

*Colloid Research  
for the Nevada Test Site*

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## COLLOID RESEARCH FOR THE NEVADA TEST SITE

by

Ernest A. Bryant

### ABSTRACT

Research is needed to understand the role of particulates in the migration of radionuclides away from the sites of nuclear tests at the Nevada Test Site. The process of testing itself may produce a reservoir of particles to serve as vectors for the transport of long-lived radionuclides in groundwater. Exploratory experiments indicate the presence of numerous particulates in the vicinity of the Cambric test but a much lower loading in a nearby well that has been pumped continuously for 15 years. Recent groundwater colloid research is briefly reviewed to identify sampling and characterization methods that may be applicable at the Nevada Test Site.

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#### I. THE ROLE OF PARTICULATES IN GROUNDWATER TRANSPORT

Anyone who has seen a mountain stream during spring runoff has observed the transport of particulate matter in water. Clearly particulate transport in surface water is a mechanism of major importance in reshaping mountains and even continents. The importance of suspended particulates in the subsurface transport of materials by groundwater is not so easy to observe or assess.

In recent years, as we have become more aware of the problems engendered by the subsurface disposal of waste materials, the general question of material transport by groundwater has become a focus for environmental concern and thus for scientific enquiry. Most of the relevant research has been aimed at understanding the transport of dissolved materials and the interactions of such dissolved materials with the geologic medium. Less is known about naturally

occurring particulate suspensions in groundwater, and such systems are often more complex than solutions. Perhaps because of their complexity, until recent times relatively little effort had been spent studying them. Although substantial knowledge has been accumulated about particulates, much of it is anecdotal in nature and we can not yet make predictions about particulate behavior with any certainty.<sup>1,2,3</sup>

Particulate transport can occur only when the particles are suspended in the groundwater. Stable suspensions, called colloids, are generally favored for small particles with a surface charge of the same polarity as that of the surrounding medium. Stability is lowered by high ionic strength and is often a function of pH and other variables. If the environment changes so that a suspension becomes unstable, it may sorb on the walls or coagulate (flocculate). Suspensions can also be lost as a result of filtration during flow in porous material.

The particulates in groundwater may be inorganic or organic or a combination of the two. Clay particles tend to be small and thus are more readily suspended than some other inorganic materials.<sup>1,2</sup> Humic substances are also frequently found as suspended particulates in groundwater.<sup>2,4</sup> Sometimes humic substances form coatings on inorganic particles.<sup>1,2</sup>

Radioactive materials are currently the focus of much environmental concern, primarily in regard to the safe disposition of radioactive waste by burial. Concern has also centered on the transport of radionuclides in solution. However, the potential also exists for groundwater transport of radionuclides either as pure colloids (plutonium, for example, forms a stable colloid) or sorbed on or incorporated into other colloidal species. Humic substances, which are known to strongly sorb several actinide elements and also to form stable colloidal suspensions, under the right circumstances, should be expected to act as a powerful vector for the transport of some radioactive materials;<sup>4</sup> the same may be true of chemically reactive inorganic materials such as clays.

There are several often-cited examples of colloidal transport, one being the Lawrence Livermore National Laboratory's interpretation of data from the Cheshire site at the Nevada Test Site (NTS).<sup>5</sup> At Los Alamos, researchers reported that americium had migrated a considerable distance from a waste-stream out-fall, much farther than anticipated.<sup>6</sup> They presented evidence that the americium was in particulate form instead of being in solution. Researchers at the Chalk River Nuclear Laboratories in Canada have reported<sup>7,8</sup> that cesium migrated away from buried test blocks of nuclear waste more rapidly than anticipated and, therefore, concluded that it moved in particulate form. Similar evidence exists for migration of inorganic and organic particulates at other sites.

The Department of Energy (DOE), Office of Health and Environmental Research has been supporting studies of the role of colloids in groundwater transport (Subsurface Transport Program, Dr. Frank Wobber). Research topics and goals have been identified in a series of seminars held at Manteo, North Carolina: colloid sampling and characterization; colloid generation and deposition; colloid mobility; and waste management/remediation demonstration.<sup>9,10,11</sup> The DOE's Yucca Mountain Site Characterization Project has also supported study of the potential for colloidal transport at the proposed nuclear waste repository at Yucca Mountain. In addition, there is active research in the field of colloid

transport at a number of other institutions throughout the world. J. I. Kim at the Technical University in Munich has been investigating the role of humic substances, as has G. Choppin at the University of Florida. P. Vilks at Whiteshell, Canada, has been studying particulates in groundwater in the vicinity of uranium deposits.

## II. COLLOID RESEARCH FOR HRMP

In view of the colloid research going on elsewhere, one might question whether colloid research should be supported with the limited resources of the Hydrology/Radionuclide Migration Project (HRMP). There are, however, special circumstances related to nuclear testing that may justify assigning a high priority to such research. First, an enormous amount of crushed rock is produced at each explosion site. Second, large amounts of drilling mud (montmorillonite clay) are introduced during routine drillback operations. Third, there are no engineered barriers to isolate the radioactive debris from groundwater.

The crushed rock and clay may provide an abundant reservoir of colloidal particles to be transported, and some of these may be highly sorptive in nature. Much of the residual plutonium and other radioactive species are incorporated in glassy debris, and, under the influence of groundwater, this debris may produce colloidal radioactive particles.<sup>12</sup> As mentioned above, there is some evidence that colloids played a role in the redistribution of radioactive material around the Cheshire site.

Thus the following three issues appear to be especially relevant to the possibility of colloid transport at the sites of underground nuclear tests:

- (1) the extent to which the crushing and/or the drilling mud provide a unique source of particles,
- (2) the extent to which any of the particles in the groundwater near a test point migrate through the geologic medium surrounding the test cavity, and
- (3) the extent to which any migrating particles can act as transport vectors for radionuclides.

## III. HRMP-SPONSORED COLLOID INVESTIGATIONS AT LANL

Over the years some experimental data have been obtained about particulates at the Cambric site. Exploratory experiments have been conducted with material from both Cambric wells (RNM-1 and RNM-2S). The experimental methods, results, and conclusions are presented below.

In 1983, a 300-ℓ volume of water from RNM-1 was filtered through 0.4-μm-pore filters and a 3770-ℓ volume from RNM-2S was filtered through 0.05-μm-pore filters. Particulates collected on the filters were studied with a scanning electron microscope (SEM) and found to contain a variety of different elemental compositions with silicon being a predominant element. It was reported that particle diameters ranged from <0.0017 μm to tens of micrometers (no explanation

was given for the presence of the particles smaller than the filter pore size).<sup>13</sup>

S. Goldstein and J. Fabryka-Martin reported on the character of particulates at RNM-1.<sup>14</sup> In 1988, an 80-ℓ water sample from the chimney zone of well RNM-1 was brought to Los Alamos in a polyethylene bottle and was subsequently processed with a Millipore Pellicon ultrafiltration system [100 000 nominal molecular weight limit (NMWL) filter] to produce a 250-mℓ concentrate. Portions of the concentrate, which was noticeably cloudy, were centrifuged onto carbon films supported on copper grids for transmission electron microscope (TEM) analysis. A blank was processed in a similar manner and proved to be quite clean. A more detailed account of the experiment is given in Appendix A.

The TEM samples were too concentrated for good resolution of individual particles, but both amorphous and crystalline particles were present. Results of the TEM assay appear in Table I. The crystalline particles were tentatively identified as clay, whereas the chemical composition of the amorphous material was consistent with apatite (a phosphate). Both the clay and the amorphous material might have been introduced as drilling materials. The natural phosphate concentration is quite low in NTS waters. Selected TEM micrographs and x-ray spectra are included in Appendix A.

Table I RNM-1 Particle Analysis by TEM

Sample	Major	Minor	Structure
General area scan	Si	Ca, S, P, K, Fe	
Particle	Ca, P	Mg	Amorphous, 0.05-0.1 μm
Nearby material	Si, Ca, P, S	Fe, Mg	Amorphous
Particle	Mn, Si	Ca, S	Amorphous, 0.05-0.1 μm
Particle	Ca, P	Mg	Amorphous, 0.3-0.4 μm
Large particle	Si, K, Al	Na	Crystalline, 0.5 μm, blocky
Nearby material	Si, Al, Mg	Fe, K	Amorphous
Particle	Si, Al	Ca, Fe, K	Crystalline, 0.5 μm
Particle	Si, Al, K	Ca, Fe	Crystalline
Particle	Ca, P	Mg	Amorphous, 0.3 μm
Particle	Ca, P		Amorphous, 0.15 μm
Particle	Ca, As	Si, Mg?	Amorphous, <0.1 μm

In December 1990, only a week after the pump at RNM-2S had been restarted (it had been shut down in September), E. Bryant and D. Rokop used filters at RNM-2S both to collect particles of different sizes for weighing and to concentrate



particulates from a fairly large volume of water for subsequent characterization studies. The filtration was done on-site to minimize the probability of chemical changes during the process.

Several filter stacks with pore sizes from 8- to 0.015- $\mu\text{m}$  diameter were used. Unfortunately, the data were compromised by experimental difficulties. Reduction in flow rate for the 1- and 0.1- $\mu\text{m}$  filters indicated the presence of particulates in the water, but the maximum weight gain for any of the filters was 0.6 mg and some of that was apparently caused by growth of algae on the filter substrate during two long, unplanned storage periods at NTS. It was not possible to achieve a sustained flow through the smallest filter. This problem was later attributed to clogging by fine particles that formed when the water was cooled by the winter air as it flowed from the well-head to the filter apparatus.

The filter stacks were subsequently dried and weighed along with a blank stack. The measured particle concentrations were very low for all particle sizes, but a gain in weight was observed for all the filter membranes, including the stack of blanks. The blank-corrected loadings (0.01 mg/ $\ell$  for 0.1 to 1  $\mu\text{m}$ , -0.005 mg/ $\ell$  for 1-8  $\mu\text{m}$ , and 0.01 mg/ $\ell$  for >8- $\mu\text{m}$ -pore filters, respectively) were not much smaller than the 0.05 mg/ $\ell$  standard deviation for all weight gains (uncorrected for blank), suggesting that the (corrected) values are comparable to the associated uncertainties. The very low loading values, if real, are interesting because it has often been suggested that rapid pumping may result in the generation of particles; in this case, after 15 years of pumping, there seem to be very few flow-generated particles.

Raymond and Bryant used SEM to examine particulates from the RNM-2S 100-to-1 concentrate of particles greater than 300 000 NMWL. Attempts to examine submicrometer particles directly on filter substrates were abandoned because of poor resolution. SEM pictures for a sample evaporated on a carbon substrate were of good quality and showed that particles in the 0.1- to 10- $\mu\text{m}$  range were mostly calcium minerals (probably calcite, the carbon and oxygen being invisible to SEM). There were occasional silica particles and other agglomerates containing a mixture of cations (calcium, sodium, potassium, silicon) and anions (sulfur, chlorine). In addition, there were highly crystalline particles (calcite?) that appeared to have recently precipitated (during storage or evaporation?). A selection of representative SEM micrographs and SEM/energy dispersive x-ray analysis (EDX) spectra appear in Appendix B. It is interesting that, in contrast to the RNM-1 particulates, the RNM-2S particulates did not contain phosphorus.

Photon correlation spectroscopy was applied to a series of samples from the above concentrate by Triay, Mitchell, and Bryant. In some cases, the aliquot had first been filtered through a Nuclepore filter to remove larger particles. Results indicated (1) the presence of particles with a wide range of sizes but concentrated in the 0.1- to 1- $\mu\text{m}$  range, and (2) the transient presence of very small (~1-nm) particles. Further details appear in Appendix C.

The clogging of filters in the cold December air at the NTS and the appearance of transient fine particles at Los Alamos are thought to be a reflection of a dynamic equilibrium between particles and solution. At Los Alamos, the particle concentrate had been stored in a cleanroom that was exceptionally warm. Apparently, the warm water dissolved some of the particulate material, which then precipitated as fine particles after being removed from the warm area. The fine

particles had disappeared from the suspension, possibly by recrystallizing to larger particles, by the second day after removal from the clean area.

The geochemistry speciation code PHREEQE was used by Goldstein and Fabryka-Martin<sup>14</sup> to calculate saturation indices for water from well 5B, which is not far from the Cambrian site. As indicated in Table II, a number of minerals had positive indices, indicating the potential for precipitation of particles. Further details appear in Appendix D.

**Table II** Saturation Indices for Water from Well 5B, Frenchman Flat, NTS

Mineral	Saturation Index	Mineral Formula
<b>Apatites</b>		
Carbonate apatite	12.8	$\text{Ca}_{9.5} \text{Mg}_{0.14} \text{F}_{2.48} (\text{PO}_4)_{4.8} (\text{CO}_3)_{1.2}$
Hydroxylapatite	-1.5	$\text{Ca}_5 (\text{PO}_4)_3 \text{OH}$
Fluorapatite	7.6	$\text{Ca}_5 \text{F} (\text{PO}_4)_3$
<b>Feldspars</b>		
Microcline (K-spar)	4.4	$\text{KAlSi}_3\text{O}_8$
Sanidine	3.9	$\text{KAlSi}_3\text{O}_8$
Anorthite	-0.2	$\text{CaAl}_2\text{Si}_2\text{O}_8$
<b>Micas</b>		
Muscovite (K-mica)	10.2	$\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2$
Pyrophyllite	7.9	$\text{Al}_2\text{Si}_4\text{O}_{10}(\text{OH})_2$
<b>Zeolite</b>		
Laumontite	4.8	$(\text{Ca}, \text{Na})_7\text{Al}_{12}(\text{Al}, \text{Si})_2\text{Si}_{26}\text{O}_{80} \cdot 25\text{H}_2\text{O}$
<b>Clays</b>		
Kaolinite	6.5	$\text{Al}_4(\text{Si}_4\text{O}_{10})(\text{OH})_8$
Nontronites	21-28	$\text{Fe}_4\text{AlSi}_6\text{O}_{20}(\text{OH})_4$
Illite	4.4	Mica-like clay mineral
Montmorillonite	3.9	$(\text{Al}, \text{Mg})_8(\text{Si}_4\text{O}_{10})_3(\text{OH})_{10} \cdot 12\text{H}_2\text{O}$
Chlorites	5-31	$(\text{Mg}, \text{Fe})_3\text{Si}_4\text{O}_{10}(\text{OH})_2\text{Mg}_3(\text{OH})_6$

In summary, several exploratory experiments have indicated the presence of a substantial particle concentration at RNM-1 and a very small particle concentration at RNM-2S. Particles at RNM-1 include clay (may be drilling mud) and amorphous materials. At RNM-2S, calcite appears to be a major constituent.

#### IV. FUTURE COLLOID RESEARCH

Efforts on colloid research must be well-focused if the nuclear-test-related issues identified above are to be addressed. Colloid research is presently hampered by a lack of well-defined and tested methods for sampling and characterizing colloidal suspensions. Until such time as adequate protocols have been developed, any research is likely to be somewhat frustrating for the scientists and the sponsor because of the many false starts that will inevitably be involved. This consideration is particularly important for the HRMP program because of its limited funds and the very natural impetus to spread them out over a wide variety of tasks.

The particle number density, size distribution, and/or chemical composition in and around a nuclear test must be compared with those from areas that are free from such tests to determine whether or not a nuclear test provides, through crushing or drilling mud, a unique source of particles. A major problem is to devise reliable methods to sample and measure numbers, size, and composition because the available methods all have limitations and pitfalls abound.

The groundwater-colloid literature is replete with examples of sample collection protocols that produced unintended changes in the samples. For example, the oxidation of ferrous to ferric iron by exposure to air can result in new precipitation, or the loss of CO<sub>2</sub> can result in an increase in pH with consequent changes in the dynamic particle-solution equilibrium. As was observed in our exploratory experiment, the change in temperature when the water left the pump apparently caused a change in solubility.

After the sample has been collected, it is often necessary to concentrate the particulates, or at least to separate them from the solution, before carrying out the desired measurement or experiment. However, dynamic light-scattering techniques can sometimes be used to count and measure the size of particulate suspensions without concentration.

Centrifugation and filtration are two techniques available for sample concentration. Centrifugation can be used for particles with diameters larger than about 500 nm but is increasingly ineffective for smaller particles. Particles can be centrifuged directly onto a substrate suitable for microscopy (SEM, TEM, electron scattering for chemical analysis, etc.). Filtration can be used to collect particulates directly on the filter membrane or, in the case of tangential-flow ultrafiltration, in a concentrated suspension.

Problems with filtration include chemical sorption of trace metals on the substrate, clogging of pores, and accumulation of salts in ultrafiltration concentrates. Clogging of pores limits the amount of material that can be collected and may change the effective pore size during filtration. Tangential-flow and stirred-cell filters reduce but do not eliminate this problem. Ultrafiltration, required for particle sizes in the 1- to 10-nm range, suffers from salt retention and poorly defined retention vs size characteristics. Special filtration techniques, for example gel-filtration, may be useful in special circumstances. Filter substrates are generally not ideal platforms for microscopy so it becomes necessary to transfer particulates from the filter to some other substrate or to alter the substrate.

An ideal method would allow size distribution and number-density measurements to be made on particles directly in the groundwater without concentration. Photon correlation spectroscopy is applicable to fairly dilute suspensions, but is not yet fully developed and accepted as an analytical technique. At best, it can provide a measure of number density vs particle size but without chemical selectivity. The 100-to-1 concentrate from RNM-2S was readily measured with this technique, but the unconcentrated particulates in groundwater might not be. Deconvolution of the measured scattered-light correlation spectrum is still more of an art than a science; it generally requires information or assumptions, in addition to the correlation spectrum itself, to provide a unique result.

Many, if not all, colloidal particles are in dynamic equilibrium with the water in which they are suspended. A general problem in sampling, filtration, centrifugation, or any other experimental procedure is maintenance of the chemical/physical conditions of the groundwater so as to minimize precipitation and dissolution of particles during the collection, filtration, and measurement process. In practice, this is very difficult to accomplish and one or more compromises must be made.

After some study, Fabryka-Martin recommended for use at Cambria a protocol based on work by P. Vilks in a study of particulates near uranium mines in Canada. It is proposed to collect an unfiltered 50-ℓ sample, anaerobically and at very low flow rates. At Los Alamos, in a controlled atmosphere, the particles would be split among several fractions according to particle size: >450 nm, ~10 to 450 nm, and ~1 to ~10 nm.

Samples would be analyzed using SEM/EDX, x-ray diffraction, atomic absorption spectrometry, inductively coupled plasma spectrometry, carbon and uranium analyses, and possibly alpha-spectroscopy for the uranium decay series radionuclides. The protocol is described in more detail in Appendix E.

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## APPENDIX A - PARTICULATES AT RNM-1

Steve Goldstein and June Fabryka Martin

### Procedure

Water was collected from RNM-1 on 8/29/89 and stored in a 20-gal. polyethylene container. No special precautions were taken to minimize contact with air and subsequent experimental work was conducted in a laboratory without special air filtration to avoid dust contamination.

On 10/11/89, 11 ℓ of RNM-1 water were filtered through a Millipore Pellicon ultrafiltration system using a 100 000 NMWL filter. This filter corresponds very roughly to a diameter of 6.9 nm for a spherical particle. After filtration, 250-mℓ of colloid concentrate was collected at a concentration factor of ~50. Although the inlet water was clear, the colloid concentrate was distinctly cloudy after shaking.

Following processing of the RNM-1 water, the system was flushed with 5 ℓ of deionized water. A blank colloid concentrate was then prepared by processing 2.4 ℓ of deionized water to form 250 mℓ of concentrate. This concentrate remained clear after shaking.

Four samples from the RNM-1 colloid concentrate were prepared for transmission electron microscopy (TEM). Portions of the concentrate were ultracentrifuged onto copper grids covered with a carbon film. Each centrifuged portion consisted of 3 g of concentrate, which was centrifuged at 50 000 rpm for 2 hours. The samples were loaded with one to three portions each. Only one sample, prepared from 9.9 g of concentrate loaded in 3 portions, was analyzed (on 10/25/89). It was found to contain too much material for optimal TEM analysis. A blank, prepared from 3 g of the blank concentrate, was also analyzed.

### Results

Each colloidal particle appeared in the TEM photos as a small dark dot centered in a circle of amorphous material, which in turn occurred in an amorphous background matrix. Considerable overlap of particles was evident and was a result of having loaded too much material. Ideally, colloid particles should not overlap so that a size distribution and density estimate (colloidal particles/cm<sup>2</sup>) can be made. Nonetheless, some useful information was gained. Altogether, nine of the small dark particles were scanned as well as the material surrounding them (see Table I in this report for a summary of the TEM results). A representative TEM photo and x-ray spectrum appear in Figs. A1 and A2.

### Discussion

The deionized water blank showed that the carbon film itself was quite clean, with possibly minor (negligible) amounts of silica and iron. One particle was seen and found to be composed of phosphorus, sulfur, and iron.

The particles separated from RNM-1 water fall into two major categories. One category consists of amorphous particles composed primarily of calcium and phosphorus; these may be like apatite in composition; that is,

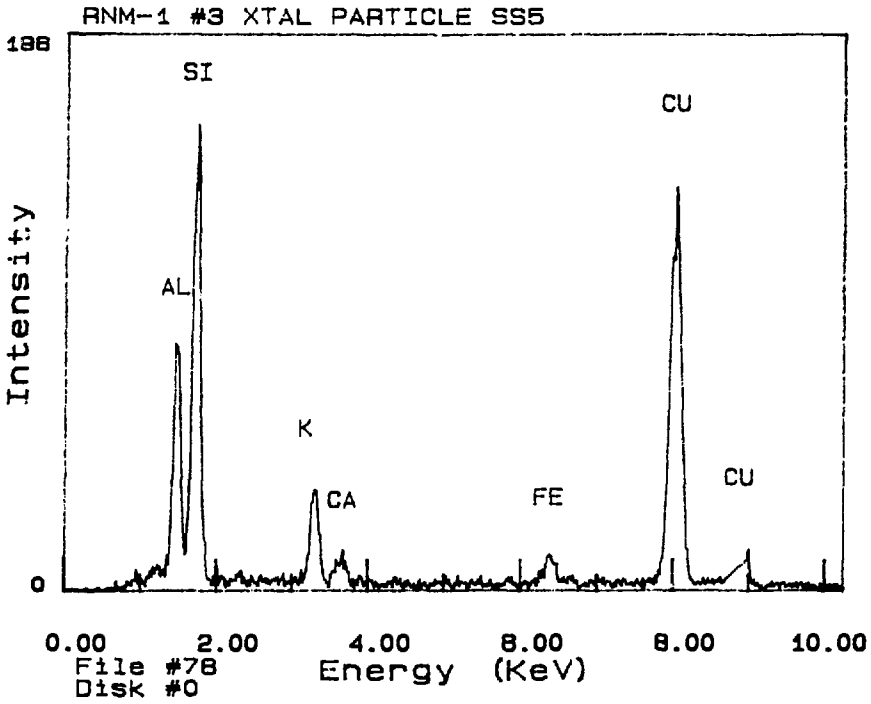
$\text{Ca}_5(\text{F,Cl,OH})(\text{PO}_4)_3$ . An interesting variant in this group is the particle with arsenic instead of phosphorus. Substitution of arsenic for phosphorus in an apatite structure is not unexpected because the major species of arsenic in groundwater are  $\text{H}_2\text{AsO}_4^-$ ,  $\text{HAsO}_4^{2-}$ , and  $\text{H}_3\text{AsO}_3^0$ , (Ref. 1), which are quite similar to the phosphate species. These colloids are probably derived from the drilling mud because phosphate levels in Nevada Test Site waters are generally almost nil. Phosphate could be present in this well from detergent, which is commonly added as a lubricant during drilling.

The second group of particles consists of relatively large crystalline particles in which silica, aluminum, potassium, and magnesium are the major elements. These may be K-spar, kaolinite, K-mica, or zeolite, and could be derived from the host rock. Some clays have a large capacity for absorbing water, thereby providing a mechanism to form colloids from clay particles in the rock. Drilling mud generally contains montmorillonite and sometimes sepiolite, and thus it could also be a source of clay particles.

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10/27/89 1: 30 PM

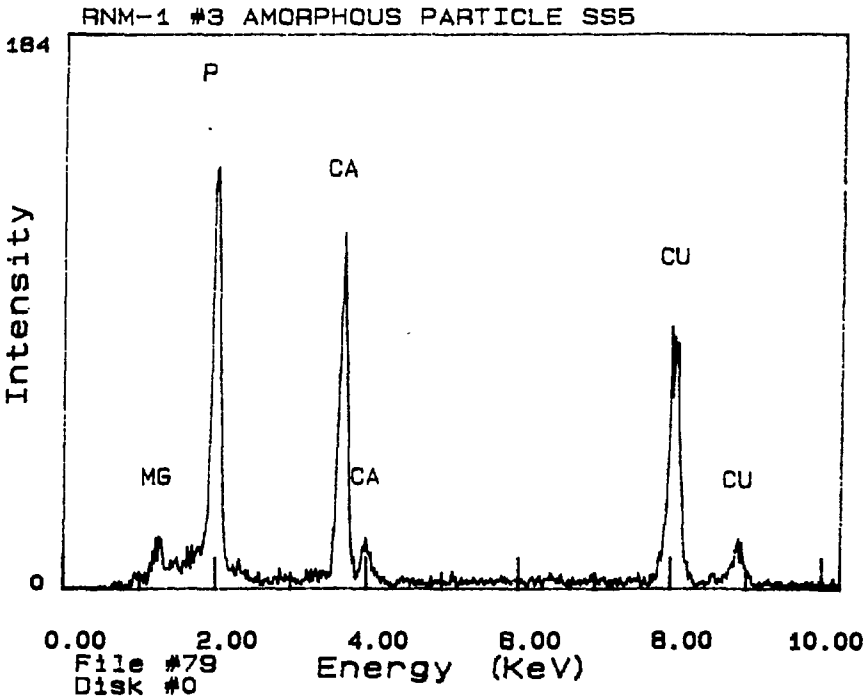


Fig. A1. Representative x-ray spectra of particles from well RNM-1.





Fig. A2. TEM micrograph of particles from well RNM-1.

APPENDIX B - SEM/EDX ANALYSIS OF PARTICULATES FROM RNM-2S

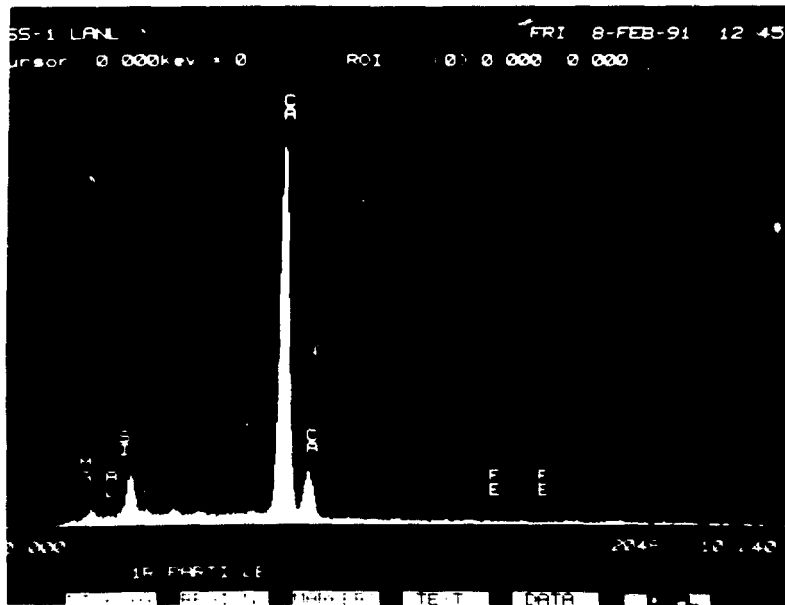
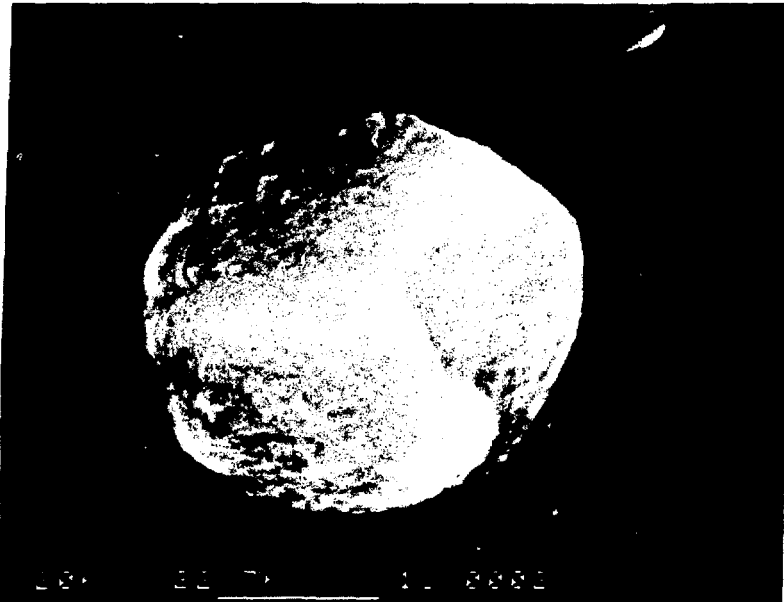


Fig. B1. A 2.5- $\mu$ m calcite particle from well RNM-2S.

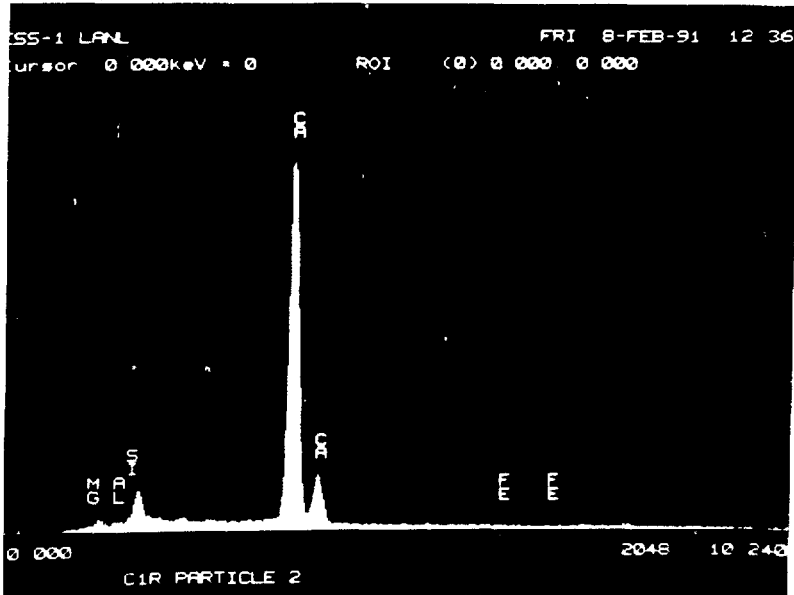
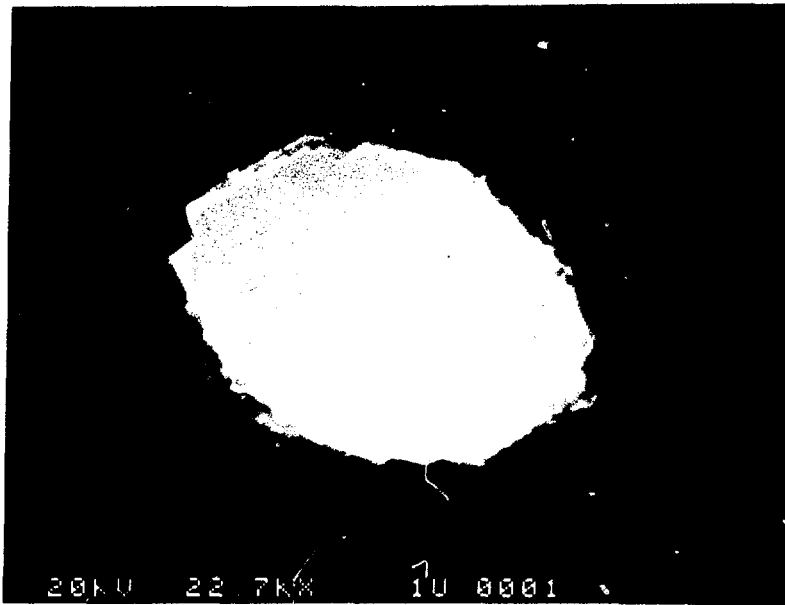


Fig. B2. A 2.5- $\mu\text{m}$  recrystallized calcite from well RNM-2S.

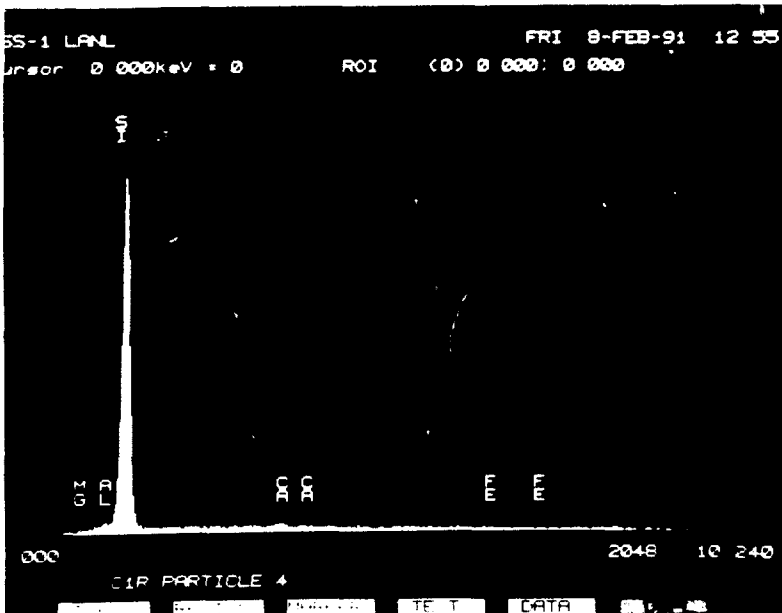
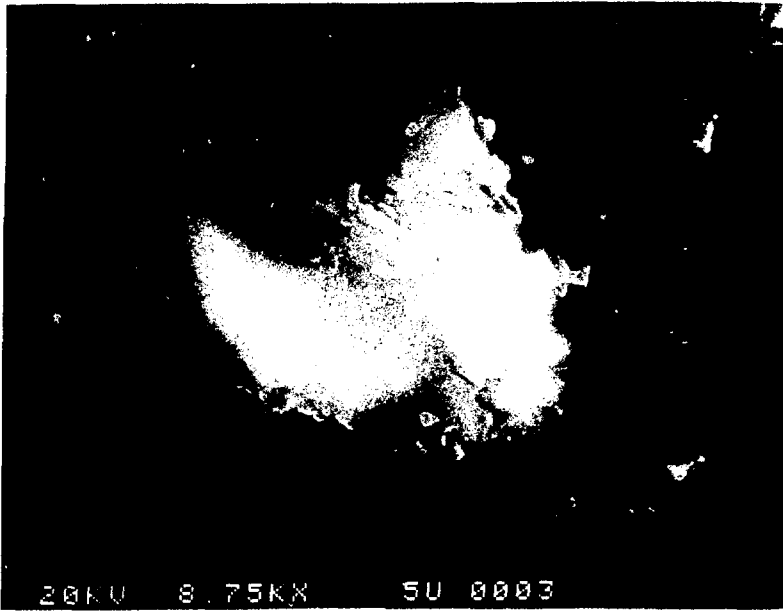


Fig. B3. A 6- $\mu\text{m}$  silica particle from well RNM-2S.

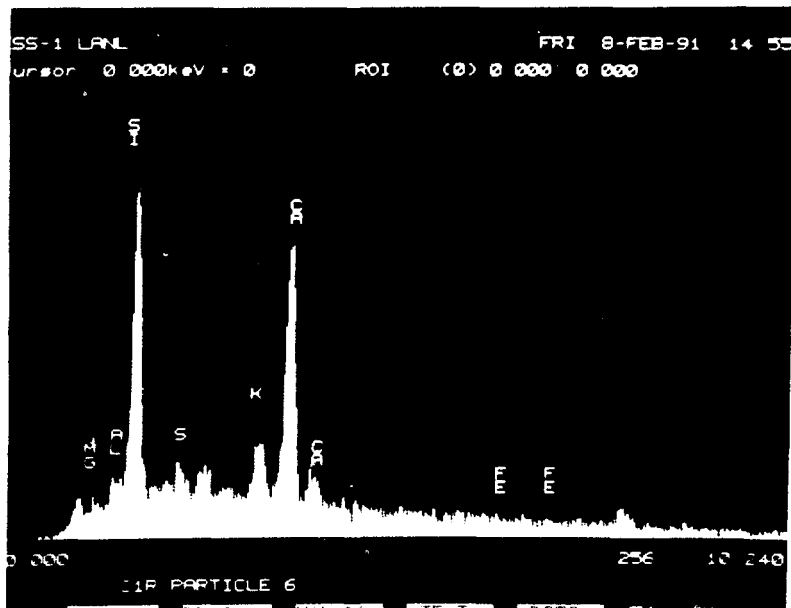
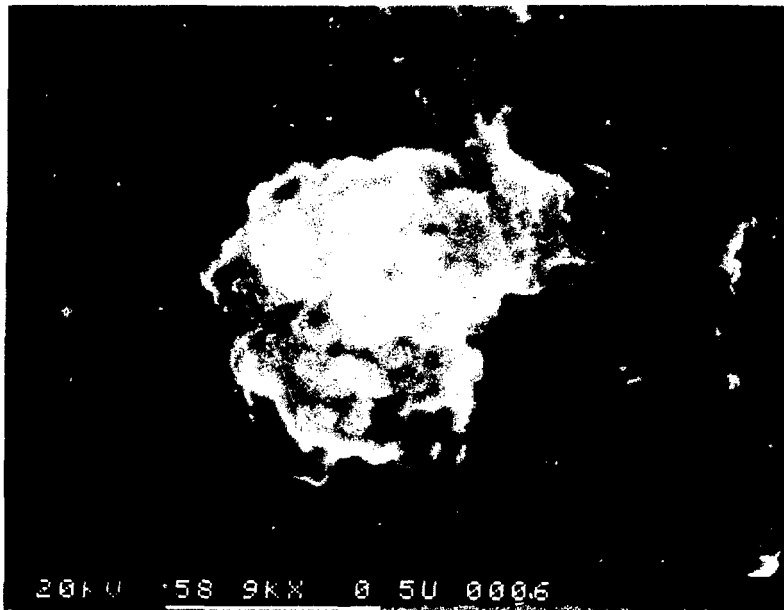


Fig. B4. A 0.7- $\mu\text{m}$  particle from well RNM-2S.

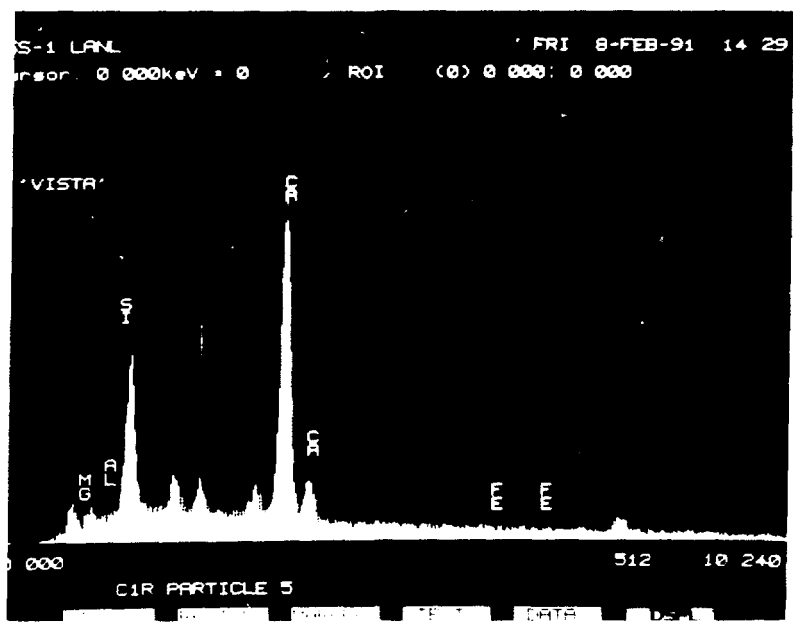
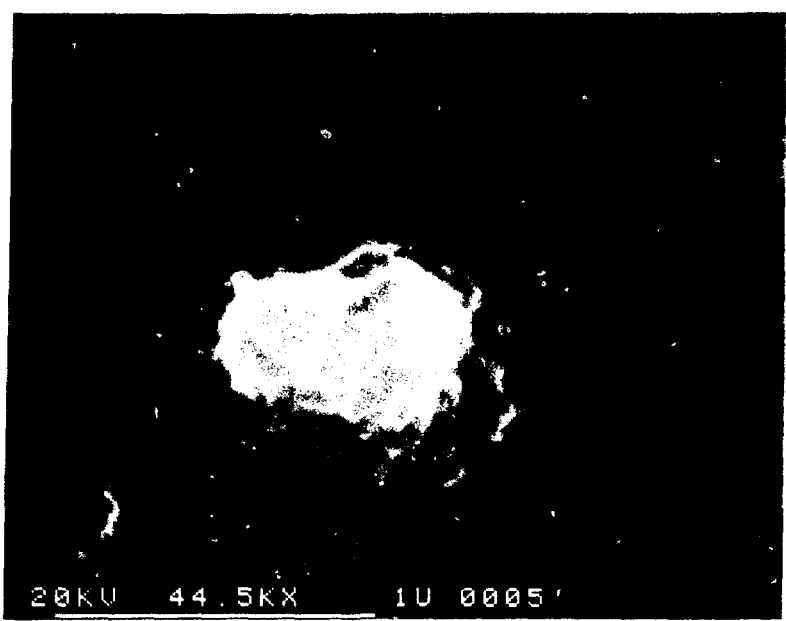


Fig. B5. A 0.6- $\mu\text{m}$  particle from well RNM-2S.

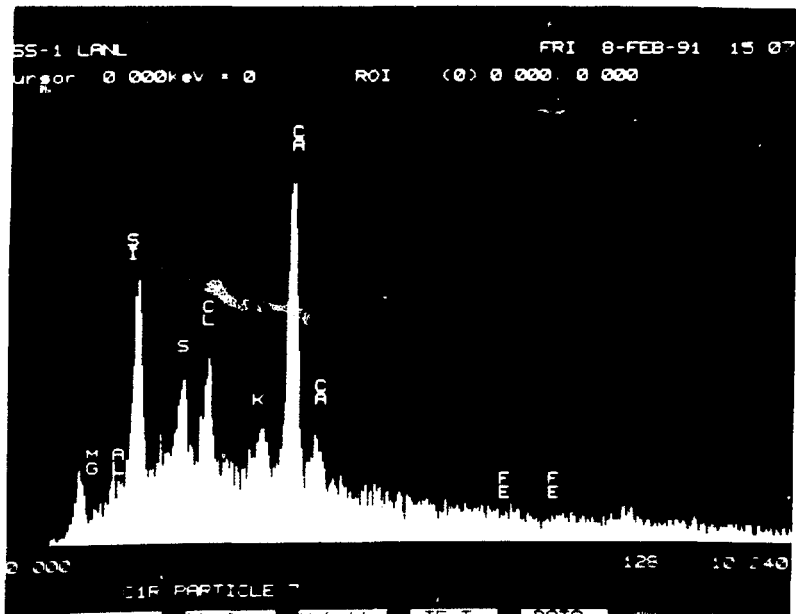
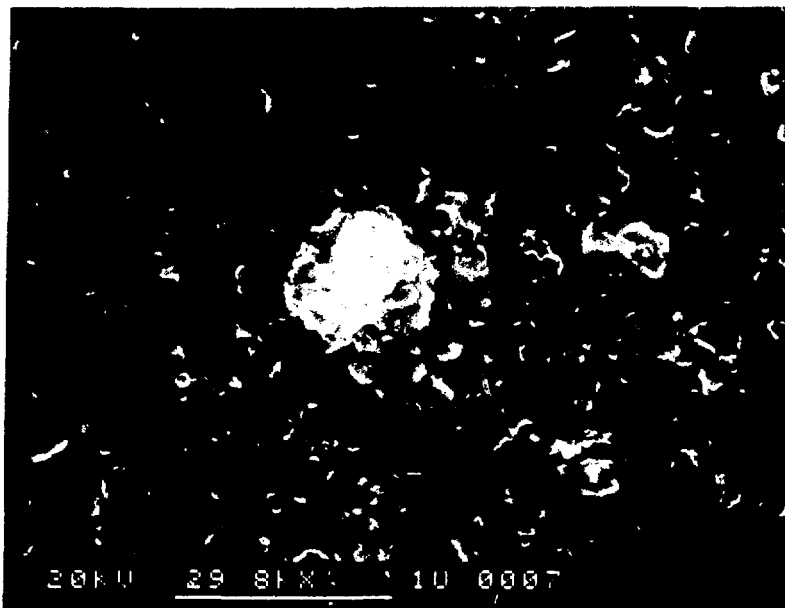


Fig. B6. A 0.5- $\mu\text{m}$  particle from well RNM-2S.

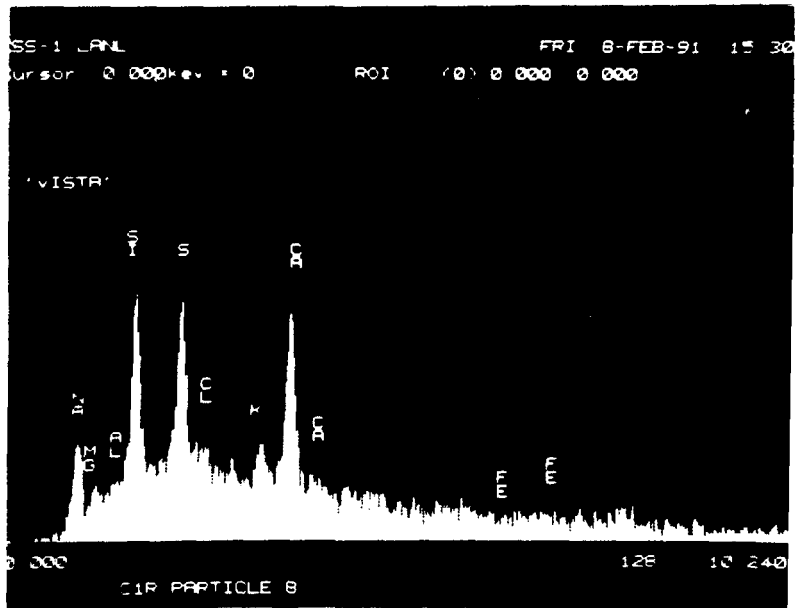
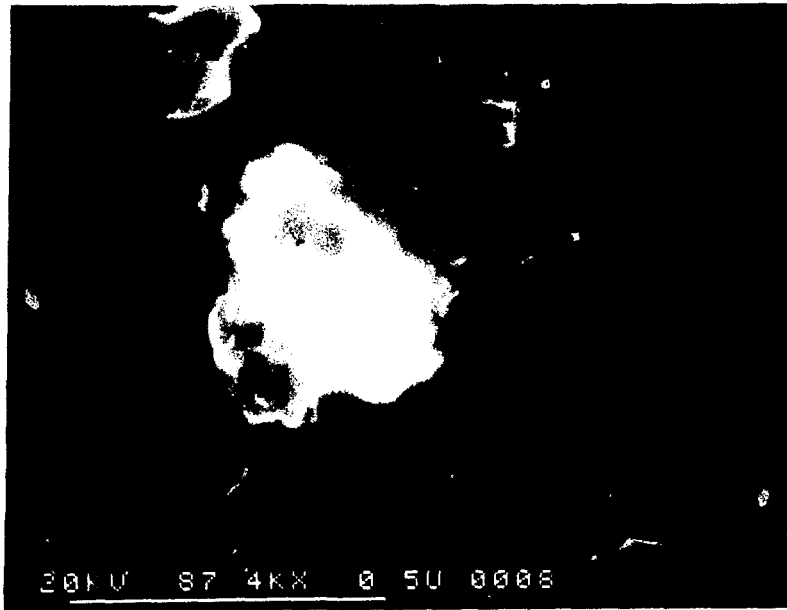
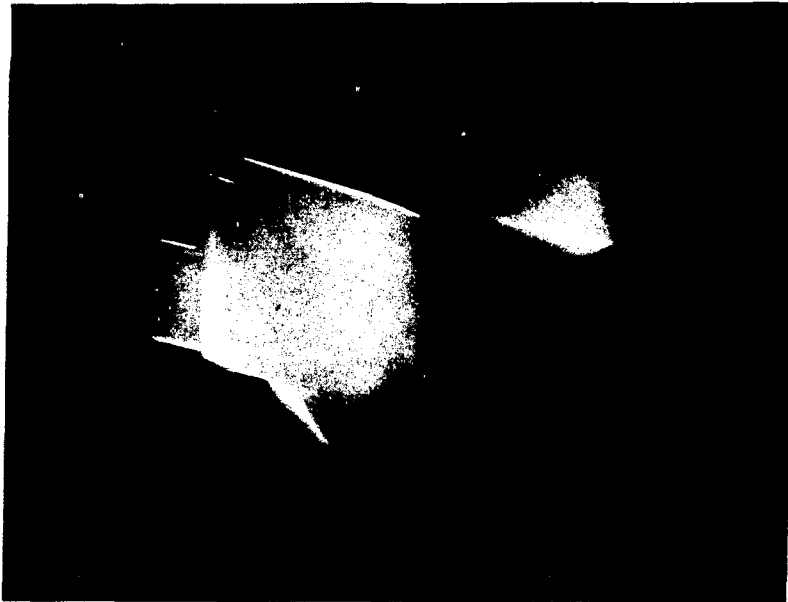


Fig. B7. A 0.3- $\mu\text{m}$  particle from well RNM-2S.





**Fig. B8.** Recently crystallized particle from well RNM-2S.

## APPENDIX C - PHOTON CORRELATION SPECTROSCOPY RESULTS FOR RNM-2S PARTICULATES

E. A. Bryant, Inez Triay, and Alan Mitchell

The auto-correlation photon spectroscopy (APS) technique can be used to characterize the size distribution of particles suspended in water. A mathematical inversion process is required to obtain the size distribution from the measured correlation spectrum. In general, additional constraints must be used to achieve this inversion for poly-disperse distributions (see, for example, Ref. 1). A nonnegativity constrained regularization<sup>2</sup> was employed for analysis of data from Cambic samples.

In a cleanroom, aliquots were removed from the ultrafiltered concentrate and passed through 0.5-, 5-, or 8- $\mu\text{m}$  pore Nuclepore filter membranes directly into the APS cuvettes. Blank samples were prepared with pure water in the same manner. After the cuvettes were sealed, they were taken from the cleanroom to the instrument room for analysis. The ambient temperature in the cleanroom was about 26°C, as a result of a ventilation problem, whereas the temperature in the instrument room was about 22°C; these circumstances apparently produced unanticipated results.

Autocorrelation spectra were collected with a Brookhaven Instrument Corporation Digital Correlator and the spectra were subsequently inverted to obtain particle size distribution information. The first sample analyzed typically exhibited a fairly strong signal for correlation times that corresponded to particles with ~1-nm diameter in addition to a distribution of particles in the 0.1- to 1- $\mu\text{m}$ -diameter range. Subsequent reanalysis of the same sample on the following day failed to detect the 1-nm particles, and they were also not seen in the second or third sample analyzed on the first day. These transient particles were probably formed when the sample cooled after being removed from the warm cleanroom but recrystallized to larger particles fairly quickly thereafter. Spectra for samples that had passed through the 5- or 8- $\mu\text{m}$  filters exhibited a larger number of particles in the 1- to 10- $\mu\text{m}$  range than did samples filtered with 0.5- $\mu\text{m}$  filters, which lends some confidence to the mathematical inversion process. The spectrum for an unfiltered sample indicated the presence of "dust" particles  $\geq 32 \mu\text{m}$  in diameter.

The autocorrelation technique will be useful for particle analysis with Cambic water but will require careful attention to details. A detailed procedure should be devised and tested.

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## APPENDIX D - CHEMICAL MODELING

June Fabryka-Martin and Steve Goldstein

One way to screen for chemical identity of colloidal particles is to calculate mineral saturation indices for the water. This approach establishes the presence or absence of a thermodynamic basis for the existence of colloidal particles of a specific mineralogy. The geochemical speciation program PHREEQE was used to calculate saturation indices for various minerals based on the chemical composition of water from nearby Well 5B, as reported by Schoff and Moore.<sup>1</sup> Altogether, four sets of analyses are available for this well; the one chosen for calculations was the only one for which the phosphate level was above zero. The accuracy of the chemical analyses is not known.

Based on the PHREEQE calculations, precipitation of a given mineral is favored for a saturation index  $SI > 0$ , and dissolution is favored for  $SI < 0$ . In the following summary of the results, only those minerals with elemental compositions matching that of the colloid analyses are listed. Except those for the apatites, mineral formulas were taken from Dana's Manual of Mineralogy, not the PHREEQE database. The calculations indicate that conditions in the vicinity of RNM-1 are favorable for precipitation of fluorapatite, carbonate apatite, kaolinite, K-spar, K-mica, laumontite, and numerous clays, as indicated in Table II in this report.

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## APPENDIX E - PLAN FOR COLLOID COLLECTION AND ANALYSIS

June Fabryka-Martin

Objectives: To develop a method for isolating colloids from groundwater without changing their size distribution, charge, and composition.

To characterize colloids with respect to size distribution and composition.

Approach: Collect groundwater sample in field, process sample in the laboratory to isolate colloidal fractions, analyze fractions by various techniques to characterize composition as function of size class. The approach outlined below is based upon that described by Peter Vilks et al.<sup>1,2</sup> Information is taken from these sources without further citation; some details and minor modifications were obtained by personal communication.\*

Suggestion: Sample same well at same time as Lawrence Livermore National Laboratory or Desert Research Institute to compare methods and results.

### FIELD COLLECTION

Equipment: 50-ℓ polyethylene carboy with inlet and check valve  
nitrogen tank  
connections  
thermometer  
pH meter and probe  
alkalinity kit  
Eh probe  
conductivity meter  
dissolved oxygen probe

#### Procedure:

- (a) Calculate the well volume and pump at least 3 well volumes before starting to collect sample. This will typically take at least several hours; for the Cheshire sample, for example, Buddemeier et al.<sup>3</sup> pumped for about 5 to 6 hours before collecting their sample.
- (b) Monitor the pH, conductivity, and temperature in the unfiltered water; don't sample until these values are constant. While waiting flush the carboy with nitrogen gas for ~30 minutes using the check valve to allow air to escape.

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\* Personal communication with Peter Vilks, Atomic Energy of Canada Ltd., Whiteshell Nuclear Research Establishment, Pinawa, Manitoba.

These measurements should be made in a flow-through cell in a controlled-atmosphere glove box. The effect of deviating from this procedure can be judged at least roughly from the stability of the pH measurements.

- (c) Collect an unfiltered sample and obtain final values for pH, Eh, conductivity, alkalinity, and dissolved oxygen. Attach the outlet hose to the carboy and collect the water sample. An unfiltered sample is preferred so as to reduce the risk of air contamination resulting in oxidation of Fe(II) and to avoid colloid losses to prefilters loaded with particles. To avoid disturbing the sides of the hole and creating more colloids or suspended particles, pumping rates should be as low as feasible; Vilks et al.<sup>1</sup> pumped at 0.25 to 1 ℓ/min, whereas Gschwend<sup>4</sup> recommends a rate of 0.1 mℓ/min.
- (d) When 50 ℓ have been collected, seal the carboy cap (it should be airtight) and transfer the carboy back to Los Alamos for laboratory processing. Maintain constant temperature as nearly as practical. A drop of a few degrees may result in particulate formation.

#### COLLOID SEPARATION PROCEDURE

The Pellicon flow-through system (Millipore) is used to separate colloids from the unfiltered 50-ℓ sample, ideally within 24 hours after field collection.

The filtration system should be set up inside a nitrogen-filled glove box at atmospheric pressure to avoid air contamination. The oxygen content of the glove box should be no more than several hundred parts per million. Also, if the P-CO<sub>2</sub> content of the water is known, the glove box could be filled with the appropriate CO<sub>2</sub> atmosphere. This approach was used by Ivanovich et al.,<sup>5</sup> who collected water from an Australian uranium ore deposit in a holding tank with a gas supply of a mixed N<sub>2</sub>/CO<sub>2</sub> atmosphere slightly above 1 bar to prevent contamination with air oxygen. The mixture contained 2.1 +/- 0.1% CO<sub>2</sub> to maintain the carbonate equilibria in the water. The percentage of CO<sub>2</sub> was calculated from measured pH and alkalinity results.

The procedure is designed to produce several concentrates of different particle sizes, to minimize salt retention during ultrafiltration (affects water with low salt content) and to eliminate problems of irreversible membrane fouling by suspended particles. At the end of colloid concentration, samples should be transferred into airtight borosilicate glass and PVC bottles without exposure to atmospheric oxygen.<sup>5</sup>

S. Goldstein, Los Alamos National Laboratory, has suggested that blanks should be run both before and after the groundwater sample and that 2 ℓ subjected to the following procedure should be adequate.

Filtration sequence (see Fig. E1.)

- (a) Filter ~40 ℓ of water through the Pellicon system with a polysulfone 100 000 NMWL (~10-nm cutoff) filter membrane packets separated by retentate screens. Discard the first few hundred milliliters to

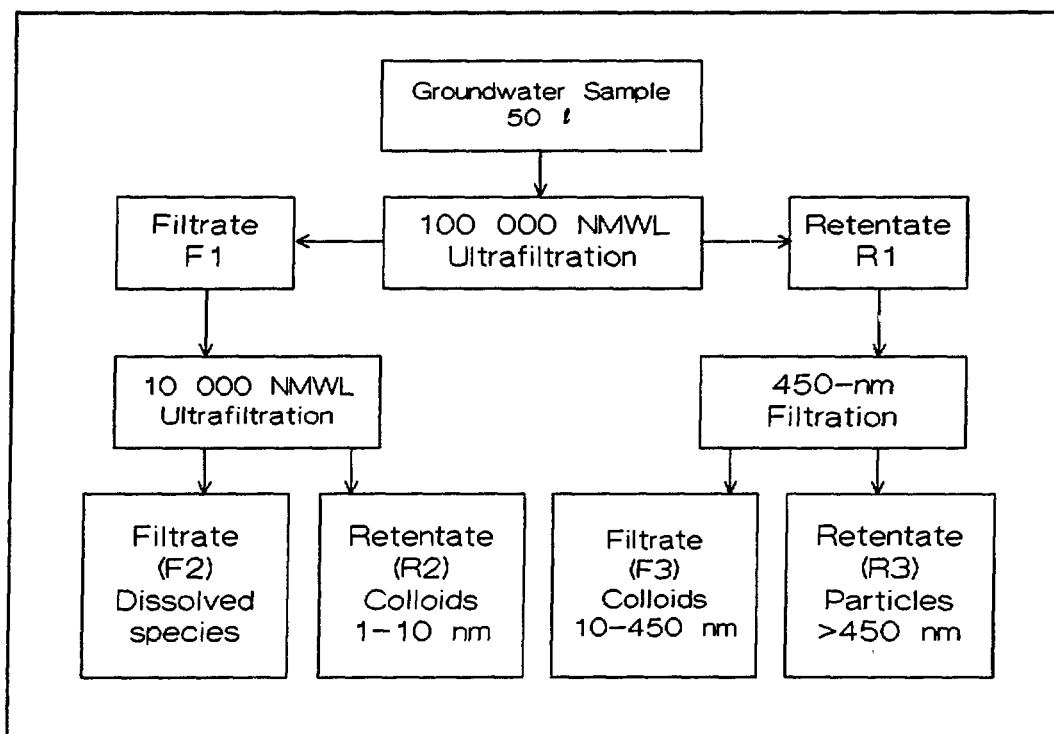


Fig. E1. Schematic for the proposed filtration sequence.

clean out the system, then circulate the water through the system until ~3 l of retentate are obtained. (The use of a multiple-membrane cassette instead of a single-unit membrane cassette was an improvement introduced by Vilks et al.,<sup>2</sup> who found the multiple-cassette system easy to dismantle for cleaning.) Label retentate R1 and filtrate F1.

- (b) The retentate R1 produced from the 100 000 NMWL filtration is filtered with a Minitan tangential flow filtration system (Millipore), equipped with a 450-nm cutoff Durapore membrane. This produces a filtrate F3 containing colloids between 10 and 450 nm as well as a retentate R3 enriched in suspended particles >450 nm.
- (c) Ten liters of the filtrate F1 produced from the 100 000 NMWL filtration is filtered through the Pellicon system with a polysulfone 10 000 NMWL filter cassette. This produces a filtrate F2 containing only dissolved species and a retentate R2 enriched in colloids with a size range of ~1 to 10 nm.
- (d) Split the samples for various analyses. The filtrate and retentate samples to be used for chemical and radiochemical analyses are preserved by acidifying to pH 1 with HCl.

## SAMPLE ANALYSIS

### (1) Size distribution

- (a) Samples F3 and R3. Pressure-filter 30 to 50 ml of particle concentrates through a series of 25-mm Nuclepore polycarbonate filters with cutoff sizes of 5000, 400, 100, 50 and 10 nm. Filter to air dryness and determine particle concentrations by the weight of the material deposited on the filters. Use a polonium charge eliminator to eliminate static on the membranes. Nuclepore is highly recommended because of the negligible water retention; cellulose nitrate membranes are a poor choice because of weight variability. In addition, Ivanovich et al.<sup>5</sup> notes that Nuclepore filters have uniform pore sizes and, hence, well-defined cutoff sizes, whereas other commonly used filters have a sponge-like structure.
- (b) Sample R2. The presence of colloidal material in the 1- to 10-nm range is indicated by autocorrelation spectroscopy and its composition is determined by chemical analysis of the colloid concentrate.\*

### (2) Particle compositions

- (a) Samples F3 and R3. The material deposited on Nuclepore membranes is examined by scanning electron microscopy with energy dispersive x-ray analysis (SEM/EDX) to identify particle compositions and to visually evaluate the effectiveness of the particle size separation. SEM/EDX has a resolution of ~50 nm, but charge effects caused by the filter membranes may degrade the resolution. Use of a conducting coating is advisable.
- (b) Sample R3. Filter ~50 ml of retentate R3 through a 450-nm silver membrane to prepare a sample for x-ray diffraction analysis.
- (c) All retentate samples and filtrate F2.
  - Determine major and trace element compositions by atomic absorption and inductively coupled plasma spectrometry. Correct for the effect of salt retention.

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\* The concentrations of elements associated with particles will be increased in the various retentate samples relative to the filtrate sample F2. The relative extent of increase in elemental concentrations can then be used to determine the particle compositions. However, this approach may be compromised if salts are concentrated along with the particles. This salt retention can be quantified by measuring conductivity in filtrates and particle concentrates. The effect decreases with increasing ionic strength and is insignificant in waters with more than 1 g/l of dissolved salts. Vilks et al.<sup>2</sup> estimates that the fraction of dissolved salts concentrated with colloids can be as high as ~4.

- Determine inorganic and organic carbon by the Astro 2001 carbon analyzer.
- Determine uranium by fluorometry using the Scintex system.
- Radioisotopic analyses. Dry to residue for gamma-counting. Use chemical separation to produce samples for alpha spectrometry (uranium, thorium, and radium), if desired.

#### OTHER CHARACTERIZATION METHODS MENTIONED IN LITERATURE

Buddemeier et al.<sup>3</sup> measured cation contents by inductively coupled plasma optical emission spectroscopy and anion contents by ion chromatography. Dried ultrafiltrates and retentates were analyzed by x-ray fluorescence to obtain elemental compositions as well as by x-ray diffraction to determine dominant mineralogy.

Ramsey<sup>6</sup> suggests ultrafiltration, centrifugation, and various light scattering techniques (static light scattering, photon correlation spectroscopy, electrophoretic light scattering, and fiber optic light scattering).

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