 38%
90Sr-90Y (116 MCi) 60%
151Sm (1.0 MCi) 1%
Total in All Tanks 190 MCi
Chemical Inventory**
~Total Other MT %
Phosphate 5,400 2
Hydroxide 21,000 7
Nitrite 12,000 4
Aluminum 7,600 3
Carbonate 9,000 3
Sulfate 3,200 1
Silicon 1,100 <1
Other 9,600 3
Other (69,000 MT) 24%
Sodium (48,000 MT) 16%
Nitrate (52,000 MT) 18%
Water (123,000 MT) 42%
Total in All Tanks 292,000 Metric Tons
*Data Source: DOE/RW-006, Rev. 13 1996 Integrated Data Base
**Tank Characterization Database, 9/97
CHG00501-11

Some facts we don't hear so much about

- Hanford is a good geological repository site[note1]
- Its radwaste is extremely dilute (not very "high"): ~90 tonnes[note2] initial FP ended up in about 292,000 tonnes of waste (0.03 wt%)*
- It's waste is now "cold": Current FP inventory includes 0.4 tonne (3.6E+7 Ci) of heat-generating $^{137}Cs/Ba$ & 0.25 tonne (3.3E+7 Ci) $^{90}Sr/Y$ or ~1.3 watts/tonne waste[note3]
- It's mainly comprised of sodium salts[note4]: 48,000 tonnes Na in 53 million gallons = 10.4 moles/liter which means that conversion to "salt stone" would roughly treble its volume.[note5]
- It also contains lots of other stuff (aluminate, chromate/ chromite, sulfate, phosphate etc.) incompatible with high loading in borosilicate glasses (BSG)

Hanford's Waste is "Low" Level[note]

	Class C LLW Limits*	∑Hanford Waste**
^{90}Sr	4600 Ci/m^3	219 Ci/m^3
^{137}Cs	7000 "	142 "
^{63}Ni	700 "	0.38 "
^{14}C	8 "	0.023 "
^{99}Tc	3 "	0.15 "
^{129}I	0.08 "	0.0003 "
All α > 5 yr $t^{1/2}$	1100 nCi/g	441 nCi/g
		sum fractions = 0.51

*Upper Class C limits: Tables 1 & 2, 10 CFR§61.55 (from NRC website)

**assumes 19 yr decayed "global" figures of Table ES-2 HNF-SD-WM-TI-740 Rev OB, 1998 & a total of 322,000 tonnes (dry basis) or 55 million gallons of waste

Why Iron Phosphate Glass[1-4] (Fe-P)?

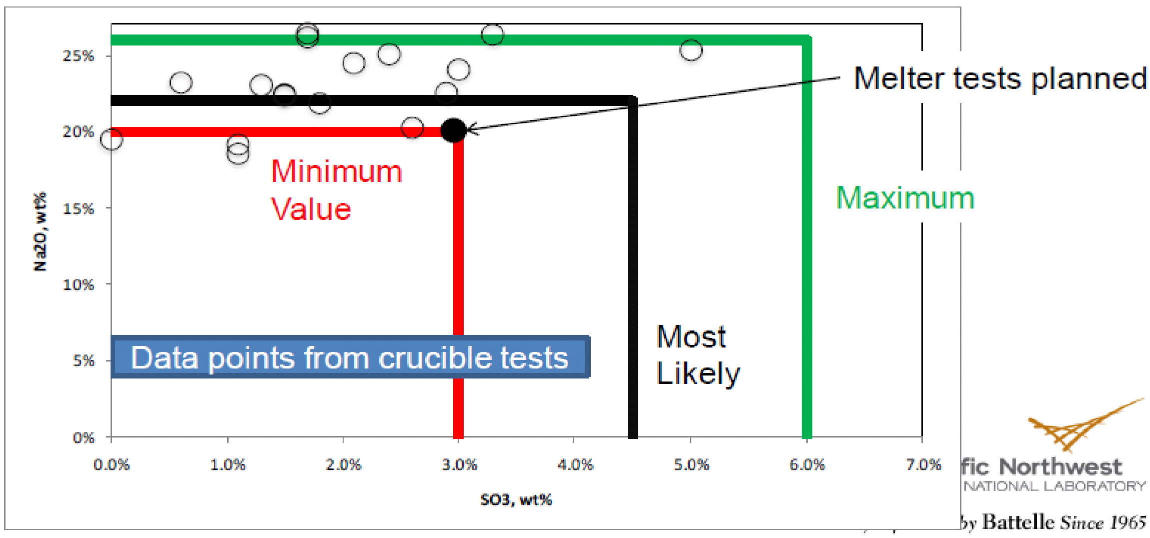
- More leach resistant than BSG glasses[note1]
- Easier to make (lower melting point & viscosity)
- Already thoroughly studied/characterized for application to many DOE radwaste streams
- Accommodates much higher concentrations of "problematic" (for BSG) waste constituents[note2]
- Compatible with "mag phosphate" grout

1. Huang et. al., Journal of Nuclear Materials, 327(2004) 46-57R.
2. Robert Leerssen, "Fe Phosphate Glass for the Vitrification of INEEL SBW and Hanford LAW", MS Thesis, UMR (now MST), 2002
3. Darryl Siemer, "Improving the Integral Fast Reactor's Proposed Salt Waste Management System", Nuclear Technology, 178(3), 2012, pp 341-352.
4. Sevigny, et. al., "Iron phosphate glass-containing Hanford Waste Simulant," PNNL 20670, August 2011

Why Iron Phosphate... cont.

Phosphate Glass Loading Estimates*

- Insufficient data to refine loading estimates, however, based on preliminary assessments:
  - 20 wt% ≤ Na_2O ≤ 26 wt% → most likely 22 wt%
  - 3 wt% ≤ SO_3 ≤ 6 wt% → most likely 4.5 wt%
  - No halide, phosphate, or chromate limits



fic Northwest NATIONAL LABORATORY
by Battelle Since 1965

*Vienna et al. 2010, http://srnl.doe.gov/techex_2010/pdfs/S02-04.pdf

Why Iron Phosphate...cont.

- 48000 tonnes Na = 2.09E+9 moles Na
- Which equals 6.47e+10 grams Na_2O
- @22 wt% loading* that's 2.94E+11 g Fe-P
- @2.9 g/cc, that's ~27 million gallons of glass (~one-half of the tank volume)

*slide 6, Vienna et al. 2010, http://srnl.doe.gov/techex_2010/pdfs/S02-04.pdf

WHY “STIR MELTER”[note]

• **Mechanical mixing is apt to lessen semivolatile (e.g. Tc , Cs, & I) loss relative to bubbler mixing (current plan)**

• **Mechanical stirring greatly accelerates the melting process (smaller/cheaper melters could be used)**

•**They readily handle multiphasic (“chunky” or “stringy”) feeds – non stirred melters can’t (simplifies feed preparation)**

•**They have already been utilized/proven for several DOE/SRS projects[1,2]**

•**This project would <u>not</u> require high temperature melters or “advanced” glasses**

1. **Marra C., “Vitrification of Simulated Radioactive Rocky Flats Plutonium Containing Ash Residue with a Stir Melter System”, Westinghouse Savannah River Company, WSRC-MS-96-0442.**
2. **D. F. Bickford et. al., TTP SRI-6-WT-31, Milestone XXX, Milestone C.I-2 Report: Functional Test of Pour Spout Insert and Knife Edge, WSRC TR 99 DO232, Rev 0, 1999.**

Why "Ceramicrete"

Putting the glass back into Hanford's tanks will require that it be rendered "pumpable" – a good way to do this would be to make "aggregate" (marbles[1], "gems"[2], or cullet) which could be slurried with a suitable "grout" (cement + water + clay?) & pumped with conventional equipment

Fe-P glass is compatible[3] with magnesium phosphate based grouts because they share a common chemistry (components, pH, etc).

Ceramicrete has been extensively studied as a stand-alone waste form[4]

[1]Germany's PAMELA ("Vitromet") process would have embedded HLW phosphate glass marbles in a molten lead "grout"

[2] Energy Solutions/VSL radwaste-to-glass "gems" project (Picket et al., "Vitrification and Privatization success", **WSRC-MS-2000-00305, Rev. 1,** 1995)

[3] Borosilicate glasses are incompatible with the OPC/flyash-based grouts usually specified for EM work

[4]Cantrell & Westsik, "Secondary Waste Form Down Selection... CERAMICRETE", PNNL-20681, August 2011.

Glass to Aggregate Examples

1995 report ("Vitrification and Privatization Success") describing the conversion of 670,000 gallons of "mixed" DOE radwaste to glass "gems" for $13.9 M

http://sti.srs.gov/fulltext/ms2000305r1/ms2000305r1.html

Another 1995 report comparing cullet, "gem", marble & monolithic options for DWPF's HLW glass

http://www.osti.gov/bridge/servlets/purl/274186-Majyqw/webviewable/274186.pdf

Today's real-world price for glass "gems" or marbles, any size, is $2.49/# - free shipping!

http://www.mcgillswarehouse.com/c/119/38

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*since SRS/DOE assumed that DWPF's glass would be shipped off to YM, the simpler-to-produce & ~35% less voluminous monolith option was deemed "best"

WHY REUSE HANFORD'S TANKS?

- Tanks are not "evil"
- They've already been paid for[note1]
- Steel-lined concrete tanks are apt to more durable than a plastic-lined pit (aka "tumulus")[note2]
- This waste management scenario would simultaneously "remediate" them[note3]

Ball park calculations

- 22 wt% Na_2O waste loading into Fe-P would put 48,000 tonnes of Na (100% of Hanford's waste) into ~3.0E+11 grams of glass
- Assuming 2.9 g/cc & 3785 cc/gallon, that translates to ~27 million gallons of glass
- Assuming 36% void volume[note] for randomly dumped spherical aggregate, this translates to needing about 42 million gallon's worth of "grouted" glass ball storage/repository space...well under the already-paid-for tankage)

note

Real World Glass Costs*

- Most fiber glass (aka “wool”) is produced with electric melters
- In 1999, the USA made 3.04 million tons of it for $4.8 billion
- Average inflation rate between 1999 & 2012 was ~ 3%

Consequently, making 27 M gallons (326,000 tons) ***should*** cost

$$\$4.8*(326{,}000/3{,}040{,}000)*1.03^{(2012-1999)} \approx \$0.76 \text{ billion}$$

*http://www.nrel.gov/docs/fy02osti/32135.pdf

Additive Cost Ballparking

- Experience suggests that component ratios of 1 Na:1.1P:0.4 Fe (and/or Al) would be OK for a mostly sodium salt-based Fe-P. This means that vitrifying 100% of Hanford's waste would require ~265,000 tonnes of fertilizer grade phosphoric acid (~3% of annual US consumption) and ~73,000 tonnes of iron ore - at current bulk commodity prices (~$600 & $2/ton) that adds up to ~$180 million

- At present, "Ceramicrete" is a specialty cement costing considerably more than OPC – however, its bulk manufacture would probably reduce cost to about $500/ton which would add another $10-15 million to the "additive" cost

Summation

DOE EM/ORP should "study" this proposal because...

1) Nuclear power's future prospects depend upon what today's leaders do with existing reprocessing wastes & how much money they spend (the institution's viability is at stake)

2) That's important because the implementation of a sustainable* "nuclear renaissance" could head off otherwise almost inevitable environmental degradation[note 1]

3) The USA can't afford to continue to waste $billions[note 2] on politically correct but technically unrealistic EM boondoggles

4) This proposal is simultaneously "doable", quick, affordable, and both technically & environmentally correct

* *"sustainable" means breeder reactors which means more reprocessing waste*

Extra slides

Formulation Guidelines[1,2]

"normalized release of Al, K, Na, and P from Fe-P is about 10 times less than B, Li, Na, and Si release from EA borosilicate glass"

"Na_2O and K_2O behave similarly in a glass"

"intermediate metal cations, such as Al^{+3}, Fe^{+3}, Cr^{+3}, and Zr^{+4}, are believed to form O–Me–O–P bonds in these glasses that connect isolated $P_2O_7^{-4}$ (pyro) or PO_4^{-3} (ortho) groups at high waste loadings so as to provide a high resistance to crystallization"

"… good chemical durability when Fe (or Fe+Al)/P is between 0.4 and 0.8"

….when the O/P (or O/(P+Si) ratio is between 3.4 and 3.8"

…when Na_2O content < 23wt%

…when P/Na≥1

1. Huang et al., Journal of Nuclear Materials, 327(2004) 46-57

2. R. Leerssen, "Fe Phosphate Glass for the Vitrification of INEEL SBW and Hanford LAW", MS Thesis, UMR (now MST), 2002

THE GALVIN COMMISSION’S REPORT (1995)

“Two yardsticks are useful in judging the EM program: progress toward cleanup goals and the costs incurred”,

“The remediation program has accomplished far less than many wish. The Government Accounting Office[8], ... concluded that while "DOE has received about $23 billion for environmental management since 1989, .. little cleanup has resulted. “

" A May 1994 Congressional Budget Office (CBO) Study[9] noted that DOE "has been criticized for inefficiency and inaction in its cleanup efforts. ... [and] has been severely criticized because of the small amount of visible cleanup that has been accomplished." These conclusions are shared by many senior DOE personnel, both within and outside the program.”

“One of the consequences of the troubles has been the enhancement of a syndrome common to large bureaucracies: risk aversion. It has a name: "the Hanford Syndrome." It has become widespread and severe in the EM program. Its symptoms are an unwillingness to alter familiar behavior patterns, to stick with unproductive or failing procedures, to enhance tendencies for excessive resource allocation and regulation, and to oppose innovation. It is an important element in sustaining unproductive patterns of work.”

“The Tri-Party Agreement at Hanford, and similar ones elsewhere, have proven to constitute major constraints on remediation progress because, in many instances, they are unrealistic, not having had proper input from those experienced in actual cleanup. The milestones they incorporate, along with penalties for noncompliance, force continued activities, some of which are make-work and should be abandoned. Other activities should be delayed or modified so as to await more effective and less costly technologies. Virtually no one believes the timetables are achievable and DOE has already been forced into renegotiations...”.

“There is a marked incapacity within the Department's EM program to evaluate current and prospective technologies in a wide-ranging and competent manner based on well-assessed risks.”

www.lbl.gov/LBL-PID/Galvin-Report/Galvin-Report.html

A “Troubled” Project

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- “Despite the big money and big employment figures (typ. 10,000)..., many feel that cleanup is off-track. Very little of the radionuclide and chemical inventory has been stabilized after thirty years of effort.”
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Current Reclassification Basis of ILAW*

- Current approach for Immobilized Low-Activity Waste (that allows the High-Level Waste to be disposed in near surface facilities, rather than a deep geologic repository licensed by NRC) comes from a series of technical letters between USDOE and the NRC in the 1980's and 1990's.
- In 1993, NRC spelled out three criteria in a letter to USDOE:
  1. Tank wastes have been processed (or be further processed) to remove key radionuclides to maximum extent technically and economically practical.
  2. Wastes will be incorporated in a solid physical form at a concentration that does not exceed the applicable concentration limits for Class C [low-level waste] as set out in 10 CFR Part 61.
  3. Wastes are to be managed so that safety requirements comparable to the performance objectives set out in 10 CFR Part 61 Subpart C are satisfied.

***slide 10,"Tank Waste Final Waste Form Perspective", Suzanne Dahl, WA dept of ECOLOGY, Jan 13, 2010**