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Authors: M. M. Minor
W. K. Hensley
M. M. Denton
S. R. Garcia

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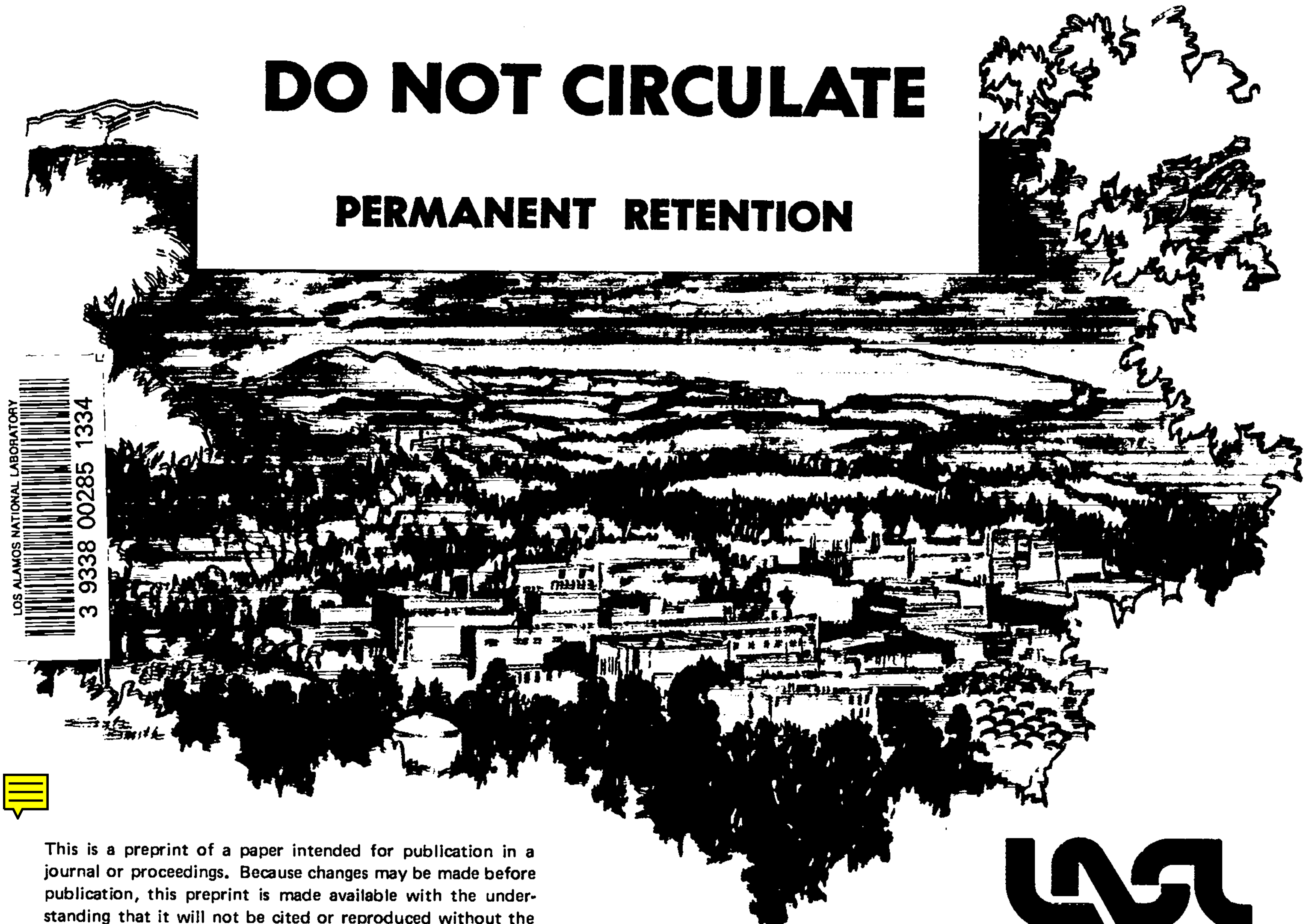

LOS ALAMOS SCIENTIFIC LABORATORY

AN AUTOMATED ACTIVATION ANALYSIS SYSTEM

M. M. Minor, W. K. Hensley,* M. M. Denton, S. R. Garcia

Los Alamos National Laboratory

(Los Alamos, NM 87545 U.S.A.)

An automated delayed neutron counting and instrumental neutron activation analysis system has been developed at Los Alamos National Laboratory's Omega West Reactor (OWR) to analyze samples for uranium and 31 additional elements with a maximum throughput of 400 samples per day. The system and its mode of operation for a large reconnaissance survey will be described.

INTRODUCTION

An automated neutron activation analysis system has been developed to analyze hundreds of thousands of stream sediment samples for a Hydrogeochemical and Stream Sediment Reconnaissance (HSSR) throughout the United States.[1] The HSSR is part of the National Uranium Resource Evaluation sponsored by the U.S. Department of Energy.

Approximately 180,000 stream sediment samples have been analyzed for uranium concentrations using delayed neutron counting (DNC) and for 31 additional elements using instrumental neutron activation analysis (INAA).

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*Present address: Battelle, Pacific Northwest National Laboratory, Richland, WA 99352 (U.S.A).

It is important to note that the HSSR is a reconnaissance program of enormous size and some parameters such as sample decay times and counting intervals required compromise to maximize sample throughput. At lower throughput, additional elements can be detected and improved sensitivity for most elements can be obtained.

The system utilizes two independent pneumatic transfer systems each having a sensitive neutron detector and four lithium-drifted germanium (Ge(Li)) gamma-ray detectors under the control of a single data acquisition computer. Samples are irradiated in the graphite thermal column of the OWR in a flux of $\approx 6 \times 10^{12}$ n/cm^2/sec and are counted immediately for delay-neutron emission to obtain the uranium assay. The samples are then routed to a delay device where the samples are allowed to decay for a preset time (typically 20 min.) before they are counted for short-lived gamma activities. The samples are then given a second irradiation and stored for later counting of the longer-lived activities at night when the reactor is shut down. System parameters are summarized in Table I.

Gamma-ray spectra along with timing and flux data and DNC data are collected and dumped to magnetic tape for subsequent data reduction by a second on-line minicomputer. Automatic analysis of data is performed by a code which identifies gamma-ray peaks and calculates elemental concentrations.

SYSTEM DESCRIPTION

1. Hardware

a. Pneumatic Sample Handling Facilities

Samples to be entered into the automated system are loaded into 4 cm^3 irradiation vials (rabbits) 1.2-cm dia. by 6.0-cm long. The rabbits are fabricated from high purity ethylene-butylene copolymer. The pneumatic system is designed to automatically load a sample and rapidly move the sample into the thermal column of the OWR and from there to the neutron detector, and later to a shielded location in front of a Ge(Li) gamma-ray detector for subsequent radioactive decay counting. The pneumatic system is summarized in Figure 1.

The individual samples are weighed and loaded into clips which can each hold fifty samples horizontally stacked. The automatic loader for each system can hold four clips and can sequentially load a maximum of 200 samples into the pneumatic system on command from the control computer. The maximum rate at which samples can be loaded into the system is limited by the rotating pick-up mechanism in the loader to approximately one sample every 34 seconds.

The samples are irradiated in a thermal neutron flux in a special irradiation port placed in the graphite thermal column of the OWR. The irradiation port contains two pneumatic tubes fabricated from 1.6-cm O.D. Aluminum tubing bent in an "S" shape to prevent gamma-ray back streaming. The tubes, which are surrounded by graphite, extend horizontally to the center of the thermal column and are approximately 100- and 110-cm from the center line of the reactor core. The ends of the tubes, which are separately vented by smaller diameter tubing, are bent downward, and the rabbit comes to rest in the irradiation position at approximately 42^0 from horizontal. A fission ion chamber is inserted in the graphite stringer between the two end positions to monitor the relative neutron flux at either irradiation position.

A delay loader was designed for use in the system to allow the irradiated samples to decay for set periods after irradiation before they are gamma-ray counted. The delay loader consists of two vertical pneumatic tubes terminated by a short section of tubing which can be switched from one vertical tube to the other. The short switch section can hold only one rabbit vial, and is connected by flexible tubing which can be pressurized to blow a sample out of the delay line. In normal operation ten samples are allowed to stack up on the input side of the delay loader and then are removed one at a time from the bottom of the stack, at the sample rate at which samples are being added to the top of the stack. Two delay loader mechanisms are installed side by side in a shield hole in the reactor room floor.

In order to route samples between the loader and reactor and the various counting stations in the system, two four-way pneumatic diverters are used in the system. Each diverter has four rigid pneumatic input tubes and four output tubes which are connected by four flexible polyethylene pneumatic tubes which can be switched to two positions at the input or output sides allowing twelve different pneumatic paths through the diverter.

After samples have been irradiated a second time, the samples are blown to a storage unloader where the samples can be stored for any length of time in the same order in which they were irradiated. The unloader consists of a circular twenty-position indexing mechanism and removable storage pipes each of which can hold fifty samples vertically. The unloader can be automatically indexed to place any one of twenty tubes under the input pneumatic line. The unloader is mounted at the top of cased hole which extends underground for shielding.

b. Control and Data Acquisition

The control and data acquisition system is normally controlled by a PDP-11/34 minicomputer. A second computer, a PDP-11/60, is used for on-line data analysis. Both computers are configured sufficiently alike that either computer can control the automated system by changing one CAMAC branch highway cable from one CAMAC interface to the other, thus samples can be run even if the PDP-11/34 must be taken down for maintenance.

All hardware in the system are interfaced to the control and data acquisition computer through CAMAC. A Microprogrammable Branch Driver (MBD) interfaces three powered CAMAC crates to the computer. The MBD is initialized and down-line loaded from the PDP-11 computer with an interpretive program. The MBD can output to and acquire data from CAMAC modules within the crates in parallel with the computer and is programmed to interrupt the PDP-11 when it has finished a process. Because of the modular CAMAC system, repair, replacement and/or substitution of modules, and configuration change or upgrade is comparatively easy and rapid.

The input and movement of samples to and from the reactor and to the neutron and gamma-ray counters is controlled in real time by a precision CAMAC timer module which generates an interrupt every second. The numerous solenoid actuated air valves in the pneumatic system are driven by solid-state optically isolated AC relays which are activated by a CAMAC output register. Photo detectors are installed in the pneumatic system to detect the passage of samples at eleven locations to detect any malfunctions. The outputs from the photo detectors are fed into a special CAMAC module which can latch the data (sample passage) until it is read.

The neutron detector used in the system is described in detail elsewhere.[2] The proportional signal from the neutron detector array is preamplified by a special preamplifier for large arrays of detectors and amplified by a standard NIM amplifier. The output of the amplifier is fed into a single channel analyzer which is set to generate a logic output for input pulses generated by delayed neutrons stopped in the detector. The neutron events are counted in a CAMAC scaler which can be cleared and read by the computer.

The neutron flux is monitored during all sample irradiations with a fission ion chamber. The bias current which is proportional to the thermal neutron flux, is fed into a current to frequency converter. The output of the converter is counted for the duration of all irradiations and is thus proportional to the integrated neutron flux in the sample.

Each multielement analysis system utilizes four Ge(Li) gamma-ray detectors with integral charge sensitive preamplifiers. The preamplified signals are fed to high-resolution spectroscopy amplifiers connected to conventional nuclear ADCs set for 4096 channel conversion gain. The digital output of the ADC is used to address a memory location in a 4096 word CAMAC memory module which is automatically incremented with each conversion. The memory modules can be cleared, read out, and enabled or disabled to accept digital conversions on command from the computer. The 4096 word store memory modules have 16-bit words and can accumulate 65K events per channel before overflowing. When a channel overflows, an interrupt to the computer is generated and the overflowing memory location is read into the data acquisition computer and stored in an overflow table.

Since the samples counted vary greatly in gamma-ray source strength, it is necessary to correct gross count rates for each sample by measuring the percent dead time of the ADC. This is accomplished by counting a 10 MHz oscillator (real time) and the same oscillator source gated off during ADC busy (live time). The real time and live times are accumulated with each gamma-ray count.

2. Software

a. Control and Data Acquisition

Two basic programs exist to acquire 1) Multielement data from short half-life isotopes, and 2) Multielement data from long half-life isotopes. The multielement analysis program for short half-life activities, SHORTS, in addition to the delayed neutron counting cycle, includes a 20-minute decay period, an 8-minute gamma-ray count period, and an additional 96 second irradiation for each sample. The program inserts a new sample into each pneumatic sample handling system every 126 seconds; a maximum of 400 samples may be analyzed in a normal day shift with the reactor at full power. Each sample cycle takes approximately 32 minutes to complete, thus there are as many as 15 samples in each system at any given time. The program executes a number of steps every second including reading photo cell latches and other diagnostic inputs, and handles asynchronous interrupts including memory module overflows. All disk and magnetic tape input or output routines are flagged for execution during the synchronous time critical loop but are actually run asynchronously at low priority background level. The program automatically backs up accumulated gamma-ray spectra to magnetic tape and stores all pertinent data including irradiation times and fluxes required by the long half-life analysis program.

The multielement analysis program for long half-life activities, LONGS, includes only a gamma-ray counting cycle for each sample. The LONGS program loads and counts up to 400 samples which have normally decayed for 14 days after being processed by the SHORTS program. The LONGS program is run at night while the reactor is shut down and processes a new sample every 360 seconds in each system. Each sample is typically counted for 1020 seconds.

b. Gamma-Ray Data Reduction

The computer program for gamma-ray data reduction, RAYGUN, is a variant of GAMANAL[3] tailored to the PDP-11 and the specific requirements of the HSSR project. Data to be analyzed are shipped to the Data Analysis Computer as a 4096 channel spectrum in which the data of the first 30 channels have been replaced by a header block of constants required for data reduction.

The RAYGUN analysis code requires approximately 24K words of PDP-11 memory in which to run. A single spectrum can be fit, gamma-ray peaks identified, and elemental concentrations calculated in approximately 17 seconds.

SYSTEM PERFORMANCE

The analytical detection of uranium using DNC has been employed for many years.[4] The standard DNC measurements in the system are calibrated for fixed irradiation (20 sec), decay (11 sec), and count (30 sec) times using samples of known natural uranium concentrations. The counting scheme is less than optimal but results in a high specific sensitivity for geological samples (e.g., a four-gram sample with 2 ppm uranium will yield 5000 net neutron counts with a statistical accuracy of 1.4%).

The INAA techniques employed are novel only in respect to the high degree of automation and analysis volume. Sample irradiation times were selected after a review of data taken on HSSR samples prior to construction of the automated system. Short and long sample irradiations of 20 and 96 seconds induce sufficient activity for sample masses greater than 1 gram and provide good counting statistics for the majority of elements of interest. Only when sample weights of less than 1 gram are used does the counting rate for several elements become marginal.

The samples are counted directly in their irradiation vials. The vials are sufficiently free of trace elements that analyses are made with no blank subtraction. Counting room background subtractions are not required for short-lived activities since contributions to the nuclides of interest are negligible. Minor contributions to ^{46}Sc, ^{59}Fe, ^{60}Co, and ^{65}Zn are present when counting long-lived activities. Provision for discrete background subtraction is incorporated in the analysis code.

At maximum throughput the automated system yields elemental assays for 13 elements from the "shorts" analysis (Na, Mg, Al, Cl, K, Ti, V, Mn, Sr, Dy, and U (by DNC)) and 19 elements from the "longs" analysis (Sc, Cr, Fe, Co, Zn, Rb, Sb, Cs, La, Ce, Sm, Eu, Tb, Yb, Lu, Hf, Ta, Au, and Th). Typical detection limits for most of the above elements are well below average crustal abundances and are below 1 ppm in several cases (See Table II). Uncertainties in the measured trace-element concentrations are usually less than 10% at concentration values one order of magnitude above the lower detection limits.

DISCUSSION

The automated activation analysis system has thus far been used mostly for high volume analysis of stream sediment samples for the HSSR. New software has been written to allow much more flexibility in the choice of irradiation, decay, and count times. These changes allow a much greater variety of sample size and matrix to be analyzed with greater sensitivity and precision with the advantages and reproducibility[5] of a fully automated INAA system. With additional gamma-ray counting at intermediate decay times (e.g., 4 days) additional elements can be observed (see Table II).

References

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TABLE I

INAA SYSTEM SUMMARY

Sample size	4 cm^3 (max)
Neutron Flux (NV)	6 x 10^{12} n/cm^2/sec (max)
Irradiation Ports	2
Irradiation Time (T_{ir})	5 sec (min)
Decay Time (T_d)	5 sec (min DNC)
	4 sec (min gamma)
Counting Time (T_c)	5 sec (min DNC)
	5 sec (min gamma)
DNC Detectors	
Type	^{3}He (4 atm., 2.5-cm dia. x 25-cm) 12 ea.
Number	2
Moderator	Polyethylene
γ-absorber	3.8 cm Pb
Efficiency	~27% (absolute)
Shielding	3.5m H_2O, 0.75mm Cd
Maximum Sensitivity	6.5×10^{-4} cts/NV/gram($T_{ir}=\infty$, $T_D=0$, $T_C=\infty$)
Detection Limit	0.01 microgram U (natural) per gram
Gamma-Ray Detectors	
Type	Ge(Li)
Number	8
Efficiency	~15% (relative to NaI)
Resolution	<2 keV FWHM
Absorbers	0.8 mm Al, 4.3 mm plexiglass
Counting Geometry	0.5- to 100-cm

TABLE II

LOWER LIMITS OF DETECTION*

Element	Detection Limit	Crustal Average	Element	Detection Limit	Crustal Average
Al	3200	8.23%	Mg	2700	2.33%
As**	3	1.8	Mn	55	950
Au	0.05	0.004	Na	1000	2.36%
Br**	4	2.5	Rb	13	90
Ba	150	425	Sb	1	0.2
Ca	1000	4.15%	Sc	0.9	22
Ce	10	60	Se	5	0.05
Cl	50	130	Sm	0.4	6.0
Co	1.7	25	Sr	110	375
Cr	10	100	Ta	1	2
Cs	2	3	Tb	1	0.9
Dy	0.7	3.0	Th	1	9.6
Eu	0.4	1.2	Ti	750	0.57%
Fe	1100	5.63%	V	6	135
Ga**	5	15	Yb	1	3.0
Hf	1.3	3	Zn	11	70
K	3400	2.09%	U	0.01	2.7
La	7	30	W**	1	1.5
Lu	0.1	0.5			

*All values are given in parts per million except as noted. Detection limits were calculated on the basis of a typical 4-gram sample of stream sediment run at maximum throughput.

**Observation requires an additional count at 4 days.

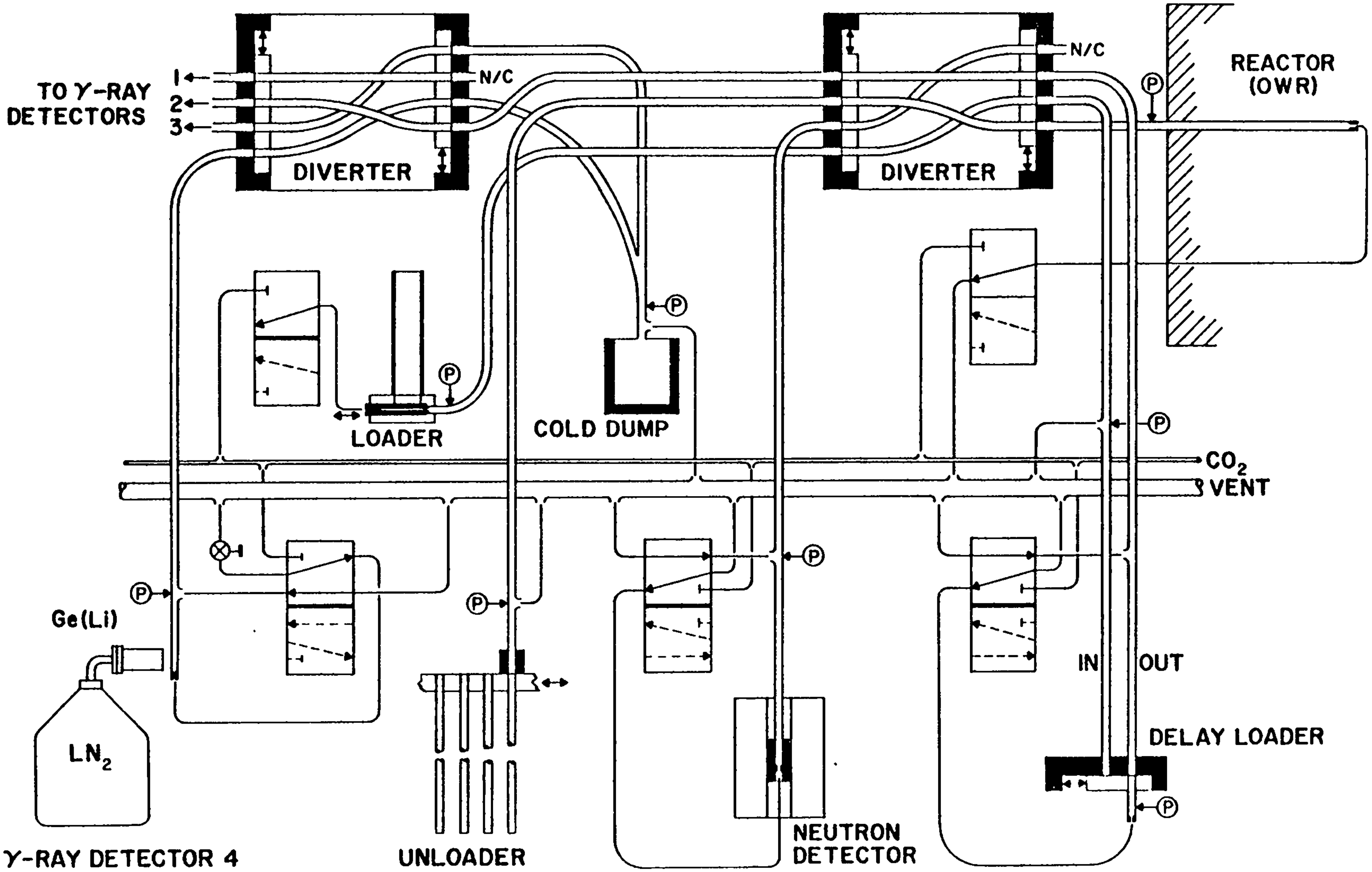
TO γ-RAY DETECTORS
1
2
3
N/C
DIVERTER
N/C
DIVERTER
REACTOR (OWR)
P
LOADER
COLD DUMP
CO2
VENT
Ge(Li)
LN2
γ-RAY DETECTOR 4
UNLOADER
NEUTRON DETECTOR
IN
OUT
DELAY LOADER

Fig. 1. Schematic diagram of the pneumatic transfer system for the Los Alamos DNC and INAA facility. The symbol P indicates the location of diagnostic photo detector units placed in the critical paths of the system.