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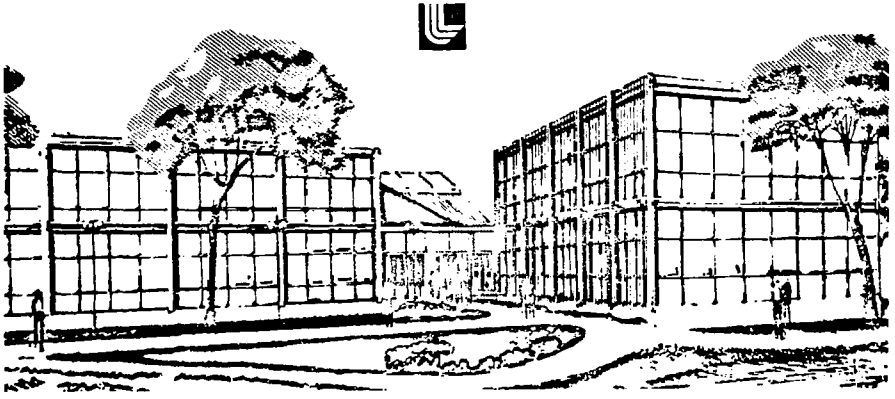

THE HYDROGEOCHEMICAL AND STREAM-SEDIMENT RECONNAISSANCE PROGRAM AT LLL*

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INTRODUCTION

The Lawrence Livermore Laboratory (LLL) is conducting a Hydrogeochemical and Stream-Sediment Reconnaissance (HSSR) survey in support of ERDA's National Uranium Resource Evaluation (NURE) program. Included in the LLL portion of this survey are seven western states (Arizona, California, Idaho, Nevada, Oregon, Utah, and Washington). Similar surveys are being carried out in the rest of the continental United States, including Alaska, as part of a systematic nationwide study of the distribution of uranium in surface water, groundwater, and stream sediment. The overall objective is to identify favorable areas for uranium exploration. This paper describes the program being conducted by LLL to complete our portion of the survey by 1981.

GEOLOGY AND SAMPLE ACQUISITION

In developing a sampling strategy for the seven western states, the region was first divided into geologic and physiographic provinces as shown in Fig. 1. Each province was then assigned a "uranium favorability" rating based on a careful consideration of geology, known uranium mining activities, and the degree of contamination. The Basin and Range Province, the Colorado Plateau, and the Northern Rocky Mountains Province were assigned the highest favorability ratings whereas the Pacific Coast region and the Great Valley of California received the lowest. Reconnaissance sampling will proceed at a rate of about 30,000 sites per year beginning with the high favorability areas and ending with the least promising areas.

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Approximately 160,000 samples will be collected at 120,000 sites over an area of 700,000 square miles (1,840,000 square kilometers). This represents an average site density of about 1 per 5.8 miles^2 (1 per 15 km^2). Clearly, such a sampling program is not designed to locate individual ore deposits. The results should, however, identify potential uranium districts and large anomalous areas associated with poorly exposed or low-grade uranium occurrences.

Each province will have a unique set of geological, hydrological, and climatic variables that will influence the dispersion of uranium. Orientation surveys will therefore be conducted in each province to select appropriate field, analytical, and interpretive methods for use in the reconnaissance surveys. Specific objectives of the orientation studies will be:

- To validate sample-collection and field-measurement methods.
- To provide "background" values for uranium and associated trace elements.
- To determine optimum sample types and establish site-selection criteria for reconnaissance surveys.
- To check the adequacy of laboratory analytical techniques.
- To determine the magnitude of temporal variations in uranium content and composition of surface waters and groundwaters.
- To evaluate the applicability of various geochemical characterizations for delineating anomalous uranium occurrences.

To date, seven orientation surveys have been completed - five in the northern Basin and Range Province (Nevada), and two in the Northern Rocky Mountain Province (Idaho and Washington) - and three orientation surveys are underway in the southern Basin and Range Province (Arizona).

Reconnaissance surveys will be carried out in each province in a reasonably uniform and systematic manner. However, sampling densities, site-selection criteria, sampling media, and analytical requirements will differ from one province to another based on environmental and geologic considerations.

The proposed reconnaissance sampling schedule is summarized in Table 1. Samples will be collected by subcontractors and returned to LLL for analysis. Typical sampling contracts covering 2000 to 4000 sites will be placed every few weeks when sample acquisition stabilizes at about 30,000 sites per year (FY 1978 through early FY 1981).

Composite sediment samples are collected from five points about five yards apart at locations identified on maps provided by LLL. Field personnel must see that the sample is representative of the sediment being actively transported down a particular drainage path and avoid sampling where there is evidence of wind-deposited material, local contamination, bank-collapse material, etc.

Table 1. Proposed reconnaissance sampling schedule through 1980-81.

	Completed	Projected			
	FY 1975 - FY 1976	FY 1977	FY 1978	FY 1979	FY 1980-81
No. of sites	3,000	16,000	32,000	32,000	38,000
Square miles covered	5,800	80,000	170,000	170,000	230,000
% of total area	1	12	26	26	35
Areas covered	Nev.	Nev.	Nev.	Cal.	Cal.
	Cal.	Cal.	Cal.	Ariz.	Ariz.
		Utah	Utah	Utah	Utah
		Ariz.	Ariz.	Idaho	Oregon
			Idaho	Oregon	Wash.
			Oregon	Wash.	
			Wash.		

Surface water and groundwater samples (approximately 1 liter total at each site) will be filtered in the field. One 500-ml sample will be filtered into a polyethylene bottle with a polyethylene bag liner. This unacidified sample will be evaporated to dryness in the bag, and the residue will be analyzed for uranium by delayed-neutron counting. A second sample, filtered into a 500-ml polyethylene bottle, will be split in the laboratory and analyzed for sulfate, chloride, and various elements by optical emission spectrometry. Temperature, pH, and specific conductivity are measured *in situ* on all water samples. Where appropriate, field measurements of alkalinity, bicarbonate, and Eh will be made.

Most contracts for sample acquisition will require wet- and dry-sediment and surface-water sampling. Occasionally, desert sampling involving only dry sediments will be done. Groundwater sampling in Nevada has been contracted to the Desert Research Institute, Reno, Nevada.

SAMPLE PREPARATION AND ANALYSIS

The flow of samples through LLL's HSSR system is illustrated schematically in Fig. 2. Approximately 90% of the samples will be delivered to a sample-processing contractor. The remaining 10% will be processed at LLL. We will process orientation-study samples that require special preparation, and replicates of field samples taken as part of our quality-assurance program.

The sample-processing contractor will dry and sieve the sediment samples and load approximately 1 g of material of the appropriate size fraction into a clean polyethylene vial. Water samples will be evaporated to dryness in the polyethylene sample collection bag, which will then be folded and placed in a clean polyethylene vial. Vials containing the processed sediment and water samples will be returned to LLL for analysis.

Facilities at LLL for sample processing and analysis are shown in Fig. 3. They include sample-receiving and storage areas, a sediment-processing laboratory, a clean room for water-sample handling, a pneumatic transport system for delayed-neutron counting (DNC) and instrumental neutron-activation analysis (INAA), a data processing center, and an archival-sample storage area.

Water samples collected during orientation studies or as part of the quality-assurance program will be evaporated to dryness in the LLL clean room; sediment samples will be dried, sieved, split, and weighed in the LLL sediment-preparation lab. The LLL-processed samples as well as those processed by contractors will be placed in clean pneumatic rabbits in the clean room and loaded into hoppers for DNC and INAA.

Neutron-activation analysis is the principal analytical method to be used in assaying uranium in water residue and many other elements in sediment samples. Two variations of this technique are used, one involving the detection of delayed neutrons and the other involving the detection and analysis of

gamma rays resulting from the decay of radioactive species induced in the sample by reactor irradiation.

A "rabbit" capsule (used to transport samples through a microprocessor-controlled, pneumatic transfer system) is shown in Fig. 4. To minimize contamination, rabbits are injection-molded from high-purity linear polyethylene. The internal volume is large enough to accommodate commercially available 2-dram (7.5 cc) polyethylene vials, which serve as secondary sample containers. A machine-readable code on the waist ring of each capsule permits automated sample identification.

Rabbits contain analytical samples are manually placed in a code reader to correlate rabbit numbers with sample numbers, and as many as 200 can be loaded into a special hopper. The hopper is then transferred to the identification-send station where the rabbits will be introduced into the pneumatic transport system.

Major components of the pneumatic transport system are illustrated schematically in Fig. 5. They include an identification-send station, a reactor in-core irradiation facility, a delayed-neutron counter (DNC), four lithium-drifted-germanium gamma-ray detectors, and a dropout station. By operating valves and switches in proper sequence, the microprocessor identifies each rabbit and sends it to the reactor for neutron irradiation for a predetermined time. It is then sent to the delayed-neutron counter, where the delayed-neutron intensity is measured to determine the uranium concentration. Following this measurement, rabbits containing residue from water samples are sent to the dropout station and then to archival storage. Rabbits containing sediment samples are forwarded to the delay (holding) station to allow for the decay of the more intense, shortlived isotopes.

After the delay period, the rabbit is transferred to one of the four lithium-drifted-germanium detectors where a 4096-channel gamma-ray spectrum of the activated elements is obtained. Four microprocessors act as multiple pulse-height analyzers.

After gamma counting, the rabbit is ejected from the pneumatic system into a special container at the dropout station for further decay. Seven to ten days after the initial irradiation, the sediment samples are reinserted into the gamma-counting portion of the pneumatic system for analysis of the long-lived activated elements. This counting is done on nights and weekends when the reactor is not operating.

A flow-chart summary of the DNC-INAA process is presented in Fig. 6.

Our analytical system can process 26,000 samples per year per shift (one sample every 3 minutes, 7 hours a day, 4 days per week, and 48 weeks per year). Double-shift operation will be used as required to increase the processing rate.

The analytical system described above is used to measure the concentration of uranium in water and sediment samples by DNC. It can also detect up to 35 trace and major elements in sediments by INAA.

A direct-reading, optical-emission spectrometer with an inductivity-coupled plasma source is used to measure trace-element concentrations in water samples. This instrument also has a dc-arc source which will be used to assay sediment samples for all orientation studies, for the quality assurance program, and for other special purpose applications. The emission spectrometer,* is located in the clean room. It has direct-reading channels for the 26 elements listed in Table 2. Eighteen elements can be measured in water samples - twenty-one in sediments. A minicomputer handles data processing for 400 samples per day under microcomputer control. The central unit also operates the automated sample changer and serves as a communication (I/O) link.

A Technicon I auto-analyzer is used to measure chloride and sulfate in water samples. Standard colorimetric tests are used. This system operates under minicomputer control.

Data processing for the delayed-neutron counter is fairly straightforward; simple computer codes are used to convert instrument readings to uranium concentrations. However, the 4096-channel gamma-ray spectra produced in INAA typically exhibit numerous peaks (representing many radionuclides) and require complex computer programs to convert the spectral information to trace-element concentrations. A modified version of the GAMANAL** program, which runs on the LLL CDC-7600 computers, has been used for this purpose. In the next few months, we will receive a DEC PDP 11/70 computer, which will be programmed to handle this task as well as data-base management and reporting for the HSSR program.

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*Manufactured by the Jarrell-Ash Division of Fisher Scientific Co.

**R. Gunnink and J. B. Niday, "Computerized Quantitative Analysis by Gamma-Ray Spectrometry," Vol. I. Description of the GAMANAL Program." UCRL-51061, Vol. 1 (March 1, 1972).

Table 2. Lower detection limits — emission spectrograph.

	Plasma (ppb in H_2O)	dc arc (ppm in solids)[a]
Al	5	10 000
As	40	—
Ca	1 000	2 000
Cd	1	—
Ce	—	1 000
Co	1.5	10
Cu	2	2
Fe	50	2
Ga	—	Internal standard
K	1 000	100
La	—	20
Li	1 000	—
Mg	10	2 000
Mn	—	1 000
Mo	10	20
Na	1 000	1 000
Ni	10	200
P	30	—
Pb	—	60
Si	30	10 000
Ti	—	1 000
U	60	200
V	1	6
Y	—	10
Zn	5	10
Zr	—	20
Jarrel ash quotation	#EM2919 6/2/76	#EM2295 9/22/76
Number of analytical channels	18	22

[a] Assumes 50:50 mixture of sample with graphite.

DATA-BASE MANAGEMENT

Data management for the HSSR program is divided into three interrelated data-base subsystems.

The first data-base subsystem deals with sample acquisition and field measurements. It is used to ensure unique site identifiers, to validate and store field measurements, to store and protect information on site locations, and to record the number and types of samples taken at each site. Data are entered via punched cards, a terminal, and a map digitizer. This data base can be queried interactively for verification, modification, and update, and used to generate periodic operational and summary reports.

The second major data-base subsystem catalogues the sample analysis data. It contains primarily uranium and trace-element concentrations and water chemistry data. This data base collates the results from the different analytical methods and serves to separate the analysis data from the site-location information. Data are mainly from the INAA data-reduction programs and optical emission spectrometer determinations. Additional data are entered for sulfate and chloride measurements via punched cards. This data base is accessed interactively for verification, modification, and update, and it provides input data for various analysis and graphics programs.

The third major data-base subsystem is the archival data base, which contains final data from both the site and sample analysis data bases. These data are used with statistical-analysis and graphic-display routines on a selected basis. Information in this data base is used to generate magnetic-tape and hard-copy tabular data for the raw data and interpretive, open-file reports.

The three major data-base subsystems are integrated to reduce data redundancy, to allow flexible querying and report generation, and to provide predetermined access schemes for the different users. Due to the sensitive nature of the data, controlled access is required for portions of the data base, specifically those linking uranium concentration with geographical location.

The configuration of the data-base-management system at LLL is illustrated in Fig. 7. At the present time, all data-base information is being processed by the Master Control Data Base in the LLL OCTOPUS computer system. In the future, all data-reduction, data-base-management, and reporting functions for the HSSR program will be transferred to the new DEC PDP 11/70 minicomputer.

Results of the survey will be published in "raw data" reports, listing concentrations of uranium and associated trace elements, field observations, and other analytical data for each sample type and location. Separate "evaluation" reports will relate the raw data to geology, delineate background and anomalous uranium values, and discuss geochemical relationships. These reports will be available to the private exploration sector through ERDA's open filing system.

Fig. 1. Geologic and physiographic province map of western United States.

Fig. 2. HSSR sample flow diagram.

Fig. 3. HSSR facilities at LLL.

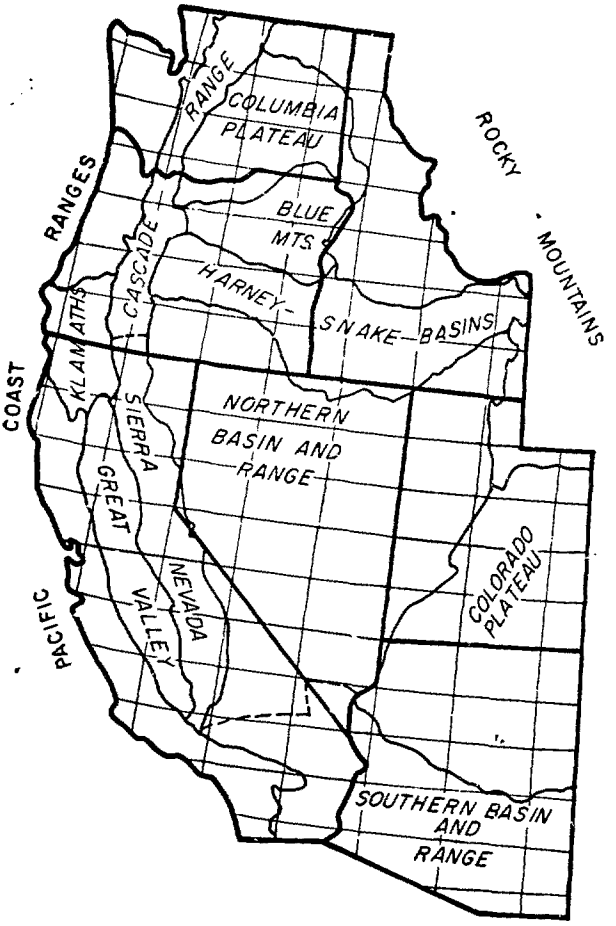
Fig. 4. Rabbit design used with pneumatic transport system.

Fig. 5. Block diagram of the pneumatic transport system.

Fig. 6. DNC-INAA process flow.

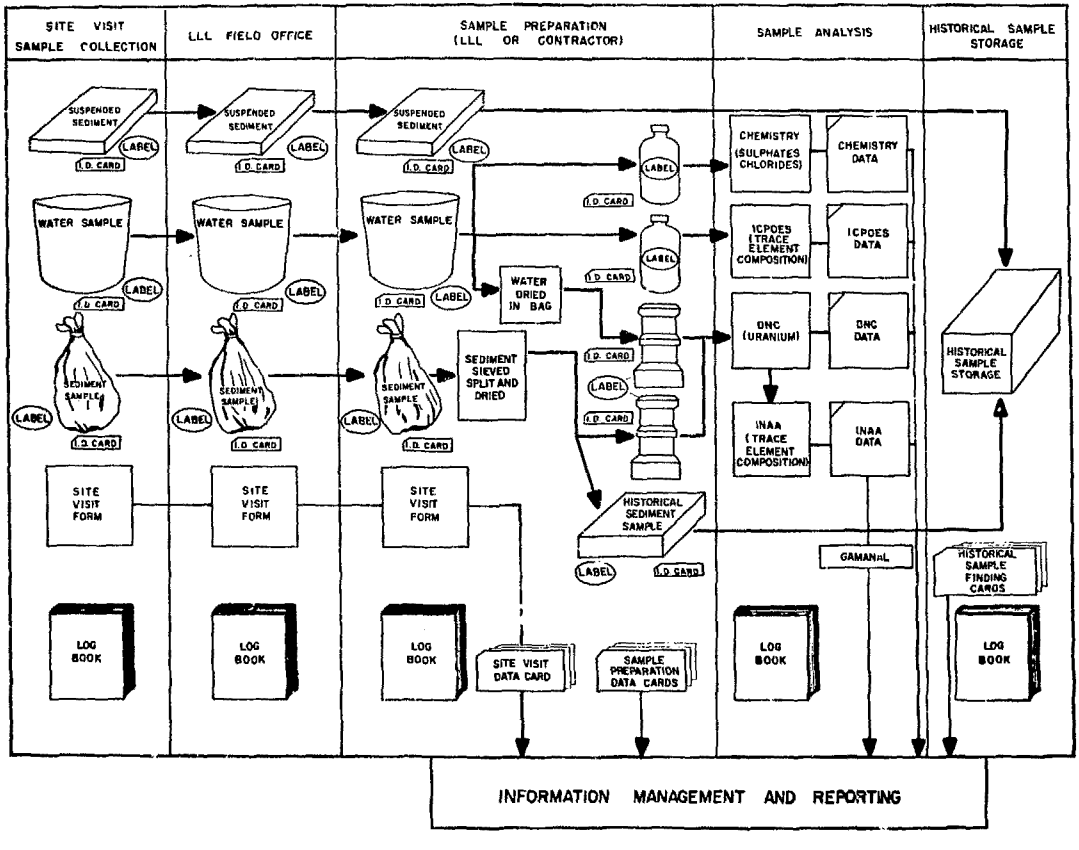
Fig. 7. Data-base-management system at LLL.

FIGURE 1



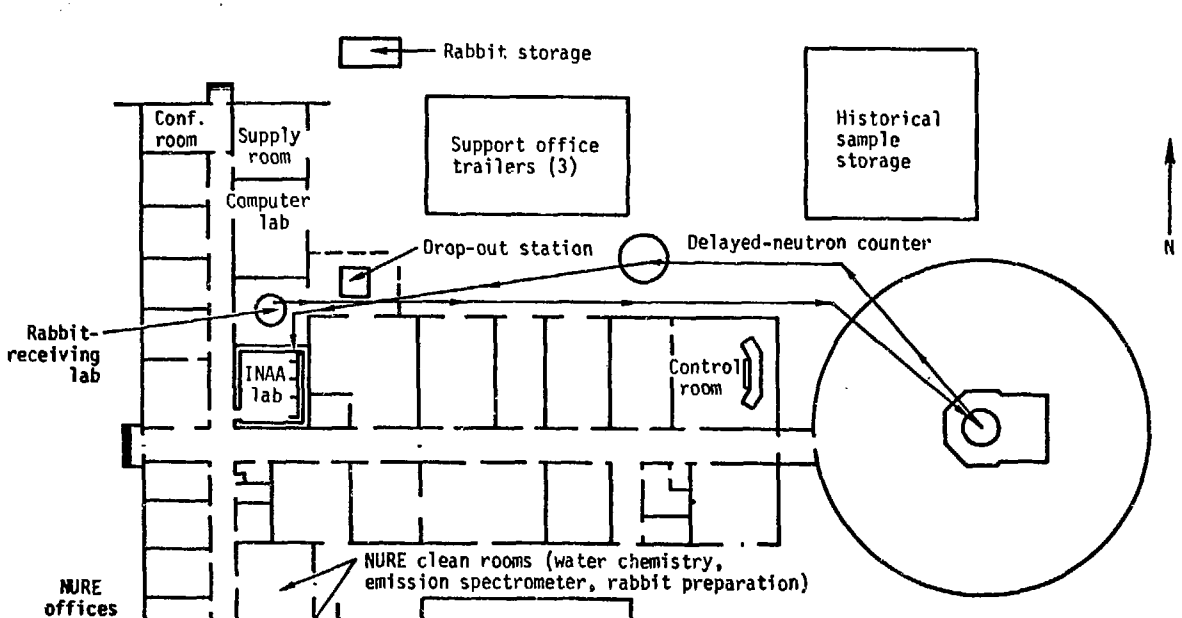
GEOLOGIC AND PHYSIOGRAPHIC PROVINCE MAP OF WESTERN UNITED STATES

FIGURE 2



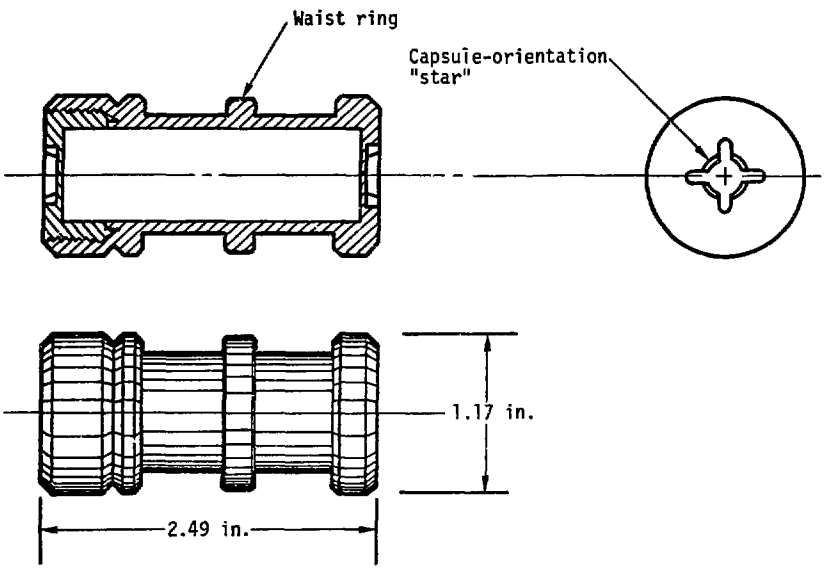
HSSR SAMPLE FLOW DIAGRAM

FIGURE 3



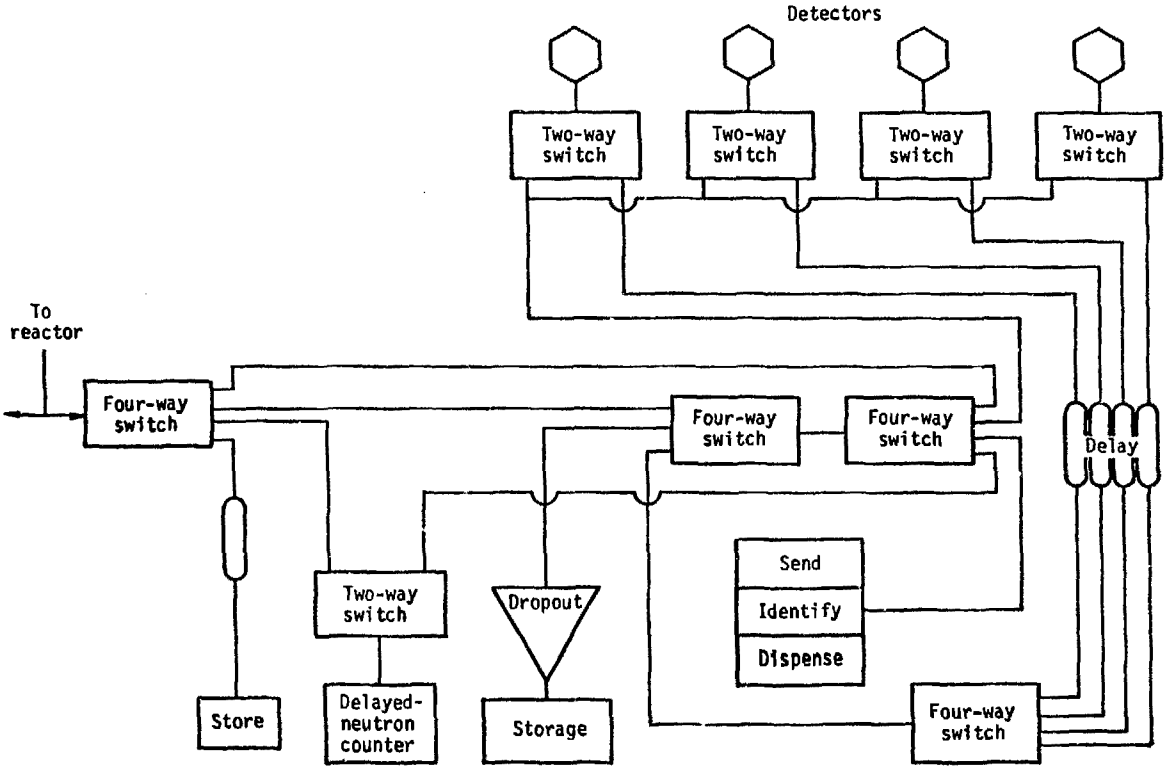
HSSR FACILITIES AT LLL

FIGURE 4



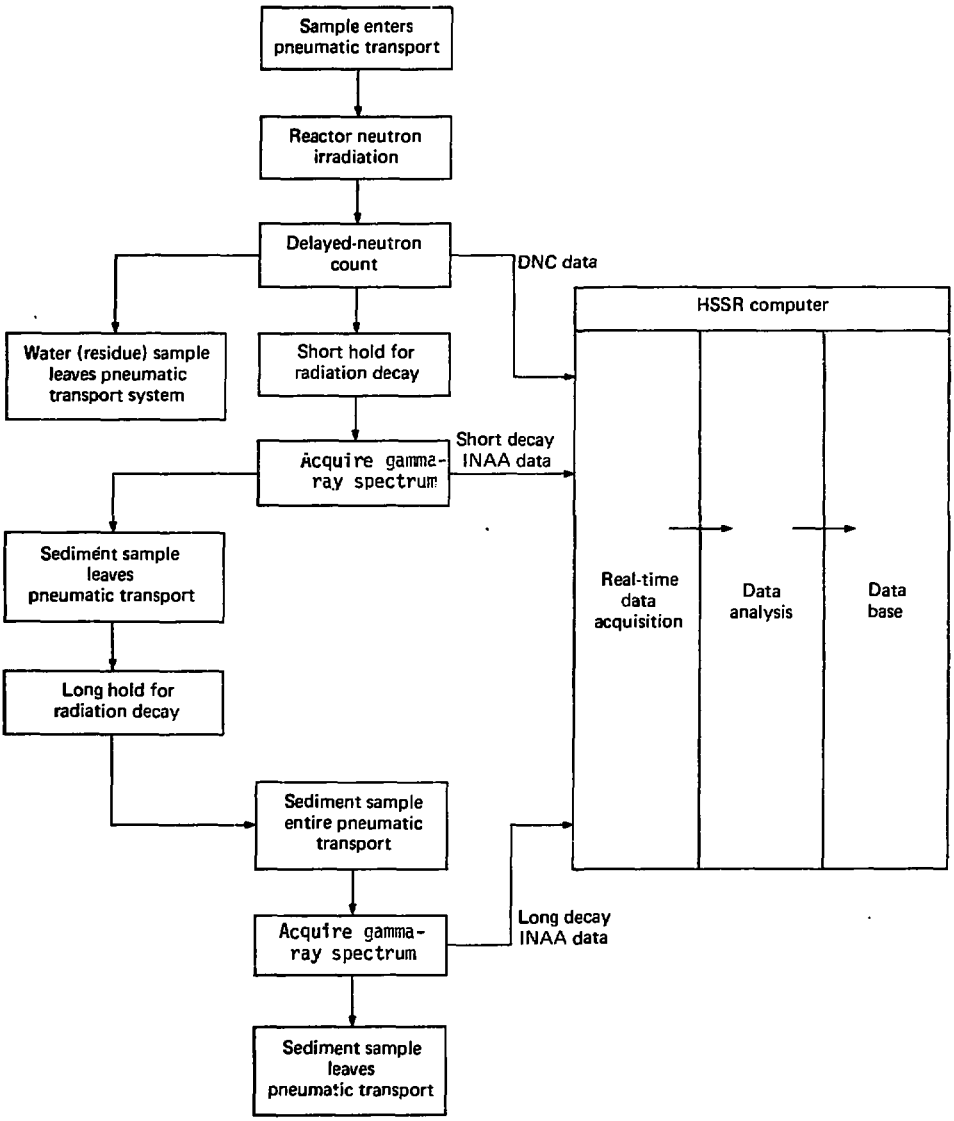
RABBIT DESIGN USED WITH
PNEUMATIC TRANSPORT SYSTEM

FIGURE 5



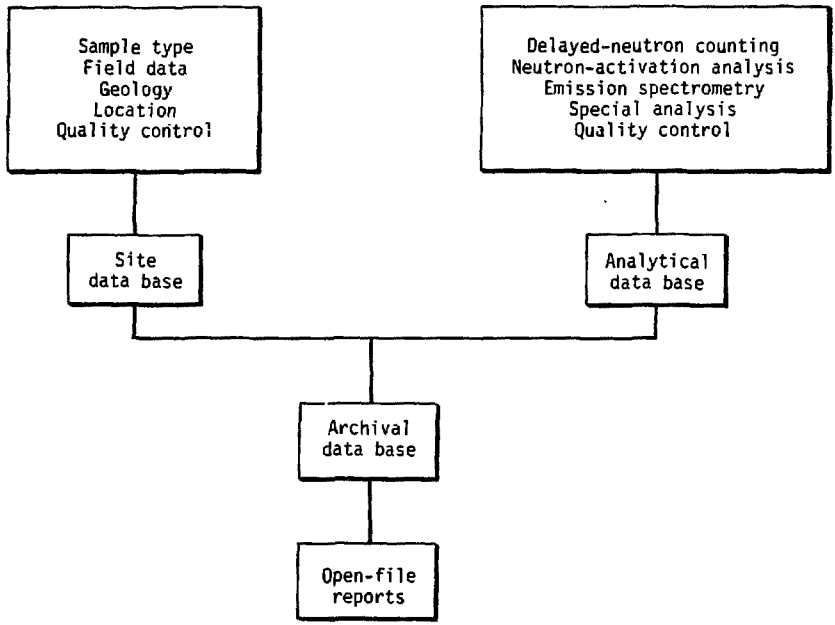
BLOCK DIAGRAM OF THE PNEUMATIC TRANSPORT SYSTEM

FIGURE 6



DNC-INAA PROCESS FLOW

FIGURE 7



DATA-BASE-MANAGEMENT SYSTEM AT LLL