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Mr. John J. Linehan, Acting Chief
Repository Projects Branch
Division of Waste Management
U. S. Nuclear Regulatory Commission
Washington, DC 20555

Distribution:

LINEHAN   HILDENBRAND
BROOKS
(Return to WM, 623-SS)

Dear Mr. Linehan:

NRC DRAFT SITE TECHNICAL POSITION, DOE HIGH LEVEL WASTE REFERENCE REPOSITORY
LOCATION, HANFORD SITE, "USE OF HYDRAZINE TO EXPERIMENTALLY SIMULATE EXPECTED
SITE REDOX CONDITIONS AND REACTIONS"

Reference: Letter, Linehan/Olson, 10/8/86, Same Subject

The reference letter contains an enclosure which is a letter from
Mr. David Brooks of the NRC staff to Dr. Sam Panno of Weston, dated
September 3, 1986, describing NRC concerns with the use of hydrazine.
The BWI review and comments on both the September 3 letter and the subject
draft site technical position are attached.

Should you require further assistance concerning this matter, please contact
Mr. J. E. Mecca of my staff on FTS 444-5038. After your review and our
submittal of draft SCP program material, further discussion of these issues
may be in order.

Sincerely,

John H. Anttonen, Assistant Manager
for Commercial Nuclear Waste

BWI:JMK

Attachment

cc: J. P. Knight, DOE-HQ, w/encl.

'87 MAR -2 A11:42

WM DOCKET CONTROL CENTER

2340

Dec 11, 1986

To: Chief, LES Branch

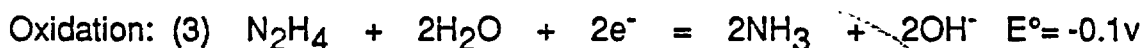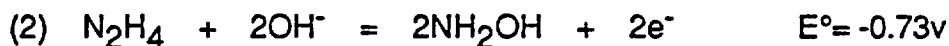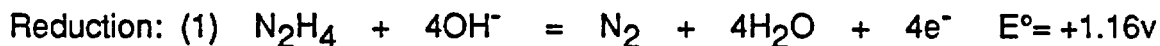Through: Chief, G&T Branch

From: M. J. Furman

NRC   DSTP ON THE "USE OF HYDRAZINE TO SIMULATE HANFORD SITE REDOX CONDITIONS AND REACTIONS"

     Following are the comments and discussion on the NRC's Draft Site Technical Position (DSTP) on the use of hydrazine in BWIP sorption experiments.   The information that was provided for evaluation consisted of two parts: A letter of 3 September 1986 to Sam Panno of Weston; and, the actual DSTP that concludes with six general concerns that are also the basis of the NRC-Panno letter.   The six concerns are the essence of the DSTP, and each of these are addressed in the following comments.   In all cases, exception is taken to each of the concerns.

     The impact of the DSTP is potentially significant, since a large part of the sorption data acquired so far involves the use of hydrazine.   These data form a lower bound for the sorption parameters.

1. DSTP Concern 1.   "Hydrazine can exhibit both reduction reactions and oxidation reactions which have widely different standard potentials. Thus experimental redox conditions may not reflect site redox conditions."

The widely different half cell potentials are a distinct _advantage_ in this case because reactions that can occur when hydrazine is introduced into the rock-groundwater system can be predicted with more confidence. All evidence points to _reduction_ of the radionuclides by hydrazine rather than oxidation. In the basic groundwaters of deep basalts at Hanford (pH ~ 9.5) the appropriate potentials are:

Reduction: (1) $N_2H_4 + 4OH^- = N_2 + 4H_2O + 4e^-$ $E° = +1.16v$

(2) $N_2H_4 + 2OH^- = 2NH_2OH + 2e^-$ $E° = -0.73v$

Oxidation: (3) $N_2H_4 + 2H_2O + 2e^- = 2NH_3 + 2OH^-$ $E° = -0.1v$

Based on these potentials, reduction by hydrazine (reaction 1) is obviously more thermodynamically favored than oxidation (reaction 3) in BWIP's basic groundwaters. Reduction of a wide variety of metal ions by hydrazine is reported in the literature [Co(III), Fe(III), Ce(IV), Ti(IV), Ag(I), Cu(II), Mo(VI), Mn(VII), Cr(VI), V(V)] in addition to radionuclide ions [Pu(IV), Pu(V), Np(VI), Np(V), U(VI), Se(IV), and Tc(IV)]. Oxidation of metal ions by hydrazine is possible _only_ under conditions far different than those that exist in the Hanford site's groundwaters (acidic pH and strongly reducing metal ions).

Since we know that reduction and not oxidation of radionuclides will occur with hydrazine, the remaining questions are (1) are the reduction reactions fast enough to be useful in these experiments? (2) what are the reduction products? and (3) are the reaction products the same as those expected in a basalt-groundwater environment? Information in the chemical literature can help provide answers to questions (1) and (2). The oxidation states for most radionuclide reduction products can be predicted with confidence from studies reported in the literature (Table I). Reduction kinetics for a number of these reactions are also available. Additional evidence for reduction is the significant increase in sorption of reducible radionuclides observed when hydrazine is present. The reduced radionuclides are more strongly bonded to mineral surfaces than radionuclides in higher oxidation states.

Table I. Oxidation States of Radionuclides Reduced by
0.01 $\underline{M}$ hydrazine in Groundwater Solution

Element	Starting species	Reduced oxidation state	References
Uranium	UO_2^{2+}	U(IV)	Kalinins and Gibson, 1959
Neptunium	NpO_2^+	Np(IV)	Keller, 1971; Koltunov and Tikhonov, 1973
Plutonium	PuO_2^+	Pu(IV) or Pu(III)	Koltunov and Zhurzvleva, 1974
Technetium	TcO_4^-	Tc(IV)	Spitsyn et al., 1983; Meyer et al., 1985
Selenium	SeO_3^{2+}	Se(-II)	Benzing et al., 1957

The NRC claims that Np(V) is not reduced by hydrazine (Kelmers et al., 1985b) and that the large increase in Np sorption observed when hydrazine is present is not necessarily due to reduction of Np(V) to Np(IV). As reported by Koltunov and Tikhonov (1973), the reduction of Np(V) to Np(IV) by hydrazine at slightly basic pH values is slow at 25 °C, but does proceed at a measureable rate. As suggested by ORNL, it is possible that dissolved iron from the basalt catalyzes the reduction. In any case, the drastic increase in sorption (Kd values of 120 - 18,100 ml/g with hydrazine and ~ 1.7 ml/g without hydrazine as measured by ORNL) must be due to reduction since the presence of hydrazine does not significantly affect the sorption behavior of radionuclides such as Am(III), Pb(II), Zr(IV), and Sr(II) that are not reduced by hydrazine. Reduction of the remaining key radionuclides is relatively fast even in the absence of iron.

Based on estimates of redox conditions in deep Hanford basalt groundwater and laboratory determinations of radionuclide redox reactions with crushed basalt, it is expected that radionuclides released from the waste will be in reduced oxidation states. Evidence for redox conditions is summarized in the Environmental Assessment for the Hanford site (DOE, 1986, pages C.5-81to C.5-86) and is based on evidence from (1) secondary mineralogy in basalt flows, (2) measurements of concentrations of dissolved redox couples, and (3) platinum electrode measurements. Identification of reduced species of neptunium and technetium in reactions with crushed basalt in groundwater has been accomplished by several laboratories (Meyer et al., 1985; and Susak et al., 1983).

There appears to be a basic misconception by the NRC concerning the reasons for using hydrazine in the sorption experiments. BWIP has never intended to "simulate expected repository redox conditions" with hydrazine. Hydrazine is a much stronger reducing agent than the Fe(II) or S(-II) minerals that probably control redox conditions in the basalts (Barney, 1981). The reason for hydrazine addition to groundwater solutions was to produce and maintain lower oxidation states of radionuclides so that the sorption behavior of these reduced species could be examined. In order to understand a complex system it is often necessary to look at component parts under well defined (i. e. controlled) conditions that do not precisely correspond to "site" conditions. The reducing minerals in basalts react so slowly with radionuclides (at temperatures expected in the groundwater flow paths) that reduction with these minerals is not practical for most laboratory experiments. Hydrazine reacts rapidly with many key radionuclides and maintains the desired lower oxidation states over the course of sorption or solubility measurements. Hydrazine does not form complexes with radionuclides that could interfere with sorption or solubility reactions and produces innocuous reaction products (N_2 and H_2O) with oxidizing agents present in the groundwater-mineral systems.

Oxidation states of some radionuclides at all points along probable groundwater flow paths may be difficult to assess with absolute certainty. Uncertainties exist in identifying redox reactions that control radionuclide oxidation states and in predicting the kinetics of these reactions. The concept of a single master Eh value that controls all oxidation states is probably not applicable because redox couples may not be at equilibrium with each other (Lindberg and Runnells, 1984) and reactions of radionuclides with redox couples may be kinetically hindered. The BWIP has attempted to bound the possible oxidation states by using conditions that are more oxidizing (air-saturated solutions) and more reducing (solutions containing hydrazine) than will exist in the natural system. The actual behavior of radionuclides is expected to be between these extremes. Laboratory measurements of radionuclide sorption where both oxygen and hydrazine are removed from the groundwater-rock systems confirm that sorption behavior is between these extremes.

2. DSTP Concern 2. "Hydrazine hydrate dissociates to release hydroxide ions. Thus experimental conditions may not reflect site pH conditions."

The NRC is concerned that addition of hydrazine will raise the pH of groundwater solutions above that observed for natural groundwaters. In the BWIP experiments, small amounts of hydrochloric acid were added to groundwater solutions to neutralize the solutions to desired pH levels. The added chloride does not affect radionuclide sorption reactions (Barney, 1984). The NRC is concerned that

radionuclides might form stable chloride complexes with the increased levels of added chloride which would indeed affect sorption. However, comparison of formation constants for chloride complexes of actinides (Apps et al., 1982) and important fission products (Sillen and Martell, 1964) with formation constants for hydroxyl and carbonate complexes shows that chloride complexation will be insignificant for Hanford groundwaters even with the added chloride. Therefore, neutralization of the hydrazine solutions with HCl is a valid method.

3. DSTP Concern 3.   "Hydrazine can react with bicarbonate anion to form carbamate anion. Thus the experimental groundwater may no longer be representative of site groundwater conditions."

The formation of hydrazine carbamate ($H_2NNHCOO^-$) by reaction of bicarbonate with hydrazine in experimental solutions as suggested by the DSTP seems highly unlikely. The carbamate has only been prepared by reaction of carbon dioxide gas with hydrazine in strongly basic solutions of sodium hydroxide (Staal and Faurhalt, 1951). The carbamate was found to decompose in water to form hydrazine and carbon dioxide. This observation is consistent with fact that carbamic acid (H_2NCOOH) and carbamate ion are not obtainable in aqueous solution, but decompose to ammonium and carbonate ions (Cotton and Wilkinson, 1962). The presence of hydrazine in experimental groundwater solutions appears to have no effect on formation of carbonate complexes of Np(IV) or Tc(IV) (Barney et al., 1985). This concern is not backed by any credible evidence and should be eliminated.

4. DSTP Concern 4.   "Hydrazine is reported to disrupt the mineral structure of clays, and disaggregate rock. Thus the rock/mineral components of the experiments may no longer represent those found under site conditions."

The DSTP suggests that hydrazine may attack secondary minerals present in sorption experiments and alter their structures. However, if secondary minerals are altered to any significant extent, sorption of radionuclides that are not reduced by hydrazine should also be affected. Sorption studies of Am(III), Zr(IV), and Pb(II), which are not reduced by hydrazine, show that there is no significant difference in their sorption behavior with basalt, secondary minerals, or interbed materials in the presence or absence of hydrazine (Barney, 1981a,1982,1984; Ames et al., 1983).

Evidence that shows hydrazine does not interfere with sorption reactions is based on (1) observations that sorption of certain radionuclides that are not reduced by hydrazine are not affected by the presence of hydrazine and (2) similar sorption distribution coefficients for reduced radionuclides whether or not hydrazine is present.

Statistically designed measurements of Am sorption on basalt, secondary minerals, and several interbed materials (sandstone and tuff) by Barney (1981 and 1982) show that there is no significant difference in sorption whether or not hydrazine is present. Because hydrazine will not react with Am(III), any interference effects of hydrazine with the sorption process would show up as different sorption behavior when hydrazine is present. Since no differences are observed, it is concluded that hydrazine does not interfere with sorption. Because other radionuclides that are reduced by hydrazine will likely sorb by mechanisms that are similar to Am(III) sorption (complexation with oxygen ligands on the mineral surface), hydrazine will not interfere with their sorption either. The sorption behavior of Pb(II) and Zr(IV), neither of which is reduced by hydrazine, is not altered by the presence of hydrazine (Ames, 1983) as shown in the following plots:

Pb(II) Sorption on Cohassett Basalt
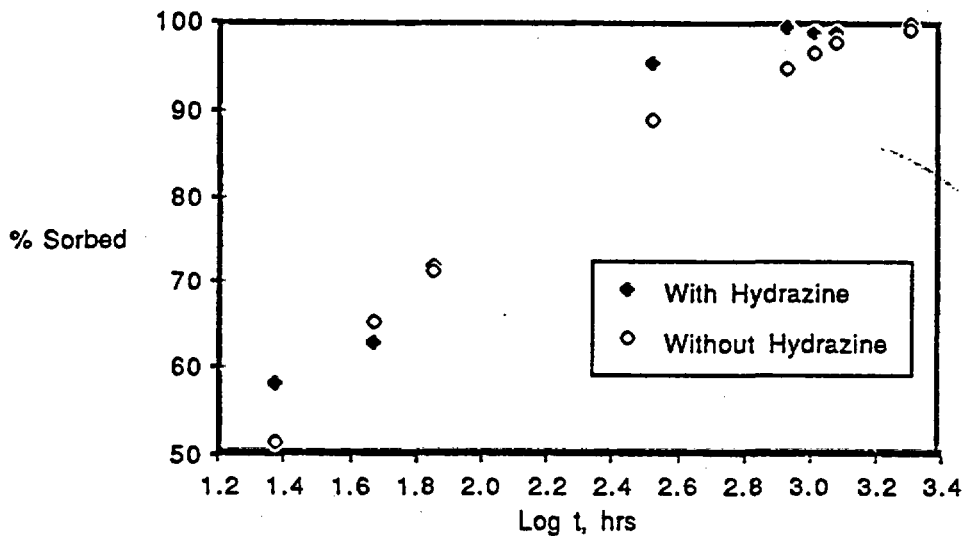
Zr(IV) Sorption on Umtanum Basalt
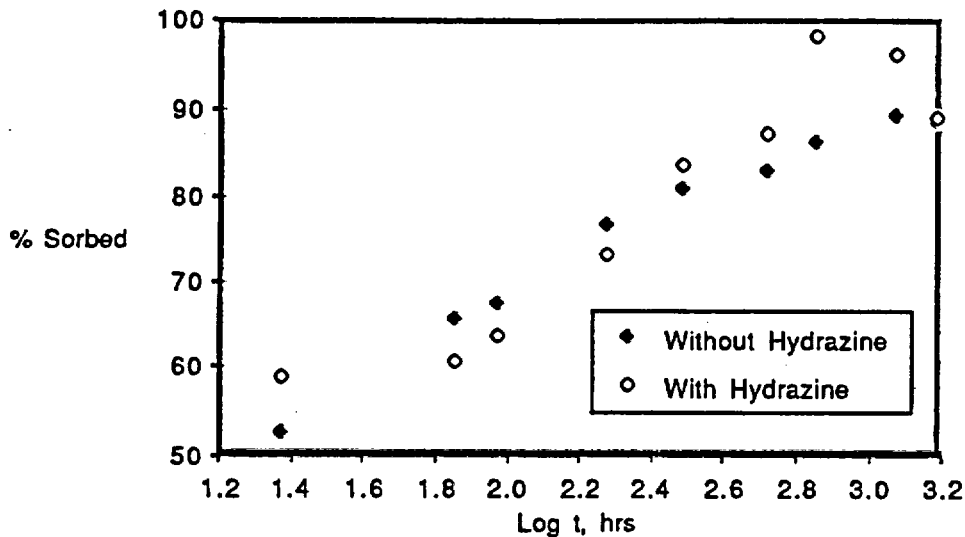
These plots show that the percentage of Pb(II) or Zr(IV) sorbed at a given time is not dependent on the presence or absence of hydrazine (within experimental error).

It is interesting to note also that ORNL-measured distribution coefficients for Tc, Np, and U sorption on basalt under anoxic conditions [when sufficient time was allowed for reduction of the Tc(VII), Np(V), and U(VI)] are essentially the same as those measured by BWIP in the presence of hydrazine (Kelmers et al., 1985d). Although they claim that the agreement may be coincidental, it provides additional proof that hydrazine does not interfere with sorption reactions.

For several radionuclides that are sorbed (at least partially) by ion exchange mechanisms such as $^{90}Sr^{2+}$, $^{226}Ra^{2+}$, and $^{137}Cs^+$, a change in ionic strength due to addition of hydrazine does affect sorption. The effect of hydrazine on sorption of these ions is to decrease sorption rather than increase sorption. Just the opposite effect is observed for radionuclides that can be reduced. Essentially none of the radionuclides considered to be "key" are sorbed by ion exchange.

5. DSTP Concern 5. "The chemistry of basalt rock/groundwater systems seems to be primarily dominated by rock components. Therefore, reactions leading to radionuclide removal from solution by

sorption or precipitation must involve heterogeneous reactions between basalt and radionuclide species in solution. Thus the addition of hydrazine (or any exogenous reducing chemical) seems likely to result in homogenous solution reactions involving radionuclides not expected under site conditions."

Another concern of the DSTP is that the rapid reduction of a radionuclide in aqueous solution by hydrazine may not accurately model the slow, heterogeneous reduction of the radionuclide that might occur in groundwater flow paths . It isn't clear from the DSTP what "heterogeneous reactions" might be occurring that would affect sorption or precipitation reactions differently than homogeneous reactions. However, in several ORNL reports (Kelmers et al., 1984b; Kelmers et al.,1984c) it is claimed that Tc(VII) and Np(IV) are reduced directly on the surface of basalts. The only evidence for this surface reaction is that more of the reduced species is found on the basalt surface than in solution after anoxic sorption experiments. This evidence is not convincing since the reduction could have taken place in solution with subsequent sorption on the surface. It is agreed that reduced species of Tc and Np are more strongly sorbed than oxidized species. Whether the reduced species are produced from homogeneous or heterogeneous reactions, it is the type of chemical bonding between surface atoms and radionuclides that will control sorption behavior of radionuclides. Recent models of adsorption of hydrolyzable metal ions on oxides from aqueous solutions point to formation of surface complexes at the water-oxide interface (Schindler, 1981; Davis et al., 1978; Hsi and Langmuir,1985). Coordinate bonding occurs between the surface oxygen ligands (e.g. from silicates or other mineral oxides) and the metal ion. For radionuclides such as Np, U, Pu, and Tc, the stability of complexes with ligands containing oxygen are greater for the reduced species [(III) or (IV) oxidation state] than the oxidized species [(V), (VI), or (VII) oxidation state]. This is consistent with observations that the reduced species of these radionuclides are more strongly sorbed than oxidized species. This type of surface bonding will prevail regardless of where the radionuclide is reduced or how fast it is reduced.

6. DSTP Concern 6.   "Hydrazine is a very aggressive chemical and reacts with plastics in the experimental apparatus. Thus complicating the interpretation of experimental data to the point that results may no longer represent site conditions or reactions."

This concern seems to be irrelevant to the intent of the DSTP. The NRC seems to be implying that hydrazine can't be used in adjusting radionuclide oxidation states in sorption experiments because we can't find materials for construction of apparatus that will not react with the hydrazine. This simply is not true and any competent

experimenter must be concerned that the apparatus used does not interfere with the processes being measured. BWIP has used Teflon, glass, polyethylene, and polypropylene containers in sorption work without any evidence of reaction with hydrazine. This concern should be eliminated from the final STP.

Specific Comments on the DSTP:

1. (Page 2, para. 2, line2) BWIP does not claim that all redox-sensitive radionuclides will be in their "least mobile state" only that they will be reduced to lower oxidation states by the basalt enviroment.

2. (Page 2, para. 2, line 7) As mentioned above, BWIP has never intended to "simulate expected repository redox conditions" with hydrazine. Hydrazine is a much stronger reducing agent than the Fe(II) or S(-II) minerals that probably control redox conditions in the basalts (Barney, 1981). The reason for hydrazine addition to groundwater solutions was to produce and maintain lower oxidation states of radionuclides so that the sorption behavior of these reduced species could be examined.

3. (Page3, para. 1, lines 3 - 11) The half-cell potential reactions for acidic conditions are not applicable to Hanford groundwater with pH values of 9 to 10. These should be eliminated. Also, the potential for the fifth reaction should be +1.275 volts.

4. (Page 3, para. 1, line 13) The references, Barney, 1982(a) and 1983, do not state that the experimental groundwater redox potential is -0.8 volts, but that this potential is a calculated value. Obviously, water would be reduced if the actual potential approached the calculated value. However, aqueous solutions of hydrazine are stable for long time periods.

5. (Page 4, para. 2, last line) Obviously, the pH can be adjusted to the in situ pH value by addition of acid.

6. (Page 6, para. 1, lines 11 -29) The discussion of technetium reduction by hydrazine leaves out some important references (Meyer et al., 1985; Spitsyn et al., 1983) that establish the technetium reaction product as Tc(IV). Also, the relevance of discussions of metallo-organic compounds and Tc(IV) organic compounds is not apparent since organics are not present in most sorption experiments.

7. (Page 7, para. 2, line 6) The statement that all BWIP sorption work has been performed using polycarbonate tubes is not true (see the more recent references of Barney and Ames) and should be eliminated.

8. (Page 7, para. 3, line 3) The statement that the use of hydrazine for characterization of radionuclide behavior will not yield any defensible data seems to be based on technical judgements stated in the DSTP that are not well supported by scientific evidence. The NRC has not provided any evidence that shows that hydrazine interferes with radionuclide sorption measurements. All of their objections to the use of hydrazine in these measurements are based on speculative assertions of problems that might exist. On the other hand, BWIP has provided evidence which addresses each of their objections.

It should be emphasized that much BWIP sorption data has been obtained without the use of hydrazine. Unfortunately, reduction reactions of radionuclides with most relevant geologic solids at temperatures expected in the groundwater flow paths from the repository are so slow that determination of sorption behavior of reduced radionuclides is simply not practical without the use of an added reducing agent. Hydrazine appears to be the best choice for reasons given in several BWIP reports. Obviously, this is not the only example of experiments in which "natural" conditions are purposely altered for logical reasons. Slow reaction kinetics create experimental problems in many areas of the repository programs and creative solutions that are scientifically sound are needed overcome these problems. The experimental alternative of using hydrazine to study the behavior of reduced radionuclides in Hanford basalts should be retained. Additional alternatives have been and will be examined in the future.

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