Nuclide-Migration Field Experiments

Program Plan

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ABSTRACT

When considering groundwater flow and radionuclide retention in the complex flow systems that can occur in geologic formations, one has a serious problem in determining if laboratory studies are being performed under conditions appropriate to natural systems and if models of nuclide transport derived from laboratory results correctly handle nuclide movement under natural conditions. These questions can only be answered by increasingly more complex laboratory and field experiments.

This document is the project plan for a program designed to begin to address these problems. The project is being carried out jointly by the Los Alamos National Laboratory, Sandia National Laboratories, and Argonne National Laboratory. The work has three principal objectives: 1) to develop the experimental, instrumental, and safety techniques necessary to conduct controlled, small-scale radionuclide migration field experiments, including those involving actinides; 2) to use these techniques to define radionuclide migration through rock by performing generic, at-depth experiments under closely monitored conditions; and 3) to determine whether available lithologic, geochemical, and hydrologic properties together with existing or developing transport models are sufficient and appropriate to describe real field conditions.

* Present member of the Steering Committee for this project.

I. INTRODUCTION

The intrinsic appeal of deep burial as a means for safe disposal of nuclear reactor waste is due to the concept that the rock overlying the repository will provide a significant barrier between the radioactive waste and man's environment. It is of considerable importance to the waste management program to understand the properties of this geologic barrier and the manner in which it will function as a barrier. Such understanding will also be the basis for predicting the performance of a repository and for identifying potential deficiencies in the models used for the predictions. Clearly such an understanding will also be essential in the process of selecting and licensing a repository and indeed in convincing the general public of the ultimate safety of such disposal.

There are, of course, many factors to be considered in selecting a particular site in a particular type of rock as the best, or at least as adequate, for the purpose. Plans to "demonstrate the safe disposal" of nuclear waste are often discussed and it is certainly a requirement for a candidate site that it be possible to construct the repository safely therein, then to emplace the waste safely. There are, nevertheless, obvious limitations to our ability to directly "demonstrate" the very long-term safety of such burial; instead we must infer such safety from a thorough study and understanding of all the factors, many of which are strongly interrelated. Each potential failure mode must be considered in terms of the probability of occurrence and associated consequences. The redundancy offered by the so-called "multi-barrier concept" plays an important role in this regard. If the safety of the system can be so overdesigned that a sequence of highly improbable barrier failures is a necessary prerequisite for the appearance of radionuclides in man's environment then a safety margin in the assessment of each individual barrier will have been provided.

As presently conceived, the waste would itself be in a chemically resistant matrix, contained in a corrosion and stress resistant canister, and surrounded by a low-permeability, highly sorptive backfill. If, over the course of time, or as a result of violent local events, all these barriers should fail, then the radioactive waste would be exposed to potential solvation and subsequent transport by groundwater. The degree to which such transport is hindered,

or retarded, by sorption on, or in, the surrounding rock matrix will determine the effectiveness of the geology as a barrier to the mass transport of radioactive waste products.

Groundwater, the water which flows through fissures, cracks, and pores in the rocks of the earth's crust, generally contains small concentrations of the elements which constitute the host rocks. As it flows there is a slow interchange between chemical species in the solution and components of the surrounding minerals. Depending on local conditions there may be a net deposition or dissolution in a given zone. Such dissolution and deposition processes are usually chemically selective in nature and are responsible for the formation of mineral deposits, some of which achieve very high concentrations of particular elements. For example, small uranium deposits exceeding 50% uranium oxide are not uncommon. Thus, on a geologic time scale, mass transport can be a very effective mechanism for moving particular elements through rock matrices and concentrating ore bodies. The mechanism is particularly active in hydrothermal regions where the combination of higher temperature with thermally induced convection of groundwater tends to facilitate the dissolution, transport, and deposition processes.

It, therefore, seems clear that transport of radioactive waste materials away from a deep geologic repository by groundwater will involve those chemical mechanisms which are at work in the similar natural geochemical transport processes, i.e., dissolution, complexation, precipitation, ion exchange, surface adsorption, hydrolysis, coprecipitation, solid solution formation, etc. Furthermore, the groundwater flow pattern and velocity, the rock fracture structure, its permeability and porosity, and other factors, such as thermal gradients, which affect the flow of groundwater, will directly influence the rate and direction of movement of the radioactive materials through the rock surrounding the repository.

Ideally, the combination of hydrological and chemical properties of a rock formation would so retard such migration that no significant quantities of radioactive waste would ever appear in the accessible environment.

In seeking the ideal (i.e., completely predictable) repository, or indeed, in seeking a satisfactory repository, it is necessary to have a mathematical model to describe the behavior of nuclides in a geologic setting in order to assess the degree to which the repository is ideal. Such models will, of necessity, incorporate many simplifying assumptions and be based on an

incomplete understanding of the detailed phenomena involved in the transport process. Such being the case one must devise a means to test the models and provide a reasonable assurance that they are valid for the purpose at hand.

Therefore, the fundamental question being addressed by the program described herein is: If the detailed physical and chemical properties of a rock and a solution are measured in the laboratory, can accurate models of radionuclide transport through fractures in that rock in the field be formulated? The understanding of which laboratory data are important and the methods of formulating models that describe in situ behavior are the principal goals of the program even though the specific results in the rock used may not be directly applicable to radionuclide transport in other rock types or even different deposits of the same rock type. The new experimental, instrumental, and safety techniques developed for the field experiments can then be applied to other in situ experiments and will be useful for planning large-scale field tests.

The objective of this document is to present a detailed experimental program to answer this fundamental question. A description of the expected results, their use, and relevance to the National Waste Terminal Storage (NWTS) effort is presented first. The specific objectives of the overall program are then given, followed by a description of a series of field and laboratory studies needed to achieve the program objectives. The integration of the technical activities into a work-flow plan that identifies the goals, interrelationships, and priorities of specific work elements are presented. The anticipated schedule for performing the tasks is then given, followed by identification of specific measures of program progress (milestones), program costs, and project management.

II. BACKGROUND

There seem to be two basically different approaches to the definition of mass transport rates based on the use of empirical evidence on the one hand and theoretical models on the other.

In principle, if one knew the fundamental chemical parameters for all the minerals, compounds, solid solutions, and simple and complex ions which each waste element can form or enter, as well as the kinetics for interconversion among them, one should be able to predict exactly what would happen chemically to each waste component in any specific circumstance. This is not

a practically achievable goal and one can reasonably expect only to identify and characterize the more important (more abundant) soluble species for each waste element and to learn something about the rates and conditions under which the species are interconverted. Likewise, one can expect to identify and characterize some of the more important minerals which can incorporate waste elements. The main utility of such a basic scientific understanding of the chemistry of waste elements in groundwater is in the construction of a realistic model framework.

Therefore, when faced with a system as complex as radioactive waste elements migrating through rock, one is obliged to make maximum use of empirical evidence to build a description of the phenomenon. There are several areas in which one can empirically explore mass transport phenomena. They include laboratory measurements of the rate and fraction of selected elements adsorbed onto various rocks and minerals and investigations of naturally occurring transport systems.

The nature of the interactions of radionuclides with the rock in an aqueous environment is of primary importance. Values are needed for the equilibrium concentrations for the reactants and products in solution and on the solid. For example, effects of hydrolysis and polymerization reactions, formation of complexes, oxidizing-reducing conditions, and radiation-radiolysis conditions must be investigated. The mechanisms and kinetics of the interactions must also be determined. These include ion exchange and ion exclusion processes, aqueous/pore diffusion, replacement reactions, physical transport (colloids and suspended matter) and filtration, and precipitation processes. The critical groundwater chemistry parameters, such as pH and oxidation-reduction potential, that affect these reactions must be identified.

The hydrology of fractured systems is a relatively new field for which a considerable amount of research will be required before it is as well understood as that of the more familiar porous media. Two methods of approach to hydrology in fractured media have been put forth. One is based on the observed regularity of many fracture systems and views the rock mass as a network of interconnected discrete fractures. In this approach, each fracture system must be characterized in detail. The other approach treats the rock mass as if it were a porous medium, in which the hydraulic characteristics of the fractures are averaged. The first approach is reasonably applied only in the near field, where the detailed identification of fracture flowpaths may be

feasible. A mixed fracture-porous medium model may be appropriate for the intermediate case where only the largest fractures are of concern. In the far field only the porous medium model will be feasible, unless there is a definite, well defined fracture flow path.

In any case, one needs an understanding of flow in a single fracture. The chemistry and groundwater composition in such a fracture is dominated by the mineralogy of the fracture surface. The hydrologic regime is then superimposed on the local chemistry. One must have knowledge of fluid velocity, hydrodynamic dispersion, and fracture characteristics such as aperture size, roughness, and length.

With a data base which includes both fundamental information and empirical data from natural and man-made transport experiments one can construct and verify a realistic model of mass transport with predictive capabilities. It seems certain that empirical data must be collected for each candidate medium; it is not clear to what extent observations for one, or several, media can be generalized so as to apply to untested media.

Our approach in this project, then, is to measure the sorption and transport of elements of interest in a variety of laboratory experiments and to investigate the behavior of these same elements under field conditions designed to represent the natural environment as closely as possible. The same rock and groundwater will be used in both the laboratory and field studies. Field tests are necessary to determine if laboratory studies are being made under conditions appropriate to natural systems and if models of nuclide transport derived from laboratory results correctly predict nuclide movement under field conditions. Field tests of rocks at depth have been limited to "well" type experiments (i.e., single hole and hole-to-hole tests). Experiments of the type described here have not yet been performed, and the procedures for in situ nuclide migration experiments are poorly developed. The "well" type field experiments sample only limited locations and do not give a complete picture of the migration behavior of radionuclides. Detailed post-flow analyses of radionuclide distributions on the fracture surface, solution-rock interactions, etc., are essential for correlating migration of nuclides under actual conditions with laboratory and modeling studies. A model, or models, for predicting mass transport will be employed in design of the experiments and in analysis of the results. This information will be interfaced with the model development efforts being performed under the NWTs program.

The field experiments will be performed in tuff exposed in G-tunnel (Fig. 1) at the Nevada Test Site (NTS). G-tunnel was selected because the formations at depth are available that will allow comparison of laboratory results with field measurements and testing of models, the support facilities already exist, mining and drilling costs and time should be reduced since the equipment and subcontractors are on site, and the tuff in G-tunnel is relatively easy to mine. Furthermore, since G-tunnel is not a potential nuclear waste repository there is no possibility that the experiments will compromise the integrity of an actual site.

G-tunnel is located in Rainier Mesa in Area 12 in the north central portion of the NTS some 100 km northwest of Las Vegas, Nevada. Rainier Mesa consists of an approximately 1500 ft thick series of thin to massive bedded calc-alkaline and peralkaline ash-flow tuffs, reworked tuff and tuffaceous sandstone, capped by a massive, welded, ash-flow tuff unit. The general stratigraphy of Rainier Mesa is shown in Fig. 2 which is taken from Ref. 1. The tuff units unconformably overlie massive paleozoic rocks in the G-tunnel area (south end of Rainier Mesa).

Thordarson² studied the hydrologic properties of Rainier Mesa and concluded that (taken from Ref. 3): 1) The regional zone of saturation occurs in Paleozoic strata several thousand feet beneath the mesa surface; 2) The welded Rainier Mesa Member and the vitric Paintbrush Tuff have only fractional interstitial saturation in contrast to the zeolitically-altered Tunnel Beds which are usually 100% saturated; 3) Vertical head measurements indicate the movement of water is generally downward; and 4) Movement of water is generally restricted to nearly vertical fractures and faults in welded and highly zeolitized units such as the Rainier Mesa Member and the Tunnel Beds.

Fig. 3 is a cross-section through Rainier Mesa which schematically shows the principal modes of water transport (taken from Ref. 3).

G-tunnel is driven horizontally at a depth of ~400 m below Rainier Mesa (Fig. 4). It is located entirely in the tunnel beds. The tunnel beds consist of medium to massive bedded ash fall tuff with occasional thin beds of peralkaline and reworked tuff.⁴ The bedded tuff units are light brown to reddish brown, fine to medium grained calc-alkaline tuff, locally with reworked tuffaceous-sandstone beds. The beds are generally zeolitized (clinoptilolite). The beds dip generally ~10° to the west, although local variations exist, 0° to ~25°. Six tuff units are encountered within the tunnel workings. From

Fig. 1. Map of the Nevada Test Site.

Era	System	Series	Formation	Member or unit and symbol
		Pliocene	Timber Mountain Tuff	Rainier Mesa Member Tar
CENOZOIC	Tertiary	Miocene	Paintbrush Tuff	Tiva Canyon Member Tpc Tp
			Stockade Wash Tuff	Tsw
			Bedded and ash-flow tuffs of Area 20	Trab
			Bedded tuff or Dead Horse Flat	Tdhh
			Belled Range Tuff	Grouse Canyon Member Tbg
			Tunnel beds	Unit 5 Tt5
				Unit 4 Tt4 Subunits AB, CD, E, F, G, H, J, K ¹
				Unit 3 Tt3 Subunits A, BC, D ²
			Belted Range Tuff	Tub Spring Member Tbt
			Tunnel beds	Unit 2 Tt2
			Crater Flat Tuff	Tcf
			Tunnel beds	Unit 1 Tt1
			Redrock Valley Tuff	Trv
			Older tuffs	Tot
			Paleocolluvium	Tc
MESOCOIC	Cretaceous		Gold Meadows Stock	Kgm
PALEOZOIC	Devonian		Paleozoic rocks, undivided	p ³ z
	Silurian			
	Ordovician		Wood Canyon Formation	CpCw
PRECAMBRIAN			Stirling Quartzite	pc ³

¹K is the youngest.

²D is the youngest.

³In some drill holes, paleocolluvium of Tertiary age (Tc) rests on Paleozoic or Precambrian rocks.

Fig. 2. General Stratigraphy of Rainier Mesa, NTS (Ref. 2).

Fig. 3. Schematic Cross Section of Rainier Mesa Showing Relative Depositional Geometry and Primary Modes of Fluid Transport. Solid Arrows Indicate Flow Through Fractures; Dashed Arrows Indicate Flow Through Pores (Ref. 2).

the tunnel portal to the back of the workings, five bedded units, known as Tunnel Beds 1-5, are penetrated while the sixth unit is the welded Grouse Canyon member (Fig. 2). Tunnel Beds 1-4 are local units, while Tunnel Bed 5 is within the Belted Range Tuff. Discrete layers range from less than an inch to more than 10 feet in thickness. Field and laboratory studies to date indicate, however, that this layering is rarely mechanically effective. It is presently planned to conduct the migration experiments along natural surfaces within Tunnel Bed 5. A general description of tuff is given in Appendix A, which is taken from Ref. 5.

Fig. 4. Section of the G-Tunnel Complex.

Preliminary examination of Tunnel Bed 5 indicates that major joints and faults, i.e., those that extend for at least one tunnel diameter, occur at one or more, while minor jointing is more frequent. At least three types of relatively planar discontinuities are evident, namely joints, faults, and bedding plane partings.

III. OBJECTIVES

The work described in this program plan for conducting field experiments of element migration has three principal objectives: 1) to develop the experimental, instrumental, and safety techniques necessary to conduct controlled small-scale, at-depth radionuclide migration experiments in the field; 2) to use these techniques to observe the movement of sorbing and non-sorbing tracers in a single fracture by performing at-depth field experiments under closely controlled and well defined conditions; and 3) to determine whether available chemical and hydrologic properties for that fracture together with existing or developing transport models are sufficient and appropriate to describe nuclide migration under field conditions.

The experiments consist of several key elements. Among these is detailed groundwater characterization, including seeps in G-tunnel and local tuffaceous wells. Site selection will depend largely on fracture flow (permeability) tests. Two migration experiments will be performed. Stable or short-lived nuclides will be used in the first experiment, in part to demonstrate that the newly developed experimental methods are proper for the objectives of the project and are safe before conducting the second, actinide-containing, experiment. It is also important to demonstrate that useful information can be obtained from field tests in which radioactive tracers are not utilized since use of radioactivity may not be allowed at some future sites that need to be studied. Study of the migration rates of actinides is one of the prime goals of this project since actinides are of appreciable concern with respect to release into the accessible environment. Both experiments include detailed, post-flow analyses of the spatial distributions of the migrating elements along the fracture and into the rock matrix, using techniques developed in the initial stages of the project. Comprehensive laboratory studies of the sorptive and migration properties of the same materials will be performed using existing and newly developed techniques. The latter include the development of laboratory techniques using large cores

or blocks of material in order to better simulate the field experiment. The suitability of various water movement tracers will also be assessed in terms of their potential to interfere with the chemical system. The one(s) selected must not interfere in any way with chemistry that occurs in the fracture-flow experiments.

IV. TECHNICAL SCOPE

The following are brief descriptions of the various program activities that must be conducted in order to achieve the program objectives identified above. The descriptions are written to show the various considerations that must be addressed in order to obtain an adequate understanding of nuclide migration in a fracture. These activities will be performed by the Los Alamos National Laboratory, Sandia National Laboratories (SNL), and Argonne National Laboratory (ANL). Additional support will be obtained from the NTS Support Organizations (NTSSO) which include Reynolds Electrical Engineering Company (REECo), Fenix and Scisson (F&S), and Holmes and Narver (H&N).

A. Site Selection

Several fracture properties will be used as criteria for selection of potential experiment sites in G-tunnel. A brief discussion of the fracture properties follows.

1) The fracture must be located in the nonwelded tuff. Consistent with the current concept of a repository in tuff, the nonwelded, highly altered tuffs compose the far-field, geologic barrier to radionuclide migration. Welded tuffs, even though they have reasonably good sorptive properties for a number of nuclides, are not generally credited as a migration barrier. Within G-tunnel, the drilling and mining techniques are easy and well established. In addition, single, planar fractures (see item 2) are only found in the nonwelded tuffs.

2) The fracture should be long, planar, and isolated from other fractures. The fracture should be long enough to accommodate the detailed characterization studies required for the experiments and to provide sufficient area for conducting the two field experiments (Secs. IV.N and IV.O), including the post-flow fracture recovery. Such requirements suggest that the fracture be about 30 m long. A planar fracture simplifies the field experiments considerably. In particular, the placement of the packers, water injection system

(Sec. IV.J), and water sampling system (Sec. IV.J) is greatly simplified, as is the post-flow recovery of the fracture (and the nuclides).

3) The fracture should have a uniform, small ($<100\text{ }\mu\text{m}$) aperture. A uniform aperture simplifies the detailed characterization studies--especially the interpretation of the permeability tests--and also simplifies the mathematical analysis of the experiments. In reality, of course, uniformity is difficult to measure in situ. Consequently, the post-test analysis (Sec. IV.I) of the flow path in the "cold" experiment (Sec. IV.N) may be the first unambiguous evaluation of fracture uniformity. The primary restriction on the fracture aperture is that it be small enough to minimize external effects (e.g., gravity), thereby allowing control of the water flow rate, perhaps to as low as 4×10^{-5} cm/sec (1 m per month). The allowable size of the aperture depends on the orientation of the fracture, i.e., horizontal or vertical. To overcome gravitational effects, the aperture of a vertical fracture would need to be much smaller than that of a horizontal fracture. It is probably impossible to determine directly the fracture aperture in the field. The aperture will therefore be inferred from the fracture permeability tests.

4) A horizontal fracture is preferable. Conducting the experiments in a horizontal fracture would minimize the primary experimental difficulties anticipated for experiments conducted in a vertical fracture, namely, maintaining controlled, sheet-water flow and providing the post-test nuclide mass balance. A horizontal fracture would virtually eliminate gravitational effects. However, the experiments can be conducted in a suitable vertical fracture (Secs. IV.N and IV.O).

The experiment sites must be out of the path of other tunnel activities scheduled within the next four years because of the anticipated use of radio-nuclides and to avoid interference. Note that a fracture with altered surfaces is not a selection requirement. The nonwelded tuffs in G-tunnel are highly altered, and contain large amounts of zeolites (clinoptilolite). The surface of either a fresh fracture or an older one through the matrix would contain minerals which are highly sorbing for a large number of nuclides.

An initial survey of G-tunnel (Fig. 5) has been carried out to locate potential sites for the field experiments. Three have been identified for further evaluation. These are the right rib of EP-1 in the Madison drift, EP-2/HF-9 drift, and HF-23/EV5-2 drift. Vertical fracture zones associated with vertical faulting is in EP-1. A single, well-defined, vertical fracture

Fig. 5. The G-tunnel Complex.

is located in EP-2/HF-9. An extensive bedding/parting plane is located in EV5-2. Since a horizontal fracture is the first choice for this experiment, the last location will be evaluated first.

The initial site evaluation will include site mapping, fracture core drilling, mineralogy/petrology, and fracture permeability measurements. The mapping is intended to determine the orientation and extent of the fractures in the vicinity. Removing portions of the fracture by core drilling will provide samples of the matrix and fracture surface for laboratory examination. The core samples, as well as hand samples removed from the site, will be used in the mineralogy/petrology studies. Permeability measurements will be mainly of the fracture to determine the flow rates although permeability measurements on the rock matrix will also be made. The initial laboratory studies will emphasize rock interactions with available tuffaceous groundwater (Sec. IV.C) and simple sorptive tests. The sorptive experiments may be carried out using fracture-surface samples if the mineralogy/petrology indicates a significant difference between the matrix and the fracture surface.

Upon completion of initial evaluation studies, the information will be summarized and compared with the desired properties listed above. One of the sites will then be selected for the field experiments and will be subjected to detailed characterization.

B. Detailed Site Characterization

The goal of the detailed characterization of the experimental site is to determine the parameters necessary to carry out the experiments. The emphasis will be placed primarily on permeability characterization of the fracture and the matrix aimed at qualifying the fracture for the "cold" experiment (Sec. IV.N). There are three key field activities associated with the site. Each of the activities will be carried out in the same fracture but at a different location. The activities are the initial permeability test, the "cold" experiment, and the "hot" experiment (Sec. IV.O). With careful planning, the initial permeability tests and the "cold" experiment might be carried out sequentially in the same location.

A brief discussion of the site characterization studies follows.

- 1) Core drilling of the fracture will provide samples for in-depth characterization of the fracture surface and the rock matrix. It is anticipated that a large number of samples will be required. It is expected to be

able to provide core approximately 1 m long containing the fracture oriented parallel to the core axis can be provided. Later, the drill holes will be used for permeability testing, examination (television), and injector/collector system testing.

2) Permeability measurements of the fracture will be made, probably at several locations, to determine the flow uniformity through the fracture. Interpretation of these measurements will require close coordination with the mathematical modeling support and with the mineralogy/petrology studies of the fracture. Permeability measurements will be extended to include the rock matrix.

A new apparatus for measuring in situ permeability is currently being designed to replace the apparatus used in other programs, which was designed for high pressure hydraulic fracturing of rock and is not suitable for this project. The new apparatus must be capable of low pressure operation (<3.5 MPa), maintaining constant pressure (± 20 kPa), delivering large fluid volumes (~ 0.75 l/s), being well instrumented (pressure, flow rate, temperature), and being tightly sealed through use of packers.

3) In addition to careful examination of the core, the drill holes will be evaluated at depth using bore-hole television cameras. The smallest hole we can examine with the television cameras is an 9.6 cm i.d. (an "HQ hole"). The wall of the holes will be evaluated for factors such as smoothness, irregularities, voids, and inclusions which might affect the injector and collector systems' packer placement and integrity (Secs. IV.J and IV.K). Location of fractures and the size of the aperture will be noted within the resolution of the camera (~ 1 mm). All of the television scans will be retained on video tapes for comparative examination with other holes and as a permanent record.

4) Three methods of fracture identification will be used. Tunnel-wall mapping identifies large fractures and is best suited to determining orientation and extent. Core examination provides a more detailed record of the fractures at depth, in the immediate vicinity of the hole. Television examination provides the best record of in situ fractures but is limited by resolution (~ 1 mm). Each of these methods provides a body of information that is complementary and somewhat overlapping with the others. The information will be compared, analyzed, and combined to yield the best single, detailed fracture map of the site.

5) The mineralogical and petrological studies begun as part of the site selection activity will be continued. Detailed studies of the matrix and fracture surface will be carried out and correlated with the sorption studies. In particular, quantitative bulk and areal mineral contents will be determined.

6) Quantitative chemical analysis of the matrix rock and, if possible, of the fracture surface will be performed. The results will impact especially the nuclide selection and concentration used in the "cold" (Sec. IV.N) experiment.

7) An important aspect of the experiments is to assess our ability to determine laboratory data which is relevant to field behavior. Toward this goal, a variety of flow experiments in artificial and natural fractures will be carried out in the laboratory. These studies will increase in scale, starting with standard 4.75 cm diameter by 10 cm long core perhaps up to core approximately 28 cm diameter by 1 m long. Eventually, we plan a laboratory experiment using a natural fracture about 1 m long.

The techniques for removing large, fractured blocks, or fractured core for that matter, are not well established. Techniques will be evaluated (within certain time and monetary constraints yet to be determined) for removal of such samples. The techniques to be considered include use of the Alpine miner, line drilling, and various immobilization methods to prevent the two sections of the fractured samples from abrading each other. Initial block (core) removal efforts will concentrate on unfractured blocks which will be artificially fractured later in the laboratory. Once (and if) the technique has been developed, one or two fractured blocks will be removed from the site.

C. Groundwater Characterization Studies

It is desirable to use water in these studies that has a composition as close to equilibrium with the rock surfaces involved in the experiments as is reasonably possible. Non-equilibrium reactions between the natural components of the water and the rock must not compete with the sorption-desorption reactions of the nuclides being studied. It is also desirable to use a natural groundwater since the chemical species in solution are more likely to be in an equilibrium state.

Claassen and White⁶ have studied the compositions of water in the Rainier Mesa system. Water recharging in the zeolitized Tunnel Bed tuffs has passed through the overlying devitrified and vitric tuff units. Seep samples collected for several years in the Tunnel Beds (primarily B and T tunnels) were typically high in sodium concentration and some contained significant calcium. The higher calcium concentrations are associated with water in the overlying units and with water from lesser thickness of the Tunnel Beds. The high sodium concentrations may result from precipitation and ion exchange reactions occurring in the zeolitized Tunnel Beds.

Therefore, natural tuffaceous water from several different sources at the NTS will be investigated. Emphasis will be placed on water from the same or similar tuff horizons as at the experimental site (Sec. IV.A). The water samples will be analyzed by plasma-emission spectroscopy and ion-chromatography for major components and by x-ray fluorescence spectroscopy and neutron-activation analyses for minor constituents. Critical groundwater chemistry parameters, such as pH and oxidation-reduction (redox) potential, will be measured. Knowledge of the redox potential is of particular importance since reducing conditions can lead to different behavior of some elements, e.g., technetium and plutonium. The presence of Fe(II) in the groundwater may be a good indication of a reducing environment and platinum or gold electrodes may be useful for measuring the redox potential. It may also be possible to verify that the redox potential is not changing in the course of the field experiments (Secs. IV.N and IV.O) by eluting a redox couple⁷ placed in the injected solution.

A number of water seeps have been active at one time or another in G-tunnel. Currently, only two are known to be flowing. The two are likely from the same system and are located at EL6168, near HF-23/EV5-2. This dual seep has a reasonably fast flow. The source of the water in the seeps is unknown, but perhaps it is from the perched zone.

Samples of water will be collected from these seeps in G-tunnel and will be analyzed. The ceiling at each seep-location will be cleaned, exposing fresh rock, prior to such sampling. The water collection apparatus being developed by the Lawrence Livermore Laboratory⁸ may be used to collect the seep water or a simple system specific for the seeps in G-tunnel will be developed. The system will allow water to be collected in an inert

atmosphere, the components of which can be controlled, while eliminating evaporation. It is possible that sufficient water is available from these sources for the laboratory and field experiments.

The water collected in the "Water Migration/Heater" experiment will also be analyzed.⁵ It is known that the water from this experiment came from the Grouse Canyon member above the nonwelded tuff, and probably moved in the vapor phase.

There are several wells at the NTS that sample tuff aquifers.⁹ Water from Well J-13 in Jackass Flats has been used in some experiments. Water from Well 8 (Fig. 1) on Rainier Mesa is also readily available. The water from Well J-13 has a higher concentration of dissolved solids than that from Well 8, but the ratio of the concentrations of most of the major constituents is fairly constant at 1.3 ± 0.2 . The magnesium, fluoride, and nitrate are more abundant in J-13 with approximate concentration ratios of 2.3, 3.0, and 2.0 relative to Well 8; however, these components are present in abundances of only 1 to 5 ppm. Although the water from the J-13 well contains ~13 ppm calcium past experience suggests¹⁰ that the water may change composition somewhat when contacted with rock. Therefore, they will be pretreated with rock from the vicinity of the experiments for several weeks or months. The results of the analyses of these water samples will be compared with data for the seep waters.

A criterion for suitability of the water is constancy in composition (i.e., equilibrium) throughout an experiment. A test might involve passing the water through a one-to-several-meter-long column of crushed tuff, or possibly through a fracture, and then measuring the composition of the effluent.

Preparation of the water for the field experiment probably will involve shaking or stirring water from a well with the surface material of the fracture or with crushed rock from near the experiments followed by filtration. The details of this preparation will be specified after the exploratory experiments discussed above have been carried out.

D. Initial Selection of Non-conservative (Sorbing) Tracers

There are several criteria that a tracer must meet in order to be used in either the "cold" (Sec. IV.N) or "hot" (Sec. IV.O) field experiments or in the laboratory studies (Secs. IV.H and IV.L). 1) It must be soluble in the groundwater selected for use (Sec. IV.C). 2) It must be conveniently detectable and must have a very low concentration in the groundwater so that

the adsorption or exchange sites along the flow path are not immediately saturated. 3) It must be easily distinguishable from the background imposed by the groundwater, rock, and location in G-tunnel. 4) It must be sufficiently mobile through the fissure so that it does not totally remain near the injection point. 5) If it is not radioactive, it must be a reasonable chemical analog of a radioactive element that is of concern in a repository.

1. Tracers for the "Cold" Experiment. Separated isotopes of stable nuclides will be considered for use in the first "cold" (Sec. IV.N) experiment. The potential stable elements, including cesium, strontium, europium, neodymium, and thorium, as well as ^{235}U , will be considered with regard to the natural background in G-tunnel and methods (and ease) of detection (Sec. IV.I). Mass spectrometry, for example, would be a useful analytical technique; however, it would be advantageous to have at least one on-line method of detection as well. Short-lived isotopes such as ^{85}Sr and $^{95\text{m}}\text{Tc}$ would be easily detected by "on-line" techniques, and it may be possible to use such radionuclides in addition to stable tracers. This will depend on safety considerations in regard to mining and coring techniques.

Additional tracers for the cold field experiment will include a water movement tracer (i.e., iodine or bromine, Sec. IV.G) and perhaps a pseudocolloid, if the technology for studying colloid migration is sufficiently developed. As mentioned in Section IV.L, a pseudocolloid is being developed in Sweden.¹¹

2. Radioactive Tracers. There are several criteria, in addition to those discussed earlier, for selecting a radionuclide for use as a sorbing (non-conservative) tracer. 1) It must be an isotopic analog of a nuclear waste product of concern in a repository. 2) It must have a half-life long enough to survive the duration of the experiment (say 1 y). 3) Within limitations, it should have as short a half-life as possible to obtain a high specific activity. This is essential if one is to stay within the low solubility of most fission products and actinides at the pH values characteristic of most groundwaters and not affect the sorption isotherm. The optimum half-life seems to be between three months and one year. 4) In order to obtain the highest specific activity, the tracers should be prepared carrier free from fission products or cyclotron bombardments. If (n, γ) reactions are needed for preparation, the highest flux should be used to minimize dilution by stable target materials. 5) Finally, gamma-emitting radionuclides should be used to facilitate sample

analysis. Alpha counting, although it requires substantial sample preparation, has the advantage of very high sensitivity due to low background in the detector systems. High-efficiency, high-resolution germanium detectors and automated counting systems for gamma analysis are available with automatic data processing.

Table I gives a tentative list of radionuclides suggested for the "hot" field experiment which best meet the outlined requirements. A final list will be decided upon after the laboratory studies are completed and the experimental test plan (Secs. VII and X) for the "hot" experiment (Sec. IV.0) is written. Table I also includes estimates of the amounts of tracer that would be used, assuming a concentration gradient (ratio of the input concentration to the final concentration) of 10^5 and a minimum detection limit of 10 d/m per cm^2 .

E. Isotope Production

The ANL 60 in. cyclotron has long been used to provide isotopes on a production basis. A partial list of such isotopes and their production rate are given in Appendix B. In particular, ANL has been supplying $^{95\text{m}}\text{Tc}$, ^{235}Np , and ^{237}Pu for the Waste-Rock Interactions Technology (WRIT) program (Sec. VI) for some time. This machine is capable of delivering about a 50 particle- μA beam. Funding for 300 h per year has been included in the budget (Sec. VIII). As implied in Table I all of the standard production techniques have already been developed at ANL. However, in order to obtain larger quantities of the required isotopes it will be necessary to deal with heat densities of over 30 KW/ml of target. New target holders will be designed and constructed to handle such power densities. A "brute force" approach, based on a scaling up of current capabilities will be used.

The beam time necessary to produce the quantities of isotopes needed for the field experiments is also given in Table I. This assumes that the high current targets (750 μA) are available. It should be noted that a gas target assembly, a solid target assembly, and an alpha-active target assembly are needed to produce the isotopes listed in Table I. These targets are already operating in the 20-30 μA range.

Since the cyclotron costs are based on using 300 hours of beam time per year ^{236}Pu rather than ^{237}Pu will be used in the "hot" field experiment (Sec. IV.0). The estimated beam times for the other isotopes (assuming ^{242}Cm is made in the reactor) total 290 hours.

TABLE I
CANDIDATE RADIONUCLIDES FOR THE EXPERIMENTS

Isotope	Half-life	Production Method	Source ^a	Comments	Activity, Injected ^b (mCi)	Beam Time ^c (h)
¹³⁷ Cs	30 y	Fission	C	long half-life		
¹³⁴ Cs	2 y	¹³³ Cs(n,γ)	C	specific activity depends on flux of reactor		
		¹³⁴ Xe(d,2n)	P		0.7	80
⁸⁵ Sr	65 d	⁸⁴ Kr(α,3n)	C,P		2.8	12
⁸⁹ Sr	50 d	⁸⁸ Sr(d,γ)	C	short half-life; β emitter		
		⁸⁸ Sr(d,p)	P	only		
^{95m} Tc	61 d	⁹³ Nb(α,2n)	P	short half-life ^d	2.5	
		⁹⁴ Mo(d,n)	C			
		⁹⁵ Mo(d,2n)				
		⁹³ Nb(α, 2n)	P			70
¹²⁵ I	60 d	¹²³ Sb(α,2n)		short half-life ^d	0.2	
¹³¹ I	8 d	Fission	C	short half-life	1.2	
²³⁵ Np	410 d	²³⁵ U(d,2n)	P	electron capture	0.9	65
²³⁶ Pu	2.6 y	²³⁵ U(d,n)	P	α emitter only	0.5	60

TABLE I (cont.)
CANDIDATE RADIONUCLIDES FOR THE EXPERIMENTS

Isotope	Half-life	Production Method	Source	Comments	Activity, Injected ^b (mCi)	Beam Time ^c (h)
²³⁷ Pu	45 d	²³⁵ U(α ,2n)	P	short half-life	3.2	400
²⁴¹ Am	450 y		C	long half-life		
²⁴² Cm	160 d	²⁴¹ Am(d,n)	P	α emitter only ^e	1.2	
²²⁸ Ra	6.7 y	natural		β emitter only	0.7	
²²⁶ Ra	1600 y	natural	C	long half-life		

^aC implies commercial; P implies production at ANL (Sec. IV.E):

^bEstimated using a concentration gradient of 10^5 on the surface and a minimum detection limit of 10 d/m/cm².

^cAt the ANL 60 in. cyclotron.

^dPossible water movement tracer (Sec. IV.G) since usually anionic in groundwater.

^eIf the laboratory experiments indicate that curium migrates in the same fashion as americium then americium may not be used.

The ^{228}Ra can best be prepared by chemical purification from large quantities of its parent nuclide, ^{232}Th , in which it is present at equilibrium. We are currently looking for old samples of ^{232}Th to use for this purpose.

F. Alcove Design and Construction

1. Experimental Area. Reasonable isolation of the experiment site from normal tunnel traffic is desirable for safety reasons since radioactive tracers will be used in some of the experiments. All of the sites currently under consideration are located in side drifts sufficiently removed from the main tunnel to provide the required safety margin.

2. Instrumentation Alcove. A fully enclosed instrumentation alcove will be constructed to provide additional safety (and containment should a leak occur) and to limit access to the experimental site only to authorized personnel. The alcove will enclose the field and analytical instruments and the data acquisition system. It will also provide the water storage area for the experiments. The temperature and humidity in the instrumentation alcove will be controlled to protect the data acquisition system and other sensitive equipment.

The alcove design will depend on the experiment configuration, the field instrumentation, and the underground analytical capabilities. The alcove may possibly be divided into two rooms, one containing the tracers and the water injection and collection systems, the other containing the data acquisition system and the analytical instruments. The need for such a design (or others) will be evaluated later.

3. Instrumentation. The parameters to be monitored in these experiments fall into three groups, chemical, hydraulic, and radioactive. All of the parameters will be monitored in the alcove, injection hole(s), and collection hole(s), where appropriate. The chemical parameters include pH, Eh (if possible) and, possibly, the concentration of specific ions (specific ion electrodes). The long term stability of the equipment needed to perform each of these measurements must be evaluated.

Hydrologic parameters include temperature, pressure, and flow rate. Depth gauges will be placed in the collection hole to monitor the water influx rate. A depth gauge will also be used (through the data acquisition system) to actuate a drain mechanism.

Radiation detectors will be used to monitor for radionuclide leakage around the packers in the injection and collection holes. The possibility of

placing small detectors along the fracture in the collection hole will be evaluated. The injection system in the alcove will also be carefully monitored.

The exact types and quantities of instrumentation and a schematic of their location will be included in the Experiment Plan. An as-built diagram of instrument locations will be available prior to initiation of flow for each of the experiments.

Some analytical instruments may be placed in the instrumentation alcove and a fully equipped analytical trailer may be located in the G-tunnel portal area. Either of these placements would provide necessary "real time" determination of the experiment status. The on-site availability of analytical capabilities would be particularly useful in monitoring certain types of system parameters (e.g., water composition).

4. Data Acquisition System. The data acquisition system will be built around existing Hewlett-Packard 9845B programmable calculators with 480K of memory. Data are recorded at programmed intervals and stored on disk, magnetic tape, and paper printout. The system will be powered by an uninterruptable power supply.

A duplicate auxiliary system may be located in the G-tunnel portal area to serve as a backup system and to provide for data reduction. The system would also allow external monitoring of the experiment during hours that the tunnel is closed (nights and weekends).

The data acquisition system will be programmed to actuate a telephone alarm system. Conditions that might require such an alarm are, water over pressurization, a radionuclide leak, or a power outage.

Raw data will be provided at regular intervals to those who wish it (Sec. VI). With the HP 9845B system, the data can be prepared in a tabular or graphical format.

G. Water Movement Tracers

One or more water tracers will be utilized to measure water velocity and dispersion parameters. Ideally, a water movement tracer for this experiment should have a number of properties. 1) It should move at a velocity very near the velocity of groundwater. 2) It should not sorb on the geologic medium (R_d or $K_d = 0$), nor should it move significantly faster than the water. 3) It should not change the characteristics of the hydrologic system, i.e., the viscosity and density of the water, and permeability of the rock. 4) The

tracer should be used and detectable in low concentration, and the natural background must be still lower. Simple detection methods and detection in the field, possibly on-line, are desirable. It may be desirable to have several tracers that can be used sequentially in the same experiment. 5) The tracer must not interfere in the transport-retardation processes to be measured. It must not complex or interact with species in solution or sorption sites to the extent that these processes could be altered.

All of these properties except the last have been considered in many published studies (e.g., Ref. 12). The last requirement for a water tracer is unique for experiments in which retardation or transport of other species is to be measured.

It will be necessary to explore the use of both radioactive and non-radioactive water tracers for this work. In the "cold" experiment (Sec. IV.N) a non-radioactive tracer may be required; however, a short-lived tracer may be permissible. The tracer used in the "cold" experiment may be used in later in situ experiments near repository sites where longer lived radioactive tracers could be prohibited. In the "hot" experiment (Sec. IV.O), a radioactive water tracer may be more desirable.

1. Survey of Possible Water Movement Tracers. A variety of types of tracers are found in the literature: (1) isotopically tagged water molecules, (2) halides, (3) dyes, (4) non-reactive gases and compounds, and (5) others. The pertinent properties of these tracers are briefly reviewed below.

a. Tagged Water Molecules. Tritiated water, HTO, has been used as a tracer for many hydrologic investigations and will most likely be the first choice for the "hot" experiment. The analysis for HTO by a scintillation method is easy, fast, quite sensitive, and amenable to on-line use; however, a distillation step probably will be required to separate the tritium for analysis when other radionuclides are present.

Heavy water, HDO or D_2O , is a possible water tracer. The low relative isotopic abundance of deuterium of 0.015% allows for a range of dilution of several orders of magnitude if ~100% D_2O is injected. Analysis of deuterium is possible by infra-red spectroscopy to 0.1 mole % D_2O .¹³ Mass spectrometric analysis of hydrogen gas is much more sensitive. Photo-disintegration of deuterium and counting of the neutrons¹⁴ is not suitable for this application since many milliliters of water are required for the analysis as well as an energetic gamma-ray source.

Water can also be tagged with ^{18}O or ^{17}O (relative natural abundances 0.2% and 0.04%, respectively) and mass spectrometric analysis can be used. Since oxygen exchange between water minerals is a possibility,¹⁵ it will be necessary to show that the effect is negligible if oxygen isotopes are used. Cory and Horton¹³ showed that elution curves for water through a 186-cm long column of kaolinitic soil were identical for water tagged with ^2H , ^3H , and ^{18}O . The same authors also considered exchange between hydroxyl ions and hydrogen atoms of the clay minerals and water and concluded that retardation of the tracer would be of little consequence in laboratory and field studies. Kaufman and Orlab¹⁶ observed that the tritium front traveled at a slightly slower velocity than bromide through a sandy loam and attributed this to exchange of hydrogen or hydroxyl ions or to replacement of water in the minerals. Another likely explanation of the phenomenon is ion exclusion, which is discussed below. In laboratory studies of various tuffs no fractionation of ^3H and ^{131}I has been observed for columns of crushed rock 3 cm long.¹⁷

b. Halides. Chlorides, bromides, and iodides are used as water tracers either as stable or radioactive ions. The observation that some anions appear to move faster than the water front^{16,18} is attributed^{16,19} to a phenomenon known as anion exclusion, in which anions are repelled by the negative charges on minerals. Iodide is supposed to sorb more strongly than bromide or chloride.^{16,20} However, no separation of iodide and tritium has been observed on tuff columns,¹⁷ and the sorption of ^{131}I on various tuffs is zero.²¹ Investigators from Pennsylvania State University²² saw essentially identical breakthrough curves for bromide and tritiated water in flow through fractured carbonate rocks at the U. S. Geological Survey Armogosa Tracer Site for distances as great as 400 feet.

Bromide is commonly used as a stable tracer. It can be detected by a bromide electrode between 0.4 (possibly 0.08) and 80 000 ppm; however, one must investigate possible interferences. It is easily detectable at 1 ppm by anion chromatography and can be detected at much lower limits by neutron activation analysis.^{23,24} The chloride concentration²¹ of tuffaceous water from well J-13 is ~7 ppm. If the ratio of chloride to bromide is 300,¹² one would expect a bromide concentration of ~0.02 ppm in water from both Well J-13 and Well 8 based on chloride analyses.⁹ An inactive spike of 300 ppm might, therefore, be detectable at a dilution of ~10 000.

The isotope ^{82}Br (35 h half-life) might be used even in the "cold" experiment because it decays so quickly; however, the short half-life limits its usefulness for experiments involving a low flow rate. It can be produced essentially carrier free.²⁵ The isotope ^{77}Br (56 h half-life) could be used for a somewhat longer period.

For the "hot" experiments, the nuclides ^{125}I (60 d half-life), ^{131}I (8 d half-life), and ^{36}Cl (3×10^6 y half-life) might be considered in addition to tritium.

c. Dyes. Many organic dyes have been used for water tracers, but their usefulness varies with the application and geologic system. Smart and Laidlaw²⁶ evaluated eight fluorescent dyes in the laboratory and field, considering sensitivity, detectability, effects of pH, concentration, salinity, sediments, background fluorescence, photochemical and chemical decay, toxicity, biodegradation, cost, and adsorption on various sediments and equipment. A number of dyes, including fluorescein, pyranine, and rhodamine WT exhibited only small sorptive losses on inorganic minerals. Sorptive losses on organic matter and retardation of dyes in systems containing organic matter has been observed by many investigators,²⁶⁻²⁸ but this may not be pertinent in studies of systems like tuff where no organics are present. However, these dyes may have some complexing ability.

d. Non-reactive Gases and Compounds. Several types of substances fall in this category. Rare (noble) gases are sufficiently soluble for water tracing;²⁹ however, recent LASL results may indicate possible fractionation of ^{85}Kr and HTO in the transport of water from the Cambrian nuclear test to a water well 90 m away.³⁰

Investigators at the University of Arizona are using a series of chloro-fluorocarbons (freons) quite successfully as water tracers.¹² These compounds can be used at very low concentrations (few tenths of parts per billion) since they are detectable at 1 to 100 parts per trillion (10^{12}) level by electron-capture gas chromatography. Losses of dissolved gases by volatilization from open samples is an operational problem.

Perfluorocarbons, such as perfluoro-dimethyl-cyclobutane (PDCB) (boiling point, BP, $\sim 40^\circ\text{C}$), perfluoro-cyclobutane (PCB, C-318) (room temperature BP), and perfluorodimethyl-cyclohexane (PMCH) (BP $\sim 90^\circ\text{C}$), have been used at LASL as atmospheric tracers,³¹ which are all detectable by the same method, at sensitivities of about 10^{-9} g/g by routine methods and 10^{-12} g/g by state-of-the-art

methods. If the solubility of the perfluorocarbons in water is sufficiently high (like rare gases, 10 to 100 ppm), perhaps one of these may not be subject to volatilization losses and yet not exhibit chromatographic retardation behavior. Sulfur hexafluoride may also be a good tracer with a detectability of 10^{-10} g/g by the same method.

A last group of dissolved gases at extremely low concentrations are the heavy methanes, CD_4 and $^{13}CD_4$. These are also being used as atmospheric tracers.³¹ The natural abundance of methane-21 in air is $\sim 1 \times 10^{-23}$ (mole fraction), and the mass-spectrometric sensitivity for air samples is $\sim 1 \times 10^{-16}$. Although an investigation of the properties of methanes is not planned for this experiment, heavy methanes should be considered as water tracers for migration tests where larger distances are involved and the greatest sensitivities are required.

e. Other tracers. Several other radionuclides have been used as water tracers. Some of these, such as ^{137}Cs and ^{198}Au , clearly sorb and are not useful for our application. The nuclides ^{51}Cr , ^{60}Co , and ^{152}Eu are useful tracers when treated with complexing agents like EDTA. However, it is necessary to use chelate concentrations³² greater than 10^{-6} M and, therefore, this type of tracer could not be used in experiments in which the sorption behavior of other species, which could become complexed, is to be measured.

Sugars¹⁶ and microorganisms, such as yeast,³³ bacteria,³⁴ and bacteriophage,³⁵ may be useful as tracers when minimal matrix diffusion is required.

There has been work³⁶ on the use of sodium thiocyanate, sodium benzoate, pentafluorobenzoic acid (PFB), and metatrifluoro-methylbenzoic acid. This work will be followed.

2. Choice of Water Movement Tracers. The first choice for tracer is, of course, tritium as tritiated water. In the Cambic experiment³⁰ a factor of $\sim 10^8$ dilution was observed at the sampling well. The use of deuterated water as an inactive substitute will be explored; however, the limitation on dilution will be a factor of 100 to 500. A possible problem is the necessity of injecting 100% D_2O which may interact with the system under investigation differently than normal H_2O .

A property of tracers that has clearly not been studied is the non-interference in sorption and transport process, and the most promising tracers will be evaluated in this regard. Laboratory experiments will be performed comparing migration of the sorbing nuclides used in the experiment with and

without the tracer of interest. The easiest experiments will be with identical columns of crushed material using tritium as the water tracer for one column and the tracer under investigation for the other. Both columns will also have sorbing nuclides.

The use of iodide, bromide, or chlorofluorocarbons will be investigated initially. Iodide or bromide are simple to handle and detect; however, they have to be used at a higher concentration than the chlorofluorocarbons and may be more likely to cause chemical problems. Volatile losses of the chlorofluorocarbons will be the biggest problem. Selected dyes will be the third candidate.

No problems in regard to the LASL capability to conduct the tracer investigation are envisioned. Tritium is counted routinely by a scintillation method. UV/visible and infrared spectrophotometers, fluorimeters, an ion chromatograph, and gas and liquid chromatographs are available.

H. Bench-Scale Experiments

In addition to the small-scale fracture flow experiments described in Sec. IV.L, several nuclide migration experiments will be performed using larger cores or unstressed blocks, some of which will contain a single natural fracture. The removal of these cores or blocks from their original location is discussed in Sec. IV.B. Increasingly larger cores or blocks will be used in these investigations in order to develop the experimental techniques and to assess the effect of increasing size. The largest sample (containing a path length up to 1 m long) will only be used in the final stages of this task. Here, we can only present our initial experiment design. The final design will evolve as the project progresses.

Nuclide migration experiments involving flow through the fracture in these larger-size laboratory experiments will be performed using the selected groundwater (Sec. IV.C), water movement tracers (Sec. IV.G), and stable and/or radioactive tracers (Sec. IV.D). The equipment to be used in the core studies may be similar to that described in Sec. IV.L. Another possible layout for these experiments is given in Fig. 6, which is based on a design given in Ref. 37. By sealing both sides of the fracture along the length of the block and keeping both ends open, a straight flow field can be established. A groove across the middle of the fracture plane provides for even distribution of water across the width of the fracture. A small hole drilled into this groove from below provides a well-defined tracer injection point. In effect,

GROUNDWATER + TRACER (POINT)

Fig. 6. Possible Configuration of a Block Flow Experiment.

this will provide two simultaneous migration experiments. Some development work must be done in order to have the tracer enter the flow path at the proper point. The water and tracer injection systems (Sec. IV.J) and water collection system (Sec. IV.K) would be used in later experiments in order to check the equipment. However, simpler laboratory techniques will be used in the initial studies. The groundwater will be injected for some time in order to assure saturation of the rock. The elements to be studied will then be injected from a single point (Fig. 6). Cores or blocks containing artificially produced fractures or natural fractures will be used. The water flow rate through the fracture will be controlled so that the residence time of water in the fracture is similar to that expected in the field experiments (Secs. IV.N and IV.O) can be used. Later experiments will allow the effect of flow rate, and thereby the effect of reaction kinetics (Sec. IV.L), to be investigated. These experiments also allow careful control of the critical chemical factors (e.g., oxygen concentration) involved.

Obviously, the principal value of these larger-scale experiments is that they are an attempt to simulate the field experiments before the latter are performed. This should be very beneficial for planning the field experiments since one, hopefully, will know the approximate tracer migration rates to be expected. Therefore, the field experiments can be optimized to yield maximum information.

The controlling factor for flow in open fractures is the size of the aperture,³⁷ and the flow per unit head depends on the cube of the aperture. Therefore, these experiments can also be used to investigate the dependence of the radionuclide migration rate on flow rate by changing the pressure applied normal to the fracture. This will change the local internal flow paths and flow rates and will emphasize any kinetic effects in the rock-water interactions.

These experiments are also necessary in order to develop the post-experiment analysis techniques (Sec. IV.I). It is not entirely clear which are the optimum methods to determine the spatial distribution of the migrating elements along the flow path. Several different bench-scale tests as simulation of the field experiments will allow us to test several analytical techniques and select the best.

The predictive capabilities of the various models developed as part of the NWTs (Sec. VI) will be applied to these experiments if directives are

given to these NWTs participants by ONWI. Since the post-experiment analysis methods will be used on the blocks, and the chemical interaction kinetics, mechanisms, and parameters will be known from the laboratory work (Sec. IV.L), comparison of the predictions with the results from these experiments will allow any deficiencies in the models to be identified. This will allow the applicable NWTs models to be updated prior to the field experiments.

It is not clear that we can determine prior to the field experiments whether or not data obtained using the batch or small column sorption techniques (Sec. IV.L) are satisfactory for use in models of real field conditions. However, the results from the bench-scale experiments will indicate whether a simple extrapolation from small cores to larger systems can be made.

Finally, the new large block technique, when combined with post-experiment analyses, may be very useful in assessing the geochemical properties of a variety of rock types since it is more representative of the actual rock of interest; the block will have been subjected to much less mechanical alteration than material used in the batch or small core measurements (Sec. IV.L).

I. Analytical Methods and Flow Path Analysis

1. Tracer Detection Along the Flow Path. There are many methods that possibly may be used to detect the location of the non-conservative (sorbing) elements along the flow path. These include radiation mapping and autoradiography, high-resolution radiation-detection methods, chemical dissolution and high-resolution mass-spectrometry, DC-coupled argon-plasma emission spectrometry, instrumental neutron activation analysis, or atomic absorption spectrophotometry, X-ray fluorescence produced by X-rays, electrons (electron microprobe) or protons, and spark-source mass spectrometry. All of these detection methods, and several others, are already available at LASL, SNL, and ANL. The actual method to be used depends upon the element, its form (stable or radioactive), and the detection sensitivity required.

When radioactive tracers are used, autoradiographic techniques or radiation mapping using a well-collimated detector may be sufficient to locate the position of the tracers along the flow path. The actual identification of individual isotopes would be made by high-resolution, gamma-ray spectrometry.

More precise determination of the actual location of the tracers along the flow path may involve removing small samples for analysis. If these small samples are dissolved then they could be analysed by high-resolution mass

spectrometry, instrumental neutron activation analysis, or DC-coupled argon-plasma emission spectrometry. If isotopic enrichment is not involved, there are many means for sensitive elemental analysis of the elements. For example, X-ray fluorescence induced by X-rays, electrons (electron microprobe) or protons can be used. In each of these methods, the backgrounds may be the limiting factor rather than the sensitivities.

A useful technique that lends itself to analysis of many elements for screening purposes is spark-source mass spectrometry. It can detect as little as 10-100 ppb of most elements in milligram samples. It provides multi-element and isotopic analysis and also allows the use of enriched isotopic tracers which can be distinguished from the indigenous isotopes of the same element. (The dilution of an isotope by exchange with the same element in the substrate would be a new type of retardation effect that can affect transport modeling.) If the precision required is not achieved by the spark-source mass spectrometric technique, then classical mass spectrometric procedures could be used on selected areas.

As mentioned earlier (Sec. IV.H), development of the optimum detection methods would be done using the cores and block from the bench-scale tests. A comparison of sensitivities of some of the possible methods is given in Table II.

2. Tracer Detection in the Eluent. The analytical procedures to be used for detection of the non-conservative (sorbing) tracers that may be transported through the fracture depend upon the actual elements selected for study (Sec. IV.D). Different procedures will also be used if radioactive tracers are employed. Obviously, the same suite of analytical methods discussed for detection of the tracers along the flow path (Sec. IV.I.1) are available. The decision on the analytical method will follow careful consideration of the alternatives.

3. Water Flow Path Tracers. It would be most beneficial to the field experiments if a tracer were found which could be used to locate the actual flow paths through the rock. This tracer should sorb slightly on the geologic medium but not otherwise interfere with the transport retardation processes. Therefore, it should be easily and efficiently detectable so that low concentrations can be used. It will be detected during the post-experiment analysis (Secs. IV.I, IV.N, and IV.O) for each field experiment.

The most likely candidates are fluorescent organic dyes,²⁷ which could be injected during the final stages of the experiment.

TABLE II
SENSITIVITY OF VARIOUS ANALYTICAL METHODS

Element ^a	Neutron ^b Activation Analysis (ppm)	Copper Spark (ppm)	Spark ^c Source Mass Spec. (ppb)	Fluorescence ^d			Resonance Photon ($\mu\text{g}/\text{cm}^2$)
				X-ray	Electron ($\mu\text{g}/\text{cm}^2$)	Proton	
Cs	.0015	0.5	10	1.0	1.0	0.05	0.001
Sr	.03	0.5	10	1.0	1.0	0.05	0.001
La	.001	0.05	10	1.0	1.0	0.05	0.001
U	.0005	1.0	10	1.0	1.0	0.05	0.001

^aThe list is intended only to characterize the range and not to specify or recommend any elemental selection method.

^bFrom Ref. 38; reactor flux of 10^{13} n/cm² sec.

^cFrom Ref. 39.

^dFluorescence sensitivities are uniform for elements of $Z > 20$. Sensitivities obtained from Refs. 40 and 41.

Bromide or iodide tracers could also be used in the final stages of each of the field experiments. Radiation detection techniques could be used as the detection method.

An interesting possibility is to inject ²²²Rn (3.8 d half-life), dissolved in the groundwater, during the final stages of the experiment. The ²²²Rn would decay during the time required for transport through the fissure and the daughter products (²¹⁰Pb, ²¹⁰Bi, ²¹⁰Po) should sorb strongly on the tuff. Autoradiographic or radiation mapping techniques could be used to define the pathways. Estimates are that only a few millicuries of ²²²Rn would be required. The radon is easily collected from a radium cow by use of the stearate emanation method.⁴²

The data available in the literature will be evaluated in order to select possibilities, and an experimental program will be performed in conjunction with development of water movement tracers (Sec. IV.G).

4. Materials Characterization. The appropriate supporting information for all of the materials used in the laboratory and field studies must be gathered to be able to interpret the measurements (Sec. IV.P). Detailed chemical, mineralogical, and geochemical analyses must be performed on the solid

materials, both the matrix and the fracture filling material. As discussed above, the chemical characterization of the reactant solutions must also be made.

These materials, will be analyzed by a variety of techniques to obtain major, minor, and trace element composition, mineralogy/petrology, pH, carbonate/bicarbonate ratio, Eh, etc. Many of the techniques to be employed have already been discussed (Sec. IV.I.1).

Detailed mineralogy studies will be done. The available analytical equipment includes research petrographic microscopes, a fully automated Cameca electron microprobe, an automated Siemens powder X-ray diffractometer, and a scanning electron microscope.

Other properties to be measured on the solid phase include surface area (gas adsorption and ethylene glycol), exchange capacity, matrix porosity, matrix permeability, etc.

The groundwater composition will be measured during the course of all measurements. In particular, the Fe(II) concentration or that of other possible indicators of redox conditions will be closely monitored.

The total organic carbon concentration in the groundwater and solid will be obtained by J. L. Means, Battelle Columbus Laboratory.

J. Water and Tracer Injection System (WATIS)

Several specifications for the water and tracer injection system (WATIS) have been currently defined. This has been done without the benefit of the detailed site characterization (in particular, the permeability testing) so it is highly likely that they will undergo some refinement in the future. Nevertheless, conceptual design is now underway based on the specifications which follow.

- 1) The WATIS will allow control of the water chemistry (e.g., Eh, CO_2). This requires that the WATIS be fabricated with inert materials (stainless steel, Teflon, silicone RTV, etc.) and be air tight. Sorption of tracers on WATIS materials must be very small.

- 2) The WATIS should allow sheet flow in the fracture to be established. This specification may be controlled more by the experimental hole array and fracture orientation and aperture than by the WATIS design. However, the WATIS should be designed to minimize as much as possible the pressure gradients along the injection line into the fracture.

3) The tracers should be injected into the fracture at the midpoint of the leading edge of sheet flow; it may be more practical to inject the tracers somewhat downstream from the water injection area. In either case, the primary goal is to provide "single point" tracer injection in a minimum volume of groundwater.

4) The WATIS will allow for sequential injection of specific radio-nuclides at predetermined times during the experiment.

5) The WATIS must have leak-free packers. This is especially important when using actinides. This specification suggests that the packers be chemically sealed to the rock walls of the holes (e.g., use two-component silicone RTV).

6) The WATIS will accomodate down-hole instrumentation. Such down-hole instrumentation may include thermocouples, pressure transducers, Eh and pH probes, and radiation detectors. The ability of these devices to operate in the field will be evaluated during the conceptual design.

K. Water Collection and Multipoint Sampling System (WCAMSS)

Water collection holes will be located downstream to collect the water as it emerges from the fracture. The number of collection holes depends on the fracture orientation, one to about six for a vertical fracture and one for a horizontal fracture. The following initial specifications have been established for the water collecting and multipoint sampling system (WCAMPSS).

1) The WCAMPSS shall be designed to collect and contain the water emerging from the fracture. The WCAMPSS will also contain a means of removing the water from the hole for storage and chemical analysis. The current concept encompasses a packer system; however, depending on the inclination and quality of the holes, all the packers may not need to be water tight.

2) The WCAMPSS should include the necessary instruments. The possibility of including Eh and pH probes, thermocouples, and a water depth gauge in the pooling region is currently being evaluated. The depth gauges(s) will be a measure of water collection and when water removal is necessary.

3) The WCAMPSS will provide for point monitoring at three to five locations along the fracture. One method is to use small radiation detectors to provide on-line observation of the emergence of radioactive nuclides. Perhaps a more general sampling method is remote insertion and withdrawal of small water collectors (e.g., inert sponges) at specific locations along the

fracture. The water collected can then be analyzed for both chemical and radioactive tracers. The most useful system may include both detectors and remote water samplers.

As before, these specifications are subject to additions and modifications as knowledge of the experimental site and needs of the experiments improve.

L. Chemical Interactions

The objectives of this work element are to try to: 1) identify the chemical phenomena controlling radionuclide transport in the fracture and surrounding rock; 2) analyze and mathematically describe those phenomena in terms of fundamental processes (such as diffusion, first-order chemical reactions, etc.) and the associated parameters; and 3) identify parameter evaluation techniques so that the chemical parameters controlling radionuclide transport in field environments can be related to specific laboratory measurements.

In order to achieve the above objectives, the mechanisms, kinetics, and equilibria for the following types of phenomena will be investigated: 1) sorption and desorption of radionuclides by the solid phase (rock) and associated stagnant interstitial solution; 2) homogeneous chemical reactions with constituents of the mobile solution (groundwater) and 3) heterogeneous reactions with constituents of the immobile phase or with particulate matter suspended in the solution.

Since transport of radionuclides relative to the motion of the groundwater will be retarded by interactions with the rock, the homogeneous and heterogeneous chemical reactions to be identified and studied are those which can enhance or degrade the retardation effects. Furthermore, these are the types of reactions which must be investigated in order to define the radionuclide source term for flow experiments in both the laboratory and field. Such reactions include those which produce polymers, colloids, precipitates, changes in the oxidation states of the nuclides, or complexes with naturally-occurring or man-introduced organic or inorganic constituents of the mobile or immobile phases. Accurate values are needed for the equilibrium constants for all reactants and products in solution and on the solid. The mechanisms include ion exchange and ion exclusion processes, diffusion into the pores of the rock, replacement reactions, physical transport and filtration, and

precipitation processes. Critical groundwater chemistry parameters, such as pH and oxidation-reduction potential, that affect these reactions must also be identified.

The general investigative approach is as follows: Pertinent sorption mechanisms and chemical reactions will be tentatively identified based on the literature (e.g., Refs. 10, 21, and 43), chemical analysis of the groundwater (Sec. IV.C), and physical, chemical, and mineralogical analyses of samples from selected fracture surfaces and of samples from the surrounding bulk rock (Sec. IV.B and IV.I).

The behavior of the elements in the groundwater selected for use (Sec. IV.D) in the laboratory and field experiments will be studied in some detail. Methods must be developed for injecting these elements into the groundwater (Sec. IV.C) in a well defined oxidation state and chemical form. This is particularly important for the actinides which frequently have multiple oxidation states and a strong tendency to form polymers and colloids.^{44,45} A non-radioactive pseudocolloid will possibly be used in the early studies; use of this material is currently being developed in Sweden.¹¹

Sorption capacities and equilibrium data will be determined from batch equilibration experiments in which samples of the immobile phase are contacted with groundwater containing selected nuclides. Samples from the fracture surfaces and from the surrounding bulk rock will both be studied. The effects of possible competing chemical reactions will be analyzed using available thermodynamic and kinetic data, and, as necessary, additional experiments will be identified and performed in order to more clearly define the sorption phenomena and any competing chemical reactions. Batch and small column techniques will be used. The effects of pH (carbon dioxide concentration), oxidation-reduction potential and oxygen concentration, element concentration (sorption isotherm), and time, among others, will be addressed. Microautoradiographic techniques⁴⁶ will be used to identify the specific minerals responsible for removal of the element from solution and/or the state of aggregation of the element. All materials will be characterized in detail described in Sec. IV.I in order to have the necessary supporting information for the analysis of these studies (Sec. IV.P).

The rate of removal of the elements from solution will be obtained from additional batch and column measurements using crushed material, tablet-like

rock samples, and small rock-core samples containing artificial or natural fractures. The final distribution of the elements on the tablets and whole-rock cores will be examined using the methods described in Sec. IV.I.

Since the effect of the unconnected porosity (i.e., low permeability) of the nonwelded tuff on the nuclide migration rates and spatial distribution of the solutes in the rock matrix may be significant, studies of diffusion will be made. The experimental work required to evaluate the importance of diffusion in tuff involves several activities. The porosity and pore size distribution must be determined as must the electro-osmotic pressure and a measure of the tortuosity of the porous matrix. Several chemical properties of the non-radioactive water movement tracers (IV.G) must also be obtained. These include the ionic conductance, activity coefficient at various ionic strengths, and the equilibrium coefficients for the various ion association and complexation reactions. These will be measured using relatively standard techniques, after which experiments specifically designed to address diffusion will be performed.⁴⁶ The effect of pressure on the extent of diffusion into the tuff may also be investigated.

The sorption-desorption processes and any other pertinent chemical reactions will then be described mathematically using expressions which represent the important features of the fundamental processes (see Refs. 47-51, for example), but which can also be incorporated into available mathematical models (or modifications thereof; Sec. VI) which will be used to analyze radionuclide transport in the fracture and surrounding rock. These will include the results from sorption equilibrium and rate experiments, post-experiment examinations, and material characterizations such as pore size distributions and surface area (Sec. IV.I).

Small-scale radionuclide transport experiments will be conducted using rock-core samples containing natural or artificial fractures and systems already developed (Figs. 7 and 8, for example). The cores can be stressed in systems like that shown schematically in Fig. 7. Realistic groundwater flow rates (<50 m/yr.) will be used, and the source term will be a step or square-wave change in the radionuclide concentration in the injected groundwater. The water movement tracers and post-experiment analysis methods developed in Secs. IV.G and IV.I, respectively, will be used. This will allow transport and retardation resulting from flow through fractures and through the rock matrix to be studied in detail.

Fig. 7. Schematic of the High-Pressure Column Apparatus Having a Confining Pressure on the Rock Sample.

Fig. 8. Schematic of the Column Apparatus Having No Confining Pressure on the Rock Sample.

Laboratory radionuclide transport experiments on a larger scale are described in Sec. IV.H.

This work element, when combined with the others identified above, is a principal input to Sec. IV.P, "Experiment Analysis," and defines and evaluates many of the expressions and parameters which must be included in the mathematical models in order to predict system behavior and analyze experimental results.

M. Laboratory and Field Fracture Flow Tests

Prototypes of the WATIS and WCAMPSS will be tested in transparent pipes in the laboratory. A more elaborate system using a full-scale artificial fracture fabricated from clear lucite is being evaluated. In this scheme, water containing dyes would be used to observe the behavior of the total system.

Following successful operation in the laboratory, a limited, near-wall fracture flow test is planned in the field to test the system emplacement and to develop operating procedures for the "cold" experiment (Sec. IV.N). A limited mine back is planned for evaluation of both the operation of the system and flow-path detection procedures (Sec. IV.I). The mine/core back operation will also help develop the procedures for similar operation in the migration experiments (Secs. IV.N and IV.O). Based on the results of these tests, final design changes, if any, will be made to the injection and collection systems and fabrication of the units for the "cold" experiment will commence.

N. First ("Cold") Field Experiment

A full-scale mock-up of the radionuclide migration experiment will be performed in the selected site in G-tunnel (Sec. IV.A) using stable tracers (Sec. IV.D). It is possible that short-lived radionuclides can also be used if the mining and coring techniques (Sec. IV.B) are considered safe. This experiment will test the experimental design and the safe handling procedures developed for the use of radioactive tracers. The secondary water movement tracer, developed earlier (Sec. IV.G), will be used. Tritrated water may also be employed as a check on the behavior of the secondary water movement tracer. If the technology for studying colloid migration is sufficiently well developed (Sec. IV.L), then studies of this type will also be performed. Post-experiment analysis of the flow path (Sec. IV.L) will be done. The

results will be compared with models, which will be updated as necessary (Sec. IV.P).

Prior to initiation of the field experiment, a detailed engineering test plan will be written. The parameters for this test will have been developed earlier. Many of the methods and procedures used to perform the experiment will have been developed and documented in the earlier stages of the project: site selection (Sec. IV.A); detailed site characterization (Sec. IV.B); groundwater characterization and selection (Sec. IV.C); isotope selection (Sec. IV.D); construction of the experimental area and development of instrumentation (Sec. IV.F); development of water and tracer injection systems (Sec. IV.J); development of the multipoint water sampling system (Sec. IV.K); development of the methods for post-experiment analysis of the flow path; and optimization of the requirements from the chemical interaction studies (Sec. IV.L).

The actual configuration of the migration experiment cannot be given until the site is selected and detailed characterization is completed. Possible configurations for experiments using a horizontal or a vertical flow path are given in Figs. 9 and 10, respectively. The maximum length of the flow path will be less than 1.5 m. Prior to injection of the tracers, groundwater will be injected for some time in order to fully saturate the rock and establish steady state flow. The elements to be studied will then be injected from a single point since this will allow data to be obtained on dispersion along the flow path.

Of primary concern is the necessity for control of the water residence time in the fracture so that rates reasonably representative of natural systems can be utilized. The flow rate will be optimized so that maximum information is obtained from the experiment. The need to control the time in the field measurements is due to the possibility of having kinetic effects.⁵²⁻⁵⁴ That is, the kinetics of the adsorption and desorption processes may be important if the flow rates are too high. Obviously, this is also true in the laboratory studies (Sec. IV.L) and the kinetic information will be obtained using laboratory techniques. It has been shown⁵⁵ that kinetic effects are not important if the time for solvent flow over the experiment flow path is significantly greater than the half-time for sorption. In essence, this means that for a real repository, where far-field flow paths are several kilometers long and flow rates are less than 100 m/y, kinetic effects will

Fig. 9. Possible Experiment Configuration for a Horizontal Flow Path.

Fig. 10. Possible Experiment Configuration for a Vertical Flow Path.

probably be neglectable. However, in these experiments, one wishes to have a minimal contribution from kinetics (i.e., reasonably chromatographic behavior) since it will be much simpler to interpret the field experiments if significant kinetic effects are absent.

In order to gain information in a single experiment on a variety of elements having greatly differing retardation properties, "staged" injections of water movement tracer and migrating element(s) may be used. That is, several injections of groundwater containing one or more tracers would be made at different times during the course of the experiment. The earliest injections would be for those elements with the lowest migration rates, while later injections would be for the elements having higher migration rates. The concentration of the migrating element in the groundwater will always be significantly below the solubility limit in that water. The length of the injection period will be optimized in the laboratory studies. The migration rate information used to select the sequence of injection would also be obtained from the laboratory studies. This procedure would localize most of the migrating elements somewhere along the flow path and would give the maximum time for migration of those elements with strong retardation characteristics. A mass balance would be attempted for all tracers used in the study.

The total duration of the experiment, from initiation to termination of groundwater flow, will be about 18 weeks. This will then be followed by application of the post-experiment analysis methods (Sec. IV.I) to the actual flow path, which will be removed using techniques described in Sec. IV.B.

The results from this experiment will then be interpreted as described in Sec. IV.P.

0. Second ("hot") Field Experiment

Since the transport of actinides by groundwater is of major concern, a field experiment will be performed to determine their potential for migration under natural conditions. This experiment will also address the reproducibility of field experiments since some of the same elements used in the "cold" test (Sec. IV.N) will be used.

The same basic operations will be used for this experiment as for the "cold" experiment (Sec. IV.N). Based on information from the "cold" experiment, the earlier experimental plan will be revised as necessary in preparation for the first "hot", actinide containing, experiment. The water movement tracer will be either tritiated water (HTO) or the secondary water movement

tracer (Sec. IV.G) used earlier. Generally, the same elements (now radioactive) utilized in the "cold" test will be used with the addition of several actinides (Sec. IV.D). This experiment will be carried out in a new area at the same site. After removal of the residual workings of the "cold" experiment (Sec. IV.N), a portion of the same fracture farther in from the tunnel wall or farther down in the same drift will be used.

The duration of the flow portion of the experiment will be about 22 weeks, the length of the flow path will be less than 1.5 m, the post-test analysis of the flow path will be done, and comparison of the results with state-of-the-art models will be made. These models will have been updated by the results from the "cold" (Sec. IV.N) test. The applicability of these models to actinide transport will then be addressed.

P. Experiment Analysis

The objective of this work element is to identify, assemble, and implement the mathematical analyses required to: 1) interpret the results from the laboratory experiments (Sec. IV.L); 2) provide residence-time-distribution analyses of water movement tracers in order to define migration paths (Sec. IV.L, IV.N, and IV.O); 3) conduct pretest radionuclide transport analyses as required for experiment design (Sec. IV.N, IV.O, and VI); and 4) interpret results from the bench-scale experiments (Sec. IV.H) and from the "cold" (Sec. IV.N) and "hot" (Sec. IV.O) field experiments, and identify deficiencies in current radionuclide transport models (Sec. VI). Currently available experimental techniques, methods of data analysis, and radionuclide transport models (Sec. VI), or reasonable modifications thereof, will be utilized whenever possible. Available literature data will also be employed. The principal types of analyses which must be implemented in order to achieve the above objective are discussed below.

Equations are required which describe the batch, tablet, and whole-core radionuclide sorption and chemical reaction rate studies (Sec. IV.L). As necessary, such equations will include terms for sorption and reaction equilibria and mechanisms, molecular diffusion into the immobile phase (rock), and kinetic expressions for homogeneous and heterogeneous chemical reactions such as those producing polymers, colloids, precipitates, changes in oxidation states, and complexes with constituents of the rock and groundwater. The equations describing transport of radionuclides as particulates suspended in

the groundwater must be included. The particulates could be either radiocolloids or naturally occurring particles which sorb radionuclides.

Equations are required which describe the residence-time distribution of water movement tracers. These would include equations for ideal flow systems and various solute source terms (such as step increase, square wave, etc.). Since "ideal" fractures will not be used in the majority of the studies, particularly in the field, the appropriate equations will be modified to account for the non-ideal conditions (e.g., Ref. 37).

Comprehensive models are required which describe radionuclide transport in the selected fractures and surrounding rock. Such models must incorporate expressions describing the pertinent chemical phenomena controlling radionuclide transport as described above and, as appropriate, expressions reflecting the results of the residence-time-distribution analyses for the water movement tracers. Existing techniques will be used to interpret the results from the bench-scale (Sec. IV.H), "cold" (Sec. IV.N) and "hot" (Sec. IV.O) experiments and will also be used for pretest analyses in order to determine the experimental conditions (flow rate, radionuclide concentrations etc.) to be used in those experiments. The chemical parameters dominating radionuclide transport models will be evaluated using methods such that the parameter values are directly related to specific laboratory measurements (Sec. IV.L) which describe fundamental processes (such as diffusion, first-order homogeneous chemical reactions, etc.).⁴⁷⁻⁵⁶

The data from the bench-scale and field experiments will be compared with analogous data calculated using the available mathematical models. The adequacy of the models and laboratory techniques used to evaluate parameters will thereby be assessed. Refinements to the models and laboratory techniques will be made as necessary, and parameter values will be continuously refined using established parameter estimation techniques (e.g., Ref. 56).

The field experiments and accompanying analyses will provide the data and means to evaluate the adequacy of NWTs models under realistic conditions and, where appropriate, will provide a supplementary data capability. The adequacy of the available data base of chemical parameters will also be examined.

The data from the field experiments will be made available on a timely basis to the NWTs modeling community (Sec. VI). This will allow independent interpretation of the experimental results.

V. TECHNICAL APPROACH

The organization of all of the various program activities described in Sec. IV into a logical sequence of interrelated events needed to achieve the project objectives is shown in detail in Figs. 11-13. These detailed logic networks identify the estimated time (in weeks) for a given activity and show the interrelationships between the work elements. A description of the elements on the network charts is given in Appendix C. The time estimates are not commitments but do reflect our current projections of the time required. Of necessity, the time estimates are less precise when they are far in the future. The major milestones are summarized in Sec. VII. The logic networks will be updated periodically as more accurate projections become available.

As indicated, peer reviews will be held, at a minimum, at the major critical points in the project. Indeed, the time for the review process seems to be a major factor in the length of the project. The impact of these peer reviews is discussed in Sec. X.

VI. PROGRAM INTERFACES

The field experiments described in this program plan are an integral part of the national (NWTs) program to describe nuclide transport in geologic systems. As such, the knowledge gained by this effort must interface with existing NWTs programs. A diagram of these interfaces is given in Fig. 14.

When applicable, the laboratory methods developed by the Waste-Rock Interactions Technology (WRIT) program at Battelle Pacific Northwest Laboratories (BPNL) under R. J. Serne will be integrated with this project. This is already being accomplished since Los Alamos is a WRIT subcontractor. Developments in experimental techniques and transport phenomena will be transmitted to the WRIT program. The field experiment effort will also define some measurement needs which must be factored into the WRIT program. Samples of the material from G-tunnel will be sent to the WRIT project for use in that program.

The other major interface to NWTs efforts is to the Waste Isolation Performance Assessment Program (WIPAP) under H. C. Burkholder at ONWI. The principal goal of this program is to evaluate the performance of geologic repository concepts, primarily by modeling efforts. The three WIPAP modeling subelements are 1) the Systematic Comprehensive Evaluation of Performance and Total Effectiveness of Repositories (SCEPTER), 2) the Assessment of the Effectiveness of Geologic Isolation Systems (AEGIS), and 3) a program at the

Fig. 14. Interfaces to NWTIS Program in Nuclide Transport.

University of California at Berkeley (UCB). These programs are being performed under the direction of the following:

SCEPTER	R. B. Lantz	(Intera, Inc.)
AEGIS	F. H. Dove	(BPNL)
UCB	T. H. Pigford	(UCB)

The modeling efforts being conducted as part of WIPAP will be integrated with this field experiment project. The applicable WIPAP models will be used to predict the experimental results for the bench-scale experiments and field experiments, provided that the directives for these calculations are given

from ONWI/WIPAP. Sufficient funds are not available in this field experiment project to support calculational activities by WIPAP participants. It does seem, however, that few of the WIPAP models currently include elements that describe the chemistry that occurs when radionuclides are introduced into a groundwater-geologic media environment. Also these models are primarily regional in nature and thus may not be applicable for predicting transport in geometrically simple, single fractures. The possible model deficiencies identified in the experimental program will be communicated to the WIPAP participants, as will possible methods of correction. These interactions should create the necessary dialogue between experimenters and modelers.

This project will also be coordinated with the studies being carried out in the NNWSI project. Extensive studies of the properties of tuff in other areas at the NTS which may have bearing on the field studies are being obtained in the NNWSI. The Los Alamos and SNL are already involved in the NNWSI.

As indicated in Sec. IV.I, J. L. Means at the Battelle Columbus Laboratory, Columbus, Ohio, will perform analyses of the total organic content of the groundwater and solids used in the studies.

As indicated in Fig. 14, a nuclide migration field test in granite is being performed by the Lawrence Livermore National Laboratory (LLNL). Since Los Alamos, SNL, and LLL are all participants in the NNWSI and both the field projects are considered tasks within the NNWSI, the Nevada project will serve as the formal mechanism for cooperation and exchange of information between the two efforts.

VII. SCHEDULE/MAJOR MILESTONES

The key milestones of this project are described in Table III. From the start of the project to completion of the experiments, the critical path, based on current projections, may be summarized as follows (assuming minimal field or laboratory work in FY-1980):

Technology development and laboratory studies	19 months
"Cold" Field Experiment	16 months
"Hot" Field Experiment	13 Months
Project Summary	9 Months

These estimates include the time for the review process and for preparation of documents.

TABLE III
MAJOR MILESTONE DESCRIPTION

<u>Number</u>	<u>Date</u>	<u>Node^a</u>	<u>Title</u>
1	5/16/80	8320	Availability of Funds.
2	8/80	8340	Complete the Peer Review of the Draft Program Plan.
3	9/80	8314	Transmit the Project Quality Assurance Plans to ONWI.
4	9/80	8355	Transmit the Final Program Plan to NV and ONWI.
5	6/81	8446	Complete the Summary Site Selection Letter Report.
6	5/82	8525	Complete the Experimental Test Plan for the "Cold" Field Experiment.
7	5/82	8527	Complete the Peer Review of Experiment Plan for the "Cold" Experiment.
8	9/83	8570	Complete the "Cold" Experiment Draft Report.
9	9/83	8565	Complete the Revision of the Experimental Plan for the "Hot" Experiment.
10	10/83	8567	Complete the Peer Review of the Revised Experimental Plan for the "Hot" Experiment.
11	3/85	8605	Complete Preparation of Comprehensive Nuclide Migration Field Experiments Report.

^aSee Sec. VII, Figs. 11-13.

VIII. BUDGET

The funding from the Department of Energy, Office of Waste Isolation, needed to achieve the goals of this program according to the schedule given in Sec. VI is given in Table IV by fiscal year (FY) and by organization. The values include an 8% per year inflation factor. The funding level given in Table IV has little contingency for uncertainties and a major uncertainty is the true inflation rate. The actual schedule of the various activities is also

TABLE IV
ESTIMATED BUDGET REQUIREMENTS (\$1000)

<u>FY</u>	<u>TYPE^a</u>	<u>LASL</u>	<u>SNL</u>	<u>ANL</u>	<u>NTSSO</u>	<u>Total</u>
1980	O	79	80	30	61	250
	C	0	0	0	0	0
1981	O	326	507	231	158	1222
	C	195	213	94	0	502
1982	O	459	586	224	191	1460
	C	100	100	22	0	222
1983	O	486	558	185	140	1369
	C	100	100	25	0	225
1984	O	508	550	213	163	1434
	C	0	0	0	0	0
1985	O	30	15	15	0	60
	C	0	0	0	0	0

^aO indicates operating costs; C indicates capital equipment costs.

rather uncertain since the schedule (Sec. V) goes so far into the future. Also, events beyond our control may make the availability of support personnel and equipment uncertain due to programmatic commitments at the NTS. Unknown difficulties in the field operations themselves can severely impact program activities.

Included in the budget given in Table IV is about \$50K per year for Quality Assurance (QA). This low figure comes about because existing QA Program Plans have been developed and implemented as part of the NNWSI; the NNWSI QA plans will be used for this project.

A listing of the capital equipment needs that have been currently identified for this project for FY-1981 is given in Table V. Additional items undoubtedly will be needed as the project develops. The items of equipment listed are especially important since they will be required throughout the entire course of this program and must be ordered early because of long delivery times. The ability to meet the projected milestones depends critically on timely acquisition of this equipment.

IX. TECHNICAL CONTROL

This program will be treated as a generic task within the NNWSI project of the Nevada Operations (NV) Office of the Department of Energy (DOE). The

TABLE V
FY-1981 CAPITAL EQUIPMENT NEEDS

<u>Los Alamos Scientific Laboratory</u>		<u>Estimated Cost</u> <u>(\$1000)</u>
1.	Radioactive flow monitor	10
2.	Process controller	20
3.	Controlled-atmosphere enclosure and accessories	30
4.	Gas chromatograph and electron capture detector	20
5.	Water preconcentrator	10
6.	Solvent delivery and pressure pumps	20
7.	UV/VIS spectrophotometer	20
8.	Data reduction equipment	20
9.	Pulse-height analyzer and electronics	25
10.	Vibrating-reed electrometers	10
11.	Hall probe - Gauss meter	10
Subtotal		195
<u>Sandia National Laboratory</u>		
1.	Flow meter and electronics	2
2.	Centrifugal pump, valves, etc.	5
3.	Pulse-height analyzer	15
4.	RTV packers (design & manufacture 20 packers)	100
5.	Injection packers (design & manufacture 3 packers)	30
6.	Flow meters (15)	8
7.	pH probes	3
8.	Eh probes (developmental)	20
9.	Data acquisition system additions and upgrading	30
Subtotal		213
<u>Argonne National Laboratory</u>		
1.	Cyclotron target holder assemblies	25
2.	Vacuum centrifugal concentrators	6
3.	High-pressure, controlled volume pumps	3
4.	Electronic modules	4
5.	Gas chromatograph	8
6.	Pressurized column holders	5
7.	Computer interface	15
8.	Remote processing equipment	10
9.	Process monitoring and fluorescence analysis electronics	15
10.	Miscellaneous	3
Subtotal		94
Total		502

Los Alamos will be the project coordinator and will represent ANL in the NNWSI program. R. M. Nelson, director of the Waste Management Program Office (WMPO), is the principal contact at NV.

The actual technical direction of the program will be done through a Steering Committee which has representatives from the three laboratories involved. All laboratories have equal responsibility. The members of this committee are:

Los Alamos	Bruce R. Erdal
SNL	J. Keith Johnstone
ANL	Arnold M. Friedman

The Los Alamos is the principal contact and spokesman for this effort and will coordinate the efforts. The steering committee is also responsible for review of technical documents prior to delivery to technical review system at each laboratory.

Quality Assurance (QA) procedures and practices will be followed in the execution of this project. The QA Program Plan (QAPP) developed at LASL and SNL for the NNWSI project will be used for this project. The ANL QAPP is similar. All these plans will be forwarded for approval by WMPO/NV.

Weekly telephone reports of project activities and accomplishments by each of the laboratories will be made to LASL and these will then be transmitted to WMPO/NV. This will mean at least weekly communication between the project participants and discussion of project needs and activities.

The main areas of responsibility for the activities are:

All:	Field experiment design Laboratory studies Bench-scale studies Analytical support
Los Alamos:	Project coordination Materials characterization Water movement tracers Post-experiment analysis
SNL:	Field experiment operations Water and tracer injection system design and fabrication Water collection and sampling system design and fabrication
ANL:	Isotope production Flow path analysis methods

Obviously, certain aspects of the program must be carefully coordinated since similar capabilities and interests sometimes exist in each laboratory. The

actual organization performing a certain task will depend on the availability of manpower when the other work elements of the program are taken into account. Also, each organization can contribute to the various areas of responsibility mentioned above. They are not exclusive. All data transferred between project participants will be entered into the QA document control system. This will ensure traceability of the information through the QA procedures implemented in each organization.

The SNL will direct all field operations within G-tunnel.

X. PROGRAM CONTROL

Several decision points have been identified in the project (Secs. V and VIII). A peer review will be scheduled at these points. The peer reviewers will be selected by WMPO/NV, and programmatic reviewers will be selected by ONWI. These peer reviews will document any project deficiencies and will be the basis for any external recommendations for changes in the project definition. Approval of such changes will be made through WMPO/NV.

Two different reports will be routinely generated in addition to the weekly telephone reports mentioned in Sec. IX. A monthly status report will be delivered to WMPO/NV by the middle of the month following the close of the report month. This will also include the actual costs for the report month and a status report on the milestone schedule. Quarterly technical reports on the project accomplishments will also be issued. Topical reports will be issued as needed and as designated in the project milestone list (Sec. VII). Similarly, publications in the open literature and presentations at major technical meetings will be made when appropriate. All of these reports and documents will go through the technical review processes at each laboratory as described in the QA Program Plans at each laboratory. The WMPO/NV will also review them for policy purposes.

Three engineering test plans, as identified in the milestone list (Sec. VII), will be generated. A safety analysis document will be delivered to DOE/NV, as required by DOE Order 5481.1, prior to initiation of any field experiment employing radioactivity.

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APPENDIX A

GENERAL DESCRIPTION OF TUFF^a

Tuff is a geological term for rocks composed principally of compacted volcaniclastic material. Such rocks may be deposited either directly by explosive volcanic events, or they may be reworked and redeposited by surface processes. Because silicic (e.g., rhyolitic) volcanism tends to be more

^aTaken from Ref. 1.

explosive than the eruption of more mafic magmas, silicic tuff tends to be much more voluminous and widespread than basaltic tuff despite the much greater volumes of basaltic volcanic rocks. In the western United States, particularly in the Basin-and-Range physiographic province, accumulations of tuff locally exceed 3000 m in thickness, and individual units may be tens of kilometers in lateral extent.

Eruptions of silicic magmas vary in volume from a few cubic meters to a massive catastrophic event depositing hundreds of cubic kilometers of ash and lava in a single eruption. On eruption, silicic magmas most commonly contain <10% primary crystals (phenocrysts) and incorporate <10% lithic fragments, so that generally >80% of the material deposited is glass. Eruption may occur as a relatively small quiescent flow of very viscous lava that commonly quenches to form an obsidian. However, in large eruptions the great majority of material is ejected explosively as fine ash, shards, and pumice because of the high viscosity and high volatile content of silicic magmas.

Large explosive eruptions commonly begin with ejection of pumice, shards, and ash that are deposited downwind of the vent as a cool unsorted layer of low-density material termed an air-fall or ash-fall deposit. As eruption proceeds and increases in intensity, clouds of incandescent ash propelled by evolving gases are formed over the vent and collapse downslope, depositing heated material in thick blankets. It is this latter type of deposit that is volumetrically most significant. As eruption then proceeds, the rocks overlying the magma chamber collapse to form a caldera, which may then also fill with thick sequences of volcaniclastic deposits. Materials deposited above ~500°C will compact and weld by viscous deformation of the glass. Compacted material that cools rapidly (i.e., weeks to months) may remain glassy, while that which cools more slowly will crystallize to cristobalite, quartz, and a mixture of feldspars.

The net result is a complex deposit up to several hundred meters thick which has cooled as a single unit, and is commonly referred to as a cooling unit. A schematic cross section through such a deposit is given in Fig. A-1. Such a deposit commonly has a core of welded material, most of which may have devitrified (i.e., crystallized) to quartz and feldspar with or without cristobalite. At the base of the welded zone is a layer of densely welded material that has not crystallized but remains a dense glass called a vitrophyre. Welding decreases outward from the core so that the welded zone

Fig. A-1. Schematic Cross Section Through an Ash-Flow Tuff Cooling Unit with Typical Bulk and Grain Densities.

is surrounded by zones of material decreasing in density, thermal conductivity, coherence, and strength. Commonly at the base of the ash-flow deposit is an unsorted, nonwelded, horizon of loosely aggregated pumice and ash called the air-fall unit. Because the surface material is also loosely aggregated, it is readily reworked by surface processes and may be redeposited by streams and ponds or by volcaniclastic mudflows (lahars). Such processes give rise to sorted, bedded deposits called bedded tuff.

Because glass is thermodynamically unstable, it commonly alters to various crystalline phases. At elevated temperatures quartz, cristobalite, and feldspars crystallize, as occurs in welded zones. If the porosity is high and water saturation occurs, glass may alter completely to zeolites in as little as 10000 years even at a temperature below 100°C. If saturation is not achieved, or if the porosity is low as in vitrophyres, alteration will not occur and the material may remain glassy for millions of years. There is no evidence that material once crystallized to quartz and feldspar will later alter to zeolites under conditions common in these rocks, although feldspars may alter slowly to clays, particularly in the presence of acid groundwaters.

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APPENDIX B

THE ARGONNE 60 in. CYCLOTRON

The products and yields produced at the ANL 60 in. cyclotron are summarized in Table B-I. The yield values are targets containing 100% of the listed target isotope. For natural elements or compounds, correct accordingly. The yield per hour depends on the current that can be held on target and the decay rate of the produced isotope.

TABLE B-I
PRODUCTS AND YIELDS

<u>Half-life</u>	<u>Product</u>	<u>Target</u>	<u>Nuclear Reaction</u>	<u>Yield ($\mu\text{Ci}/\mu\text{A}\cdot\text{h}$)</u>
53.6 d	^7Be	^7Li	(p,n)	20
53.6 d	^7Be	^7Li	(d,2n)	9.4
110 m	^{18}F	^{16}O	(α ,pn)	5,000
110 m	^{18}F	^{16}O	(^3He ,p)	7,000
2.6 y	^{22}Na	^{24}Mg	(d, α)	~2
15.0 h	^{24}Na	^{27}Al	(d,p α)	1,200
21.3 h	^{28}Mg	^{26}Mg	(α ,2p)	~50
14.3 d	^{32}P	^{32}S	(n,p)	?
87.2 d	^{35}S	^{37}Cl	(d, α)	~2
37.2 m	^{38}Cl	^{36}S	(α ,pn)	~10,000
12.42 h	^{42}K	^{40}Ar	(α ,pn)	~400
162.7 d	^{45}Ca	^{45}Sc	(n,p)	?
4.53 d	^{47}Ca	^{50}Ti	(n, α)	?
27.8 d	^{51}Cr	^{51}V	(d,2n)	~40
5.63 d	^{52}Mn	^{52}Cr	(d,2n)	~200
313 d	^{54}Mn	^{52}Cr	(α ,pn)	~1
8.5 h	^{52}Fe	^{52}Cr	(^3He ,3n)	~50
2.7 y	^{55}Fe	^{55}Mn	(d,2n)	~1
271 d	^{57}Co	^{56}Fe	(α ,p2n)	2.2
71.4 d	^{58}Co	^{56}Fe	(α ,pn)	24
6.2 d	^{56}Ni	^{56}Fe	(α ,3n)	~40

TABLE B-I (cont)
PRODUCTS AND YIELDS

Half-life	Product	Target	Nuclear Reaction	Yield ($\mu\text{Ci}/\mu\text{A}\cdot\text{h}$)
36.1 h	^{57}Ni	^{54}Fe	(α, n)	8.3
92 y	^{63}Ni	^{63}Cu	(n, p)	?
12.75 h	^{64}Cu	^{62}Ni	(α, pn)	~400
9.15 h	^{62}Zn	^{60}Ni	($\alpha, 2n$)	167
243.7 d	^{65}Zn	^{65}Cu	(d, 2n)	~4
78.2 h	^{67}Ga	^{64}Zn	(α, p)	~20
287 d	^{68}Ge	^{66}Zn	($\alpha, 2n$)	~1
26.4 h	^{76}As	^{74}Ge	(α, pn)	~150
120.4 d	^{75}Se	^{75}As	(d, 2n)	~5
56 h	^{77}Br	^{75}As	($\alpha, 2n$)	27
35.4 h	^{82}Br	^{80}Se	(α, pn)	~100
34.9 h	^{79}Br	^{79}Br	(d, 2n)	~500
54.58 h	^{81}Rb	^{79}Br	($\alpha, 2n$)	~1,000
4.58 h	^{81}Rb	^{82}Kr	($\alpha, 3n$)	~1,000
83 d	^{83}Rb	^{81}Br	($\alpha, 2n$)	~2
33 d	^{84}Rb	^{81}Br	($\alpha, 2n$)	~1
18.66 d	^{86}Rb	^{86}Kr	(α, pn)	~10
64.7 d	^{85}Sr	^{85}Rb	(d, 2n)	~10
80 h	^{87}Y	^{85}Rb	($\alpha, 2n$)	~50
106.6 d	^{88}Y	^{85}Rb	(α, n)	~0.5
61 d	$^{95\text{m}}\text{Tc}$	^{93}Nb	($\alpha, 2n$)	~2
4.3 d	^{96}Tc	^{93}Nb	(α, n)	~30
41.0 d	^{105}Ag	^{103}Rh	($\alpha, 2n$)	30
8.4 d	$^{106\text{m}}\text{Ag}$	^{103}Rh	(α, n)	~10
7.48 d	^{111}Ag	^{108}Pd	(α, p)	~5
2.83 d	^{111}In	^{109}Ag	($\alpha, 2n$)	~50
115 d	^{113}Sn	^{111}Cd	($\alpha, 2n$)	~1
60.2 d	^{124}Sb	^{122}Sn	(α, pn)	~2
2.73 y	^{125}Sb	^{122}Sn	(α, p)	~0.1
13.1 h	^{123}I	^{122}Te	($\alpha, p2n$)	250
4.17 d	^{124}I	^{122}Te	(α, pn)	~25
5.0 y	^{155}Eu	^{154}Sm	($\alpha, p2n$)	0.01
15.2 d	^{156}Eu	^{154}Sm	(α, pn)	0.8
15.2 h	^{157}Eu	^{154}Sm	(α, p)	0.9
93.1 d	^{168}Tm	^{166}Er	(α, pn)	0.5
57 h	^{166}Yb	^{164}Er	($\alpha, 2n$)	370
2.2 h	^{178}Ta	^{181}Ta	($^3\text{He}, \alpha 2n$)	60
8.13 h	$^{180\text{m}}\text{Ta}$	^{181}Ta	($^3\text{He}, \alpha$)	60
21.5 d	^{178}W	^{177}Hf	($\alpha, 3n$)	~3
21.5 d	^{178}W	^{176}Hf	($\alpha, 2n$)	~3
23.9 d	^{187}W	^{186}W	($^3\text{He}, 2p$)	30
19 h	^{181}Re	^{181}Ta	($\alpha, 4n$)	500
70 d	^{183}Re	^{181}Ta	($\alpha, 2n$)	10
38 d	^{184}Re	^{181}Ta	(α, n)	0.65
91 h	^{186}Re	Re	($^3\text{He}, 2p$) ($^3\text{He}, \alpha$)	10

TABLE B-I (cont)
PRODUCTS AND YIELDS

<u>Half-life</u>	<u>Product</u>	<u>Target</u>	<u>Nuclear Reaction</u>	<u>Yield ($\mu\text{Ci}/\mu\text{A}\cdot\text{h}$)</u>
64.1 h	^{197}Hg	^{197}Au	(d,2n)	500
23.8 h	$^{197\text{m}}\text{Hg}$	^{197}Au	(d,2n)	1,300
3.31 h	^{209}Pb	^{208}Pb	(d,p)	9,500
30 y	^{207}Bi	^{207}Pb	(d,2n)	~ 0.02
5.01 d	^{210}Bi	^{209}Bi	(d,p)	130
10 d	^{206}Po	^{207}Pb	(^3He ,4n)	10
5.7 h	^{207}Po	^{207}Pb	(^3He ,3n)	1,000
2.9 y	^{208}Po	^{209}Bi	(d,3n)	2.5
103 y	^{209}Po	^{209}Bi	(d,2n)	0.06
8.3 h	^{210}At	^{209}Bi	(α ,3n)	2,900
7.2 h	^{211}At	^{209}Bi	(α ,2n)	2,300
396 d	^{235}Np	^{235}U	(d,2n)	0.22
2.85 y	^{236}Pu	^{235}U	(α ,3n)	~ 0.05
2.85 y	^{236}Pu	^{235}U	(d,n)	~ 0.2
2.85 y	^{236}Pu	^{238}U	(d,4n) ^{236}Np	?
45.6 d	^{237}Pu	^{235}U	(α ,2n)	~ 0.18
45.6 d	^{237}Pu	^{237}Np	(d,2n)	0.15
32.8d	^{241}Cm	^{239}Cm	(α ,2n)	0.15

APPENDIX C ELEMENTS IN THE NETWORK CHARTS

The network charts given in Figs. 11-13 are work-flow diagrams which include an identification of the activities, the sequence in which they take place, the assignment of responsibilities, and estimated time needed to perform the activity. It is important to emphasize that the network charts are to be used as a plan in every sense of the word. Because of the uncertainty of research projects, the plan must have flexibility and be easily changed. The times given are not commitments, but current estimates of the time required for an activity provided the work goes smoothly with only minor delays. As more information becomes available, these time estimates will need to be revised. For more information on the approach and proper use of network charts, see Ref. 1. For a brief, but informative discussion of the philosophy and realistic use of network charts, see Ref. 2.

The various elements shown in the network charts given in Figs. 11-13 are identified in Fig. C-1. The event numbers are used to identify activities for computer generated activities and to show their interrelationship.

Fig. C-1. Elements of a Network Chart

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