REDUCING EMISSION OF ARGON-41

FROM THE MIT REACTOR

by

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ABSTRACT

In keeping with the radiation protection concept of ALARA (as low as reasonably achievable), an attempt has been made to find a practical method for reducing the emission of argon-41 from the MITR-II. Several sources of potential argon-41 production within the reactor have been identified, and an air sampling procedure has been developed to permit determination of argon-41 concentrations in these regions. Data show that 86% of the argon that was eventually emitted from the stack was initially produced in the region of the graphite reflector. The reflector is purged with a helium cover gas, but substantial concentrations of argon-41 present in the helium indicate that the gas does not successfully purge all the air from the graphite region.

Based on this information, strategies were developed to decrease argon-41 emission from the reactor. The first involves increasing the flow of helium through the graphite region; data are presented to quantitatively illustrate that increasing this flow decreases argon-41 production in the volume, hence decreasing ultimate releases to the atmosphere. Such an increase in helium flow would be costly, however, so possibilities should be investigated for the use of a less expensive purge gas.

Mass flow rate through the reactor pipe tunnel, into which the graphite helium exhausts, also has an effect on the argon-41 emission rate (though presumably has no effect on argon-41 production). Reducing suction on this region decreases stack output in the near term, but may increase argon-41 concentrations on the reactor floor and in the control room, thus increasing exposures to reactor personnel. Optimum blower settings, combined with an increased helium flow rate to limit argon-41 production, will permit determination of conditions which will limit total man-rem exposure.

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DEDICATION

To all of my friends at the MITR-II who have encouraged me, listened to me, educated me, and in general made the last year at least as much fun as it was work.

1. INTRODUCTION

1.1 Description of the MITR-II

The Massachusetts Institute of Technology Reactor, MITR-II, is a light-water cooled and moderated nuclear research reactor that operates at a maximum thermal power of five megawatts. The reactor is heavy-water and graphite reflected, and it utilizes plate-type fuel elements made of highly enriched uranium-235 encased in aluminum cladding.

The original MITR attained criticality in 1958. That reactor was heavy-water moderated and cooled and it operated until 1974. The present modifications have improved the economy, efficiency and practicality of the reactor. Two views of the reactor are shown in Figures 1 and 2, and more specifics are available in the MITR-II Operations Manual (1).

Since the MITR is a research reactor, several special features have been incorporated to maximize its experimental applications. Research facilities include a thermal column, horizontal neutron beam ports, irradiation facilities, nuclear instrumentation penetrations, vertical thimbles in the graphite reflector, in-core facilities, pneumatic tubes, a medical therapy room, and a fuel storage facility. The design of the reactor has established a variety of different neutron and gamma environments for use by experimenters; however, the design, by nature, also allows for air to enter some regions which have a significantly high neutron flux.

When air interacts with a neutron field, the argon-40 that is naturally present as 0.9% of the atmosphere is neutron-activated to

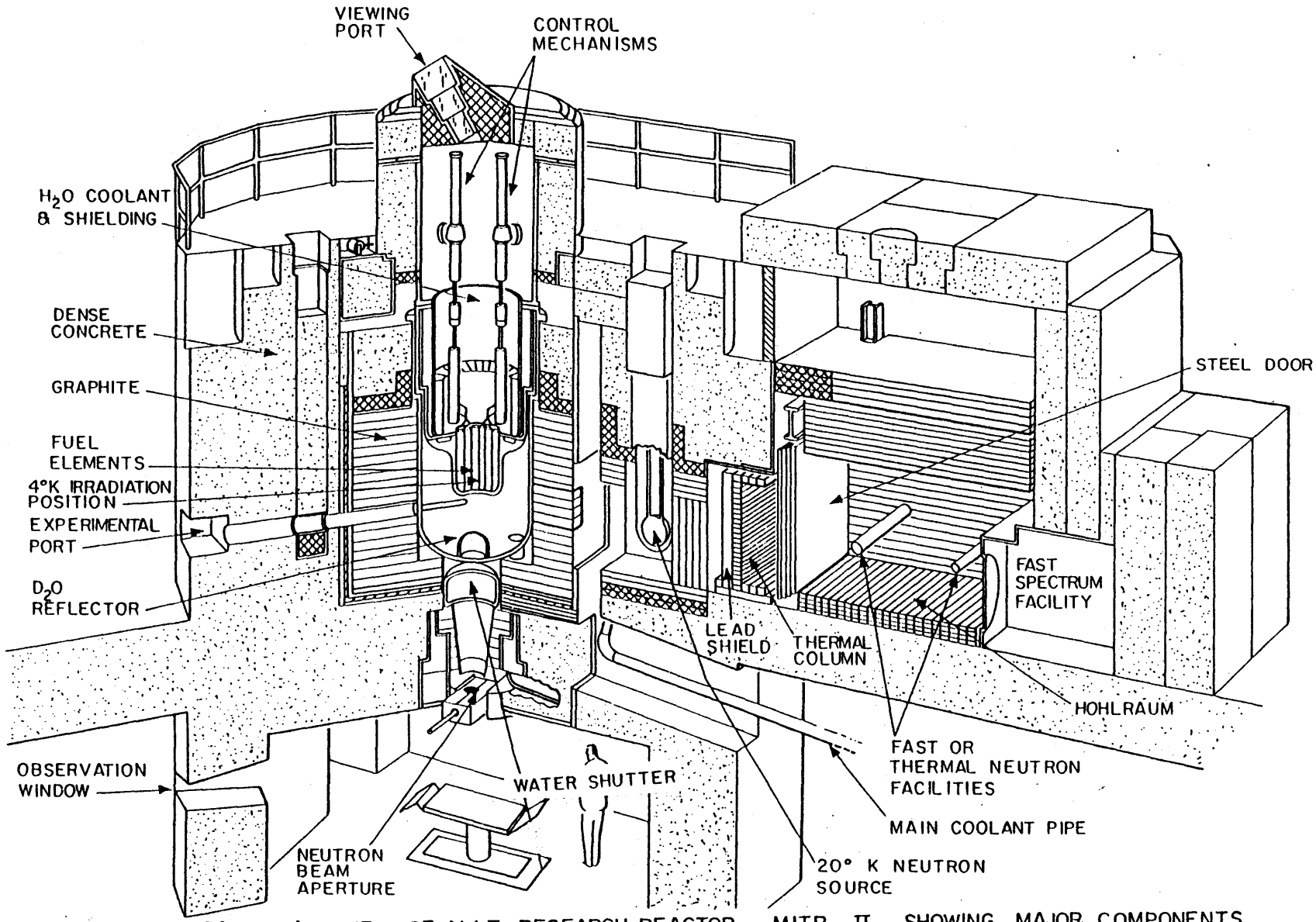

Figure 1. VIEW OF M.I.T. RESEARCH REACTOR, MITR-II, SHOWING MAJOR COMPONENTS AND EXPERIMENTAL FACILITIES

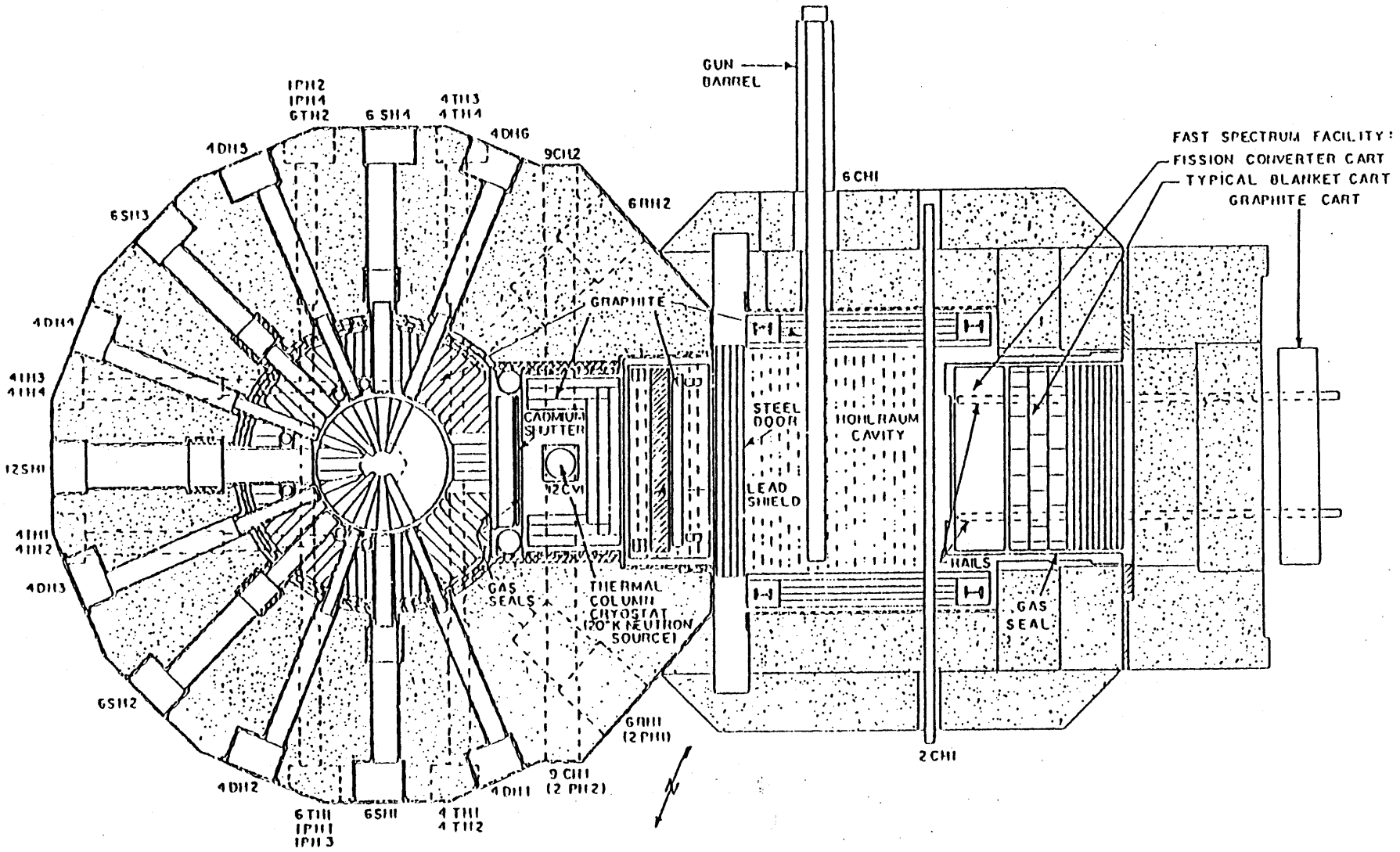

Figure 2. HORIZONTAL CROSS SECTION THROUGH REACTOR, THERMAL COLUMN, AND FAST SPECTRUM FACILITY

form argon-41. Argon-41 is a beta (1.20, 2.49 MeV) and a gamma (1.29 MeV) emmiter, with a half life of 1.83 hours (110 minutes). It is the major radioactive effluent from all research reactors, since it is impossible to eliminate all influx of air to the high neutron flux regions of the reactor. Tritium is also emitted from the MITR-II, but in much smaller quantities.

At present, the emission of argon-41 from the MITR-II results in a dose of about one mrem (millirem) per year to the maximally-exposed individual living in the vicinity of the reactor. Current Nuclear Regulatory Commission standards set the upper limit of such exposure at 500 millirem per year, so the MITR-II operates well within limits. In keeping with the radiation concept of ALARA (as low as reasonably achievable), however, one always tries to keep emissions as low as possible. Also, renewed interest in limiting emissions has come about as a result of an Environmental Protection Agency proposal, 40 CFR 61, which suggests that emissions be restricted "to that amount that would cause a dose equivalent of 10 mrem per year to any organ of any individual living nearby."(2) If such a ruling were to come into effect, the MITR-II would be operating at one-tenth of the limit, rather than at one-five hundreth.

1.2 Systems to Limit Argon-41 Production

There are three systems presently incorporated to limit the production of argon-41 in the MIT reactor. The first is an off-gas system which provides a continuous flow of air in the void space over the primary water pool. Fresh air enters the void through an

absolute filter, at the rate of 5.5 cubic feet per minute, and is discharged to the main ventilation system through a radiation monitor and a storage tank. While this system does limit argon-41 production to some extent, it also serves to control radioactive gaseous nuclides that may be released from the primary coolant and to dissipate radiolytic hydrogen formed in the coolant.

The second major system is the graphite helium system. The graphite reflector is constructed of a series of reactor-grade graphite stringers, and there are many void spaces between and around the stringers. These voids could potentially be filled by an influx of air, so the graphite is blanketed by an inert helium cover gas in order to help prevent air from entering the region. The helium is supplied to the graphite region through a constant pressure gasholder at the rate of approximately four cubic feet per hour. The helium is exhausted to the main plenum through the pipe tunnel that runs beneath the reactor.

Finally, other potential sources of air influx are purged with carbon dioxide. The carbon dioxide is supplied to a variety of experimental facilities through regulated flow meters. These facilities are: all port boxes, ports 4TH1, 6TH1, 12SH1, and 6SH4, the instrument ports, and the 3GV facilities (Figure 3). Carbon dioxide is also supplied to the space between the upper and lower annular rings, and to the two large aluminum boxes which were added to the reactor at the time of its modification in 1974. These boxes fill voids in the thermal column and lead shutter region (Figure 4) to prevent the accumulation of air in these spaces.

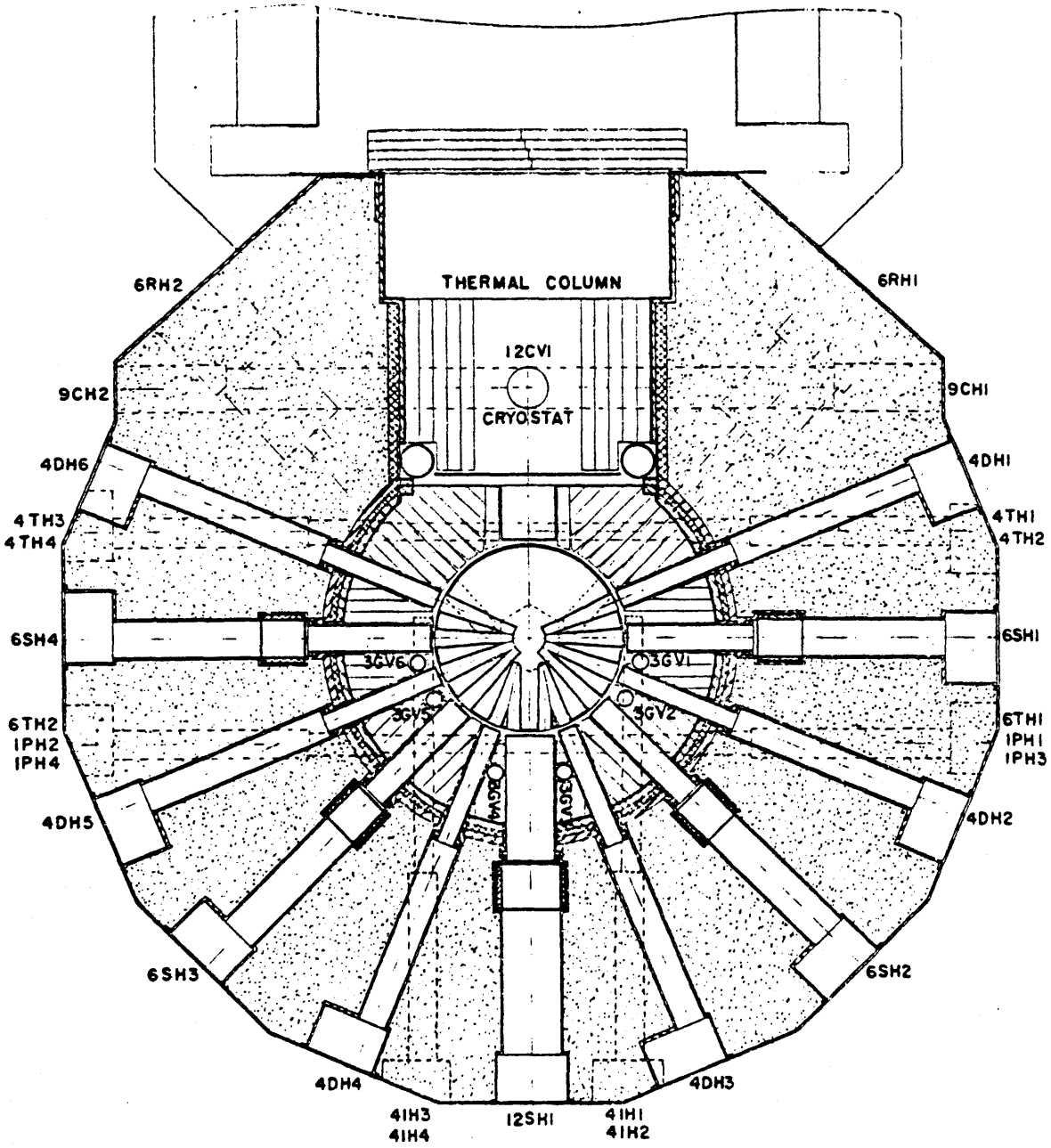

Figure 3. Another cross-sectional view of the MITR-II, showing the various experimental ports.

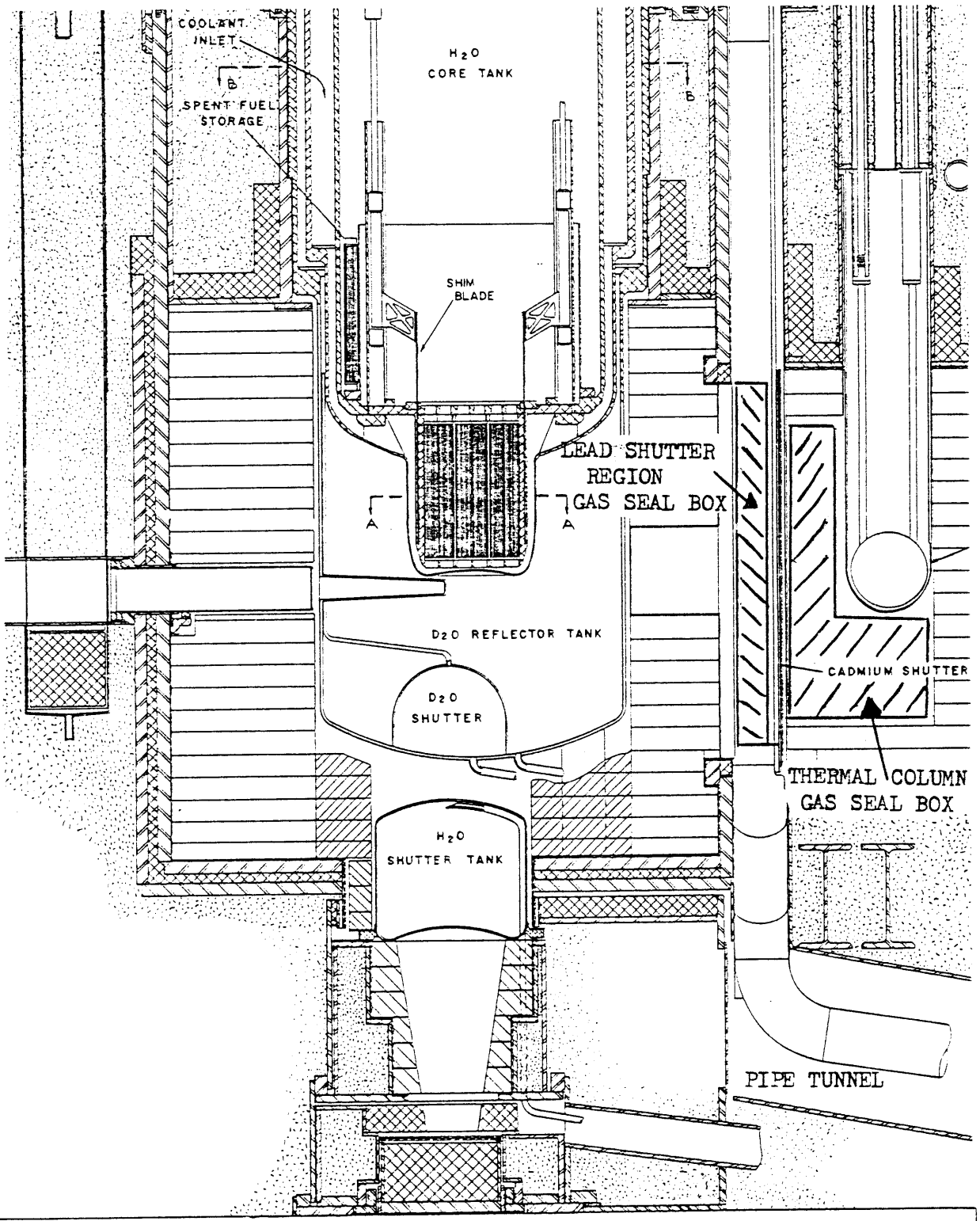

Figure 4. Cross-section of the reactor, showing the location of the lead shutter region and thermal column gas boxes. The boxes are continuously purged with carbon dioxide to prevent air from entering the spaces.

1.3 Scope of this Work

As was discussed, these systems do a good job of limiting argon-41 production, but any or all of them might be improved. It is also possible that argon-41 is being produced in some part of the reactor that is not being treated with an inert gas, such that providing a carbon dioxide or helium purge might be a simple way of improving the situation.

Although there were "feelings" among reactor operations and radiation protection personnel about what might be the major contributors to the argon-41 source term, no hard data was available to substantiate these suppositions. The scope of this thesis, then, includes the identification of all sources of argon-41 production, and the quantification of how much argon-41 is contributed by each region. This enables the most significant sources to be labeled with confidence. Although this initial work has emphasized forming a clear description of the problem, recommendations for possible means of improving the situation have also been put forth. Based on what has been learned, an outline of possible directions for future work has been developed.

2. QUANTIFICATION OF POTENTIAL ARGON-41 SOURCES

2.1 Characterizing the Ventilation System

Since all of the air in the reactor containment eventually must pass through the plenum on its way out the stack, it was at the plenum that the first gas samples were collected. Using the known concentration of argon-41 in the plenum gas, and the known flow rate of air through the plenum, the total argon-41 source term was determined, in microcuries per minute. One could then "trace back" from the plenum and quantify all of the individual components which eventually dump into the plenum.

This determination is not as straightforward as it might first appear. For example, all sources of argon within the reactor are subject to reactor power, so concentrations must be normalized to some standard power level (4.9 megawatts, for the data included in this thesis). Also, flow rates in the various ventilation ducts are not always precisely constant, and even the original determination of flow is subject to some degree of error. Measurements and air samples often assume that "equal mixing" has taken place, and this may not be an appropriate assumption, especially where plenum samples are concerned, since air is being exhausted into the plenum from many different locations. In general, because the reactor is such a complex and interconnected unit, samples may vary slightly from day to day, or even within a single day. This fact must be kept in mind when reviewing the experimental data. When error bars are indicated in the tables and figures, these represent only statistical

variations. Table 1 indicates variations in values that are due simply to typical fluctuations in reactor conditions. All these samples were taken when the reactor was running at full power, 24 hours or more after start up.

Figure 5 shows a schematic diagram which illustrates the various systems that exhaust into the plenum. (This figure is just a simplified version of the ventilation diagram that is included in the MITR Operations Manual (Figure 6).) Using a thermal velocity meter, or, where possible, a pitot tube system, air flows were determined for each of the components. Measurements were performed by personnel from MIT's Industrial Hygiene Office, and the accuracy of the instruments is approximately ±1%. However, because of the variations discussed in the previous paragraphs, these values are estimated to be within 10% of the actual values in the reactor at any given moment in time.

2.2 Sampling Procedures

Once values for the various air flows were obtained, samples were taken to obtain concentration data for each of the components. The basic sampling apparatus consists of an airtight chamber with an inlet and an outlet penetration that can be sealed. Initially, samples were collected in 333 ml polyethylene jars (Figure 7) which were then analyzed on a germanium-lithium crystal detector with a Canberra model 8180 multichannel analyzer. The gamma analysis was an important initial step, since it was possible that argon-41 would not be the only radioisotope present in any given sample, and gamma

TABLE 1

Typical variations in argon-41 concentrations at various locations in the reactor. All data is expressed in units of μCi of argon-41 per milliliter; reactor power, 4.9 megawatts; graphite helium flow rate approximately 4 cubic feet per hour. (Data taken from Reactor Radiation Protection Office files.)

DATE	STACK	REACTOR FLOOR	CONTROL ROOM
11/17/83	1.36×10^{-4}	-----	1.18×10^{-6}
11/22/83	1.23×10^{-4}	1.47×10^{-7}	8.70×10^{-7}
12/01/83	1.39×10^{-4}	1.62×10^{-7}	6.90×10^{-7}
12/08/83	1.49×10^{-4}	4.40×10^{-8}	5.16×10^{-7}
12/15/83	1.36×10^{-4}	8.80×10^{-8}	4.85×10^{-7}
12/22/83	1.46×10^{-4}	1.62×10^{-7}	9.70×10^{-7}
12/29/83	1.69×10^{-4}	7.35×10^{-8}	6.03×10^{-7}
01/05/84	1.44×10^{-4}	1.03×10^{-7}	7.20×10^{-7}
01/12/84	1.45×10^{-4}	4.41×10^{-8}	1.01×10^{-6}
01/19/84	1.36×10^{-4}	-----	1.02×10^{-6}
AVERAGE, $\pm \sigma$	1.42 ± 0.11 ($\times 10^{-4}$)	1.03 ± 0.46 ($\times 10^{-7}$)	8.06 ± 2.26 ($\times 10^{-7}$)

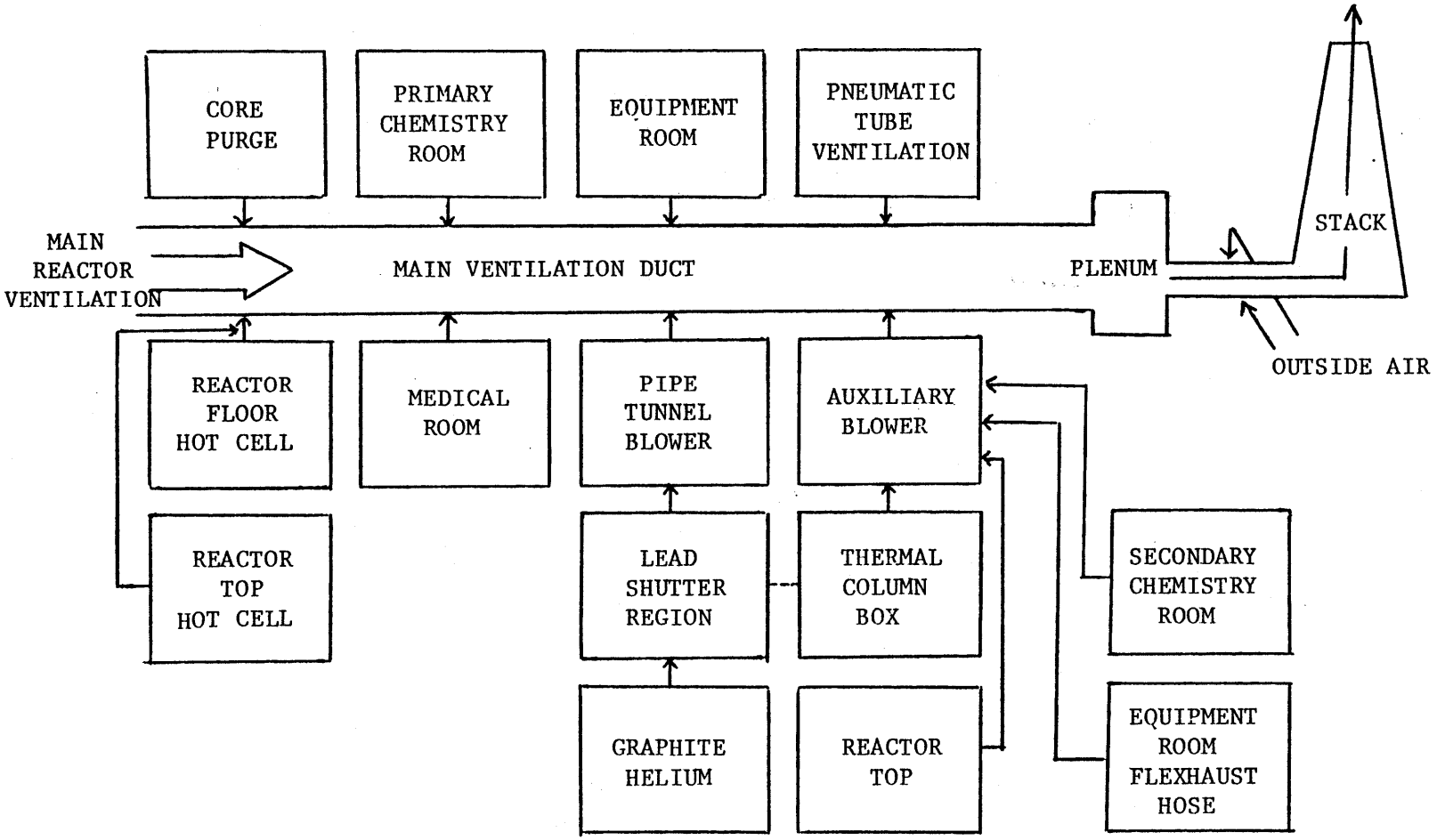

Figure 5. Schematic diagram of the MITR-II ventilation system. The dotted line between the Lead Shutter Region and the Thermal Column Box indicates a possible connection, in the event that there is a leak in the Thermal Column Box.

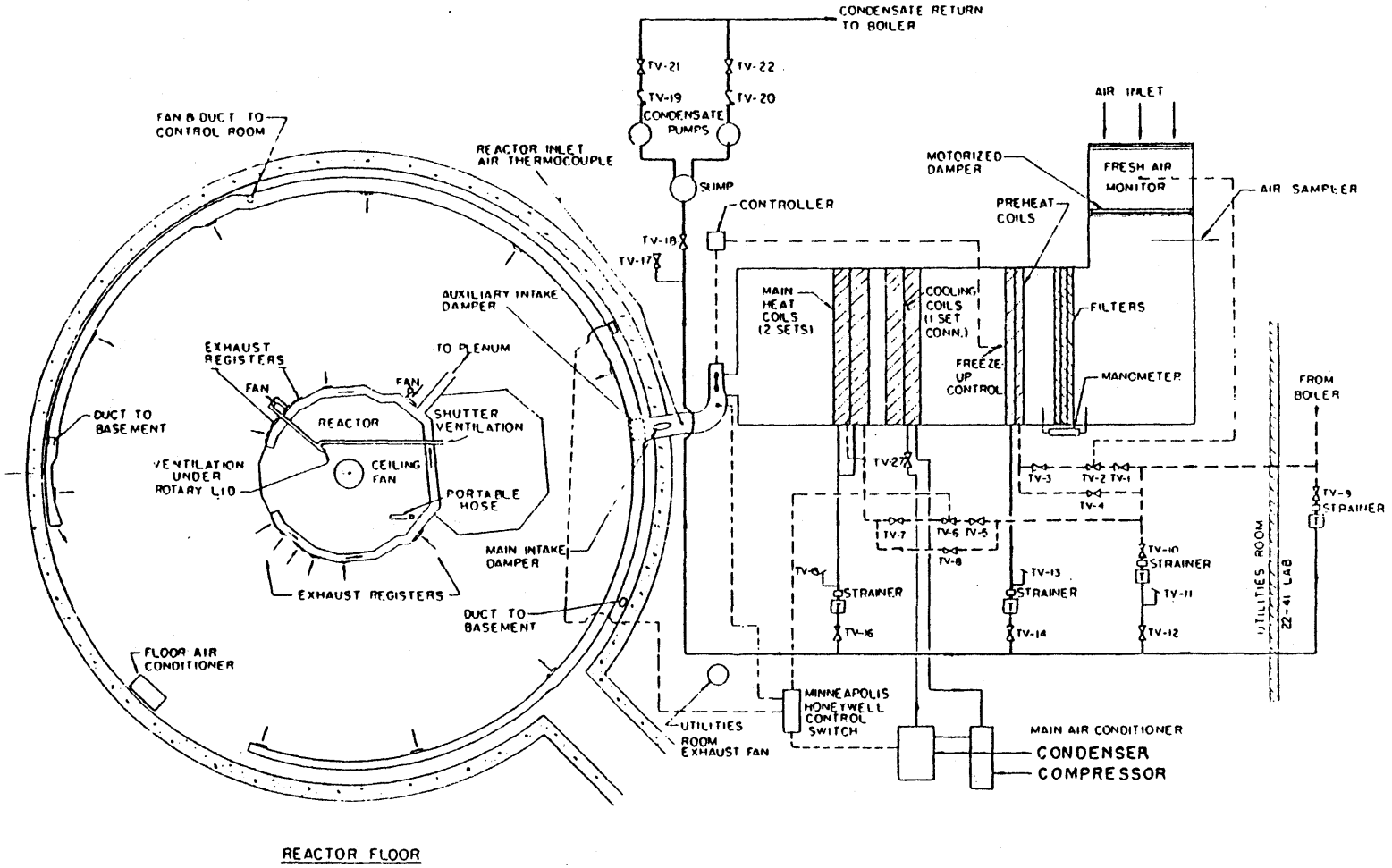

Figure 6a. MITR-II ventilation system, reactor floor plan.

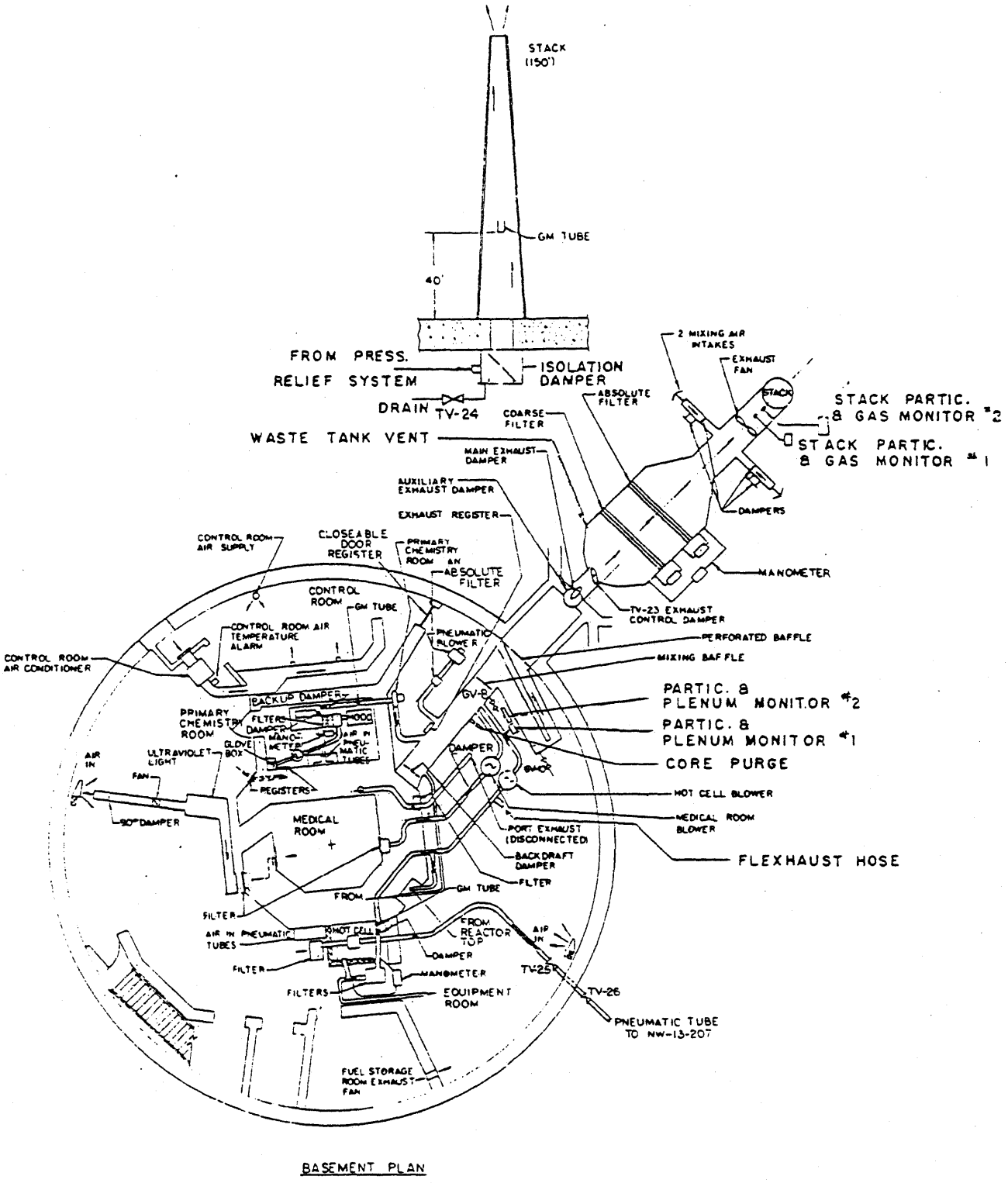

Figure 6b. MITR-II ventilation system, basement floor plan.

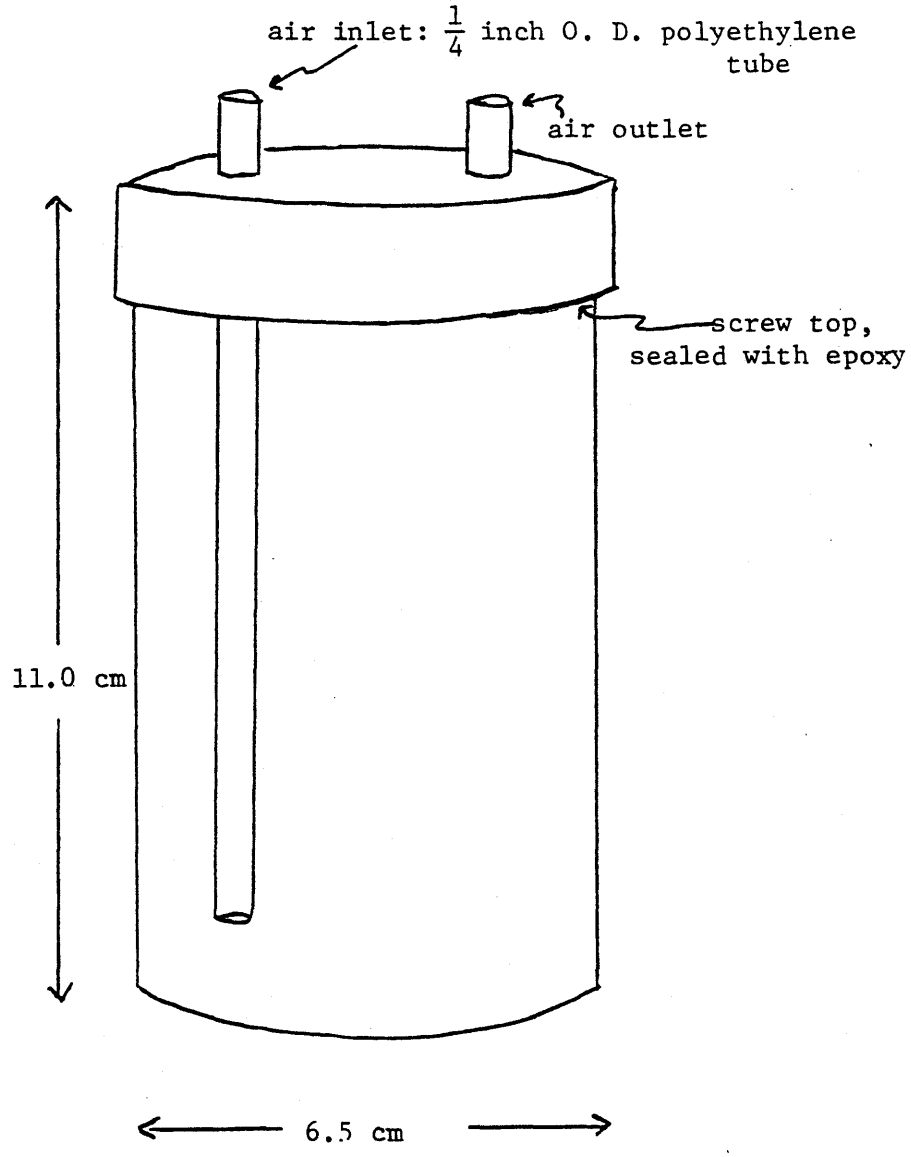

Figure 7. 333-milliliter Nalgene polyethylene jar, used to collect air samples for gamma analysis. After the jar is filled, inlet and outlet penetrations are sealed with rubber caps.

spectroscopy permits the isolation of argon-41 activity. The total activity in the jar was calculated, and then the concentration could be determined by simply dividing the total activity by the volume of the jar, and correcting for radioactive decay. Calibration procedures are detailed in Appendix A, and the total counting efficiency for this geometry is 0.022%.

A second, more sophisticated sampling chamber provided more sensitive results, but was only useful for samples which contained beta activity from only argon-41. This chamber (Figure 8) is a sealed stainless steel canister, 1125 ml in volume, which houses a one-cm diameter beta-sensitive Geiger tube. The cable to the tube is external to the canister, and can be connected directly to a preamp (Mechtronics Nuclear #502003), an amplifier (CI amplifier, model 1415), and a scaler (Tennelec TC 545A counter/timer, serial number 360), or to a count rate meter (Baird Atomic ratemeter model 441A, RPL #237) to determine the activity of the argon-41 in the canister. In both cases, the high voltage supply was set at 840 volts. A cross-correlation with the 333 ml jar gamma calibration permitted the determination of sample concentration from knowledge of net counts per minute in the canister.

For all sampling operations, the basic procedure was the same. In some locations, such as for the plenum and the core purge gas, a samplng station already exists. For other ducts, holes were drilled in the ductwork and a short piece of 1/4 inch outer diameter copper tubing was fed into the hole. The tubing was connected to the sample container via 1/4 inch outer diameter heavy-walled rubber hose. Duct

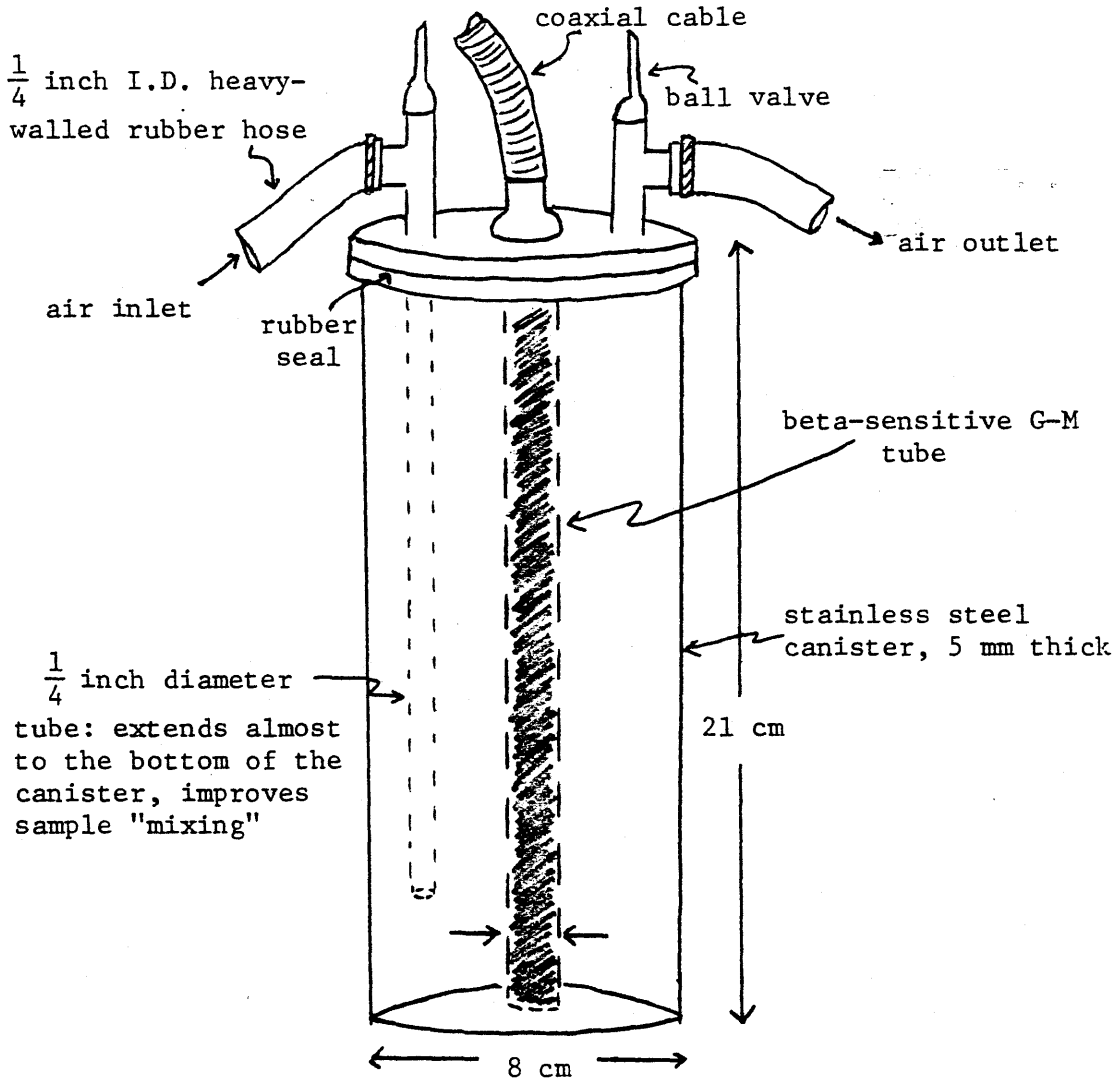

Figure 8. RPO Chamber #2. This 1125-milliliter stainless steel canister is used for collecting air samples for beta-analysis. A beta-sensitive Geiger-Muller tube, manufactured by Anton Electrical Labs, Inc., is mounted inside the canister. The associated coaxial cable passes through the lid of the canister and can be connected directly to a scaler or count rate meter.

tape was used to seal the ductwork as completely as possible. Air was then continously pumped through the sample chamber at the rate of 1.6 liters per minute. Portable air pumps were employed, manufactured by Bendix, type C115, serial number 1302 for all samples except the graphite helium and core purge samples, and serial number 1304 for the helium and core purge. In all cases, the pump was continuously run until a complete air exchange occurred (at least two minutes for the 333 ml containers, and three minutes for the 1125 ml canister). The exception to the 1.6 liter per minute pumping rate occurred for the plenum samples, which are pumped at about 10 liters per minute, since there is already a sampling station in place that uses the plenum pump as a means to draw air through the canister, and then re-exhaust it back into the plenum. Sampling configurations are illustrated in Figure 9.

2.3 Concentration Data

Using the procedures described above, concentrations were determined for each of the components that exhausts into the plenum. Multiplying these concentrations by the air flow rates and appropriate conversion factors yields a source term value, in microcuries per minute, for each component. These results are summarized in Table 2. All indicated activities are for argon-41. Beta counting was used to obtain increased sensitivity for low-activity samples, but only after gamma analysis showed that argon-41 was the only source of activity. It was immediately clear, after the initial round of samples was analyzed, that the vast

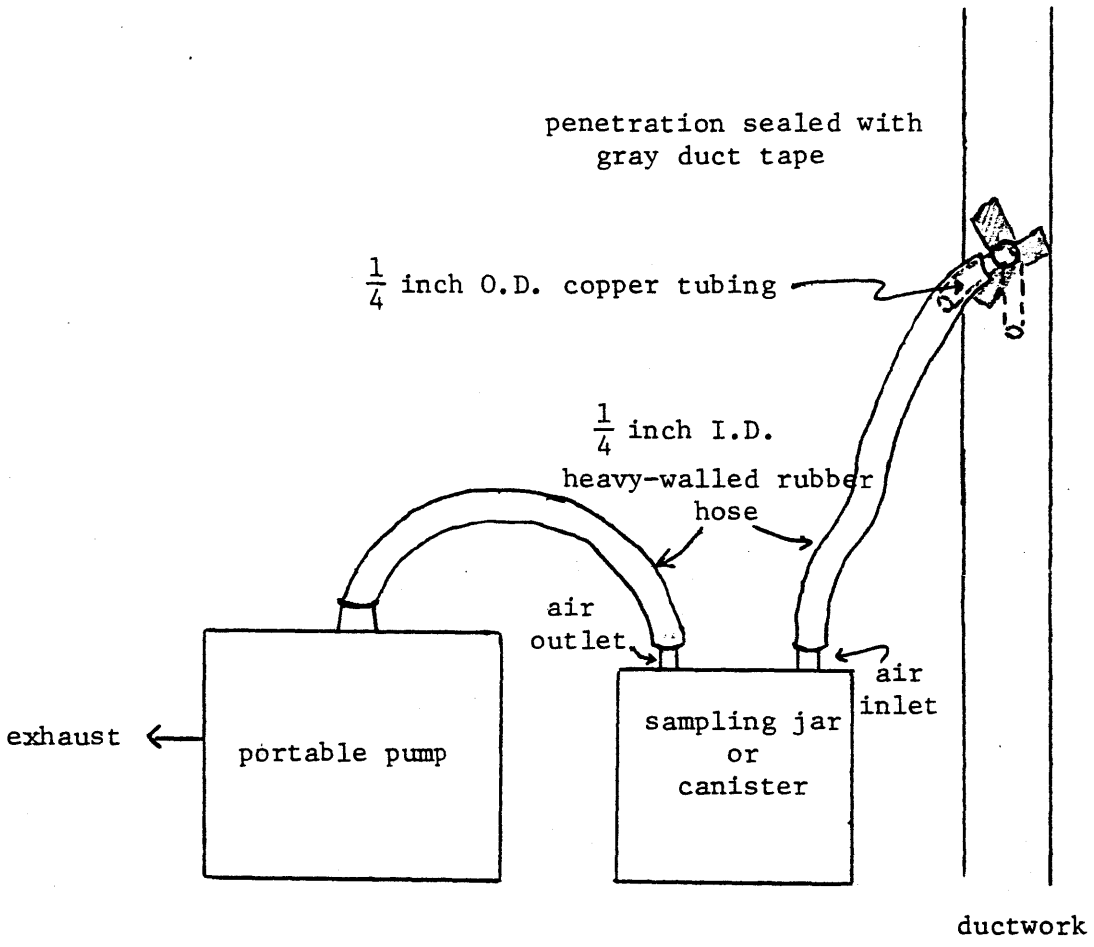

Figure 9. General configuration for procuring a grab sample. The pump would simply exhaust to the room with the exception of the plenum samples, where air was exhausted back into the plenum, and the graphite helium samples, where air was exhausted into the sucker hose on the reactor top.

TABLE 2

Contributions to the argon-41 source term in the MITR-II. Estimates of error in sample concentrations are based solely on counting statistics. Note that 86% of the total source term is contributed by the pipe tunnel blower (*).

SOURCE	AIR FLOW RATE (ft^3/min)	SAMPLE AR-41 CONC. (μCi/ml)	SOURCE TERM (μCi ^{41}Ar/min)
CORE PURGE	5.5 ± 0.3 (c)	$7.20 \pm .07$ ($\times 10^{-3}$) (d)	$0.112 \pm .006$ ($\times 10^{4}$)
PRIMARY CHEMISTRY ROOM	766 ± 38 (a)	$3.20 \pm .45$ ($\times 10^{-7}$) (e)	$0.001 \pm .000$ ($\times 10^{4}$)
EQUIPMENT ROOM	880 ± 47 (b)	$7.42 \pm .49$ ($\times 10^{-6}$) (e)	$0.018 \pm .002$ ($\times 10^{4}$)
PNEUMATIC TUBES	----	special	small pulse of argon when opened
REACTOR FLOOR HOT CELL	450 ± 23 (b)	$1.99 \pm .30$ ($\times 10^{-7}$) (e)	$0.000 \pm .000$ ($\times 10^{4}$)
MEDICAL ROOM	587 ± 29 (b)	$4.89 \pm .59$ ($\times 10^{-6}$) (d)	$0.008 \pm .001$ ($\times 10^{4}$)
PIPE TUNNEL BLOWER	32 ± 2 (a)	$4.43 \pm .04$ ($\times 10^{-2}$) (d)	$4.01 \pm .20$ ($\times 10^{4}$) *
AUXILIARY BLOWER	739 ± 20 (b)	$1.56 \pm .03$ ($\times 10^{-4}$) (d)	$0.326 \pm .019$ ($\times 10^{4}$)
MAIN REACTOR VENTILATION	1650 ± 165 (c)	$1.83 \pm .92$ ($\times 10^{-7}$) (e)	$0.001 \pm .0005$ ($\times 10^{4}$)
Σ INPUTS =	5110 ± 180	------	$4.48 \pm .20$ ($\times 10^{4}$)
ACTUAL PLENUM DATA =	5100 ± 510 (b)	$3.23 \pm .06$ ($\times 10^{-4}$)	$4.66 \pm .47$ ($\times 10^{4}$)

(a) Data obtained with a Kurz Air Velocity Meter, model 440, serial number 314
(b) Data obtained with an F. W. Dwyer standard 18-inch pitot tube, MIT ID# 7620-391
(c) Data obtained from reactor operations records
(d) Sample analyzed for gamma component with GeLi detector
(e) Sample analyzed for beta component with beta-sensitive G-M tube

majority of argon-41 that is exhausted into the plenum is produced by some region serviced by the pipe tunnel blower, which draws air through the pipe tunnel that runs beneath the reactor core.

Backtracking from the pipe tunnel blower on the ventilation diagram, three potential sources of argon-41 were isolated. First, any argon generated in the lead shutter region (Figure 4) would appear in the pipe tunnel blower ductwork. Furthermore, if there were a leak in the thermal column box, this might also appear. But, the primary source of activity was found to be in the helium cover gas that blankets the graphite reflector. While this cover gas is intended to purge air from the system, it obviously does not do a complete job. The problems associated with the presence of even a small amount of air are understandable since, near the core, the helium/air flows through a relatively high flux region, of about 10^{13} neutrons per square centimeter per second. This situation permits efficient generation of argon-41.

With the knowledge that the helium cover gas is the main source of argon-41 generation, work was then carried out to find a means for decreasing the influx of air to this region.

3. REDUCING ARGON-41 IN THE PIPE TUNNEL

3.1 Adjusting Graphite Helium Flow Rate

To observe the effect of helium flow rate on the argon concentrations in various parts of the reactor, helium flow was increased and resultant samples were taken. Before discussing the specific results, several general facts should be presented.

3.1.1 General Observations

(1) No flow meter is attached to the grahite helium, nor could a suitable one be obtained during the course of these experiments. Flow is therefore calculated based on the refill rate of the helium gas holder, which refills after each time 20 cubic feet of helium has been exhausted.

(2) The data which follow were taken during a week that began with a "flushing out" of the graphite region by putting helium through at the rate of one refill per hour (i.e. 20 cubic feet per hour) for at least three cycles prior to reactor start-up. Normally, the helium is cycled at the rate of one refill every 4.5 - 5.0 hours, or about 4 cubic feet per hour. The data illustrate that in the 24 hours following start-up, flushing out the system reduced argon-41 concentration in the plenum by a factor of 1.2, even though helium flow rate was returned to normal.

(3) Concentrations of argon-41 are intimately related to the time

that has passed since the helium flow was adjusted. Originally, it was thought that 4 to 6 hours would be a sufficiently long equilibrium time for the reactor to reach steady state after a change in flow rate was made, since a complete volume change of air within the containment itself takes only 3.6 hours. After initial samples yielded suspect results, this presumption was investigated more closely. Figure 10 shows the argon-41 concentration in the pipe tunnel as a function of time after the helium flow rate was changed from 10 to 20 cubic feet per hour. Whether this variation would be observed after all such changes is uncertain, but re-examination of early data indicated that, indeed, a trend was found to exist if one divided the samples into two groups---those taken within 10 hours of the change, and those taken at greater than 18 hours after the change. Data presented herein are a summary of data which were obtained after the reactor had approached equilibrium, 20 to 22 hours after the helium flow rate was changed.

The fact that the reactor takes so long to equilibrate seems to indicate that there might be pockets of air within the graphite region that take time to "migrate" into the main helium flow; or, that the helium is not evenly dispersed throughout the graphite region. This is related to the next point:

④ There is a sampling station at the top of the graphite region (valve SV-20, see Figure 11) that is used for procuring grab samples of helium. When helium samples were taken from this region during the course of experimentation, two facts were noted:

(a) The relationship between argon concentration in the grab sample

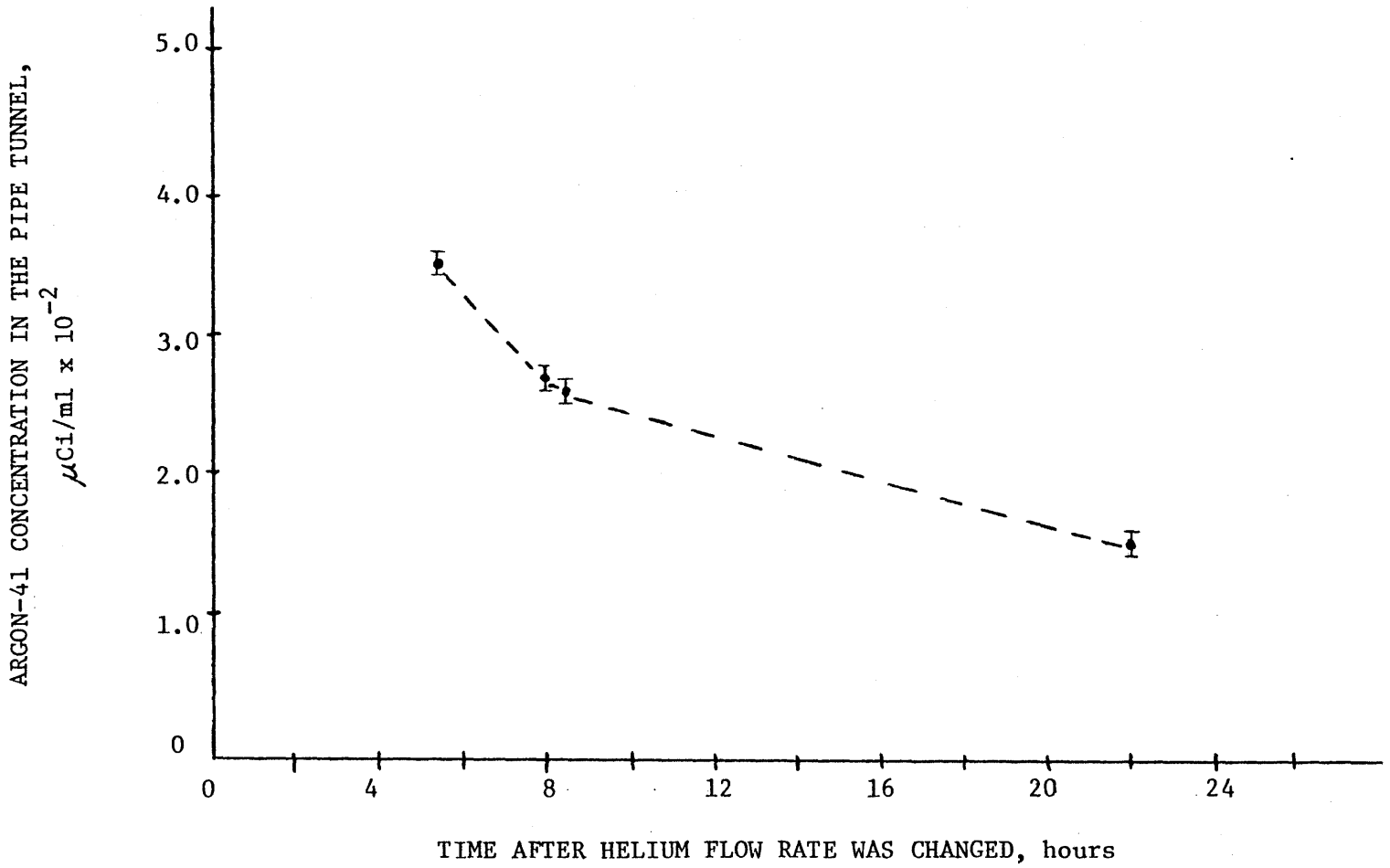

Figure 10. Argon-41 concentration in the pipe tunnel as a function of time after the helium flow was changed from 10 cubic feet per hour to 20 cubic feet per hour.

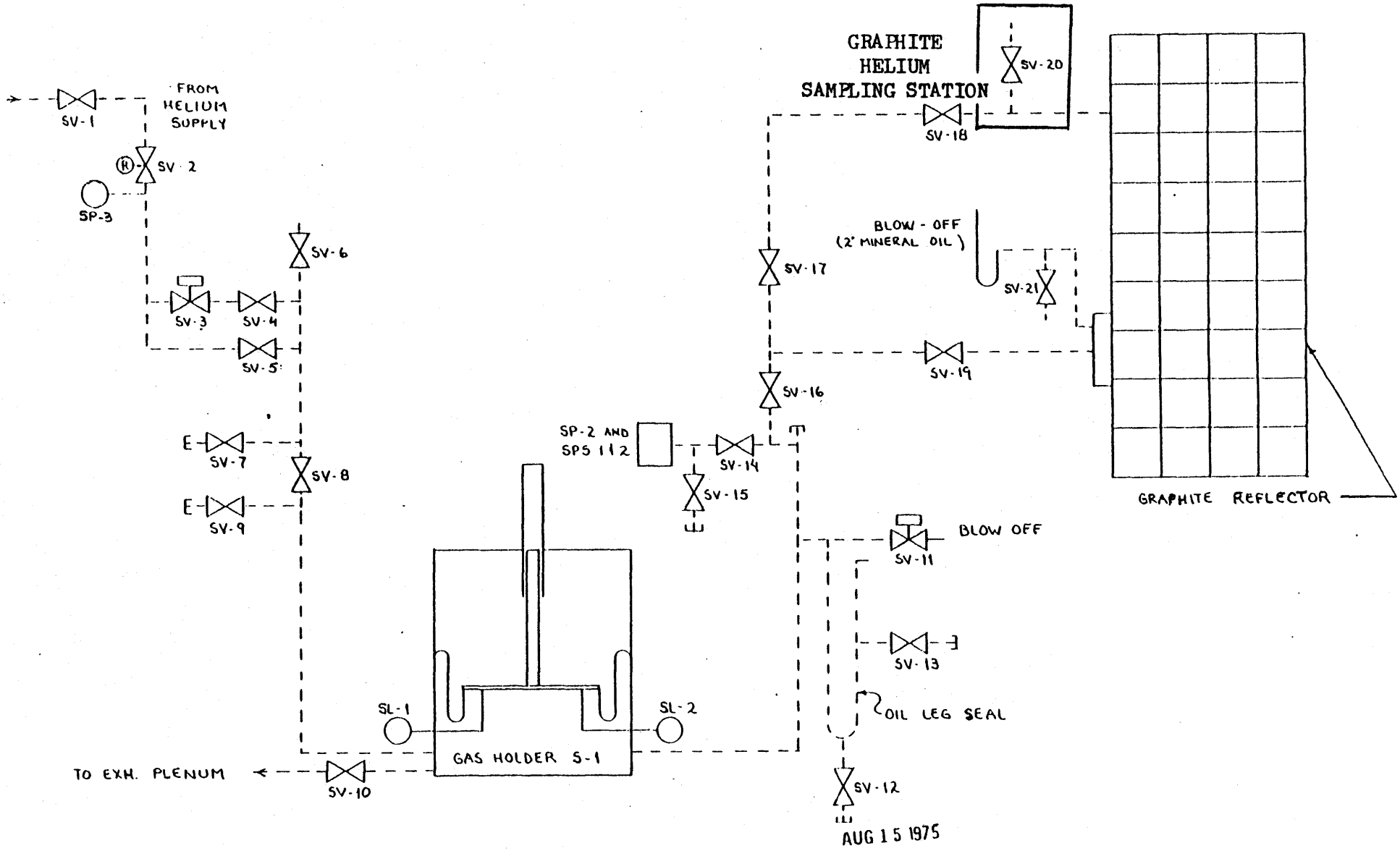

Figure 11. GRAPHITE HELIUM SYSTEM

and helium flow rate was not linear (Figure 12). Since the relationship indicated by the pipe tunnel blower samples is clearly a linear one, this indicates that these helium samples are not representative of the true average argon-41 concentration in the helium cover gas.

(b) Also, if one considers the total output of the reactor to be about two curies of argon per hour, then this sample accounts for only about 10% of the stack output.

In summary, this seems to indicate that perhaps helium does not flow efficiently (or evenly) throughout the graphite stringers that comprise the reflector.

(5) An unusual fact was observed by noting the brush recorder output of the stack gas 1 monitor in the reactor control room. This monitor registers the counts per minute that are detected by a pancake Geiger tube located in the base of the reactor stack. If one marks the appropriate times associated with refilling of the helium tank, one sees a dramatic rise in stack gas 1 counts within 15 minutes after the tank refills (Figure 13). This was seen for refill rates of 8.7 to 10.0 cubic feet per hour, and implies that perhaps the increased pressure of the gas holder immediately after refill may provide enough driving pressure to push out additional air from the small cracks and voids in the graphite region. This was not observed for helium flows of 20 cfh.

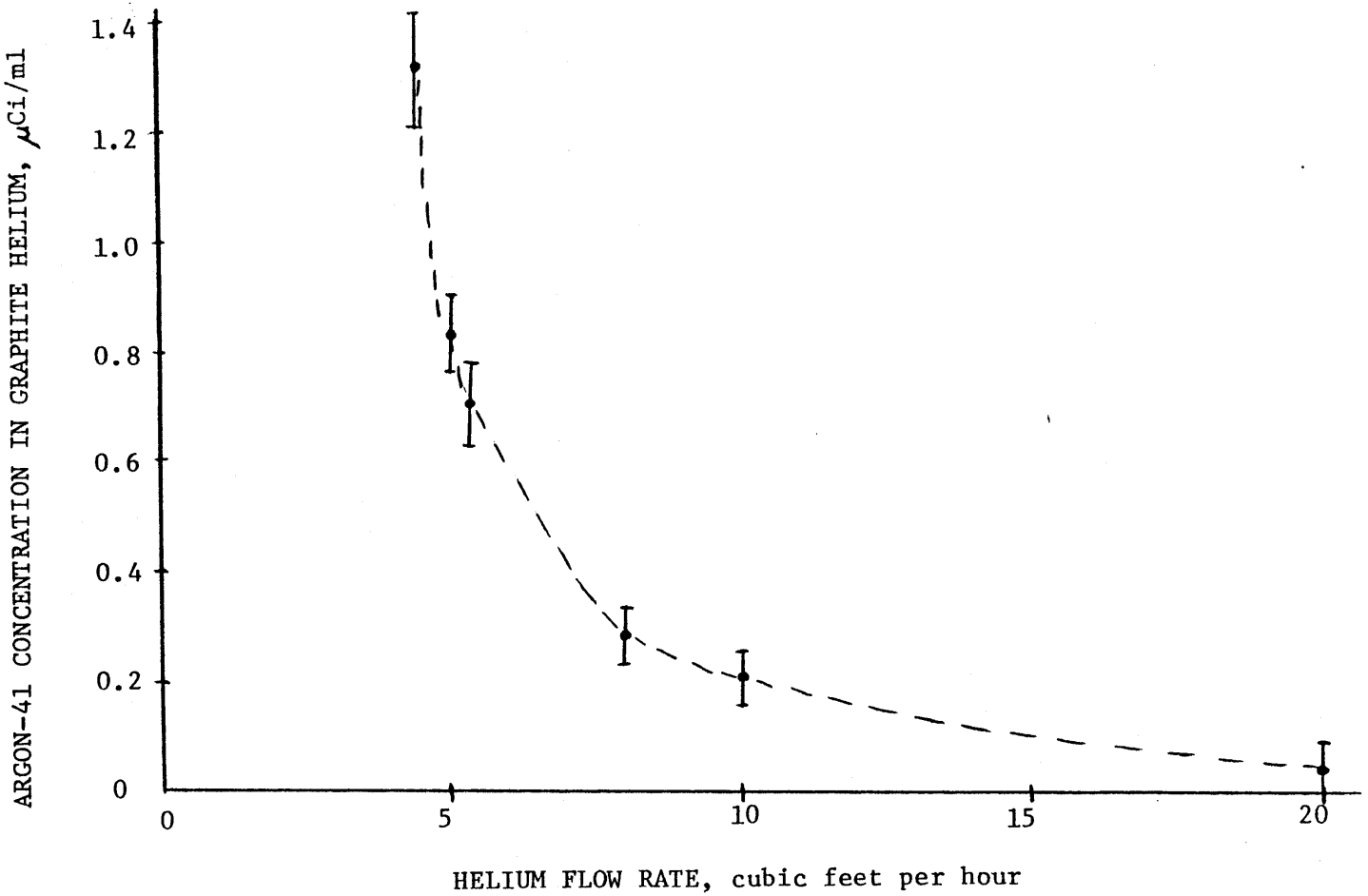

Figure 12. Argon-41 concentration in the graphite helium as a function of helium flow rate. Samples were taken at the SV-20 sampling station on the reactor top.

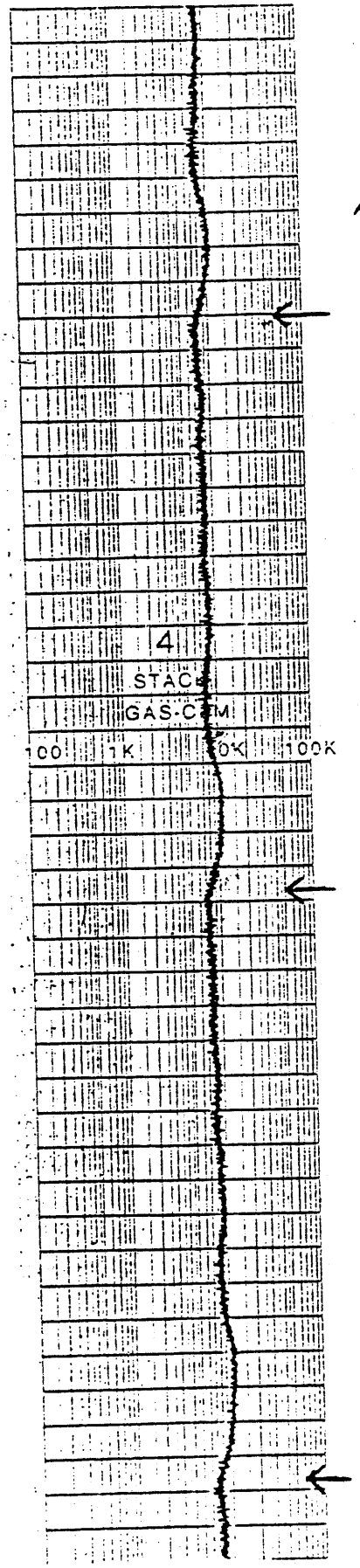

Figure 13. Actual data from the Stack Gas 1 brush recorder clearly shows an increase in stack counts shortly after the helium tank refilled (refill times are indicated by the arrows). The helium flow rate was 10 cubic feet per hour while this data was being recorded.

3.1.2 Specific Results

With the above discussions in mind, the results can be viewed with a critical eye. These data are important since they illustrate one way that the argon-41 production in the MITR-II can be significantly reduced.

Figure 14 shows the relationship between helium flow rate and the argon-41 concentration in the pipe tunnel. As the flow of helium through the graphite region is increased from 4 to 16 cubic feet per hour, there is a linear decrease in the concentration of argon-41 in the pipe tunnel. The data are fit by a line defined as:

$$\text{concentration} = -.16(\text{flow}) + 3.84$$

Figure 15 shows a similar reduction in plenum argon-41 concentration with increasing helium flow rate. This is to be expected, since the pipe tunnel is the main argon-41 contributor, and since the pipe tunnel exhausts directly into the plenum. The slope of this curve-fit line is -.14, a value that compares well to that for the pipe tunnel blower data.

The consequences of increasing helium flow are quite significant. For example, Figure 15 illustrates that doubling the helium flow rate from 5 to 10 cfh reduces argon-41 concentration in the plenum by a factor of about 1.4. A further increase of the flow, however, yields an even more effective reduction: a doubling of flow rate from 10 to 20 cubic feet per hour reduces the concentration by a factor of approximately 3, from 2.0 to 0.6 microcuries per milliliter. Clearly much can be gained by increasing helium flow as much as possible, at least within the flow range of 5-20 cfh for

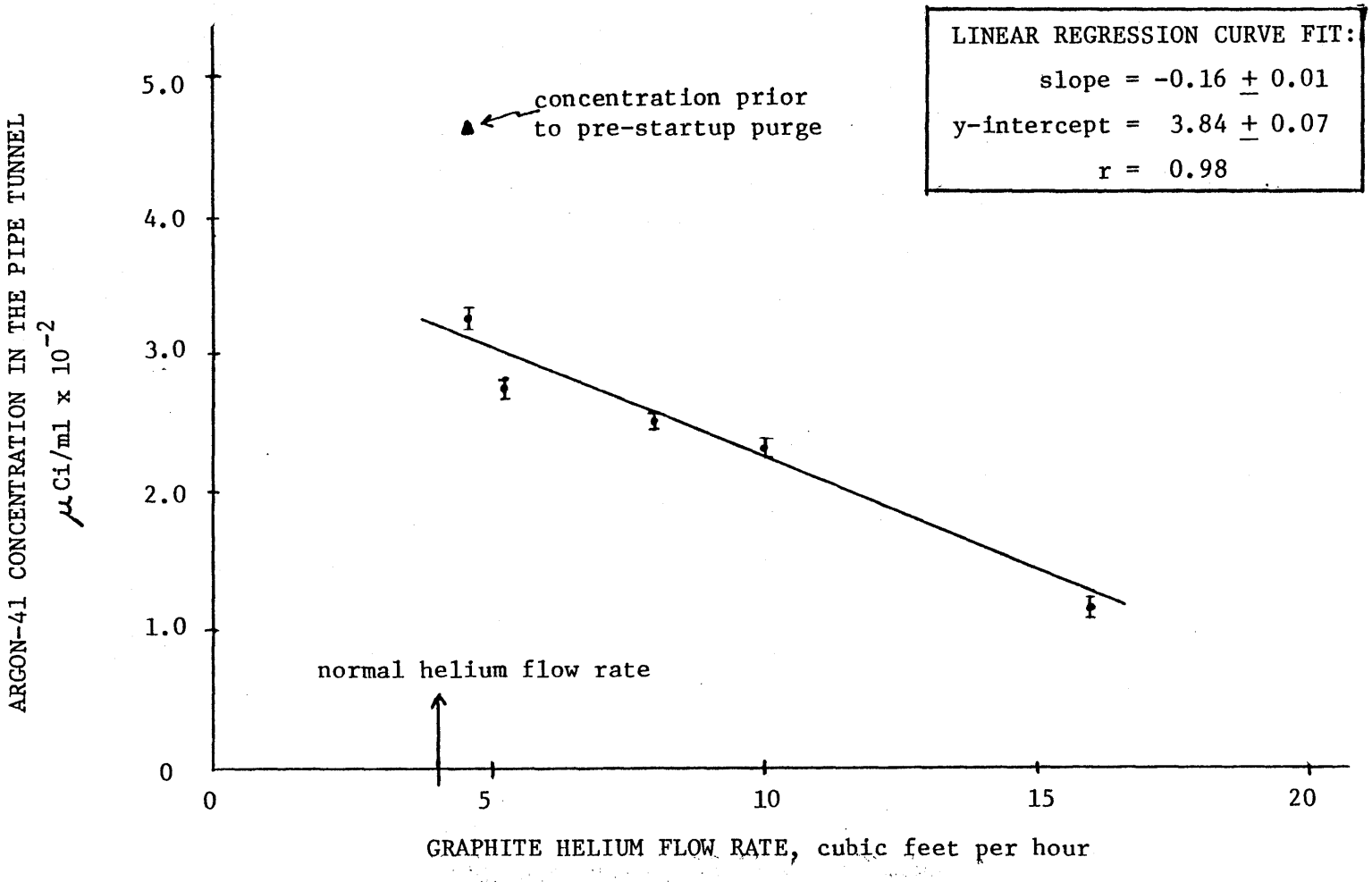

Figure 14. Argon-41 concentration in the pipe tunnel as a function of graphite helium flow rate. The value for r is a measure of the goodness of fit of the line, and is calculated from $r = \frac{m^2 \sigma_x^2}{\sigma_y^2}$

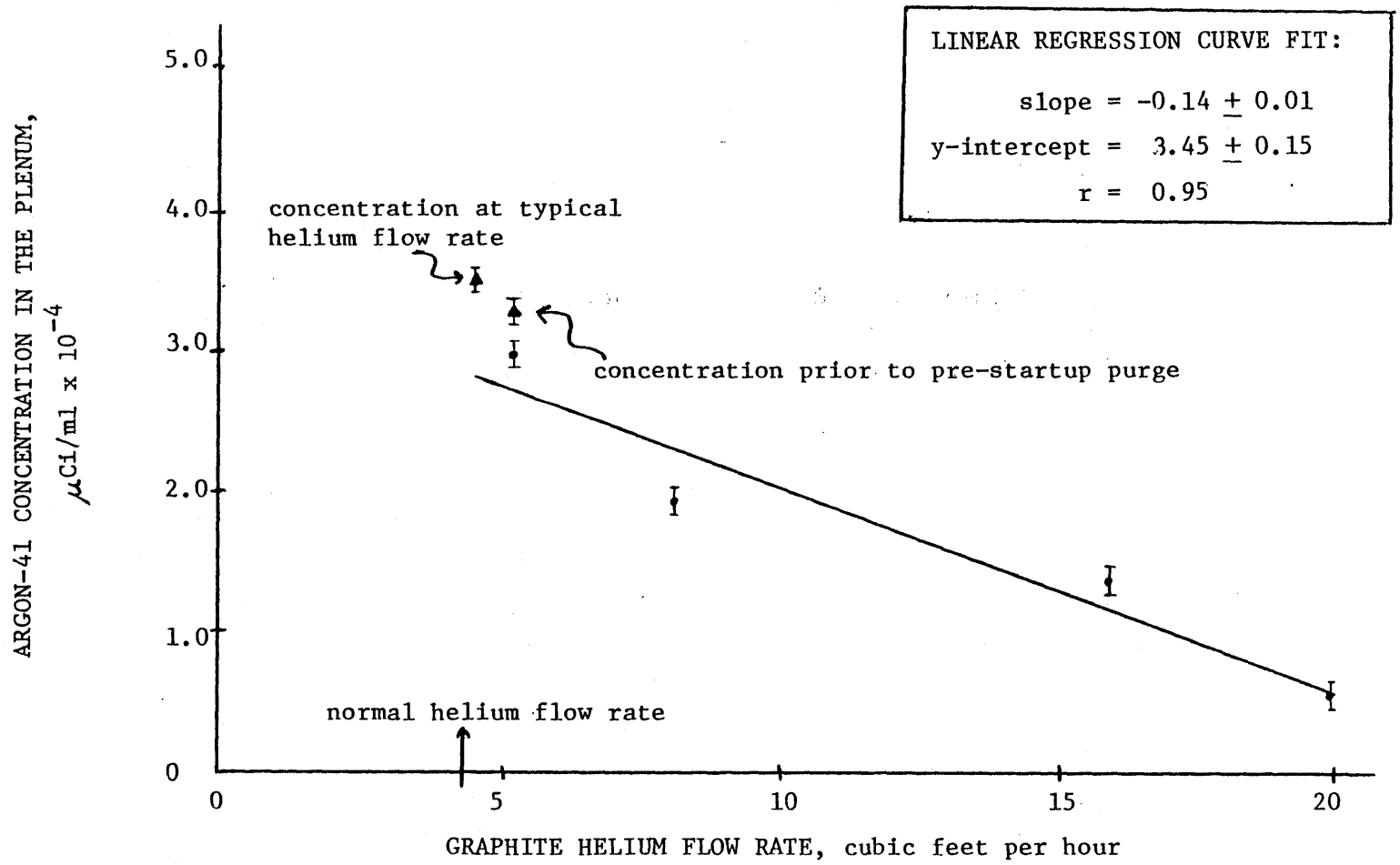

Figure 15. Argon-41 concentration in the plenum as a function of graphite helium flow rate. The value for r is a measure of the goodness of fit of the line, and is calculated from $r = \frac{m^2 \sigma_x^2}{\sigma_y^2}$.

which data is available.

Samples were also taken to determine the effect of increased helium flow on argon concentrations in the control room, equipment room, and on the reactor floor. Equipment room samples followed the same trend as those for the plenum and for the pipe tunnel (Figure 16) and no significant change in reactor floor and control room concentrations was observed.

3.2 Effects of Pipe Tunnel Blower Flow

During the past years of the reactor's operation, radiation protection and operations personnel have noted that if the pipe tunnel blower flow is decreased, a significant increase in argon-41 concentration is noted on the reactor floor and in the control room. Accompanying this increase is a decrease in stack gas concentration. Presumably, if the blower does not exhaust the air from the graphite region quickly enough, the argon is able to migrate through the region and escape into the containment.

This thesis research attempted to investigate this phenomenon by using a variety of pipe tunnel blower settings. These "settings" are actually measures of the differential pressure across the orifice of the valve which is located in the ductwork that immediately precedes the blower itself. Figure 17 shows the values of flow rate in the pipe tunnel blower as a function of the differential pressure. The manometer is connected such that an increased delta p implies a decreased flow rate.

As the settings were varied between 2.5 and 3.0 inches of water,

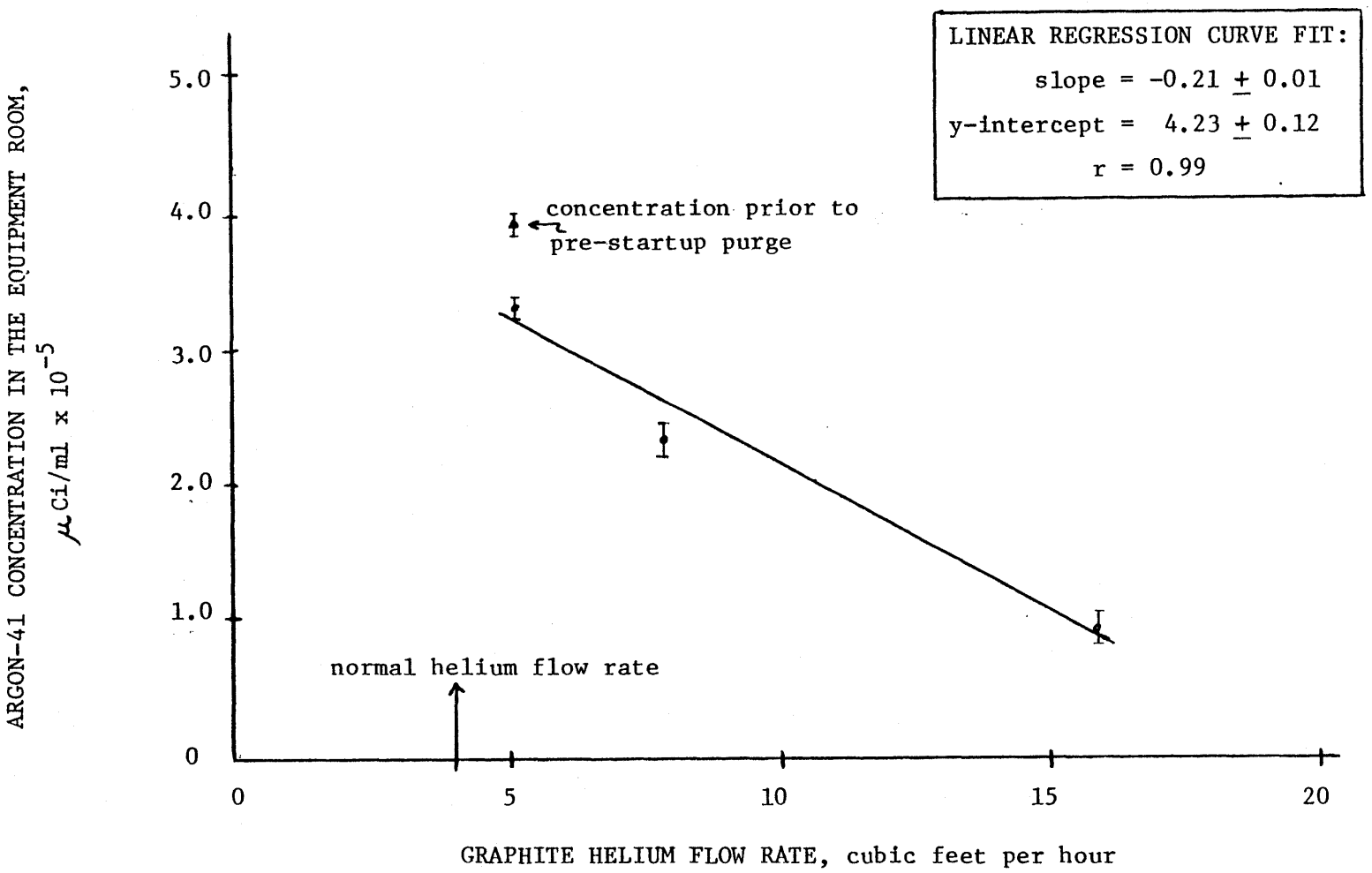

Figure 16. Argon-41 concentration in the equipment room as a function of graphite helium flow rate.

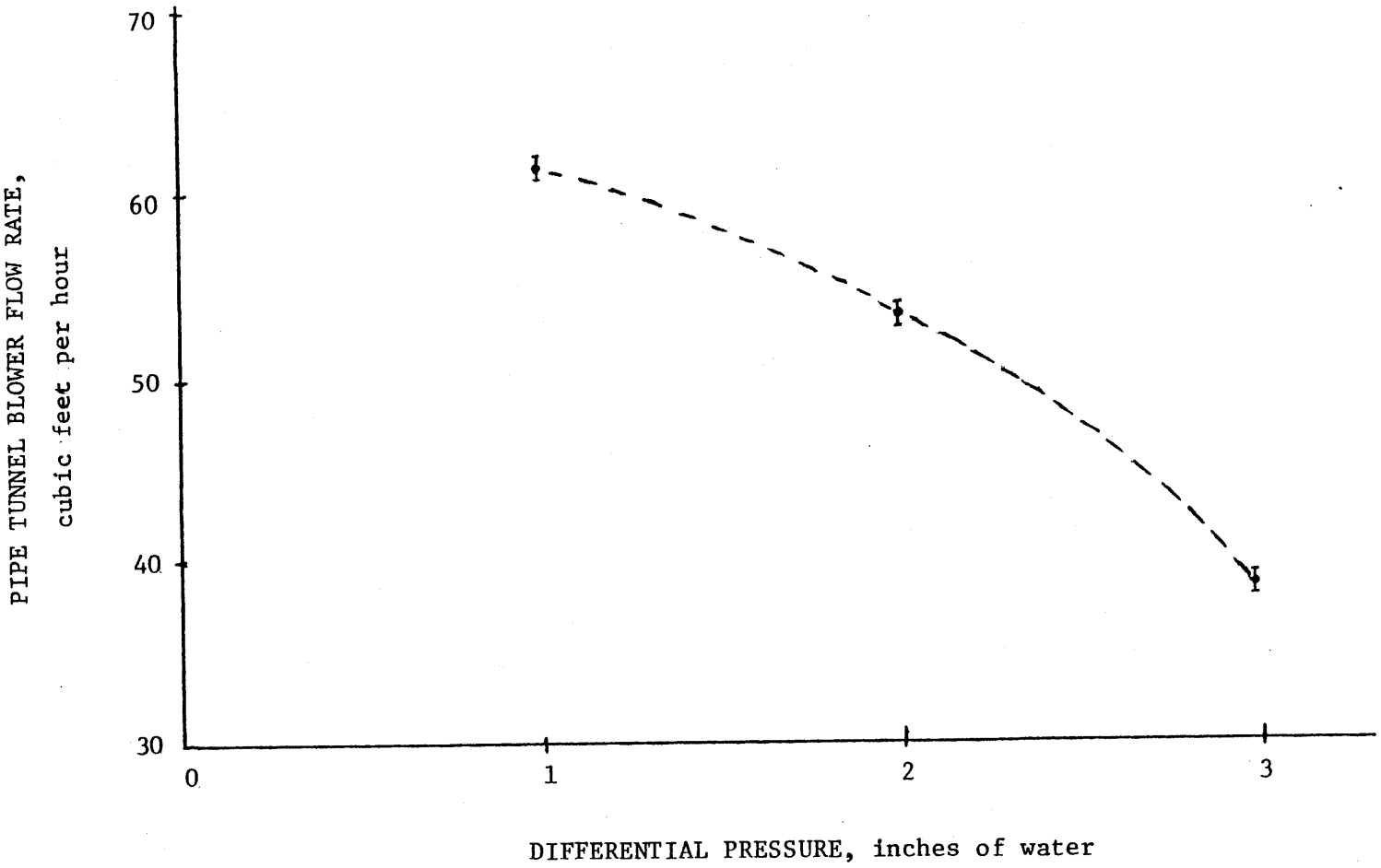

Figure 17. Pipe tunnel blower flow rate as a function of pressure drop across the blower valve, as measured with an in-line manometer.

grab samples from the control room, reactor floor, and plenum were taken and analyzed for the presence of argon-41. Samples were taken five hours after the blower setting was changed. Longer equilibrium times were not used in order to comply with a request by reactor operations staff that experimentation go on only during the day, in case the argon-41 concentraton in the containment were to rise dramatically.

The results for the plenum samples are shown in Figure 18. There is a clear decrease in plenum concentration with decreased pipe tunnel blower flow. Data from the other samples are contained in Table 3 for reference, but no trend was found in these numbers.

In retrospect, one must apparently wait for a period of time greater than five hours in order to allow the reactor to come to equilibrium. While one might expect to see a change in plenum concentration almost immediately (since the pipe tunnel blower dumps directly into the plenum), it will take longer before the effects are seen on the reactor floor or in the control room.

In summary, the stack output of argon-41 can be significantly decreased by decreasing pipe tunnel blower flow. However, this procedure is not recommended until further measurements can determine the effect of such an action on the reactor floor and control room argon-41 concentrations.

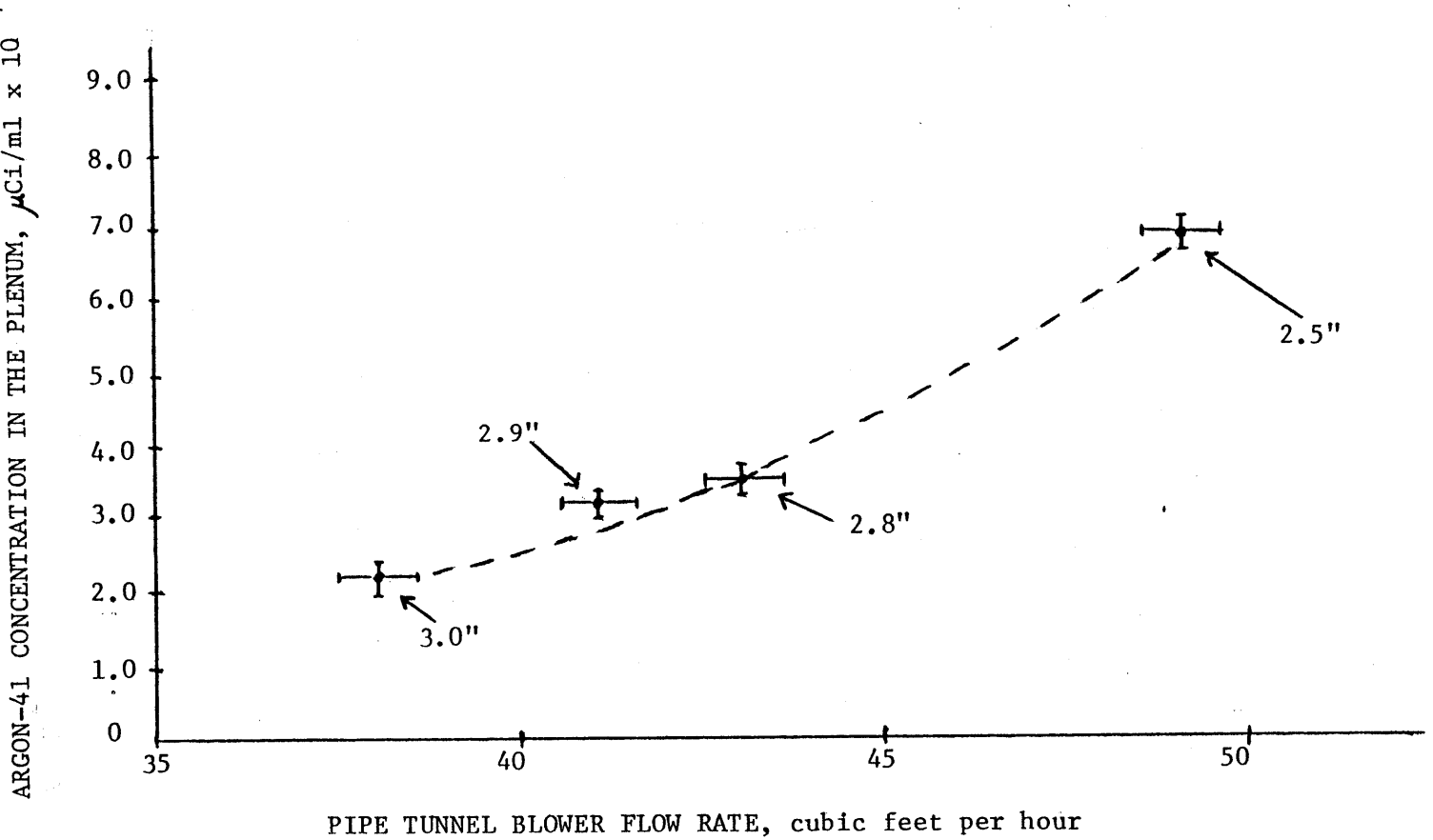

Figure 18. Argon-41 concentration in the plenum as a function of pipe tunnel blower flow rate. The numerical label associated with each data point indicates the corresponding setting of the pipe tunnel blower for each flow rate. This setting is a measure of the differential pressure across the blower valve, and is recorded in units of inches of water.

TABLE 3

Concentrations of argon-41 in various locations as a function of pipe tunnel blower setting. Values for pipe tunnel flow rates are approximate, based on Figure 17.

BLOWER SETTING (inches of water)	BLOWER FLOW RATE (cubic feet/hr)	REACTOR FLOOR (μCi/ml)	EQUIPMENT ROOM (μCi/ml)	CONTROL ROOM (μCi/ml)
2.8	43	2.09×10^{-7}	2.69×10^{-5}	6.83×10^{-7}
2.9	41	3.00×10^{-7}	3.85×10^{-5}	8.87×10^{-7}
3.0	38	1.31×10^{-7}	1.99×10^{-5}	3.02×10^{-7}

4. DISCUSSION

4.1 Summary of Results

The experimental work recounted herein clearly identifies the pipe tunnel, and more specifically, the graphite helium, as the main source of argon-41 production in the MITR-II. A linear relationship was found between graphite helium flow rate (in the range of 4 to 20 cfh) and plenum concentration. This relationship illustrates that increasing the helium flow has a significant impact on the reduction of argon-41 production.

Decreasing the flow in the pipe tunnel blower is another means for reducing argon-41 emission out the stack, but more experiments need to be performed to determine the long range effects of such an action on the argon-41 concentration on the reactor floor and in the control room.

4.2 Altering Helium Flow: Cost vs Benefit

What are the potential disadvantages of increasing helium flow? The main problems are those of helium availability and cost. Helium is a scarce natural resource, and its current retail price is $38.81 per 285 cubic feet (3). (This price includes a $6.23 surcharge per gas bottle.) The present helium flow rate in the reactor is about four cubic feet per hour, totaling 35,040 cubic feet per year, at a total annual cost of $4772. This total is based on the current practice, which permits the helium to flow seven days a week, even

when the reactor is shut down. Increasing the flow to 20 cubic feet per hour would decrease the argon-41 output by a factor of approximately five, but with a subsequent additional cost of $20,000 per year at current prices. Since helium is becoming increasingly scarce as a natural resource, this price could increase considerably in the future.

Several alternatives may be considered to offset these disadvantages and yet still result in a decreased argon-41 output. One possibility is to decrease or turn off helium flow when the reactor is not operating (i.e., on weekends or holidays). Air could be flushed out of the graphite region on Mondays by running through several cycles of helium flow at a fast rate. This could result in a 28.5% cost savings, if the flow were reduced to zero during shutdown. One would have to consider the possibility, however, that reducing the flow to zero may permit the accumulation of moisture in the graphite region.

A second possibility is the incorporation of another, less-expensive gas instead of helium. The current price of helium is $38.81 per 285 cubic feet. Carbon dioxide, for example, can be purchased for only $9.85 per 50 lb bottle (50 lb = 437 cubic feet at STP). The price per cubic foot of helium is therefore 13.62 cents compared to only 2.25 cents for carbon dioxide---approximately six times geater. The graphite region could be purged with carbon dioxide at a rate five times the current helium flow for $718 dollars less per year than the current helium expenditure.

There are, of course, many considerations in choosing an

alternate gas. As part of this thesis research, an initial literature search was performed to gain some insight as to the feasibility of such a change. The literature references are included in the bibliography at the end of this thesis. Although no definite conclusion has been drawn, it seems worthwhile to discuss some of the information that was discovered.

4.3 Use of an Alternate Gas

The choice of any gas to be used in a nuclear reactor must depend on the cost of the gas, its physical properties, and its reactivity with the materials with which it will interact. Several gases have been used in contact with graphite; these include air, helium, hydrogen, carbon dioxide, oxygen and nitrogen. In the case of the MITR-II, air is clearly not an acceptable substitute for the helium cover gas. Oxygen is unattractive since it produces nitrogen-16 through an (n,p) reaction. Furthermore, the reaction between oxygen and carbon is exothermic, making self-sustained burning possible in the event of high temperatures (4). Hydrogen is not practical owing to its explosive properties. The remaining alternatives are carbon dioxide and nitrogen.

In the literature, carbon dioxide is by far the most widely-discussed gas for use in nuclear reactors. The main concern associated with the use of carbon dioxide is the oxidation of the graphite. The overall reaction is endothermic, and results in the formation of carbon monoxide:

$$C + CO_2 \longrightarrow 2CO$$

The rate of this reaction increases substantially with temperature, and decreases as the concentration of carbon monoxide increases (5). However, there is some indication that the temperatures that would be reached in the MIT reactor (of the order of 300 C) are not sufficient to cause concern: "No gasification, hence no loss of carbon, occurred at this temperature (500 C)" (6) and "graphite weight losses in pure carbon dioxide are difficult to detect at temperatures below 625 C." (7)

This information points to carbon dioxide as a promising possibility. However, through further research it was found that graphite may be significantly oxidized by carbon dioxide "at modest temperatures" (8) in a radiation field. When carbon dioxide is irradiated in the presence of carbon, the solid is first oxidized until the steady state condition is reached. This is followed by the primary dissociatons of carbon dioxide and CO, yielding CO, C, and O. Secondarily:

- carbon and carbon suboxides are deposited on the graphite surface
- these deposits interact with O or O_3 to regenerate CO_2
- oxygen atoms at surfaces react to form O_2 and O_3
- oxygen reacts with deposited carbon to form CO,
- and with CO to form CO_2
- heterogeneous oxidation of graphite by oxygen atoms occurs.(9)

Hence, in the presence of radiation, carbon dioxide dissociates, greatly increasing the number of active species available to attack the carbon, and "although the thermal attack of carbon dioxide on graphite at temperatures below about 600 C is sufficiently slow to

cause little concern to the reactor design engineer, a measurable and significant reaction takes place in the presence of high-energy radiation." (10) No quantitative data was found regarding the extent of this enhancement where the MITR-II is concerned.

Regarding the use of nitrogen as a purge gas, nitrogen is relatively inert under most conditions. The formation of cyanogen is the major concern:

$$2C + N_2 \rightleftharpoons C_2N_2$$

but this reaction is not thermodynamically feasible in the temperature range of 300 to 3000 K (11). Under irradiation, if air is present in even small amounts, potentially troublesome nitrous oxides may result.

Yet another consideration in the use of an alternate gas is the possibility that the different molecular weight of helium (8) versus carbon dioxide (44) and nitrogen (28) may have an important effect on the distribution of gas through the graphite region. Helium may have an advantage in that its smaller molecular weight permits the gas that enters the reflector at the bottom to percolate up to the top.

More research needs to be performed before a decision is made concerning the utilization of an alternate purge gas for the graphite reflector region of the MITR-II, but the cost of helium makes such an investigation relevant.

5. FUTURE DIRECTIONS

As outlined in the introduction to this thesis, this research is an initial step in identifying, characterizing, and subsequently reducing the argon-41 source term in the MIT reactor. A substantial amount of groundwork has been laid: the principle producers of argon-41 in the reactor have been identified and quantified, and it is clear that one means to reduce argon-41 production is to increase the flow of inert gas that blankets the graphite reflector. Adjusting the flow on the pipe tunnel blower will not reduce argon-41 production, but might be employed to reduce stack output.

As with most projects, this work has opened up at least as many avenues as it has closed. Future work can be divided into several different categories:

GRAPHITE HELIUM: examine long-term effects of increased flow rates; look at the possibility of using a more effective and/or less expensive cover gas; investigate flow of the gas through the graphite region: why does a sample taken from SV-20 (at the top of the reactor) appear to be unrepresentative? Consider feeding helium into the region from the bottom only (i.e., close valve SV-18): does this alter the results?

PIPE TUNNEL BLOWER: expand work to investigate whether reactor floor and control room concentrations of argon-41 actually do increase when pipe tunnel blower flow is decreased.

LEAK DETECTION: attempt to isolate locations of air influx into the reactor---particularly into the graphite region---and attempt to seal

these holes.

OTHER SOURCES: after improving the situation in the graphite helium purge area, it might be reasonable to consider the other 15% of the argon-41 source term that does not come from the pipe tunnel. In particular, one could examine the areas that are purged with carbon dioxide to determine whether the maximum possible amount of air is actually being purged from these regions.

This project has provided the author with an unusual opportunity to work on a practical health physics problem in a real reactor environment. Although the problems of argon-41 production and emission in the MITR-II are not yet completely understood, the work presented herein has included key data which have helped to identify and ultimately reduce the argon-41 source term.

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APPENDIX: CALIBRATION PROCEDURES

In order to permit conversion of sample counts per second to concentration of argon-41, a calibration method was devised which utilized the 333 ml polyethylene jar and the germanium-lithium detector.

Because it is impractical to work with a standard composition of radioactive gas, a liquid cobalt-60 source was used to calibrate for this geometry. A standard cobalt-60 solution was obtained from New England Nuclear Co., and carefully diluted to form a stock solution of 0.95 microcuries per milliliter ($\pm$2%). Next, an approximation was introduced. Cobalt-60 gamma rays, with energies of 1.17 and 1.33 MeV, bracket the energy of 1.29 MeV that is emitted from argon-41. The height of the polyethylene jar is 11 cm, so it would not be accurate to simply fill the jar with a solution of known activity and then analyze it on the GeLi detector, unless a correction were made for self-absorption of the gamma rays in the water. One can simplify the matter, however, by utilizing the fact that, at 1.2 MeV, the attenuation of a gamma ray in a single centimeter of water is approximately 2%.

The 333 ml jar was figuratively "divided" into eleven equal slices, each one centimeter in thickness. Twenty milliliters of water were added to ten milliliters of standard solution in the poly jar, resulting in a one-centimeter layer of solution that had an activity of 0.95 microcuries. Eleven different measurements were made, adjusting the distance between the jar and the detector so that the sum of the resulting counts would be the same as that for a jar

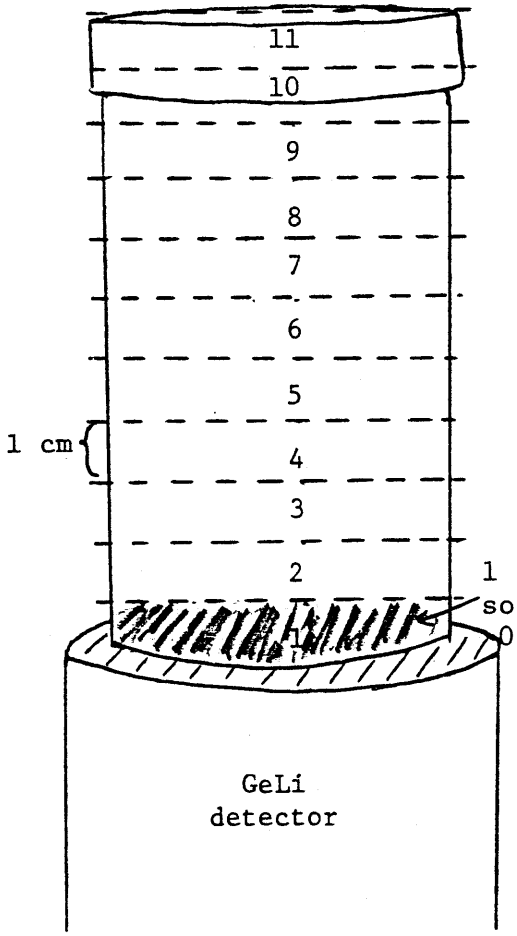
filled with cobalt-60 solution (11 x 0.95 = 10.45 microcuries total) with minimal attenuation (see Figure A1).

From this calculation a value of 85.2 counts per second per microcurie was obtained, resulting in an efficiency of 0.022% for this sample geometry. Any gas sample in one of these polyethylene jars could then be counted on the GeLi detector, net counts in the argon-41 peak determined through use of the associated multichannel analyzer, and the resultant activitiy in the jar calculated. Sample counts were also corrected for decay time between actual sampling and resultant counting. Dividing by the volume of the jar yields the concentration in microcuries per ml. For example, suppose a 30-second count of a sample yields 10,897 net counts in the argon-41 peak. The calculation of concentration would then proceed as follows:

$$\frac{10,897 \text{ counts}}{30 \text{ seconds}} = 363.23 \text{ counts per second}$$

$$\frac{363.23 \text{ counts}}{\text{second}} \cdot \frac{85.2 \text{ cps}}{\text{microcurie}} \cdot 333 \text{ ml} = 1.28 \times 10^{-2} \text{ microcuries per ml}$$

Hence, the concentration of the gas sample is 0.0128 microcuries of argon-41 per milliliter of gas.



When bottom of jar is placed at each dotted line, total counts per minute for each slice:

11	1363.1
10	1584.4
9	1811.6
8	2218.9
7	2723.9
6	3436.9
5	4388.3
4	5680.8
3	7516.4
2	10662.4
1	12060.8

Σ = 53447.5 counts per minute

= 890.8 counts per second

$$\frac{890.8 \text{ cps}}{(.95 \times 11)\mu\text{Ci}} = \boxed{\frac{85.2 \text{ cps}}{\mu\text{Ci}}}$$

Figure A1. The poly jar is moved relative to the detector to obtain a total number of counts per "slice." Total counts represent what one would expect if the jar were filled with cobalt-60 solution and there were minimal attenuation of the gamma rays in the water.