

STUDY ON THE COLLOIDS GENERATED FROM TESTING OF HIGH-LEVEL NUCLEAR WASTE GLASSES*

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ABSTRACT

The generation of colloids in the interaction of high-level nuclear waste glasses with groundwater at 90°C has been investigated. The stability of the colloidal suspensions has been characterized with respect to salt concentration, pH, time, particle size, and zeta potential. The compositions and the morphology of the colloids have also been determined with transmission electron microscopy (TEM). From our test results combined with earlier ones, we conclude that the waste glass may contribute to the colloid formation by increasing ion concentration in groundwater, which causes nucleation of colloids; by releasing radionuclides that adsorb onto existing groundwater colloids; and by spalling colloidal-size fragments from the surface layer of the reacted glass. The colloids are silicon-rich particles, such as smectites and uranium silicates. When the salt concentration in the solution is high the colloidal suspensions agglomerate. However, the agglomerated particles can be resuspended if the salt concentration is lowered by dilution with groundwater. The colloids agglomerate quickly after the leachate is cooled to room temperature. Most of the colloids settle out of the solution within a few days at ambient temperature. The isoelectric point is at a pH of approximately 1.0. Between pH 1 and 10.5, the colloids are negatively charged, which suggests that they will deposit readily on positively charged surfaces. The average particle size is largest at the isoelectric point and is smallest around pH 6.

INTRODUCTION

Colloids are a potential vehicle for the transport of radionuclides and other hazardous contaminants in a waste repository. However, colloidal studies to date (1-4) have been concentrated on pseudocolloids, which are formed by adsorption of radionuclides onto existing groundwater colloids, and real colloids, which are formed by solubility-limited hydrolysis and polymerization of actinides. Previous work at Argonne National Laboratory (5-7) has indicated that during the interaction of waste glass with the groundwater, the metastable glass may transform into more stable phases, which become a new source of colloidal material. The colloids formed in such a system display a wide size distribution, ranging from 3.8 nm to 1 μm (6). The typical colloids, by definition, consist of particles between 1 nm and 1 μm in diameter.

The work reported here is a continuation of our previous effort to characterize the colloids formed during waste glass dissolution under repository-relevant conditions. Our objective is to provide information that is pertinent to 1) resolving issues concerning the contribution of colloids to the radionuclide source term and 2) providing input to assist in designing an engineered barrier system that will restrict colloid transport in waste repositories. Here we report some preliminary results of our studies on 1) colloid generation, 2) stability of the colloidal suspensions, 3) effects of pH on the particle size distribution and charge (zeta potential), and 4) the mineral composition of the colloids.

EXPERIMENTAL

Materials and Methods

The colloids were generated through interaction of either simulated SRL 131S glass or actinide-doped SRL 131A glass with J-13 well water at 90°C. The initial pH of the J-13 water was 7.68. The glass and J-13 well water compositions were reported previously (8). The test procedure used is a modified Product Consistency Test (PCT), which is described in more detail elsewhere (9). In each test, the requisite quantity of 100-200 mesh powder glass was immersed in a volume of J-13 well water in a Teflon vessel. The volume of well water depended on the desired SA/V (ratio of surface area of glass to volume of solution). The test vessel was tightly sealed and placed in a 90°C oven. After termination of a test, the glass leachate was analyzed for colloids, and measurements were made to determine pH.

All the solutions used in this study were prepared without filtration. There was no effort to control the uptake or release of CO₂ from the colloidal solution because the relevant repository condition is supposed to be CO₂-saturated. After settling at room temperature overnight the colloidal solution was carefully withdrawn from the test vessel with a syringe. Care was exercised to avoid collecting small flakes of glass floating on the top of the leachates or the precipitates entrained in the solution at the bottom of the vessels.

Size Measurements

The size of the colloids was measured with a dynamic laser light scattering system (Malvern Instruments System 4700°C photon correlation spectrometer). A 35 mW HeNe laser was used as the light source in the Malvern spectrometer.

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The autocorrelation function was measured at 25°C and a light scattering angle at 90°. A 64 channel correlator was used in the parallel (geometrically spaced channels) and serial (equally spaced channels) modes. However, most of the data was collected in the serial mode. The correlation function was analyzed using CONTIN (10-12), a model independent data analysis program that inverts the time correlation function to give a distribution of relaxation times. The colloidal suspension generated from the interaction of glass with groundwater is polydisperse. It is usually characterized by a smooth and broad peak, or by multiple less-smooth peaks, in the CONTIN size distribution plots. The former was usually chosen in our data analyses to derive the average particle size as a function of pH and time. Thus, our emphasis is on the relative particle size, not on the absolute size values.

Zeta Potential Measurements

A Pen Kem Laser Zee™ Model 500 was used to measure the electrophoretic mobility in terms of zeta potential. It uses about 25 mL of solution for each measurement. The zeta potential measurements were made immediately after the pH of the solution was adjusted to the desired value by addition of ultrapure nitric acid or NaOH solution. Usually, four to six measurements were performed on each test solution to derive an average zeta potential. The solution pH usually drifted to lower values during the zeta potential measurement. However, the pH of the colloidal solution in the measuring chamber during the measurement is expected to be close to the value before measurement. We used average zeta potential values because some particles (much less than 10%) were observed to have opposite to most of the colloids. This is probably a characteristic of the waste-glass colloidal system, because nuclear waste glass usually contains more than 20 elements and the groundwater also has a complex composition.

Transmission Electron Microscopic (TEM) Examinations

The microscopic analyses of the colloidal sample were performed with a JEOL 2000FXII TEM operating at 200 kV, equipped with two energy dispersive X-ray detectors. A small drop of colloid solution was wicked through a "holey" carbon grid with ethanol (13). The ethanol was used to attract the water through the copper grid, similar to the action of ultrafiltration. Controls were run to determine possible particulate contamination routes. Soluble salts, such as sulfates and sodium chloride, were often found to precipitate on the grid. This was due to small droplets of leachate evaporating on the surface of the carbon film of the grid. Contaminants on the as-received grids were usually easily recognized in the microscope since they were large particles, and were often single crystals.

RESULTS

Colloid Generation

Our first concern was to generate colloidal solution with enough volume for this study. The first four tests (designated A-1, A-2, A-3, and A-4) at SA/V of 11,400 m⁻¹ were carried out with SRL 131S glass. After 80 days at 90°C, the four test vessels were removed from the oven, and about 1 mL leachate was carefully withdrawn from each vessel to check for colloids using the laser light scattering system. The results of several measurements of the leachates indicated that the concentra-

tions of the colloids were so low that the counting rates, counts per second (CPS), of the light scattering measurements were close to the background level (i.e., similar to the CPS of the starting J-13 well water). This indicates that almost no additional colloids in the leachate were produced by glass reaction.

In earlier testing of SRL 202U glass in J-13 groundwater (7), it was found that the amount of colloids in the leachate usually increases steadily, and then decreases with time. The duration from initial colloid increase to drop to a very low concentration depends on the SA/V of the tests. These durations were 300 days for SA/V = 340 m⁻¹, 200 days for SA/V = 2000 m⁻¹, and 100 days for SA/V = 20,000 m⁻¹. The very low concentration of colloids observed for SRL 131S at SA/V = 11,400 m⁻¹ is, therefore, consistent with the observations for SRL 202U at SA/V = 20,000 m⁻¹.

To better understand the above effect, a dilution experiment was performed. It consists of four tests (designated A-1-1, A-1-2, A-1-3, and A-1-0) using the leachate; reacted glass powders; precipitates from the terminated test, A-1; and fresh J-13 water. Table I gives the test parameters.

These dilution tests were carried out at 90°C for 4 days. At the end of the tests, about 1.0 mL of solution was withdrawn from each vessel. The CPS values of the laser scattering measurements were 2,000, for Test A-1 and 11,000, 10,500, and 12,000 for Tests A-1-1, A-1-2, and A-1-3, respectively. Thus, the CPS increased about 5-6 times over the original A-1 solution. However, the CPS for Test A-1-0 only increased by a factor of two (to 4,000). These values indicate that there are sufficient colloids in A-1-1, A-1-2, and A-1-3 for size and zeta potential measurements, but not in A-1-0 or A-1. In a separate dilution test similar to A-1-3, where the test duration was 5 days, the CPS value measured was 60,000, which is a factor of 30 increase over that of A-1 solution.

To prepare enough volume of the colloidal solution for our subsequent studies, all the solutions withdrawn from A-1-1, A-1-2, and A-1-3 were combined together, and this colloidal solution was designated A-1-ALL. This solution was measured to have a CPS value of 10,500.

Two tests, B-1 and B-2, were carried out with SRL 131A glass in J-13 well water at 90°C for 560 days at SA/V of 2000 m⁻¹. At the termination of these tests, the CPS values of the leachates were measured to be around 22,000, which is suitable for this study.

Stability of Colloidal Suspension vs. Time

The stability of the colloidal solution of B-1 and B-2 was monitored at room temperature by determining the size and the amount of colloids as a function of time. The leachates were transferred directly from the test vessel into a measuring vial for light scattering measurement. The first measurement at room temperature was performed about 1 hour after the test was terminated. The mean colloidal particle size from B-1 vs. time is shown in Fig. 1. The data indicate that, when the leachate was at room temperature, the colloids began to agglomerate. The mean particle size increased to about 1250 nm by 1.25 days after the test termination. Then, these large colloids began to settle out of solution, and after five days only small colloids remained in solution. When the measuring vial was shaken, the large agglomerates were resuspended in solution, but quickly settled out again, as shown in Fig. 1. The mean colloid size measured before shaking was nearly the same as that one hour after shaking, indicating that only the small colloids, about 600-700 nm, are stable in solution.

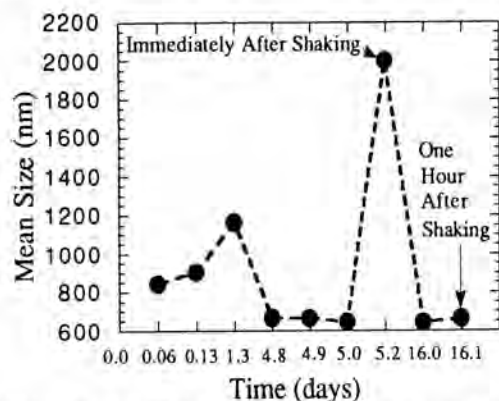


Fig. 1. Mean colloid size vs. duration (in days) at room temperature for test B-1 solution. The line is a guide for the eye.

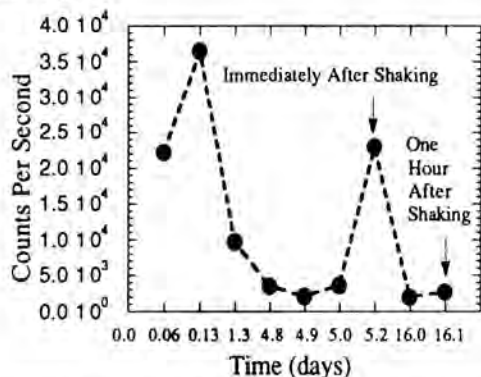


Fig. 2. Counts per second measured by light scattering system on the colloid solution vs. duration (in days) at room temperature for test B-1 solution. The line is a guide for the eye.

The CPS value of B-1 solution, shown in Fig. 2, is a measure of the amount of the colloids, which depends on their number and sizes. The data in Fig. 2 display the same trend as that in Fig. 1. The CPS value increases as the colloids become larger and decreases as the colloids settle out of solution. The CPS values decrease from the initial value of about 22,000 to about 2000, which is the same value as the blank J-13 well water. Thus, most of the colloids generated from glass interaction with groundwater settled out of the solution after about five days at room temperature. The colloid stability for B-2 solution is in general agreement with the results of B-1 solu-

TABLE I
Parameters for the Dilution Tests

	A-1-1	A-1-2	A-1-3	A-1-0
Leachate (mL)	1.0	2.0	3.0	25*
Reacted Glass and Precipitates (g)	1	1	1	17
Fresh J-13 Water (mL)	32.0	31.0	30.0	10.0

* This is the leftover solution from test A-1 after removing leachate for size measurements and for the new tests (A-1-1, A-1-2, and A-1-3).

TABLE II
Zeta Potential (Z.P.) Measurements on A-1-ALL

pH (before)	pH (after)	Z.P. Measurements, mV						Z.P. (mV) (average)	STD
		1	2	3	4	5	6		
1.06	1.01	0	1	2	0			0	1
2.77	2.66	-36	-40	-42	-46			-41	4
3.99	3.93	-34	-44	-50	-60	-35	-42	-44	9
6.33	6.07	-72	-65	-60	-66			-66	4
7.48	7.26	-23	-24	-25	-26			-25	1
9.14	9.01	-18	-19	-11	-17	-16	-13	-16	3
10.50	10.40	-44	-41	-42	-45			-43	2

tion and most of the colloids in B-2 also settled out of solution in about five days.

Zeta potential and Size at Different pH

The effect of solution pH on zeta potential and particle size was studied using 60 mL of the A-1-ALL solution. The original A-1-ALL solution had a pH value of 9.14, a zeta potential of -16 mV, and a mean particle size of 638 nm. Then, ultrapure nitric acid was added to half of this solution to adjust the solution pH downward, and this solution was labeled as A-1-ALL-D. The other half was used to adjust pH upward (labeled as A-1-ALL-U) by addition of 1M or 10M NaOH solution. The measured zeta potentials of the leachate over the pH range of 1 to 10.5 are presented in Table II and Fig. 3.

As shown in Table II, the solution pH usually drifted to a lower value during zeta potential measurements. The isoelectric point, as shown in Fig. 3, was around pH 1, and the colloidal particles were always negatively charged when the pH of the solution was above 1.

The size of the colloids was expected to be closely correlated with solution pH and zeta potential. The particle size in the A-1-ALL solution was measured at each pH after the corresponding zeta potential measurement. Figure 4 shows the measured size variations with solution pH. The size of the colloids is largest at the isoelectric point, pH 1, second largest at pH of 9.14, and smallest around pH 6. These trends are similar to those observed for zeta potential (Fig. 3).

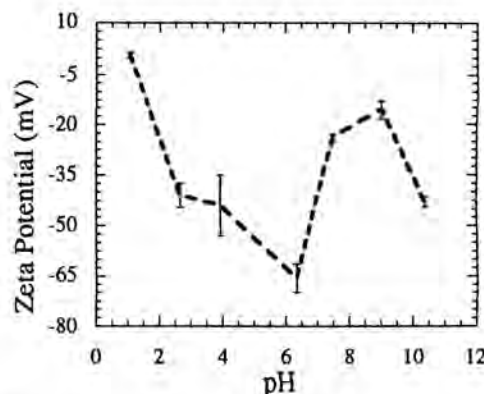


Fig. 3. Zeta potential of A-1-ALL solution as function of pH. The line is a guide for the eye.

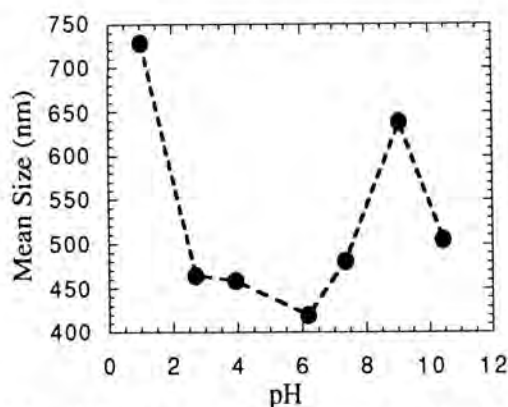


Fig. 4. Mean size of A-1-ALL solution as function of pH. The line is a guide for the eye.

TEM Examinations

The mineral composition and the morphology of the colloids were assessed with TEM. The analysis on the B-2 solution was performed after this solution was subjected to the stability vs. time study described above. Light scattering measurements had demonstrated that shaking the solution resuspended the precipitated materials for a relatively short time.

The B-2 solution was, therefore, shaken before being sampled for TEM analyses. A number of different types of particles were observed during TEM analyses. There were large amounts of clays which exhibited the characteristic turbostratic oblique textured electron diffraction (OTED) pattern and EDS composition, typical of smectite minerals (14), as seen in Fig. 5.

The clays displayed basal spacings on occasion, which is why the OTED is only slightly elliptical at 35° of tilt (Figs. 5a and 5b). Large particles, sometimes exceeding 2-3 μ m in length, of a uranium silicate were also found. Electron diffraction and EDS composition (Fig. 6) of these phases was characteristic of weeksite. The uranium phases were found with particles of clay, as seen in Fig. 6b. The same weeksite phase was also observed in the surface layers of the reacted glass in the B-2 test as shown in Fig. 6a.

Two other colloidal solutions were examined with TEM: A-1-ALL-U, which was produced by adjusting the pH of A-1-ALL upward to 10.5, and A-1-ALL-D, which was produced by adjusting the pH of A-1-ALL downward to 1.0. The TEM analyses were performed one week after these two solutions were used in the particle size-zeta potential-pH study, i.e., these solutions had been at pH's of 1.0 and 10.5, respectively, for more than one week. The small droplets of the colloidal solution used in TEM analysis were taken from

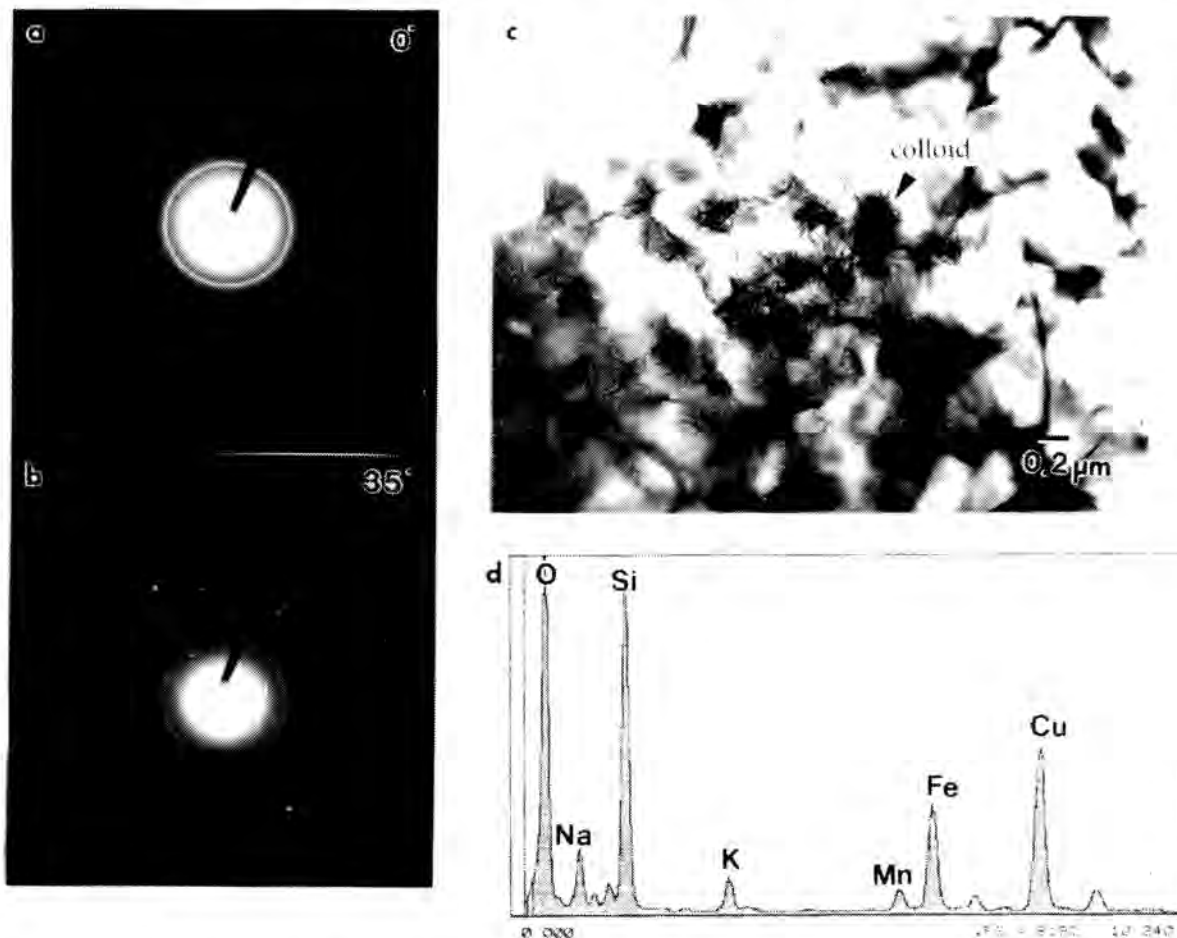


Fig. 5. Transmission electron micrograph and selected area diffraction (SAD) patterns of smectite clay colloids found in B-2 leachate. (a) SAD taken at 0° tilt, (b) SAD taken at 35° tilt, shows some signs of texturing, (c) micrograph of the colloids, and (d) EDS compositional analysis of the Smectite Phase.

undisturbed colloidal solutions. The A-1-ALL-D sample had virtually no colloidal particles present, while the A-1-ALL-U sample contained a large amount of colloidal matter, including smectite clay and silicon phosphate phase. A similar silicon phosphate phase had also been identified in the surface layers of the reacted SRL 202U glass (16).

DISCUSSION

Effect of Salt Concentration

It is well known that the addition of electrolytes to a colloidal system can compress the diffuse counter-ion atmosphere (electric double layer) toward the surface of the colloids, causing the colloidal particles to agglomerate by van der Waals attraction and eventually settle out of solution (17). The time period during which the colloid concentrations of SRL 202U leachates initially increased and then decreased to very low concentration was observed to decrease as the SA/V changed from 340 to 20,000 m^{-1} (7). Tests done at higher SA/V result in more glass reaction and higher salt concentrations in the leachate because more glass surface area is available for glass reaction. The salt concentration in higher SA/V tests, then, reaches the concentration level which can flocculate a colloidal suspension sooner than in the lower SA/V tests. It is likely that the high concentration of salts produced in the

80-day leachate of SRL 131S at SA/V = 11,400 m^{-1} (samples A-1, A-2, A-3, and A-4) had already flocculated most of the colloids, which resulted in the low CPS values observed by light scattering measurements.

This salt effect was verified by the dilution experiments. In these tests, the colloid solution was diluted with fresh J-13 well water and was equilibrated with the precipitated colloids for several days at 90°C. When the salt concentration was lowered by this dilution, the compression on the double layer of the colloids was reduced. The expanded double layer decreased the van der Waals attraction and increased the repulsive force between the colloidal particles, which resulted in resuspension of the flocculated colloids in solution. This was evidenced by up to a factor of 30 increase in CPS value of the leachates in the dilution experiment.

This salt effect is important because a repository setting is likely to be in a nearly static condition with minimal groundwater in contact with the glass. The glass reaction in such a situation will produce a leachate with high salt content and high solution pH. Under this condition, the colloid concentration will likely be minimal, as will the colloidal transport of radionuclides. On the other hand, if a large amount of groundwater with a low salt content contacts the glass reaction site, the precipitated colloids may become resuspended. Alternatively, if the salt content is high in groundwater (such

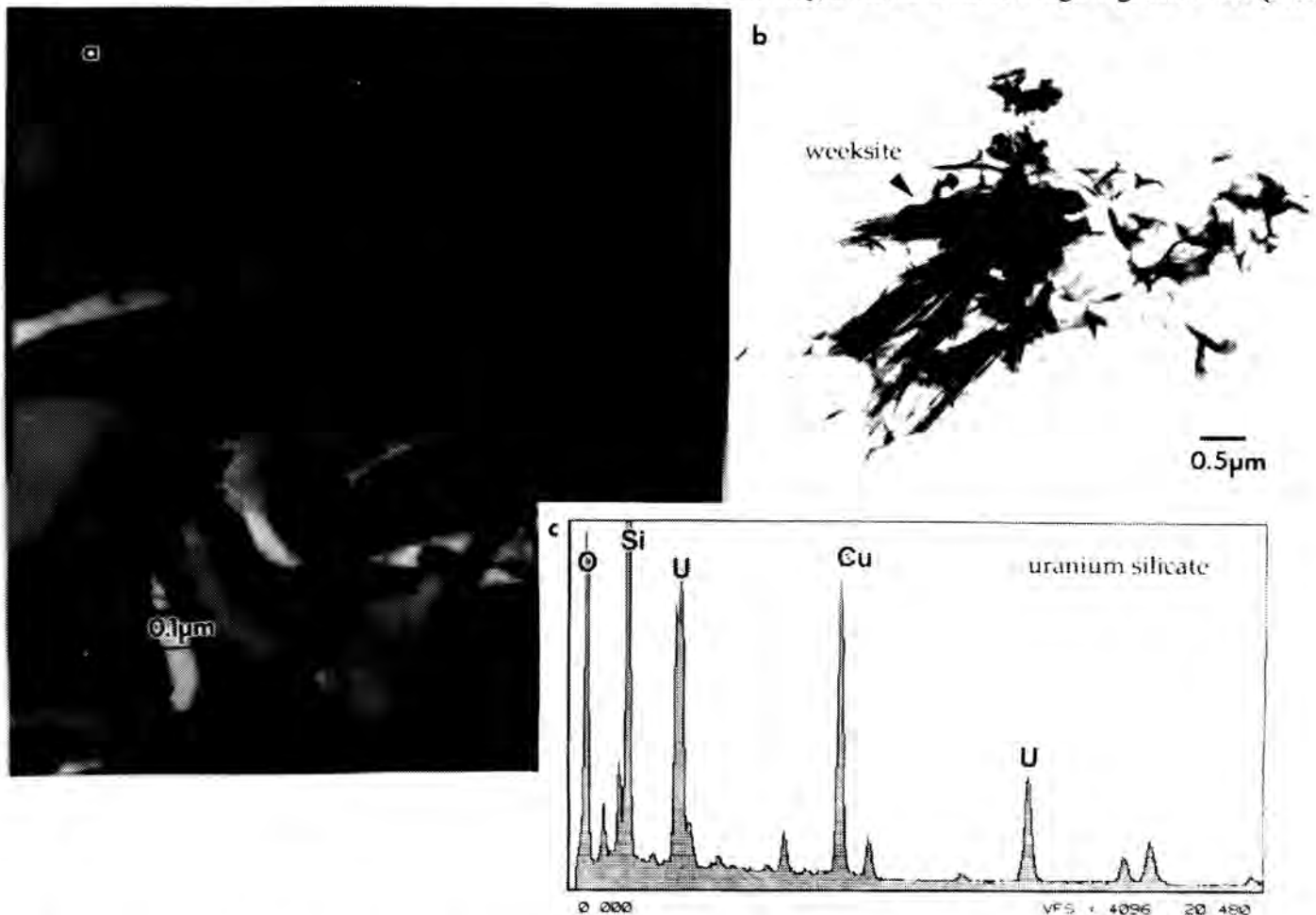


Fig. 6. Transmission electron micrograph of the uranium silicate phase (wecksite) found in (a) surface layers of reacted glass of B-2 test, (b) B-2 leachate (note attached clay material). (c) EDS compositional analysis of the uranium silicate phase.

as in a salt repository), the formation of colloids may be prohibited.

Effect of pH on Colloidal Charge and Size

The leachate pH range resulting from leaching of waste glass in groundwater, especially under static conditions, varies from near neutral to 13 as glass composition and SA/V change. Moreover, the charge carried by the colloids strongly depends on leachate pH. As a result, the charge at a particular pH is important in understanding the colloid behavior. It will decide what surface (positive or negative) will attract the colloids. The particle size of the colloids also changes with solution pH. At a certain pH such as the isoelectric point, the net charge of the colloids becomes zero, and the colloids agglomerate quickly. The charge and size of the colloids may have a very large influence on their stability, transport properties, and interactions with the glass surface, canister wall, and packing materials.

The mineral compositions of the colloids influence their zeta potentials, which vary significantly with solution pH, as shown in Fig. 3. The TEM examinations on these waste glass colloids revealed mainly Si-rich phases, such as smectites and uranium silicates. The richness in silica makes these colloids similar to a pure silica colloidal system.

The diagram of stability vs. pH of a pure silica colloidal system (Fig. 7) can help to explain the trends seen in Figs. 3 and 4. When the solution pH increases from 1 to 6, the negative charge on the colloid increases rapidly, probably due to deprotonation from Si-O-H, Al-O-H, and M-O-H (where M = metal ions) sites. Between pH 6 and 9.14, the negatively charged colloids may adsorb some positively charged hydrolyzed cations, such as $M(III)OH^{2+}$, $M(II)OH^+$, because many metal cations can be easily hydrolyzed in this pH range. This reduces the negative charge on the colloids. At pH 10.5, most hydrolysis products of metal cations are neutral or negatively charged, and the pH is also close to the pK_2 of silica and the negative charge on the colloids increases again (Fig. 3). The presence of other elements in the colloids from our experiments gives them the observed deviations from the pure silicon colloidal suspension. For example, the isoelectric point is at pH 1 for the glass leachate, instead of at pH 2 for pure silica colloids (18).

The size of the colloids was expected to be closely correlated with solution pH and zeta potentials (Figs. 3 and 4). In

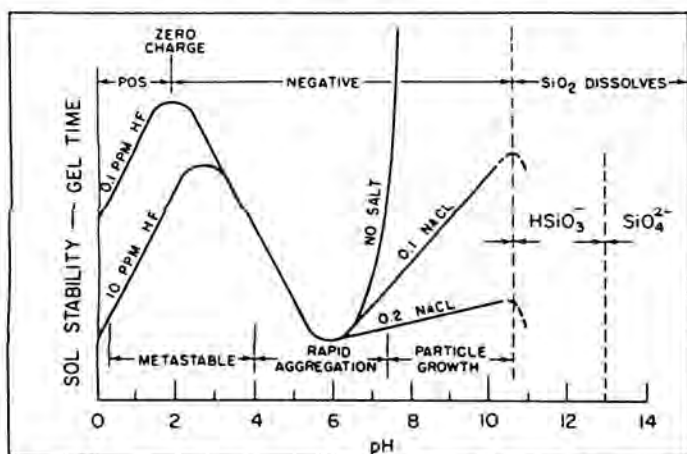


Fig. 7. Effects of pH on colloidal silica-water system (18).

our tests with waste glass colloids, the colloids have zero charge at pH 1, and the colloid particle size grows due to van der Waals attraction, which draws two particles together at the moment of collision. This produces the largest size observed for the A-1-ALL solution. At the intermediate pH around 6, the colloids are negatively charged, and they collide much less frequently and less effectively owing to the increasing similar charges on the colloids. The smallest size of the colloids was observed at this pH. As the pH is further increased toward 9.14, the particles are less negatively charged, and they repel each other less effectively than around pH 6, resulting in size growth due to increased collisions. The second largest size was observed at pH 9.14, which is consistent with the region labeled as "particle growth" in Fig. 7.

Therefore, both particle size and zeta potential contribute to the stability of the colloids. When the size becomes too large to be suspended in the solution, the colloids precipitate. For instance, the largest mean colloid size for the A-1-ALL solution was observed at pH 1, immediately after pH adjustment. However, these large colloids at pH 1.0 (as A-1-ALL-D) settled almost completely out of solution within one week as determined by TEM. When the zeta potential of the colloids is opposite to the zeta potential of the glass surface, the colloids may be adsorbed onto the glass surface to form a precipitated surface layer (19). This may also happen when the two are similarly charged but have low potentials. If the colloids have the same sign as the glass surface and relatively high zeta potential, they may be repelled from the glass surface and grow in the solution (19).

Source of Colloids

Bates et al. (5) have demonstrated that the colloids generated by the interaction of nuclear waste glass with groundwater were found mainly from spalled glass surface layers and from solubility-limited precipitation. The result from this study suggest that the waste glass contributes to the colloid formation by leaching, which increases the ion concentration to the level at which nucleation of the colloids and adsorption of radionuclides on existing colloids occurs. The sudden appearance of the colloidal uranium silicate phase in the leachates at various SA/V's and the similar phase observed both in the surface layer (Fig. 6b) of the reacted glass and in the leachates (Fig. 6a) also suggest that the colloids may be formed from fragments of spalled surface layers of the glass. Modeling the colloidal transportation of radionuclides, the sources of the colloids should not only be from solubility-limited precipitation and adsorption of radionuclides onto existing groundwater colloids, but also from the altered waste glass surface layers. The observation that the colloidal particles are often elongated or planar is also interesting, since most models of colloidal migration assume a spherical geometry for colloids, as do programs used in light scattering to determine size distributions of colloids.

CONCLUSION

The generation of colloids in the interaction of high-level nuclear waste glasses with groundwater at 90°C has been investigated. The stability of these colloids was determined with respect to salt concentration, pH, time, particle size and zeta potential. The composition, morphology, and source of the colloids were also characterized with TEM. The waste glass may contribute to the colloid formation by increasing ion concentration in the leachate, resulting in nucleation of

colloids, by releasing radionuclides that adsorb onto existing groundwater colloids, and by spalling colloidal size fragments from surface layers of the reacted glass. The colloids are mainly silicon-rich smectites and uranium silicates. High concentration of colloids can only be produced in the glass leachates, where salt concentration is low i.e., low SA/V (340 and 2000 m⁻¹) for relative long reaction time or high SA/V (11,400 and 20,000 m⁻¹) for relatively short reaction time. A colloidal suspension can be flocculated if the salt concentration is high. However, the flocculated colloids can be resuspended if the salt concentration is lowered by groundwater dilution. The colloids agglomerate quickly when at room temperature. Most of the colloids can settle out of the solution at ambient temperature in about five days. The colloids have an isoelectric point at pH around 1.0 and are negatively charged between pH 1 and 10.5. The size of the colloid particles is largest at the isoelectric point and grows even at pH 9.14. The size change observed can be explained by the solution pH and the zeta potentials.

The implications of the results from this study are as follows. First, in a glass-reaction dominated repository setting, where salt concentrations are likely to be high, the colloid concentration is likely to be low, and the colloid transport of radionuclides will be minimal. Second, when a large amount of groundwater contacts the glass reaction site, the precipitated colloids may be resuspended, and colloid transport may become important. Third, at ambient temperature, the colloids may agglomerate quickly and settle out of solution in a short time. Fourth, under most pH conditions (1.0-10.5), the colloids are negatively charged, and any positively charged surfaces may adsorb or precipitate these colloids. Finally, the sources of the colloids in a waste repository come from both existing colloids in the groundwater and contribution from the waste forms.

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