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several orders of magnitude lower yet. The substantial drop in nitrate diffusivity between the first and later leach intervals indicates that ~90% of it had been "microencapsulated" within sodalite/cancrinite cages – the remainder leached as it would from conventional grout.

Table I: ANS/ANSI 16.1 Leach Test Performance

Today's most popular HLW leach test is the "product consistency test" (PCT)[27] because it is relatively simple and quick to do. Since it involves a week-long exposure of finely powdered sample material to a relatively small amount (10 x as much) of hot (90°C) water, it generates an estimate of the material's gross solubility under conditions apt to cause saturation. Table II compares several "representative" radwaste-type glasses[28] with several HCs with respect to dissolution of their most soluble common component, sodium. For "easy" components such as ^{90}Sr, the HC's would have outperformed the glasses by a greater margin.

Table II: Comparison of HCs with glasses on the PCT test

Most of the attention now being paid to INEEL's reprocessing wastes is focused upon the <10% which had not yet been calcined by the time (1991) ICPP/INTEC lost its reprocessing mission – and which still hasn't been. Because this "sodium bearing waste" (SBW) contains a higher proportion of thermally-stable alkali nitrates (which melt but don't decompose at practical calcination temperatures) than did the other liquid waste streams, it cannot be efficiently processed in the existing calciner unless a reducing agent (e.g., sugar) is first dissolved in it[7,8] – an option that DOE-ID has rejected. Table III gives the results of a TCLP (EPA Method 1311, SWP 846) leach test applied to an HC made with a sugar-calcined SBW simulant that had been doped with unrealistically high levels of several RCRA metals. The simulant was calcined as follows: After 38 grams of sucrose per mole of nitrate had been dissolved in the liquid, it was then slowly added to a stainless steel beaker situated on a maximum-temperature hotplate. Then that beaker was placed into a muffle furnace preheated to NWCF's normal operating temperature (500°C) to burn out residual elemental carbon. The HC formulation consisted of 30 wt % of this calcine, ~1% sodium sulfide, a small amount of household lye to provide "free" hydroxide (the sodium in the calcine itself was present as a ~1:2 mix of sodium aluminate plus sodium carbonate), plus sufficient water to make a "stiff" modeling clay-like dough. This was rolled into a ball, wrapped with tin (not aluminum!) foil, and than autoclaved for two hours at ~200°C. Table III lists regulatory limits along with the concentrations of "characteristic" metals in both the calcine and the TCLP leachate.

TABLE III: TCLP Results: Sugar-calcined "sodium bearing waste" specimen

HYDROCERAMICs vs "REGULAR" GEOPOLYMERIC CONCRETES

Hydroceramics are geopolymeric[1] concretes designed to minimize solubility of the "aggregate". In order to achieve the quick-set characteristics needed for construction-work, commercial geopolymeric cements are usually activated with alkali polysilicate(s), not with alkali hydroxide(s) and often contain substantial proportions of CSH-forming components (e.g., granulated blast furnace slag) too[29]. Table 4 compares PCT leach performance of three "geopolymeric" concretes (same formulation, different curing conditions) activated with sodium silicate and a similar "hydroceramic" activated with hydroxide alone. The waste simulant represents the soluble fraction of the caustic-neutralized liquid waste present in tank #44 at DOE's SRS site [~11.5 M sodium hydroxide, 1.5 M sodium nitrate, 1.13 M sodium nitrate, 0.4 M sodium aluminate, 0.2 M sodium carbonate, plus a trace of cesium chloride.] A 10:1 mix of "Troy clay" plus powdered vermiculite was used for all of them. 1.1 grams of a 37% NaOH was added to the "hydroceramic" formulation (10 grams of the clay mix plus 5 grams of the simulant). The "geopolymeric" formulation (11 grams of the clay mix plus 5.06 grams of the waste simulant) was activated with 2.5 grams of liquid sodium silicate ("water glass", ~38% solids, $SiO_2:Na_2O$ wt. ratio of 3.22:1). While the physical characteristics of all of the concretes appeared to be identical, the leach results indicate that polysilicate does not reform clay into salt-fixing minerals as effectively as does hydroxide alone. Because of their

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excellent durabilities and chemical compatibility with HCs, the best use for conventional geopolymeric cements in this context would be as construction and/or backfill materials.

TABLE IV: PCT leachability of geopolymeric vs hydroceramic concretes

These leach tests indicate both that the chemical durability of HCs is equivalent to that of vitrified materials and that the leaching of individual constituents is not determined by congruent matrix dissolution. Single-phase waste form materials such as "WV-39-2" glass tend to release everything at the same concentration-normalized rate – HCs do not. Like many rocks, HC concretes consist of assemblages of physically interlocked crystalline minerals possessing differing intrinsic solubilities. Due to the rapid, batch-nature of the process used to make them, the porosity (~15-25 %) and total surface areas (~15 m^2/g) of HCs are more like those of conventional concretes than natural rocks or glasses. HCs match the performance of glasses on leach tests because their lower intrinsic solubility compensates for their greater surface areas. Because the cation fixation sites in conventional grouts tend to be saturated with calcium ions (portland cement is ~65 wt% CaO), HC's generally outperform them with respect to the leachability of "easy stuff" too (e.g., ^{90}Sr). This may also explain why the MCC-1 leachability of Cs from HCs is about ~ 2 orders of magnitude lower than it was from the original FUETAP formulations[23].

SUITABILITY AS A DISPOSAL FORM

The US federal government's decision to confound disposal of its own waste with that produced by the commercial nuclear power industry constitutes another reason why it has failed to honor its promises to people living near its reprocessing facilities. Due to DOD insistence that DOE's civilian waste management responsibilities not interfere with its interests in NFSI[1,39,231,232], the federal government chose to "withdraw" another ~600 km^2 of land from Nevada for today's official HLW repository modeling exercise (YM). This plus its assertion that *all* commercially-produced HLW is to be sent there plus the fact that the exact consequences of hypothesized future floods at YM are impossible to predict (YM is a heterogeneous assemblage of different types of brittle rock situated in a seismically active region) will continue to engender enough confusion/litigation/paperwork/excuses to indefinitely stall implementation of this repository too – which is why linking these two problems engenders total paralysis. The most reasonable place for the federal government to site a repository dedicated to its cold-war defense-type waste is at its cold-war defense-type test range, the Nevada Test Site (NTS). The NTS makes good sense because, a) it's already "federal land" (no new "withdrawal" required), b) it receives less precipitation than do other DOE sites, c) it possesses the USA's deepest water table, d) it has already been the object of over thirty years worth of immediately relevant hydrological research[14-15], e) it has already been irredeemably "crapped up" by ~950 nuclear "events", and, finally, f) a little-publicized example of a practical repository (~$300/$m^3$ disposal cost) for this sort of waste has already been implemented there and then exhaustively tested[16,17]. However, it is *not* necessary for DOE to wait for a repository siting decision to render INEEL waste road-ready – regardless of exactly *where* in the Great Basin region this waste might eventually go, HC waste forms would probably prove to be more durable than glasses because their mineralogical similarity to surrounding soils/rocks would provide less thermodynamic driving force for alteration.

OTHER CONSIDERATIONS

The inventors of the original FUETAP process addressed the "radiolytic pressurization" bugaboo by proving that only the chemically uncombined water in concrete serves as a potential source of radiolytic gas[25]. Consequently, the porewater in HC concretes would also be baked out before the canisters were sealed. [Sellafield's "historic waste" solidification facilities sidestep this issue by venting their canisters through tiny filters.] Due to local stakeholder insistence that DOE-ID pay *some* attention to HLW management scenarios which would render all of the waste road-ready (not just "volume-reduced" fractions), several paper-engineering studies of the HC solidification process have been commissioned since 1996[30,31]. However, because no programmatic funding was allocated for laboratory or pilot plant studies, the process' main liability is "immaturity" – the same handicap that demoted FUETAP to "runner up" status in a independent review of solidification technologies for INEEL twenty years ago[32]. While it is reasonable to assume that an HC process could be implemented in a straightforward fashion, some

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important questions need to be answered before a real facility is designed. For example, "would a few month's worth of curing at the ambient-pressure boiling point of water at INEEL (zero gauge pressure) make an HC concrete comparable to one produced within two hours in a 200 psi autoclave?" If the answer is "yes", the process would be safer/simpler/cheaper to implement.

CONCLUSIONS

The "hydroceramic alternative" is especially attractive at INEEL for these reasons:

1) INEEL has not yet officially committed to vitrification.
2) Because INEEL calcines do not contain excessive concentrations of soluble salts, it would be possible to satisfy the "sodalite formulation" rule-of-thumb with high waste loadings.
3) Since two of the three elements making up HC binder phases (Na & Al) are high-percentage constituents of INEEL calcines, there is no need to separate them (or anything else) prior to solidification. This means that everything would be prepared for offsite disposal – the promise made to INEEL stakeholders. {A primary goal of the "volume reduction" practiced at WVDP and SRS is to transfer those elements to "low level" fractions.}
4) Straightforward changes to the existing calcination facility would permit it to efficiently calcine the remaining liquid waste[6,7] – either alone or (preferably) after it has been slurry-mixed with existing calcines[31]. The latter scenario would consolidate all INEEL reprocessing wastes into a single, relatively homogeneous, feedstream ideally suited for HC solidification.
5) It would also be a good way to deal with other INEEL wastes. For example, INEEL must find some way to dispose of ~1000 metric tons of radioactive NaOH generated by reacting metallic sodium reactor coolant with water. Since this just happens to be the amount of "activator" required to turn ICPP/INTEC's calcines into HC-type concrete, coprocessing them would solve both problems. If the changes to the existing calcination facility alluded to above were to be implemented, virtually any sort of liquid or particulate waste (e.g., contaminated soils) could also be accommodated.
6) It is probable that a formal proposal to implement an HC-type solidification process would satisfy INEEL stakeholders[12,35].
7) Finally, if a future generation of US taxpayer deems it to be both politically expedient and affordable, HC-type concrete monoliths could be hot-isostatically-pressed into "vitrified" monoliths without removing them from their original canisters[24-36]

Two months ago, DOE released a five-volume Draft Environmental Impact Statement (DEIS) describing proposed management scenarios for INEEL HLW[37]. Unfortunately, "command influence" has rendered this document useless as a tool for decision-making. For example, its authors were told to assume a "disposal fee" of $850,000/m^3 for any "high level" waste forms produced – which, of course, constitutes an overwhelming bias for schemes which invoke "volume reduction" of already-calcined waste. Let's look at numbers: if INEEL's 320 MTHM's worth of reprocessing waste (which represents 0.46% of YM's "capacity") were to be converted to 13,000 m^3 of HC-type concrete via the "Direct Cement Waste Option", DOE would charge US taxpayers $11 billion for disposal; if the same waste were to be dissolved (which would require about 200,000,000 gram-moles of nitric acid), chemically separated, and converted to 470 m^3 of high-level glass and 30,000 m^3 of low-level "grout" (the "Full Separations Option")", DOE would charge them *only* ~$400 million for the same service.

The National Research Council released its review[38] of INEEL's HLW management program one month before the DEIS was issued. While the NRC report again challenges the validity of many of DOE's assumptions, it eventually concludes that, under the present circumstances, it would be "best" to abrogate the latest promises made to Idaho; i.e., to not calcine the remaining liquid waste and to just let the existing calcines decay away in the binsets for a few hundred more years.

In this writer's opinion, that's precisely the conclusion that DOE had been hoping for. It is also unnecessarily defeatist because if DOE were willing to eschew some of its "symptoms"[4], it could keep those promises. "Sugar calcination" of SBW was discovered at Argonne National Laboratory in the late 1950s and tested in INEEL pilot plants 35 years ago[7] and, again, in a lab-scale system, 4 years ago. The

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same approach was reinvented, pilot-plant tested, & then heartily recommended by Hanford subcontractors in 1995[8]. BNFL routinely implements sugar-calcination of SBW in its rotary calciners at Sellafield. Its virtues relative to the "high temperature" (~600°C) approach espoused by DOE-ID include: the bulk of the nitrate is converted to elemental nitrogen rather than to toxic (& visible) NO_x, one third as much of the solid-forming "cold" additive (aluminum nitrate) is required, a lower percentage of troublesome "fines" are produced (they tend to plug the offgas system), and calcination of the remaining waste would take half as long and produce half as much calcine. In light of this, DOE-ID's arbitrary rejection of sugar calcination because of "safety considerations" simply suggests that it does not want to keep its promise. Similarly, its refusal to devote programmatic research funding to the alternative solidification technology described in this paper (and also, inaccurately, in its DEIS) suggests that the federal government is no more committed to rendering INEEL's calcines road-ready now than it was twenty years ago.

Because radionuclides have finite lifetimes and US reprocessing waste will never pose an *immediate* hazard to the public-at-large, it will always be possible for the federal government to make a plausible *technical* case for more "temporary delay". Management of this waste is not a purely "technical issue" because it also involves "people issues" such as missions, money, promises, careers, ethics, and institutional credibility. The viability of the US nuclear power industry requires tangible proof that the federal government's waste management bureaucracy is willing to deal with its own garbage.

ACKNOWLEDGMENT

Without the technical input of my colleagues at Pennsylvania State University's Materials Research Laboratory, Professors Barry Scheetz, Michael Grutzeck, and Delia Roy, the "hydroceramic alternative" would not have been developed. Without the vision provided by that institution's founder, Professor Rustum Roy, the need for it would not have been realized.

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Figure 1: MCC-1 Performance of an HC vs Glasses and a HIP Glass-Ceramic

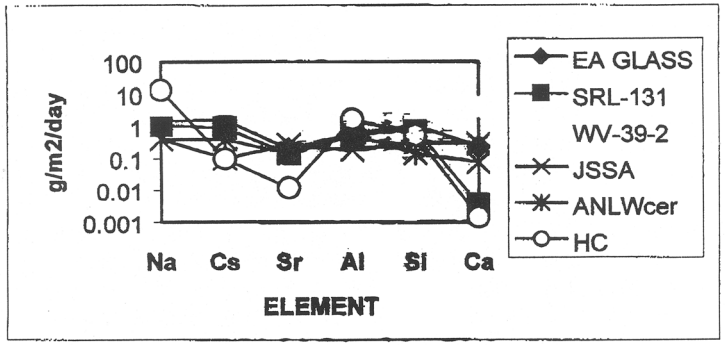

Table I: ANS/ANSI 16.1 Leach Performance*

Interval	Sodium		Cesium		Zirconium		Strontium		Chromium		NO_3^-	
(hrs)	ppm	-logD	ppb	-logD	ppb	-logD	ppb	-logD	ppm	-logD	ppm	-logD
2.83	32	9.55	<2	>13.8	33	15.3	3	14.1	0.28	10.6	23	8.3
5.7	20	9.7	<2	>13.5	68	14.4	2	14.1	0.21	10.6	5	9.3
15.3	28	9.8	<2	>13.9	140	14.1	7	13.5	0.076	11.8	2.3	10.4
19.5	13	10.3	<2	>13.7	<10	>15.3	1	15.0	0.05	11.1	0.9	11.1
22	22	9.8	<2	>13.9	100	14.2	6	13.4	0.03	12.4	1	10.9
35.8	21	10.1	<2	>13.5	<10	>16.4	1	15.2	0.02	13.0	1	11.1
25.5	15	9.9	<2	>13.7	<10	>16.0	3	14.2	0.02	12.6	0.4	11.5
36	14	10.2	<2	>13.7	<10	>15.2	1	14.9	0.01	13.4	0.9	11.0
LI	9.9		13.7		>15.4		14.3		12.0		10.4	
Total % Leached	8.26		<0.0099		<0.0025		0.015		1.2		10.5	

the <> figures in this table are based upon detection capabilities of the analytical instrumentation: ICPAES for all metals except Cs, graphite furnace AAS for Cs, and ion chromatography for nitrate

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Table II: Comparison of HCs and glasses on the PCT test

MATERIAL	% Na_2O	mg/l Na in leachate	% Na dissolved
EA GLASS	16.9	1720	13.7
PUREX GLASS	12.1	941	10.4
SRL-131	12.9	931	9.7
HC#1 $NaAlO_2$/NaOH/TROY clay	16.7	718	5.8
HC#2 NaOH, $NaNO_3$ (25% of Na)/TROY clay	12.6	513	5.5 (2.6% of the NO_3^- had also leached)
HC#3 38% alumina calcine/NaOH/DEA/TROY clay	13.1	554	5.7
HC#4 46% zirconia calcine/NaOH/TROY clay	12.4	558	6.1
HC#5 30% sugar-calcined SBW/TROY clay*	12.6	925	9.9
HC#6 NaOH/ Englehard Metakaolinite, 9-hr cure @ 200 °C	16.3	229	1.9 (ANSI 16.1 LI_{Na} = 11.6)

* This particular HC violated the "sodalite composition" rule of thumb - too much carbonate

TABLE III: TCLP Results: Sugar-calcined "sodium bearing waste" specimen

Analyte	Found(ug/g)	Limit (ug/g)	Calcine (ug/g)
As	<0.002	5	10.8
Ba	0.35	100	48
Cd	0.13	1	1372
Cr	0.023	5	950
Hg	<0.01	0.2	<0.01*
Pb	<0.1	5	1500
Se	<0.002	1	6.9
Ag	<0.1	5	1510

* Mercury was not added to the liquid simulant because it would have been lost during subsequent calcination. In a properly-implemented real calcination system, mercury would be recovered from the off gas.

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TABLE IV: PCT* leachability of geopolymeric vs hydroceramic concretes

	Hydroceramic	Geopolymer	Geopolymer	Geopolymer
Cure Conditions	200°C, 2 hours	200°C, 2 hours	90°C, 4 days	~20°C, 4 days
pH of leachate	10.7	11.3	11.7	12.3
% Na leached	7.1	9.6	21	52
% Cs leached	0.086	0.060	0.18	2.0
% nitrite leached	26	36	51	71
% nitrate leached	14	46	57	71

*samples crushed to pass 100 mesh screen (150 micron)-- no lower size limit, powders leached with 10x as much 90°C distilled water,

ACIdoc

HLW & FD EIS PROJECT - (AR) PF
Control # DC-81

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March 12, 2000

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Attention: Public Comment: Idaho HLW & FD EIS

RECEIVED APR 21 2000

Mr. Wichmann,

Please accept this as my formal written commentary on DOE/EIS-0287D, the Idaho High-Level Waste and Facilities Disposition Draft Environmental Impact Statement dated December 1999.

A fully acceptable solution to the problem of what to do with radioactive waste has never been implemented or even discussed. I will here present my thoughts on the subject.

A. Repository Location

81-1 X1 (7) Because waste radioactive materials must be isolated from the biosphere and because water transport is the principal mechanism for migration (after carefully excluding tectonic activity), a truly dry location with no access to a water table must be chosen.

The current U.S. repository sites fail to meet the dual site-selection criteria: no tectonic activity and no water. In fact, no U.S. locations at all meet both these criteria. Have you seriously considered locations outside the United States? I would like to point out that according to the global seismic hazard map on the web at http://seismo.ethz.ch/GSHAP/ there are large regions in Africa that appear to be low seismic risk and presumably quite dry. In fact a line all the way across that continent at 20 degrees north latitude appears free of seismic hazard. I suggest serious negotiations (and serious resources) be engaged in this region for repository selection, characterization, and implementation.

I feel the Yucca Mountain site is totally unacceptable as a high-level waste repository due to the tectonic hazard there. The close proximity, geologically, to the phreatic eruption site at Ubehebe Crater in Death valley shows what I mean. This class of volcano has the potential to blow hundreds of cubic miles of earth into the sky, as it did just up the road, at the Crowley Lake / Mammoth Lakes area on the east side of the Sierra Nevada.