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ABSTRACT

Fission gas release from spherical BeO-based fuel elements designed for the conceptual ABORIGINE reactor and from 200 micron diameter fuel particles of UO_2-ThO_2 solid solution has been measured by in-pile sweep capsule testing in the temperature range 600°C to 1000°C.

Release parameters (R/B ratios) of 10^{-7} to 10^{-6} were measured on several fuelled spheres, and 10^{-4} to 10^{-2} on bare fuel particles. Two spheres with bonded cladding maintained good gas retention at 1000°C for fast neutron doses (6×10^{20} nvt) which were three times that required of the ABORIGINE fuel element; however, the fission burnup (1.1 percent• U + Th) was a factor of nine below the design burnup.

It has thus been shown that BeO coatings on fuelled BeO spheres can in principle retain fission gases to the high standard required for ABORIGINE. Further work would be needed, (a) to assess the reliability of the fabrication process, (b) to test to high burnups fuelled spheres designed so that the coating is subjected only to fast neutron damage and which therefore should not crack from burnup effects, (c) to eliminate other possible cracking mechanisms such as thermal stress, and (d) to measure the release from cracked spheres under a range of conditions and thus to determine the allowable cracking rate more accurately.

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BERYLLIUM MODERATED REACTORS; BERYLLIUM OXIDES; BURNUP; COATED FUEL PARTICLES; CRACKS; DECONTAMINATION; FABRICATION; FAST NEUTRONS; FISSION PRODUCTS; FLOW RATE; FUEL ELEMENTS; GAS FLOW; HIGH TEMPERATURE; IRRADIATION; KRYPTON 85; KRYPTON 87; KRYPTON 88; RARE GASES; RELIABILITY; SPHERES; THERMAL CYCLING; THORIUM OXIDES; URANIUM DIOXIDE; VERY HIGH TEMPERATURE; XENON 133; XENON 135

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1. INTRODUCTION

The Australian Atomic Energy Commission studied a 200 kW(e) beryllia moderated nuclear reactor known as ABORIGINE for which there were two conceptual adapations of the basic design. One was an air-cooled system with an open cycle and the other a carbon dioxide cooled system with closed cycle. Obviously, the fuel element for either system must be capable of retaining fission products to a high degree so as to avoid hazardous contamination of the environment in an open cycle system or the build-up of high levels of radioactivity in a closed cycle coolant circuit.

As part of the fabrication programme designed to improve the fission product retention properties of the fuel element, the release of gaseous fission products was measured using both in-pile and post-irradiation techniques. Attention was confined to the noble gases because they are convenient to handle experimentally and, with the radioiodines, are the most troublesome as regards contamination of the coolant in a gas-cooled reactor. This report describes the in-pile sweep capsule experiments; the post-irradiation experiments were reported earlier (Roman, Randall and Hanna 1969).

The proposed fuel element was a sphere of beryllia (the moderator) fuelled with a solid solution of urania stabilised with thoria. The exact diameter was not specified but would be in the range 2 cm to 3.8 cm. The fuel was contained in a kernel as a dispersion in BeO, and this kernel was clad with an unfuelled layer of beryllia to retain fission products. The four stages in the development of this design were:

(i) Coarse (200 micron) fuel particles of UO_2-ThO_2 solid solution dispersed in, and bonded, to a matrix of BeO. This fuelled kernel was surrounded by, and bonded to, an unfuelled outer shell of BeO about 1.5 cm thick.

(ii) As in (i) but with the unfuelled outer shell increased in thickness to 4.6 mm and the kernel diameter reduced accordingly.

(iii) As in (ii) but with fine (less than 10 micron) fuel particles.

(iv) As in (iii) but with a porous friable BeO layer, about 0.7 mm thick, separating the kernel from the unfuelled shell. This structure has become known as the shell-and-kernel design.

The reasons for the change to fine fuel particles and the introduction of the porous BeO layer have been given in detail by Walker and Hickman (1967) and Hickman et al. (1968), and are discussed in relation to the observed behaviour of shell-and-kernel spheres by Hanna and Reeve (1972). Briefly, fine fuel particles, compared with coarse, give improved mechanical properties both before and after irradiation and the porous layer is designed to accommodate radiation-induced swelling of the kernel and to reduce β-particle bombardment of the cladding.

Obviously, the fission product retention properties of a fuel element of this design are very dependent on those of the BeO. Hence, it was considered necessary to measure the retention of both bare fuel particles and fabricated fuel elements in order to assess adequately the part played by the beryllium oxide. Fuel particles (previously sintered at 1700°C) were incorporated into BeO by isostatic pressing followed by sintering at 1350 to 1575°C. Metallographic examination showed that fuel particle microstructure did not change during this treatment.

The fuel particles and the spheres used in this programme were prepared using techniques described elsewhere by Reeve et al. (1966).

2. EXPERIMENTAL PROGRAMME AND SPECIMEN PREPARATION

The experimental programme was determined largely by the sequence in which specimens became available and, therefore, depended very much on the success of developments in fabrication techniques. It began with a series of experiments in rigs designated X-124 and X-160 (see Sections 4.2 and 4.3) from which data were obtained on bare fuel particles and on spheres which were considered to be the best available at that time. When a later series of rigs, designated X-132, became available, the post-activation diffusion (PAD) programme was well established and materials could be assessed before committing them to sweep capsule testing. The specimens used in the X-132 series reflected the improvements made in fabrication and sphere design and the ultimate objective of that series was to measure the fission gas retention in a sphere with all the final design features of the fuel element to the design burnup and fast neutron dose (10 atom per cent uranium plus thorium, and 2×10^{20} nvt > 1 MeV).

The fabrication of fuel particles and fuelled spheres with bonded cladding has been described by Reeve et al. (1966) in a report concerned specifically with the preparation of specimens for the X-124 experiments.

A more recent report by Reeve and Ramm (1969) describes the fabrication of spheres with fine fuel particles and porous buffer zones.

All spheres were prepared from Brush UOX grade beryllia. Those irradiated in rig X-124 contained no sintering additions but all those for the X-132 rigs had additions of 0.1 per cent magnesia. The PAD programme (Roman, Randall and Hanna 1969) had shown such an addition to be beneficial in achieving high fission gas retention.

Details of the dimensions, compositions, and structures of all specimens subjected to sweep capsule testing are given in Tables 1 and 4.

3. EXPERIMENTAL TECHNIQUES

The in-pile sweep capsule technique has been used extensively throughout the world and has been well described by Carroll (1965). Briefly, the specimen undergoing irradiation is heated either by nuclear or electrical heating under a flowing gas which entrains volatile fission products and carries them to a point remote from the reactor core where they can be sampled and analysed quantitatively.

Although helium has been used most frequently as the sweep-gas (for example, Carroll 1965) any gas can be used provided it is chemically compatible with the specimen and experimental equipment, has a low activation cross section, and does not interfere with the analysis of the volatile fission products. Carbon dioxide has been considered a possible coolant gas for the closed cycle version of ABORIGINE and was also the proposed coolant for an earlier high temperature gas cooled reactor concept (Roberts 1964). Hence, it was chosen as the sweep-gas for experiments relating to the development of the beryllium oxide based fuels for both systems. Cylinder gas with a moisture content not greater than 20 p.p.m. was used, so as to minimise the mass transfer of BeO at the irradiation temperature.

The sweep-gas circuit is shown schematically in Figure 1. Gas was fed from the cylinders at a pressure of 20 p.s.i.g. to a reducing valve where pressure was reduced to 5 p.s.i.g. It then passed through copper tubing to the irradiation rig, over the specimen and back to the sweep-gas console where it was treated for removal of fission gases. Flow control valves and flow meters were placed near the end of the gas line so that the greater part of the line and the specimen capsule was at 5 p.s.i.g. and so that a rapid response was obtained to manipulation of the flow control valve. A bursting disc designed to rupture at

15 p.s.i.g. and a charcoal filter intended to prevent accidental back-flow of active gas were located on the inlet side of the irradiation rig.

3.1 Separation of Sweep-Gas from Fission Gases

Separation of the fission gases was achieved by bubbling the sweep-gas into a solution of potassium hydroxide (350 gram per litre) in which the CO_2 dissolved, leaving the impurities and fission gases to rise to the surface, where they passed directly to the active ventilation system or were collected by displacement of water from a glass flask.

In the first two experiments the solution was contained in a one-litre stainless steel vessel with a large spherical base and a narrow neck about 15 inches long. The sweep-gas was injected through a sintered glass disc at the bottom of the vessel and a spring loaded ball valve was fitted close to the injection point to stop 'suck-back' of the solution when the gas flow was stopped. These vessels proved very unsatisfactory owing to frequent blockage of the glass discs and crystallisation of potassium carbonate on the seat of the non-return valves. A further disadvantage was that the passage of gas from the disc and through the solution could not be seen.

An improved dissolution column (Figure 2) was introduced during experiment X-124-3. It consisted of a stainless steel tank (9 litre capacity) with one transparent side made of Perspex. Immersed in the solution in this tank was a helix of soft PVC tubing wound around a 1 inch diameter Perspex rod about 15 inches long. At the lower end of the helix was a Perspex chamber of about 50 ml capacity into which the sweep-gas was fed through a hypodermic needle passing through a rubber seal. The gas bubbled to the top of this chamber (inverted cone) and then passed into the helix in which the final traces of CO_2 dissolved as the bubbles travelled slowly upwards. The outlet of the helix was near the top of the tank but still below the level of the KOH solution. Hence, it was possible to place either an inverted, water-filled flask (as in Figure 2) over the outlet to collect the emerging gas, or a loose fitting tube to conduct the gas to the active exhaust system.

The small chamber at the bottom of the helical column was approximately half-filled with mercury and the gas was injected well below its surface. This was a very effective non-return valve, preventing 'suck-back' of KOH solution into the gas line when the gas flow was stopped. However, to prevent mercury itself from sucking back

along the gas line (for example, during accidental evacuation of this region of the gas line during commissioning of a new experiment), a barometric seal was provided by making the gas line rise vertically for a distance of at least 30 inches above the bottom of the chamber. Except for one occasion when a large deposit of potassium carbonate grew on the end of the hypodermic needle, this device worked satisfactorily throughout the experimental programme and was far superior to mechanical valves.

The passage of the gas up the dissolution column caused some circulation of the KOH solution within the tank. Solution was forced ahead of the gas bubbles and overflowed from the top of the helix. A tube adjacent to the hypodermic needle in the top of the lower chamber allowed new solution to replace that which was forced upwards but it was essential that the end of the tube was just above the surface of the mercury and on the other side of the chamber so that bubbles did not escape by this route. However, in addition to this inherent circulation, a mechanical stirrer was placed in the tank and was operated daily to ensure adequate stirring of the solution. At a gas flow rate of 50 cm^3 per minute for 24 hours a day, 9 litres of the 350 gram per litre KOH solution would last for over seven days and replacement was possible on a regular weekly basis.

3.2 Measurement of Sweep-Gas Flow Rate

Three devices were used to measure gas flow rates. First, a calibrated rotameter was fitted adjacent to the control valve and was used for routine checks and when making adjustments to the flow rate. Secondly, a pressure cell, consisting of a bellows and differential transformer monitoring the pressure drop across a short capillary was used to give a continuous record of the flow rate. Thirdly, accurate measurements of the flow rate were made each time the gas was sampled for analysis using the device illustrated in Figure 3. This device was a glass tube, graduated at 0 to 50 cm^3 volume, and contained inside a larger diameter Perspex tube. Gas flowed up the glass tube and then down the Perspex tube and on to the dissolution column. When a measurement was required a small volume of teepol-water mixture was injected into the bottom of the glass tube from a syringe-type reservoir below the tube. Bubbles generated by the gas were timed as they traversed the graduated section of the tube. Transit times could be measured accurately with a stop watch and the precision of the overall

measurement was determined by the precision of the calibration of the glass tube (burettes proved very suitable, giving a precision of about one per cent at a flow rate of 25 cm^3 min^{-1}).

3.3 Sampling of Sweep-Gas for Fission Product Analysis

Provision was made in the sweep-gas line for two sampling techniques which enabled a wide range of sample sizes to be taken, depending upon the fission gas content of the sweep-gas. The results from samples taken by both techniques were in very good agreement.

For high fission gas release rates, small samples were taken by drawing gas through a hypodermic needle into an evacuated 15 cm^3 serum vial with a self-sealing rubber cap (after Carroll 1965). The needle was on a branch of the gas line close to the dissolution column (see Figure 2) in the manner shown in Figure 4. The serum vial was placed over the needle and evacuated by venting it into an evacuated 1 litre flask. By manipulation of appropriate valves, it was then isolated from the evacuated flask and allowed to fill with sweep-gas. This was repeated twice and the vial withdrawn from the needle and removed for analysis.

Where fission gas release rates were low, larger samples were taken by collecting the gas at the top of the helical dissolution column. In this way only the fission gases and impurity gases from the CO_2 were sampled (as distinct from the needle sampling technique). Tests on a simplified dissolution column demonstrated that the loss of fission gases by dissolution in the KOH solution amounted to only 0.1 per cent.

Of two techniques used to collect gas the simplest was to allow the gas to displace water from an inverted 5 cm^3 sampling flask with a conical neck. Gas was collected for a suitable sampling time or until the gas level reached the shoulder of the bottle. It was then moved from the gas outlet and stoppered before raising the neck above the surface of the KOH solution. To minimise counting errors due to absorption of the gamma radiation in the residual liquid in the sampling bottle, it was necessary to inject air until the bulk of the liquid was displaced and the volume of gas was just great enough to permit insertion of the stopper without displacing some gas.

The above early procedure was later replaced by a more precise technique in which the gas was collected in a Pyrex burette of about 17 cm^3 capacity (Figure 5) whose tap had been replaced by a two way stop-cock. A hypodermic needle was sealed with epoxy resin into one outlet of the stop-cock and it was possible to place a serum vial over

the needle and evacuate it through the other outlet without disturbing the sample collected in the burette. The burette was calibrated so that V1, the volume between the lowest graduation and the stop-cock, and V2, the volume of the stop-cock plus needle were measured.

A measured fraction of the gas collected in the burette was transferred to a 15 cm^3 serum vial for counting as follows. The volume of gas in the burette was made up to between 16 and 17 cm^3 by injecting air when the neck of the burette was held below water. The gases were mixed by shaking the burette (still containing some liquid) and the total volume of gas read on the scale with the burette inverted. The serum vial on the hypodermic needle was then evacuated through the two way stop-cock and, with the unstoppered neck of the burette immersed in water the cock was rotated to allow the gas to vent into the vial, thereby drawing liquid into the burette. The residual volume of gas was then read on the scale. Both readings of gas volume were made at atmospheric pressure by locating the burette on the cone of the pressure equalising device shown in Figure 5 and adjusting the movable column until the liquid levels in the column and the burette were the same. The serum vial was also removed from the needle while the gas was held at atmospheric pressure. The fraction of the sample transferred to the vial was given by

$$F = \frac{(S1 - S2) - V2}{S1 + V1},$$

where S1 and S2 are the initial and final scale readings, and V1 and V2 are defined above. The value of F was used to correct an analysis result to the value expected from the total sample.

3.4 Analysis of Fission Product Samples

Gas samples were analysed for radioactive fission gases by gamma ray spectrometry; no attempt was made to measure the stable fission gases. Samples were counted with a Packard Model 16 multichannel analyser connected to either a sodium iodide detector or a lithium-drifted germanium detector, both of which had been calibrated for the sample geometry in use as described by Hanna, Walker and Beach (1967).

The sodium iodide detectors, both a well crystal and a 2 inch cylindrical crystal, were used in experiments X-124-1 to X-124-4 inclusive. The solid state detector was used exclusively in all subsequent experiments. The radioactivity in the gas samples consisted of five isotopes of krypton and xenon with gamma energies between 0.08 MeV and

0.4 MeV, and ^{41}Ar arising from activation of the ^{40}Ar in the sweep-gas. The superior resolution of the germanium detector greatly simplified the evaluation at peak intensities for the five fission gas isotopes.

The peak intensities were used to calculate the release rate (atoms per second) of each fission product in the sample and subsequently the ratio of the release rate to a birth rate, commonly referred to as the R/B ratio, which is used as the release rate parameter in this type of experiment. The birth rate was calculated from the known ^{235}U content of the specimen and the thermal neutron flux; the latter was obtained from tabulated data or from cobalt wire monitors. Generally data were accumulated for a full reactor programme (24 days in HIFAR) and processed through an IBM-360 computer to give a tabulated and graphical record for the specimen being studied.

3.5 Design of Specimen Capsules

The irradiation rigs used for these experiments were designated X-124, X-160 and X-132. Rig X-160 was identical in design to X-124 which it replaced when the X-124 heater burned out. The designs of specimen capsules in these rigs are described below.

3.5.1 The X-124 and X-160 Capsules

The X-124 capsule was designed for use in a 2V facility in HIFAR and could accommodate four spheres of 0.5 inch diameter, or a small can of fuel particles disposed in an unsintered carrier powder. The two designs are shown in Figures 6a and 6b. The specimen capsule was electrically heated by a furnace winding in the outer section of the rig. This outer section was a semi-permanent insertion into the irradiation facility but the inner section was renewed each time new specimens were loaded.

Spheres were held in the specimen capsule by a series of stainless steel spacers and rings and the thermocouples were led through these to touch the specimen surfaces. Fuel particle samples were irradiated in admixture with a coarse, unsintered alumina powder; the mixture was contained in a small stainless steel can with a porous end-cap cut from a sintered stainless steel disc. The thermocouple in this case was located in a pocket in the lower end-cap of the specimen can.

With both types of specimen the sweep-gas was fed to the capsule through the outer of two concentric tubes welded to its top. On entering the capsule, the gas passed down the outside of the spacing rings and then upwards over the specimens, leaving the capsule through the inner tube.

Maximum surface temperatures of 850°C were obtainable with a loading of four spheres. Centre temperatures were estimated to be 50°C higher. However, with the fuel particle specimens, the smaller amount of gamma heating permitted a maximum centre temperature of only 800 to 810°C.

A cobalt wire flux monitor was placed near the centre of the capsule.

3.5.2 The X-132 Rigs

The X-132 rigs were designed for the irradiation of single spheres of 2 cm diameter in a hollow fuel element in HIFAR. Unlike the X-124 rigs no electrical heating was provided and specimen temperatures were obtained by nuclear heating (predominantly gamma heating in the specimen and specimen holder); both the gas gap and the helium/nitrogen ratio of the gas could be adjusted as required. The rigs were capable of giving a surface temperature of about 1100°C but were intended to operate for prolonged periods at 1000°C.

The specimen capsule is illustrated in Figure 7. The specimen was held in hemispherical seats between two mating inconel blocks which were held centrally within a welded stainless steel can measuring $2\frac{1}{2}$ inches by $1\frac{3}{16}$ inches diameter. Three pins in each half of the inconel block kept the specimen from touching the hemispherical seats and ensured a free path for the sweep-gas around the specimen. These pins were carefully lapped for each specimen so that a rattle-free fit was obtained.

The sweep-gas entered the capsule through a tube welded to the lower end-cap, gained access to the specimen through holes drilled from the flat faces of the inconel block and emerged from the capsule through a tube welded to the upper end-cap.

Two thermocouples were led through the sweep-gas inlet tube, one to touch the specimen surface through a specimen locating pin, and the other to measure the temperature of the inconel block. Titanium and cobalt wire flux monitors were placed above the inconel block to provide measurements of both fast and thermal flux.

4. RESULTS

4.1 Fission Products Detected in Sweep-Gas

Five gaseous fission products were detected in the sweep-gas of most experiments. The products and the gamma ray energies used for their determination were as follows:

^{133}Xe	0.81 MeV
^{135}Xe	0.25 MeV
^{85m}Kr	0.15 MeV
^{87}Kr	0.4 MeV
^{88}Kr	0.19 MeV

In discussing the results in the following sections of the report, greater attention is given to the krypton isotopes. These arose from the decay of selenium and/or bromine precursors all of which have half-lives of less than 3 minutes; escape of bromine from the specimen was, therefore, very low and the formation of kryptons by decay of bromine outside the specimen could be ignored. The two xenons, on the other hand, were produced from precursors with half-lives of several hours (^{133}I, 20.8 h, and ^{133}I, 6.7 h) and there was appreciable build-up of the iodines in the sweep-gas line.

It is possible, in principle, to measure the contribution of iodine decay to the observed release rates by measuring the xenon content of the sweep-gas while the reactor is shut down and extrapolating back to the moment of shut down. The true xenon release rate should then be the difference between the observed value at power and the extrapolated value. Such corrections can only be applied with accuracy to the values observed just before shutdown or to times before shutdown when the amount of iodine in the gas line was in equilibrium with the iodine release rate from the specimen. Where the temperature has been varied during the operating programme it is extremely difficult, if not impossible to apply corrections to the early stages of the programme, owing to lack of knowledge of the iodine release rates.

4.2 Fission Gas Release from Fuel Particles, X-124 Series

The results of measurements on bare fuel particles are summarised in Table 2. Results are presented in order of increasing burnup which corresponds with increasing temperature for batch COP-25-B2 particles and decreasing temperature for batch X-124-1,2 particles. The xenon figures have not been corrected for the release of xenon from iodine precursors outside the specimen.

Release parameters for COP-25-B2 materials ranged from 2×10^{-5} for ^{87}Kr to 5.4×10^{-4} for ^{135}Xe. With all five fission gases, there was a definite increase in release parameter between 760°C and 800°C. Below 760°C however, the parameters for ^{85m}Kr, ^{88}Kr and ^{133}Xe remained approximately constant while those for ^{87}Kr and ^{135}Xe showed an apparent

decrease as the temperature was raised. This decrease is not ncessarily directly related to temperature since, for this experiment, the burnup was increasing while the temperature was being raised.

The parameters obtained with the X-124-1,2 material ranged from 4×10^{-4} to 1.4×10^{-2}. In this experiment the temperature was deliberately reduced from the maximum obtainable (750°C) during the first reactor programme, to 710°C in the second and 650°C in the four subsequent programmes. With each of the five gaseous fission products measured, there was a marked increase in the release parameter as irradiation proceeded and no temperature effect was detected.

4.3 Fission Gas Release from 1.27 cm Spheres, X-124/X-160 Series

Fission gas release parameters for the small spheres (Table 1) irradiated in rigs X-124-1 to X-124-4 are presented in Table 3. Again, the figures for xenon release have not been corrected for the contribution from iodine decay outside the specimen.

The parameters measured in the X-124-1 experiment were in the range 10^{-7} to 10^{-6} at 840°C. Results obtained at lower temperatures have not been included because of the large scatter and consequent lack of confidence in the figures obtained. Release parameters for the X-124-2 experiment were in the range 10^{-7} to 8×10^{-5} and for X-124-3 and X-124-4 (specimens from the same batch) in the range 7×10^{-5} to 4×10^{-4}.

4.4 Fission Gas Release from 2 cm Spheres, X-132 Series

Summaries of the results obtained in the X-132 experiments are given in Table 5 and Figures 8, 9 and 10.

4.4.1 X-132-1

This experiment was withdrawn from the reactor after only two operating programmes during which the specimen accumulated a fast neutron dose of 9×10^{19} nvt and attained a burnup of 0.008 atom per cent of the heavy metals.

During the first programme the fission gases detected were ^{85m}Kr, ^{133}Xe and ^{135}Xe. Initial release parameters were 2×10^{-6} to 3×10^{-6} with a fairly large amount of scatter during the first nine days. On the tenth day the ^{133}Xe parameter rose to 4×10^{-5} and the ^{85m}Kr parameter rose from 1.1×10^{-6} to 3×10^{-6}. The parameters fell again over the next two days to values close to those prevailing before the burst, but then increased steadily until the end of the programme.

A sample taken shortly after reactor shutdown gave a ^{133}Xe content higher than any observed while the reactor was at power. The apparent release rate was equivalent to an R/B ratio of 6×10^{-5} if the reactor was assumed to be operating at the normal power. High ^{133}Xe contents persisted for the next two days, after which sampling was stopped.

During the second programme ^{87}Kr and ^{88}Kr were detected in addition to those gases detected during the first programme. The parameters following startup were 3 to 4 x 10^{-5} for the kryptons and 9 to 10 x 10^{-5} for the xenons and increased steadily until they were higher by factors of four (for ^{88}Kr) to 30 (for ^{133}Xe) after a further 11 days irradiation. At this stage the specimen temperature was reduced to 770°C with an accompanying drop in release parameters. Parameters dropped slightly over the next four days but then increased steadily until reactor shutdown.

The behaviour of this specimen is shown in Figure 8.

4.4.2 X-132-2

Following an initial reactor operating programme of only $6\frac{1}{2}$ days, this experiment remained in-pile for a further 14 programmes during which the specimen received a fast neutron dose of 6×10^{20} nvt and a burnup of 1.13 atom per cent of heavy metals.

At the start of this experiment the release parameters were in the range 3 to 10 x 10^{-8} and they subsequently increased slowly to the range 1.4 x 10^{-7} to 8 x 10^{-7}. Mid-way through the fifteenth programme there was an unscheduled reactor shutdown following which the release parameters increased by two to four orders of magnitude. This behaviour indicated that the specimen had fractured (probably due to thermal shock) and the experiment was withdrawn at the next shutdown.

Experimental scatter was very pronounced during the first three programmes; for example, the high and low for Xe-133 in programme 3 differed by a factor of five. From the fourth to fifteenth programmes the scatter was less although variations of a factor of two were observed between the high and low for a given isotope in any one programme. The scatter at the beginning and end of the experiment is illustrated in Figure 9.

4.4.3 X-132-3

Experiment X-132-3 ran for eight reactor operating programmes and attained a burnup of 0.78 atom per cent heavy metal and a fast neutron

dose of 3.3×10^{20} nvt. It was withdrawn at the end of the eighth programme to make room for experiment X-132-4.

Release parameters measured during the first programme were in the range 2×10^{-8} to 2×10^{-7}. During the remainder of the irradiation the parameters increased slowly and, just before unloading, were in the range 2×10^{-7} (^{87}Kr) to 1.6×10^{-6} (^{135}Xe).

With X-132-2, experimental scatter was comparable to that observed at the equivalent stages of the X-132-2 experiment.

4.4.4 X-132-4

Sweep-gas samples taken shortly after the X-132-4 experiment went to power, with the specimen temperature held at 690°C gave the following release parameters:

^{85m}Kr	6×10^{-8}
^{87}Kr	2×10^{-8}
^{88}Kr	4×10^{-8}

After three days irradiation, however, the release parameters had increased to the following:

^{85m}Kr	3×10^{-4}
^{87}Kr	8×10^{-5}
^{88}Kr	2×10^{-4}
^{133}Xe	4×10^{-4}
^{135}Xe	6×10^{-4}

In view of these high values and low specimen temperature the sweep-gas was shut off and the rig removed at the next shutdown. Hot cell examination showed the specimen to be cracked.

4.4.5 X-132-5

From the outset of this irradiation release, parameters were in the range 1×10^{-4} to 6×10^{-4}. In view of these high values the specimen temperature was not taken above 840°C (the minimum obtainable) and sampling was discontinued after nine days of irradiation.

On hot cell examination this sphere was found to be cracked in a manner similar to that from rig X-132-4.

4.4.6 X-132-6

The specimen in rig X-132-6 was taken to an indicated temperature of 1000°C immediately the reactor came to power. During the first six days of operation release parameters were in the range 4×10^{-6} to 3×10^{-5}

(Figure 10). On the ninth day after startup a blockage developed in the sweep-gas line and a serious downward drift occurred in the indicated temperature. The sweep-gas line was therefore isolated and the rig was unloaded at the following shutdown.

It was found subsequently that the thermocouples in this rig were slightly further from the specimen than in previous rigs and the true specimen temperature was calculated to be close to 1200°C. Examination of the specimen holders in the hot cell showed that severe corrosion had occurred and that corrosion product had blocked the path of the sweep-gas through the specimen holder.

On examination the sphere appeared quite sound and no large cracks could be seen. However, there was one localised area of surface crazing which appeared to have resulted from contact with corrosion product from the stainless steel specimen holder.

5. DISCUSSION

5.1 Reproducibility of Experimental Results

Figures 8 and 9 and Tables 2, 3 and 5 show that the release parameters measured under constant conditions of temperature and fission rate generally fell within a scatter band in which the minimum and maximum differed by a factor of about two. However, there were instances where genuine bursts or trends appeared to occur, as for example, with the X-132-1 experiment (Figure 8). To determine the reproducibility of results a special experiment was carried out utilising the X-132 sweep-gas line in which the in-pile capsule was replaced by one located out of pile and containing a porous alumina sphere impregnated with ^{131}I. Decay of the ^{131}I gave rise to ^{131m}Xe which entered the sweep-gas in the same way as the fission gases in an irradiation experiment. The ^{131m}Xe was produced at an accurately predictable rate and the amount in the sweep-gas should have decayed with the same half-life as the ^{131}I from which it was formed.

The results from this experiment are presented in Figure 11. The figures have been treated in such a way as to give an R/B ratio, with the knowledge that the ^{131}I source had an activity of 5 mCi at the beginning of the experiment. The R/B values varied by a factor of three from 8×10^{-3} to 2.4×10^{-2}. This scatter is comparable to that observed in the irradiation experiments and indicates that bursts and trends should only be regarded as genuine when they exceed this factor.

Consideration of the release rates observed in the irradiation experiments suggests that the scatter is related inversely to the magnitude of the release rate from the specimen. In the second programme of the X-132-1 experiment there was little scatter and R/B ratios increased smoothly as irradiation progressed. Similarly (as is shown in Figure 10), in experiment X-132-6 the parameters for the short-lived gases increased smoothly and appeared to be saturating while that for the longer lived ^{133}Xe was still increasing when the experiment failed. In both these cases the release rates were between 10^4 atom sec^{-1} and 6×10^6 atom sec^{-1}. In experiments X-132-2 and X-132-3, where scatter was high, the release rates did not exceed 3×10^4 atom sec^{-1} before specimen failure and, in fact, were less than 10^3 atom sec^{-1} in the early programmes of the experiments.

It is interesting to note that in the experiments of Carroll and Sisman (1966), where the experimental scatter was low, the release rates were in the range 10^5 to 10^7 atom sec^{-1}.

The rate of release of ^{131m}Xe in the simulation experiment was approximately 2×10^3 atom sec^{-1} at the end of the experiment. Thus, it was a very close reproduction of the conditions existing before specimen failure in the first three X-132 experiments.

The reasons for the observed scatter are not known. Fission gas samples were always active enough to permit counting to a statistical accuracy of better than two per cent (except for ^{87}Kr for which it was about 4 per cent). Sweep-gas flow rates were measured to about two per cent but the release rate calculation is very insensitive to small changes in flow rate for fission gases with half-lives as long as those detected in these experiments. The errors would seem then, to have occurred during sampling and as the result of inhomogeneous gas mixing within the sweep-gas line. The importance of uniform mixing can be seen from the fact that even at a release rate of 10^5 atom sec^{-1} and a flow rate of 50 cm^3 min^{-1} the concentration of a given species is only 1 part in 10^{16} by volume.

5.2 Fission Gas Release from Bare Fuel Particles

The data obtained on bare fuel particles are limited in usefulness by the low temperature range covered. Furthermore, extrapolation of the data to higher temperatures is made difficult by what appear to be the effects of burnup on release rates.

Material X-124-1,2 showed pronounced increases in release rates as burnup increased and as temperature decreased. During the last four operating programmes when the temperature was steady at 650°C, the burnup increased from 0.015 to 0.03 atom per cent uranium and the rates of release of the krypton isotopes increased by factors of three to five. No post-irradiation examination of this material was undertaken and the reasons for this deterioration are not known.

The behaviour of the COP-25-B2 material was somewhat different in that there was evidence of a temperature dependence consistent with that observed for UO_2 and some evidence of a slight decrease of release rate with increasing burnup. The release parameters for the krypton isotopes had a distinct upward trend above 750°C but with only two points for each isotope at and above 750°C it would be inadvisable to calculate a precise activation energy. However, the figures are consistent with the activation energy of 26 kcal.mole^{-1} measured by Carroll and Sisman (1966) for ^{88}Kr in single crystals of UO_2. (These workers obtained values of 51 kcal.mole^{-1} and 26 kcal.mole^{-1} on different batches of single crystals.)

In the temperature range 600°C to 750°C the parameters for ^{85m}Kr and ^{88}Kr remained approximately constant but that for ^{87}Kr decreased from 4.5×10^{-4} to about 2.3×10^{-4}. The magnitude for this decrease was greater than the experimental scatter and, therefore, appears to be significant. Carroll and Sisman (1965) have explained the temperature independent release in UO_2 by a knock-out mechanism whereby gas atoms are released when fission events occurring near the specimen surface cause localised surface evaporation. They also observed that knock-out release from unpolished materials decreased with increasing burnup and that the decrease was in the same proportion for each isotope. If this was the reason for the decrease in ^{87}Kr parameter observed here, decreases should have been observed also with ^{85m}Kr and ^{88}Kr. Hence, the observed decrease remains unexplained.

The release parameters measured on the bare fuel particles can be compared directly with those for the spheres irradiated in rig X-124. For comparison with the data from the X-132 rigs however, it is necessary to estimate what parameters would have been expected at 1000°C. Assuming that an activation energy of 25,000 cal.mole^{-1} is valid, the following values are obtained:

Batch COP-25-B2

^{85m}Kr	1.5×10^{-3}
^{87}Kr	3×10^{-4}
^{88}Kr	1×10^{-3}

Batch X-124-1,2

^{85m}Kr	10^{-2} to 10^{-1}
^{87}Kr	10^{-3} to 10^{-2}
^{88}Kr	10^{-2} to 10^{-1}

These estimates for COP-25-B2 material compare favourably with data obtained on UO_2 at 1000°C by Carroll and Sisman (1965) and Schurenkamper and Soulhier (1964).

5.3 Fission Gas Release from 1.27 cm Spheres, Rig X-124

The three batches of spheres used in the X-124 experiments gave results which indicated that fission gas retention deteriorated as material properties were changed in a direction expected to give improved retention.

Comparison of the behaviour of the X-124-1 and X-124-2 spheres directly with that of the bare fuel particles of batch X-124-1,2 shows that the release parameters are lower by three and four orders of magnitude. The behaviour of spheres irradiated in rigs X-124-3 and X-124-4 (same batch of spheres) is more difficult to assess as no measurements were made on bare X-124-3 fuel particles. In post-activation diffusion experiments this batch of particles gave release parameters mid-way between those of COP-25-B2 and X-124-1,2. It is possible, then, that its in-pile release parameters would also have been mid-way between the same two batches, and that the BeO matrix and coating reduced the gas release by about an order of magnitude. Regardless of which way the role of the BeO should be interpreted, the fact remains that the retention of these spheres was inferior to that of the X-124-1 and X-124-2 spheres.

The behaviour of the three batches of spheres correlates directly with the post-irradiation open porosity values (Table 1). Batch X-124-1 has the lowest open porosity and the highest gas retention (subject to the precision of measurement mentioned in Section 4.3) while batch X-124-3 had the highest open porosity and the lowest gas retention. Batch X-124-2 lay between these two in both open porosity and gas retention. No post-irradiation metallography was done and the nature of the open porosity (pores or cracks) is not known.

5.4 Fission Gas Release from 2 cm Spheres, X-132

The experiments with 2 cm diameter spheres demonstrated clearly that release parameters of the order of 10^{-7} can be achieved with BeO based fuels and that they can be maintained to high fast neutron doses. This value of 10^{-7} represents a decrease of three to four orders of magnitude over bare fuel particles when the comparison is based on the behaviour of COP-25-B2 fuel.

The early failures of specimens X-132-4 and X-132-5 suggested that spheres of these types had poor thermal shock resistance. However, four spare spheres from the X-132-6 batch (similar to X-132-5) survived a series of out-of-pile thermal cycles between 1000°C and 280°C without cracking, suggesting that the failures of X-132-5 (and possibily of X-132-4) specimens had another cause, such as the existence of defects which had not been detected in the pre-irradiation examination. Details of the thermal cycling tests are given in Appendix 1.

The limited data obtained with experiment X-132-6 before the shutdown necessitated by rig failure indicate that the shell-and-kernel design can give gas retention comparable to that from spheres with bonded cladding. If the observed release parameters are corrected from 1200°C to 1000°C using Carroll's activation energies of 26,000 cal.mole^{-1} and 51,000 cal.mole^{-1} values of 10^{-6} and 7×10^{-7} respectively are obtained. These are consistent with the values measured in experiment X-132-3, and in experiment X-132-2 before specimen failure.

Although this series of experiments was not successful in testing spheres to both high burnup and fast neutron dose, it has been reported elsewhere (Hanna and Reeve 1972) that four shell-and-kernel spheres have been irradiated at 1000°C to 17 per cent heavy metal burnup and 1.9×10^{20} nvt (fast) without cladding failure. All failures so far observed in shell-and-kernel spheres have arisen from macrocracking in the unfuelled shell and, on the basis of the experiments reported here, it seems highly probable that the fission gas retention will remain high until cracking occurs. The failures in this series of experiments should therefore not be taken to mean that good gas retention cannot be maintained to the required irradiation levels.

Cladding failure results in a sudden rise in release parameters to around 10^{-4}, that is, to values comparable to those for bare fuel particles. This represents a large increase in gas release and only a small fraction of a reactor core could be allowed to break. The minimum

fission product retention for safe operation of an open-circuit ABORIGINE reactor was estimated (Hanna, unpublished) from a consideration of the allowable releases of noble gases, ^{131}I, ^{137}Cs, and radiostrontiums. The following allowable parameters were obtained for the gases of interest in this work (for exhaust stack 40 metres high):

^{85m}Kr	1×10^{-5}
^{87}Kr	5×10^{-6}
^{88}Kr	8×10^{-6}
^{133}Xe	5×10^{-6}
^{135}Xe	1×10^{-6}

These target values are slightly greater than those actually achieved in experiments X-132-2 and X-132-3, and therefore allow a small margin for safety to accommodate sphere breakage.

A rough estimate of the allowable sphere breakage rate can be obtained by assuming that the whole core is at a temperature of 1000°C and that a typical fission product has a release parameter of 5×10^{-7} from an unbroken sphere (this is consistent with the X-132-2 results); the core as a whole, therefore, has a release parameter of 5×10^{-7}. Making the pessimistic assumption that a broken sphere will have a parameter equal to that for bare fuel particles (say 5×10^{-3}) a simple calculation gives an allowable breakage rate of 0.1 per cent if the effective parameter for the core is not to exceed 5×10^{-6}.

This value for breakage rate is conservative as the calculation has ignored the following facts:

(i) Much of the core would be below 1000°C and effective parameters for the unbroken and broken fractions would be lower than assumed.

(ii) Even when the shell fractures, the release parameter is unlikely to reach that of bare fuel particles. (Failure of the X-132-2 sphere led to parameters an order of magnitude lower than those expected from COP-25-B2 fuel at 1000°C).

The effect of temperature distribution in the core is difficult to assess. On the other hand, it is readily shown that a release parameter of 5×10^{-4}, rather than 5×10^{-3}, for a broken sphere increases the allowable breakage rate to one per cent.

The fission gas retention of the core as a whole is so sensitive to sphere breakage that there is no incentive to improve fission product retention beyond a release parameter of about 10^{-7}. For example, if it

is assumed that the release parameter for a broken sphere is 5×10^{-4}, increasing the retention of an unbroken sphere from 5×10^{-7} to 5×10^{-8} alters the allowable breakage rate from 1 in 110 to 1 in 100. This is obviously a small benefit for a relatively large decrease in release parameter and indicates that further improvements in the retentivity of unbroken spheres would not be worth the considerable effort required.

Work aimed at minimising the breakage rate or the release rate from broken spheres would be more worthwhile.

6. CONCLUSIONS

The aim of the work described above was to demonstrate that the BeO coating of a BeO based fuel element is capable of a high degree of fission gas retention. The original intention was to demonstrate this first at low fuel burnup and eventually to the burnups and fast neutron doses required by the ABORIGINE concept. Unfortunately the high burnup experiment was unsuccessful because of a rig failure. However, in several low burnup experiments the R/B ratios were in the range 10^{-7} to 10^{-6} and in one of these experiments the fast neutron dose achieved was three times that required for the ABORIGINE fuel element. This was a very promising result and suggested that in principle satisfactory fission gas retention could be achieved in a BeO coated fuelled BeO sphere. However the limitations of this work were:

(i) Small numbers of spheres were tested.

(ii) High burnups were not covered.

If ABORIGINE fuel element development were to proceed, further sweep capsule tests on larger numbers of spheres would be required in parallel with a fabrication programme, to assess the reliability of the fabrication process in achieving the standard required; these tests need only be to low burnup, to detect pre-existing pinholes and flaws. Fabrication and inspection standards may have to be refined as a result. However, the observed sudden rise in gas release which occurs when spheres (more particularly sphere coatings) crack suggests that the general problem of sphere or coating cracking, whether from mechanical or burnup effects, would require the major effort. The breakage rate would have to be restricted to the range 0.1 to 1 per cent, depending on the release parameter ultimately attained by a broken sphere.

The approach taken to cover burnup has been to design a fuel element in which the BeO coating is subjected only to fast neutron

damage and therefore should not crack. Sealed capsule tests on the resultant 'shell-and-kernel' or 'buffer zone' concept are described in another report (Hanna and Reeve 1972). In brief, the concept was successfully demonstrated on four spherical elements to high burnup at 1000°C.

Thus although this work has shown that, in principle, BeO coatings can retain fission gases to the standard required, further work to develop ABORIGINE fuel elements should include:

(i) Further low burnup sweep capsule irradiations to assess fabrication/inspection standards.

(ii) High burnup sweep capsule tests on 'shell-and-kernel' type fuel elements over a range of temperatures and burnups.

(iii) Study and elimination of other possible cracking mechanisms, for example thermal stress and thermal cycling.

(iv) An investigation of the fission gas release from cracked spheres at various temperatures and burnups to provide data from which the allowable breakage rate could be determined more accurately.

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8. REFERENCES

Carroll, R.M. (1965). - Nuc. Sci. and Eng. 21, 141.

Carroll, R.M. and Sisman, O. (1966). - Nuc. Applications. 2, 142.

Carroll, R.M. and Sisman, O. (1965). - Nuc. Sci. and Eng. 21, 147.

Hanna, G.L., Walker, D.G. and Beach, P.M. (1967). - AAEC/TM384.

Hanna, G.L. and Reeve, K.D. (1972). - AAEC report in preparation.

Hickman, B.S., Rotsey, W.B., Hilditch, R.J. and Veevers, K. (1968). J. Am. Ceram. Soc. 51, (2), 63.

Reeve, K.D. and Ramm, E.J. (1965). - AAEC/TM251.

Reeve, K.D. and Ramm, E.J. (1969). - AAEC/TM521.

Reeve, K.D., Clare, T.E., Silver, J.M. and Bridgeford, K.C. (1966). - AAEC/TM333.

Roman, D., Randall, C.H. and Hanna, G.L. (1969). - AAEC/E201.

Roberts, W.H. (1964). - J. Nuc. Mater. 14, 29.

Schurenkamper, A. and Soulhier, R. (1964). - CEA-R2588.

Walker, D.G. and Hickman, B.S. (1967). - J. Nuc. Mater. 24, 60.

TABLE 1

DETAILS OF SPECIMENS USED IN THE X-124 SERIES OF IRRADIATION EXPERIMENTS

Rig Number	Specimen Type	BeO Grain Size, μm	Fuel Particles Batch and Grain Size, μm	U:Th Molar Ratio	Sintering Temperature and Time	Mean Open Porosity, %	
						Pre-irradiation	Post-irradiation
X-124-5	Fuel Particles		X-124-1,2 4-7	1:10.5	1700°C 2 hr.	ND	ND
X-160-1*	Fuel Particles		COP-25-B2 20	1:10	1700°C 2 hr.	ND	ND
X-124-1	Spheres	Core 5-100 Shell 3	X-124-1,2	1:10.5	1450°C 3 hr.	0.03	0.08
X-124-2	Spheres	Core 10-100 Shell 10-100	X-124-1,2	1:10.5	1525°C 3 hr.	0.08	0.13
X-124-3	Spheres	Core 5 Shell 5	X-124-3 13-100	1:9.8	1450°C 3 hr.	0.03	0.32
X-124-4	Spheres	Core 5 Shell 5	X-124-3	1:9.8	1450°C 3 hr.	-	-

* X-160 was the replacement for rig X-124; identical design.

ND Not determined.

TABLE 2

FISSION GAS RELEASE RATE PARAMETERS FOR BARE FUEL PARTICLES, BATCHES COP-25-B2 AND X-124-1,2

Rig Number	Irradiation Temp., °C	Burnup atom per cent (U + Th)	Release Rate : Birth Rate				
			^{85m}Kr	^{87}Kr	^{88}Kr	^{133}Xe	^{135}Xe
X-124-5 (COP-25-B2)	600	0.007	1E-4-1.2E-4	4.2E-5-4.7E-5	7E-5-1E-4	1.3E-4-1.7E-4	3.5E5-5E-5
	660	0.01	1.1E-4-1.4E-4	3.6E-5-4.2E-5	8.5E-5-1E-4	6E-5-8E-5	3.5E-5-4.5E-5
	710	0.013	1.1E-4-1.5E-4	2E-5-4E-5	7E-5-1E-4	1E-4-1.3E-4	2E-5-3.5E-5
	760	0.017	1.4E-4-1.7E-4	1.6E-5-3E-5	6.5E-5-1.1E-4	2E-4-3E-4	1.4E-5-2.3E-5
	810	0.02	2.1E-4-2.7E-4	3.3E-5-4.5E-5	1.3E-4-1.5E-4	4E-4-5.4E-4	2E-5-3.5E-5
X-160-1 (X-124-1,2)	750	0.004	9E-4-1.4E-3	4E-4-6.6E-4	8E-4-1.1E-3	5.6E-4-8E-4	5E-4-7E-4
	710	0.012	1.8E-3-2.5E-3	5E-4-9E-4	1.1E-3-1.8E-3	1E-3-1.6E-3	6E-4-9E-4
	650	0.015	3E-3-3.4E-3	9E-4-1.1E-3	2E-3-2.4E-3	1.3E-3-2.4E-3	1.1E-3-1.4E-3
	650	0.022	5.6E-3-9E-3	1.4E-3-1.9E-3	4E-3-6.7E-3	5.5E-3-9E-3	2E-3 - 3.3E-3
	650	0.027	8E-3-1.3E-2	1.6E-3-2.4E-3	5E-3-8.7E-3	7E-3-1E-2	3E-3-5.6E-3
	650	0.03	1E-2-1.4E-2	2E-3-3E-3	6E-3-8.7E-3	8.1E-3-1.4E-2	3.4E-3-5.4E-3

The notation E-n is used to denote the order of magnitude of the release parameter. For example, 1.4E-4 means 1.4×10^{-4}.

TABLE 3

FISSION GAS RELEASE RATE PARAMETERS FOR SMALL FUELLED SPHERES IRRADIATED IN THE X-124 SERIES RIGS

Rig Number	Irradiation Temp., °C	Burnup atom per cent (U + Th)	Release Rate : Birth Rate			
			^{85m}Kr	^{87}Kr	^{133}Xe	^{135}Xe
X-124-1	840	0.015	3E-7 - 6E-7		3E-7 - 6E-7	3E-7 - 9E-7
X-124-2	700	0.01	5E-7 - 4E-6		1E-6 - 2E-6	1.5E-7 - 6E-7
	800	0.017	3E-6 - 4E-6		4E-5 - 6E-5	1E-7 - 1E-6
		0.024	1.5E-6 - 3.5E-6		2E-5 - 8E-5	1.5E-6 - 2.5E-6
		0.031	1E-6 - 2E-6		4E-6 - 2E-5	1E-6 - 3.5E-6
	850	0.036	1.5E-6 - 2.5E-6		1.5E-6 - 7E-6	2.5E-6 - 3E-6
X-124-3	460	0.55	1.2E-4 - 1.4E-4	2.7E-5 - 3.2E-5	2E-4 - 2.3E-4	2E-4
	650	0.24	9E-5 - 1.2E-4	2.1E-5 - 2.5E-5	2.2E-4 - 3.3E-4	1.1E-4 - 1.4E-4
	750	0.4	1E-4 - 2E-4	3E-5 - 4.4E-5	2E-4 - 4E-4	1.3E-4 - 2.8E-4
	830	0.5	3E-4 - 4E-4	5E-5 - 6E-5	3E-4 - 4E-4	3E-4 - 3.6E-4
X-124-4	600	0.12	7E-5 - 8E-5	1E-5 - 1.3E-5	1.7E-4 - 2.4E-4	5.5E-5 - 7E-5
	650	0.2	7E-5 - 8E-5	9E-6 - 1.1E-5	1.7E-4 - 2.2E-4	6E-5 - 8E-5
	750	0.35	1.1E-4 - 1.4E-4	1.4E-5 - 1.8E-5	2.2E-4 - 2.7E-4	9E-5 - 1.2E-4
	840	0.41	1.7E-4 - 2E-4	2E-5 - 4E-5	2.8E-4 - 3.4E-4	3E-4 - 3.4E-4

TABLE 4

DETAILS OF SPECIMENS IRRADIATED IN THE X-132 SERIES RIGS

Rig Number	Fuel Particles	Weight of U-235, g	Sintering Addition	Sintering Temp., °C	Sintering Time, hr.	Buffer Zone	Density g cm^{-3}
X-132-1	COP-25-B2 200 μm 7.4 vol. per cent	0.0003	-	1400	6	No	2.98
X-132-2	X-124-3 200 μm 5 vol. per cent	0.006	0.1% MgO	1450	6	No	3.06
X-132-3	X-124-3 200 μm 5 vol. per cent	0.006	0.1% MgO	1400	6	No	3.06
X-132-4	1U:1Th 5 μm 1.8 vol. per cent	0.007	0.1% MgO	1450	6	No	2.93
X-132-5	1U:1Th 5 μm 1.9 vol. per cent	0.007	-	1400	6	Yes	2.87
X-132-6	1U:1Th 5 μm 1.7 vol. per cent	0.043	0.1% MgO	1450	6	Yes	2.88

TABLE 5

FISSION GAS RELEASE PARAMETERS FOR 2 cm DIAMETER SPHERES IRRADIATED IN X-132 SERIES RIGS

Rig Number	Temp. °C	Burnup atom per cent (U + Th)	nvt > 1MeV	Release Rate : Birth Rate				
				^{85m}Kr	^{87}Kr	^{88}Kr	^{133}Xe	^{135}Xe
X-132-1	1000	0.003	4.5×10^{19}	1.1E-6-3E-6			1.5E-6-4E-5	1.2E-6-6E-6
	1000	0.006	7×10^{19}	1.1E-4	1.1E-5	5.6E-5	2.8E-4	7.7E-5
	760	0.008	9×10^{19}	5.2E-5	6.7E-5	2.5E-5	1.5E-4	1.9E-5
X-132-2	1000	0.54	2.6×10^{20}	1.3E-7-2.4E-7	5E-8-9.6E-8	8.5E-8-1.6E-7	1.6E-7-3.1E-7	2.3E-7-4E-7
		1.07	6×10^{20}	3.8E-7-4.7E-7	1.4E-7-1.9E-7	2.5E-7-3.5E-7	3.2E-7-4.5E-7	6.4E-7-8.3E-7
		1.13	6.5×10^{20}					
		After Fracture		1E-4	1E-5	5E-4	1.8E-3	9E-5
X-132-3	1000	0.21	1.3×10^{20}	1.3E-7-3.3E-7	3.4E-8-9E-8	7.3E-7-1.6E-7	1.7E-7-3.2E-7	2.8E-7-4.3E-7
		0.78	3.3×10^{20}	5.6E-7-8E-7	1.5E-7-2.8E-7	3.2E-7-4.9E-7	1E-6-1.5E-6	1.3E-6-1.7E-6
X-132-4	700	0.062	1×10^{19}	2.7E-5	8E-6	2E-5	5E-5	6E-5
X-132-5	850	0.25	2×10^{19}	3E-4	1E-4	2E-4	6E-4	6E-4
X-132-6	1200≠	0.83	1×10^{19}	1E-5	4E-6	1E-5	1E-5	3E-5

≠ Ball temperature calculated from temperature of specimen holder.

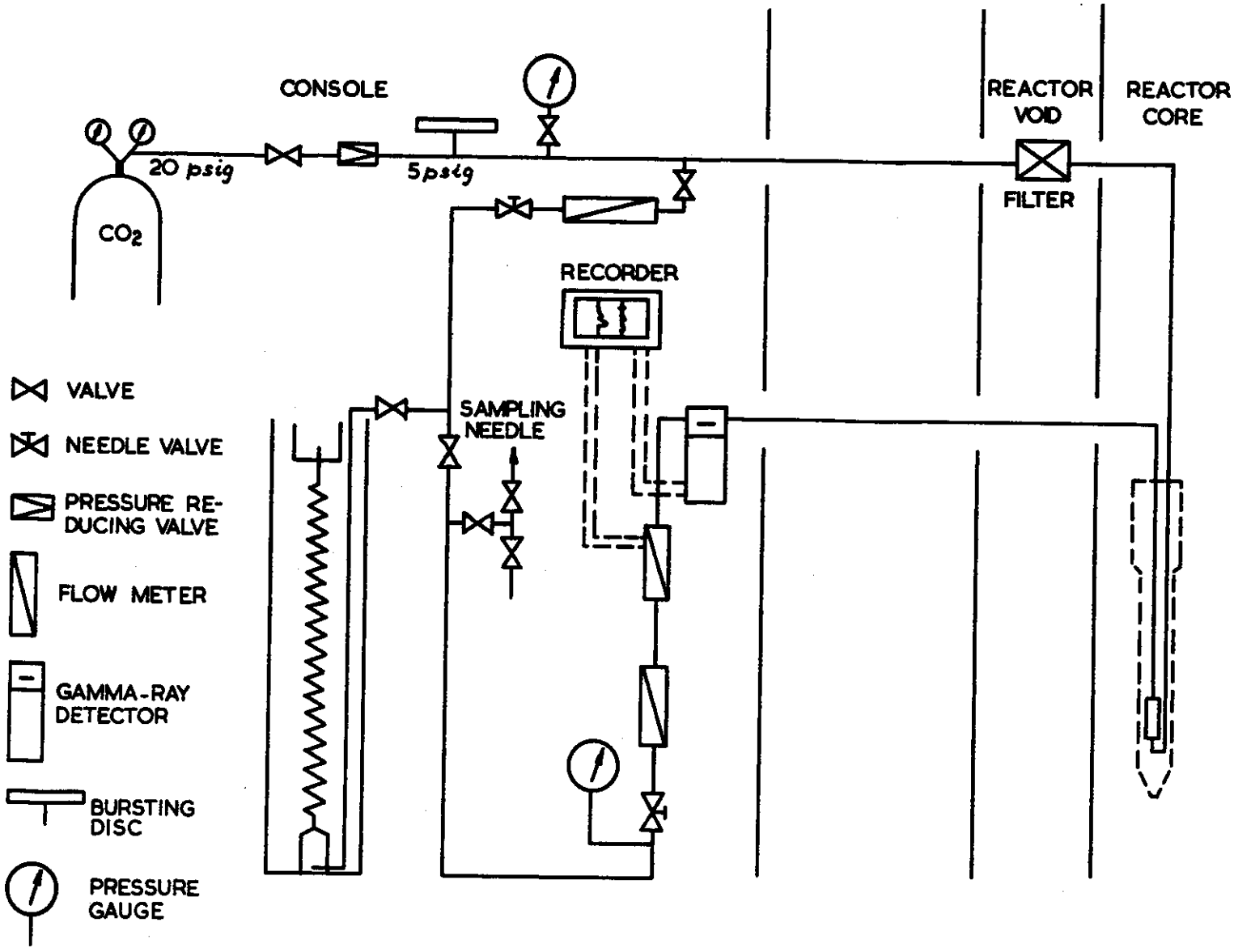

FIGURE 1. SWEEP-GAS CIRCUITS IN IRRADIATION EXPERIMENTS X-124, X-160, AND X-132

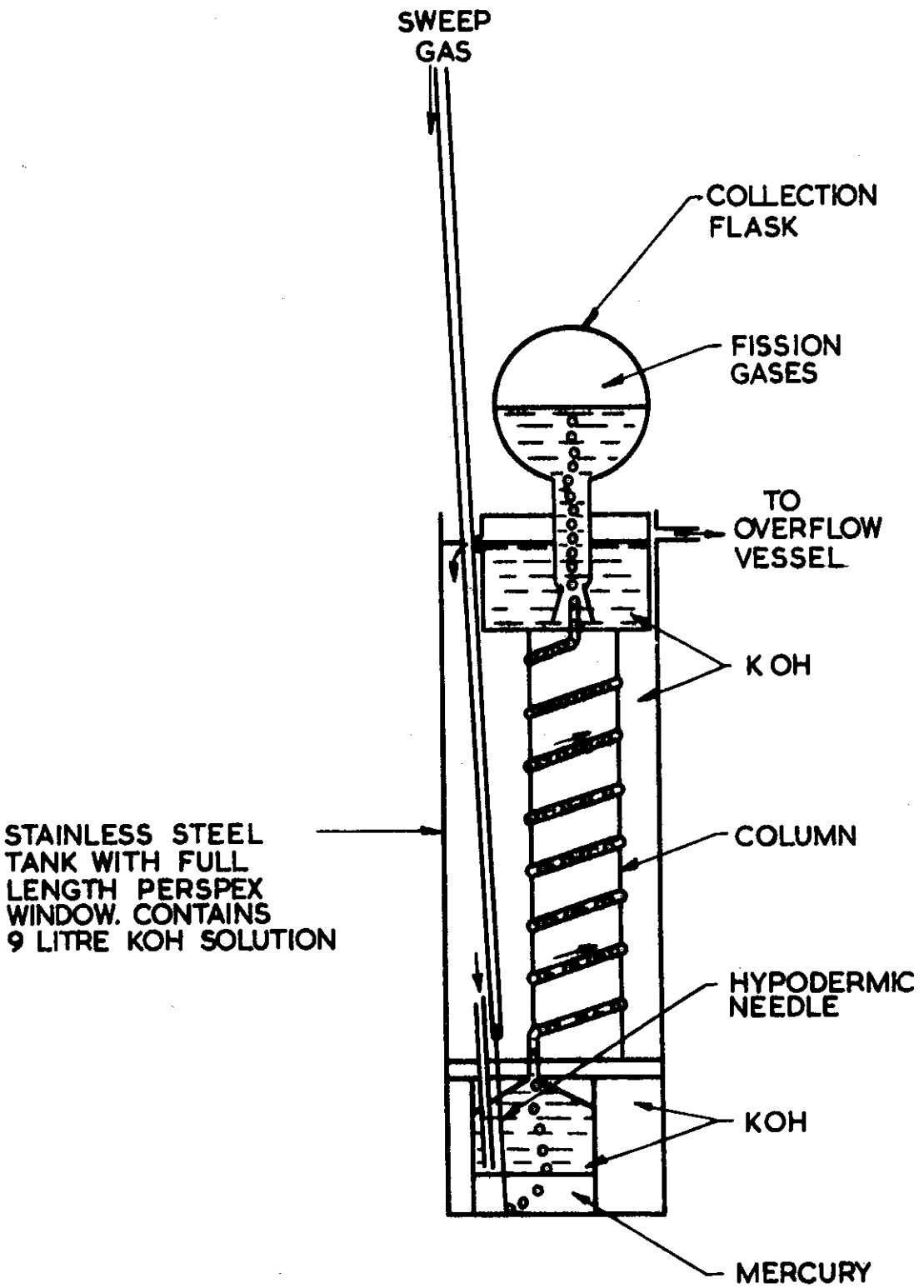

FIGURE 2. MODIFIED APPARATUS FOR DISSOLVING CO_2 SWEEP-GAS IN RIGS X-132 AND X-160

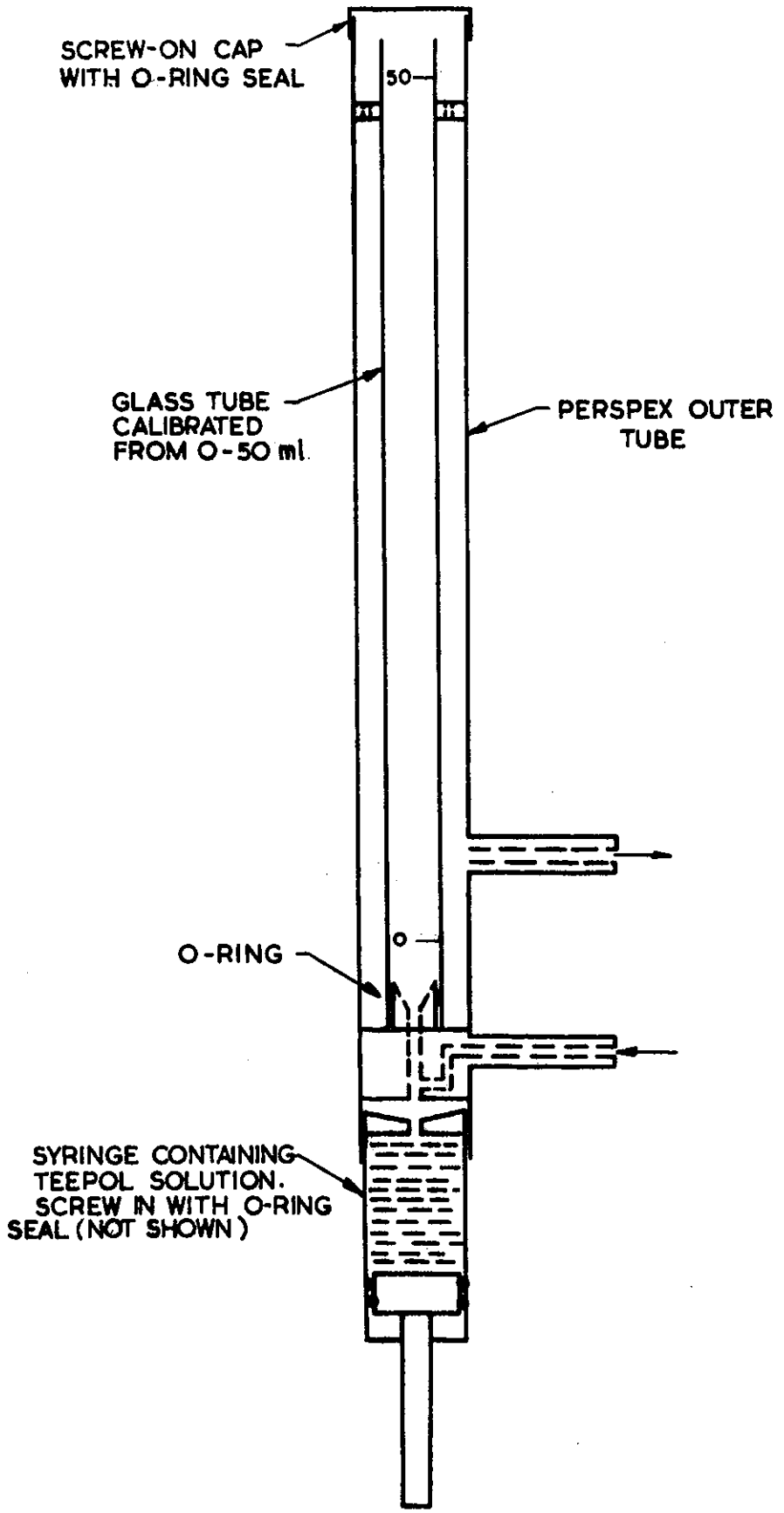

FIGURE 3. SCHEMATIC DIAGRAM OF THE BUBBLE TRANSIT DEVICE FOR MEASURING GAS FLOW RATES

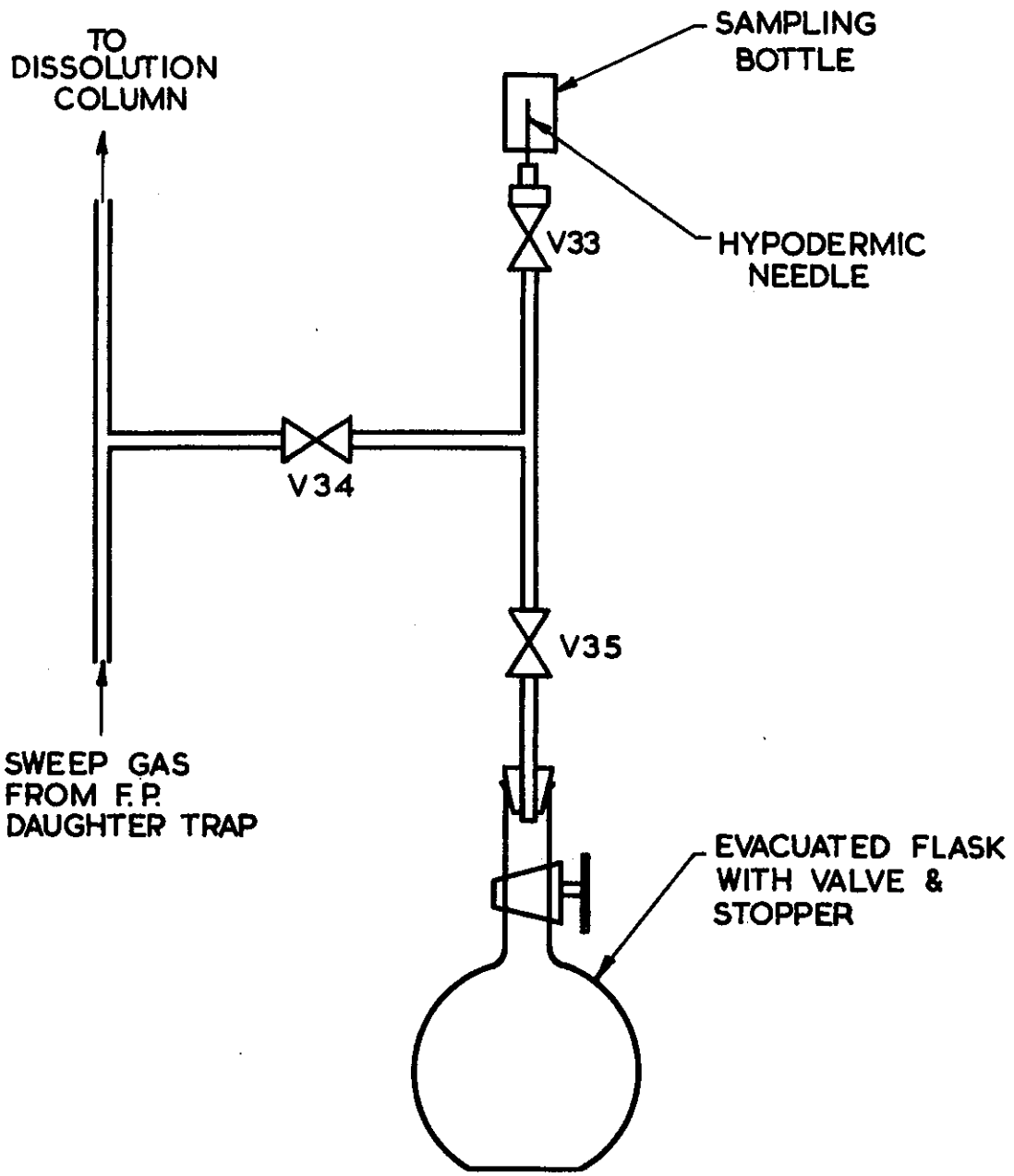

FIGURE 4. HYPODERMIC NEEDLE SAMPLING DEVICE

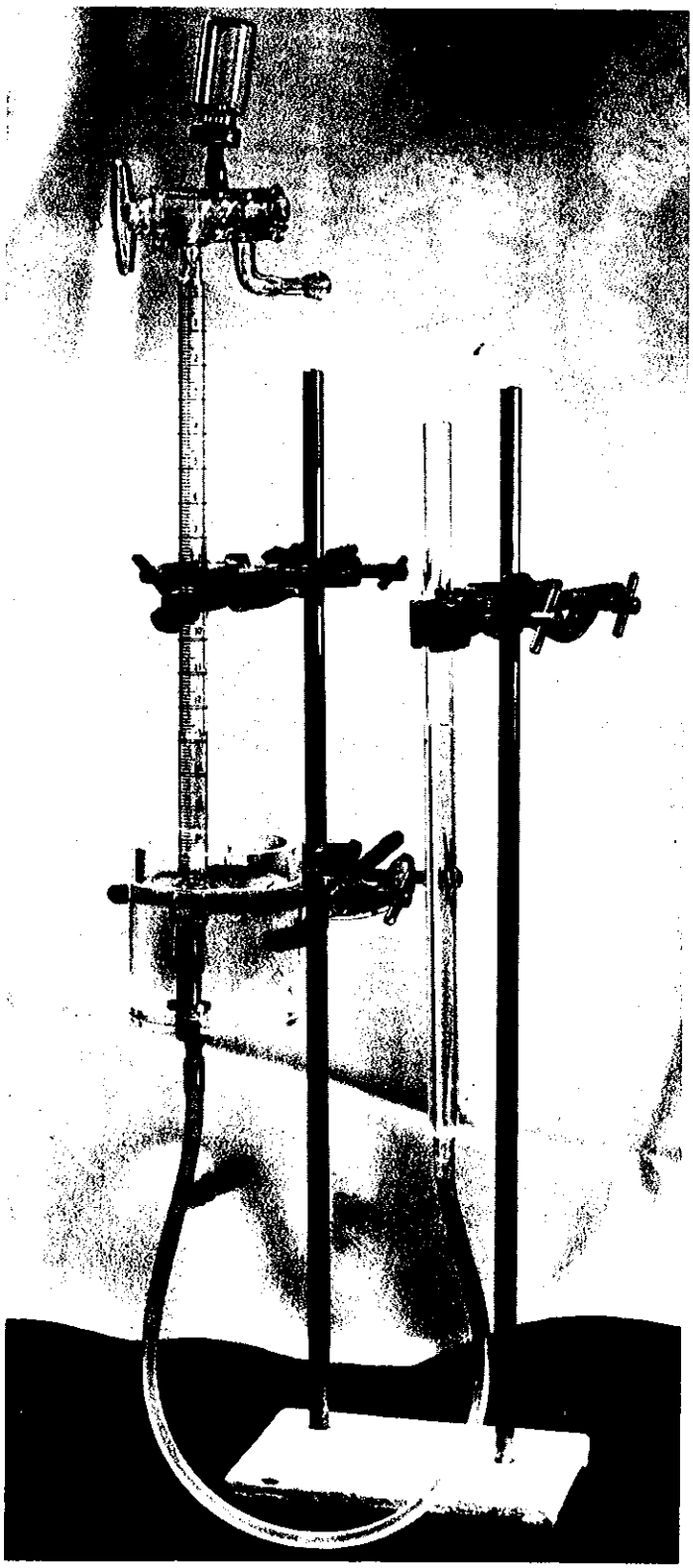

FIGURE 5 Burette gas sampler and pressure equaliser. Gas collected in the burette is transferred to the serum vial after evacuating it via the 2-way stopcock. All burette readings are made at atmospheric pressure by equalising liquid levels in the U-tube.

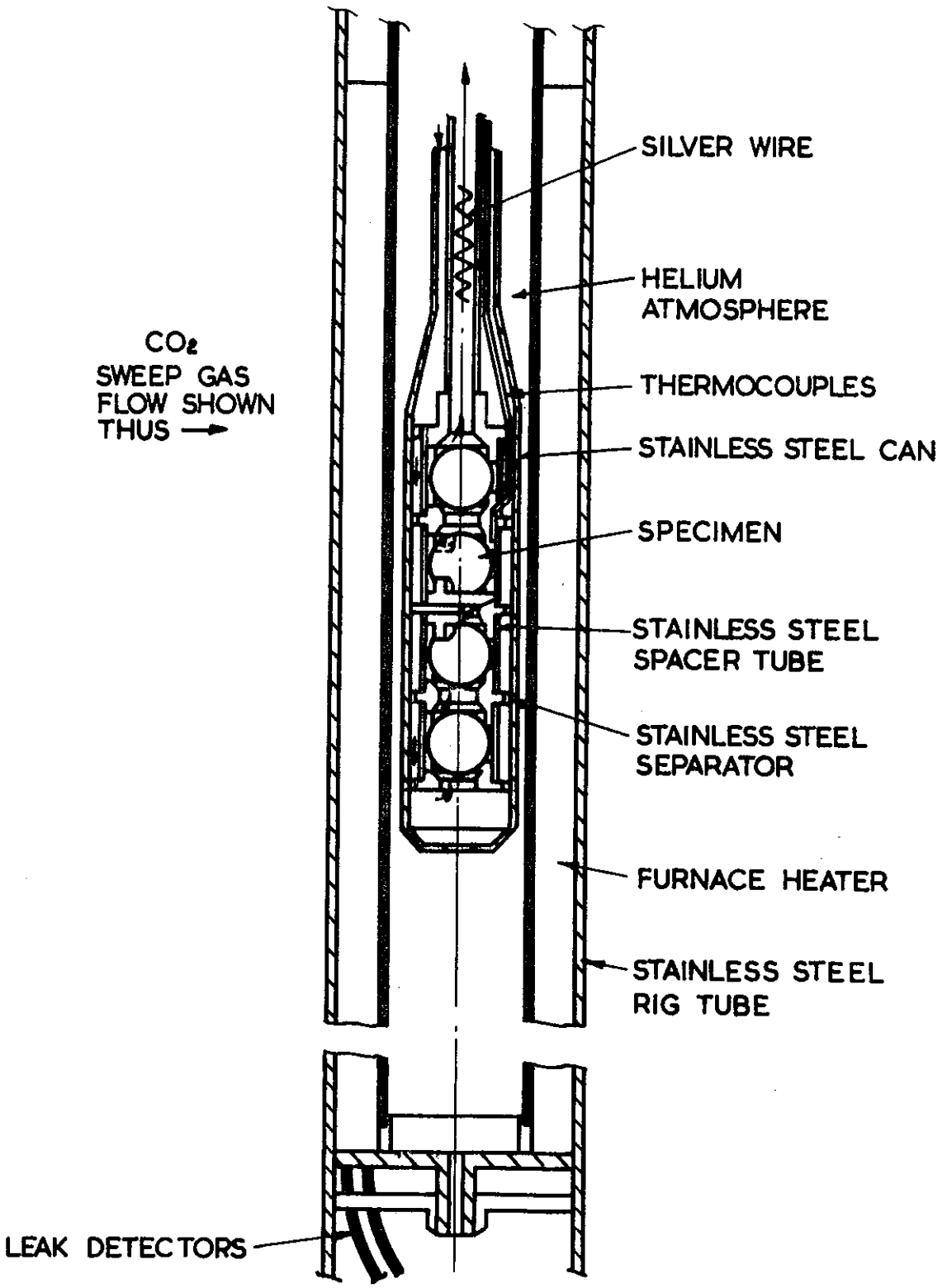

FIGURE 6a. FUEL BALL SPECIMEN CAN ASSEMBLY

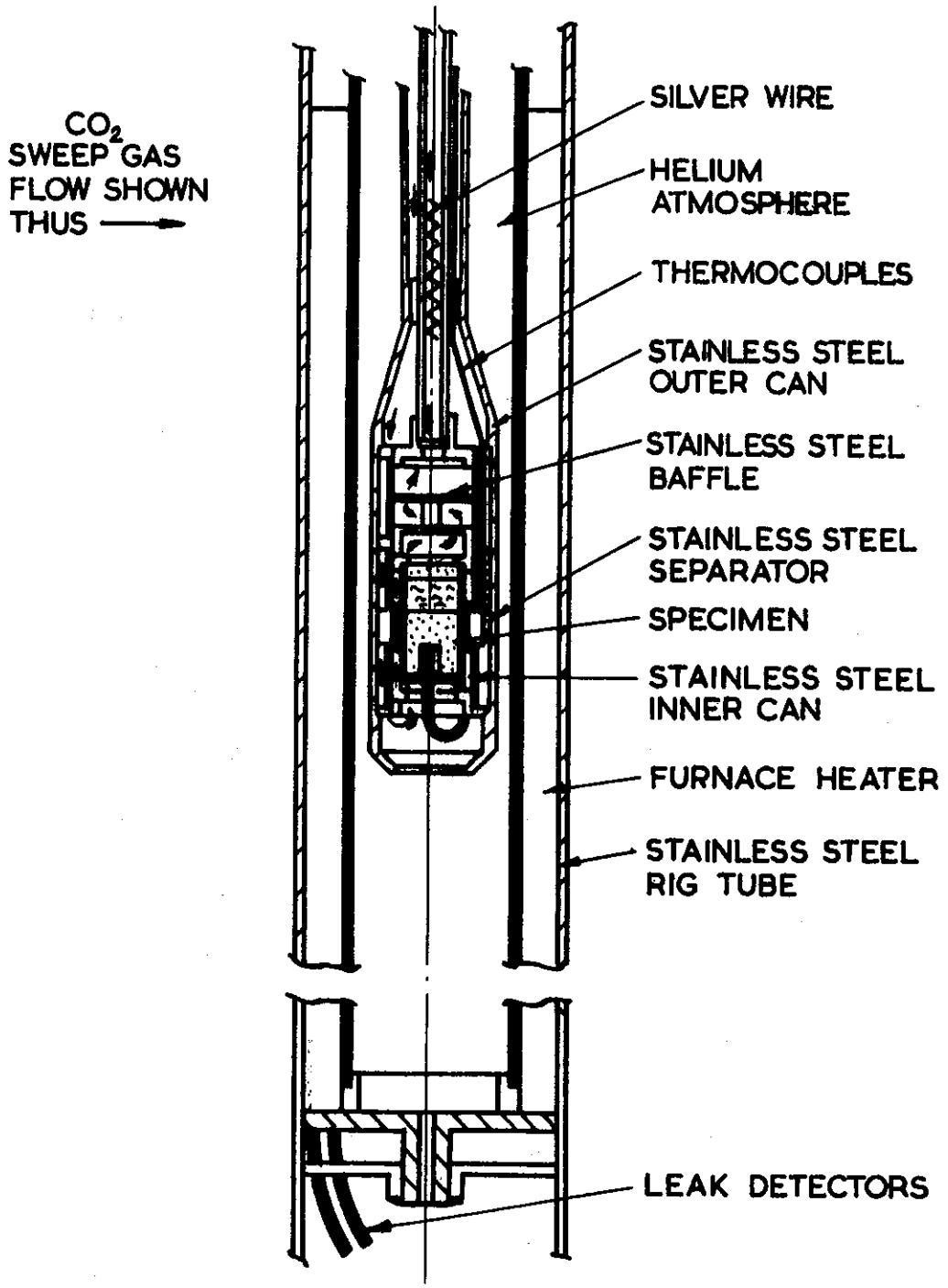

FIGURE 6b. FUEL POWDER SPECIMEN CAN ASSEMBLY

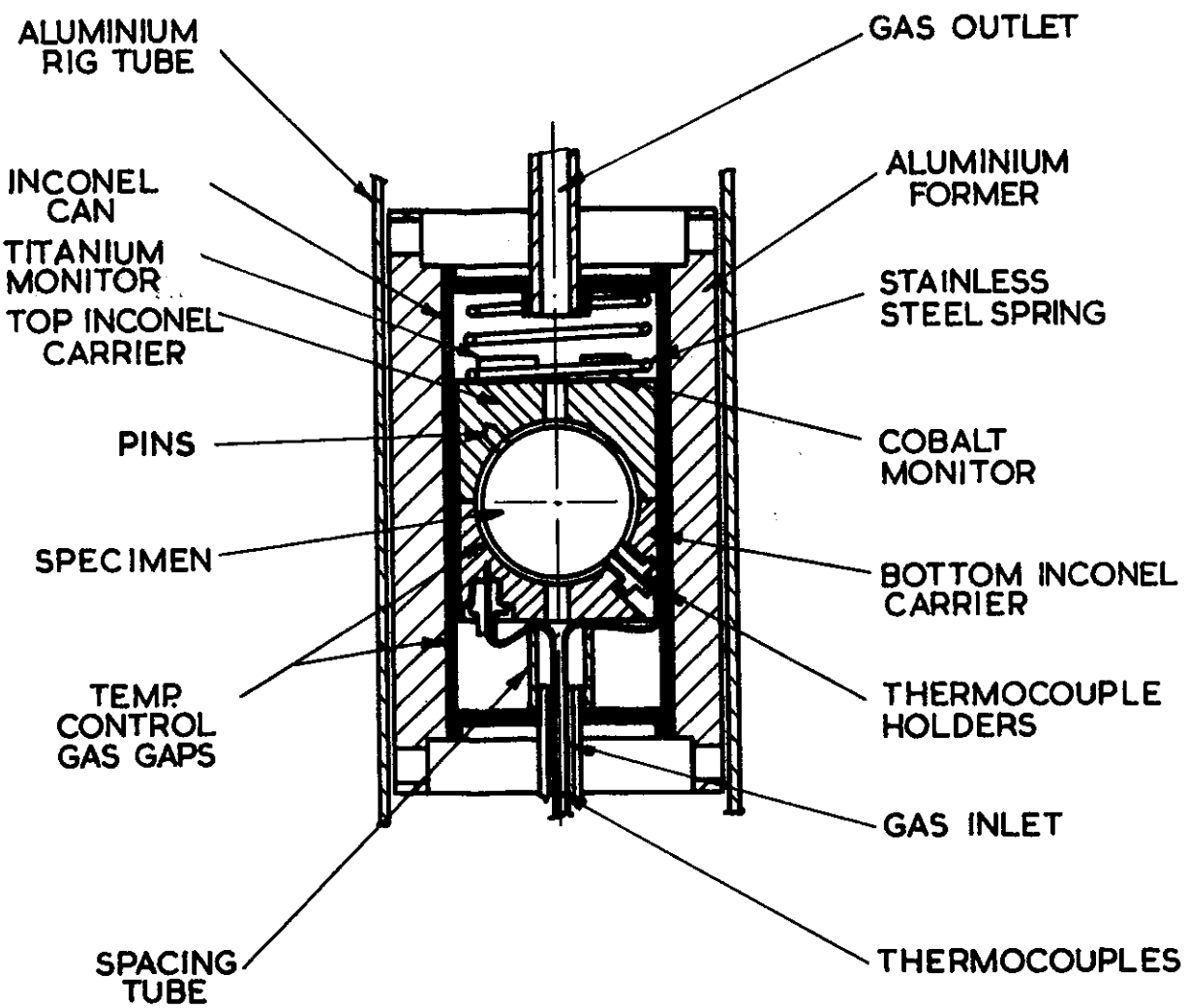

FIGURE 7. SPECIMEN CAN ASSEMBLY

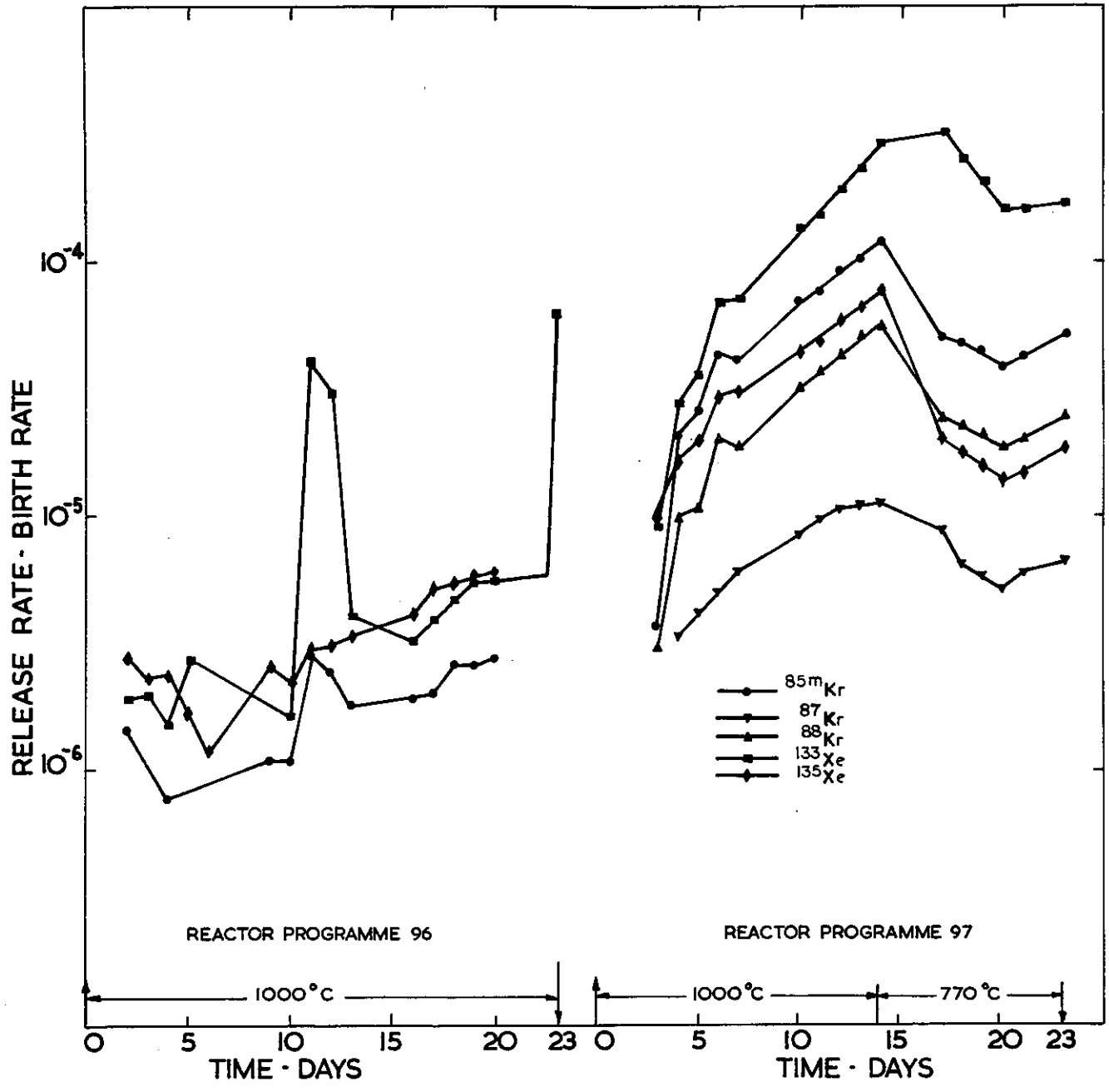

FIGURE 8. FISSION GAS RELEASE RESULTS FOR X-132

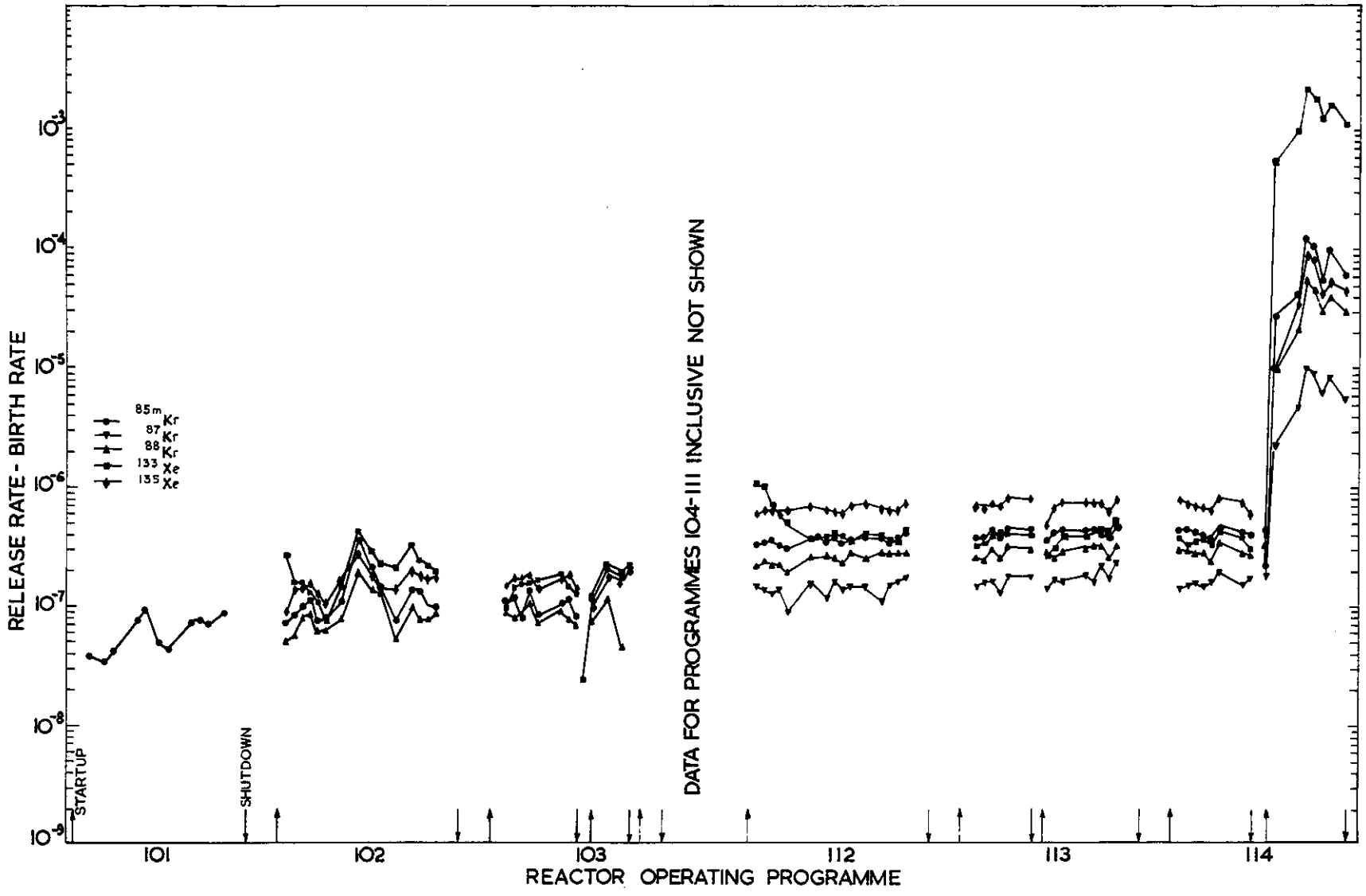

FIGURE 9. FISSION GAS RELEASE RESULTS FOR X-132-2

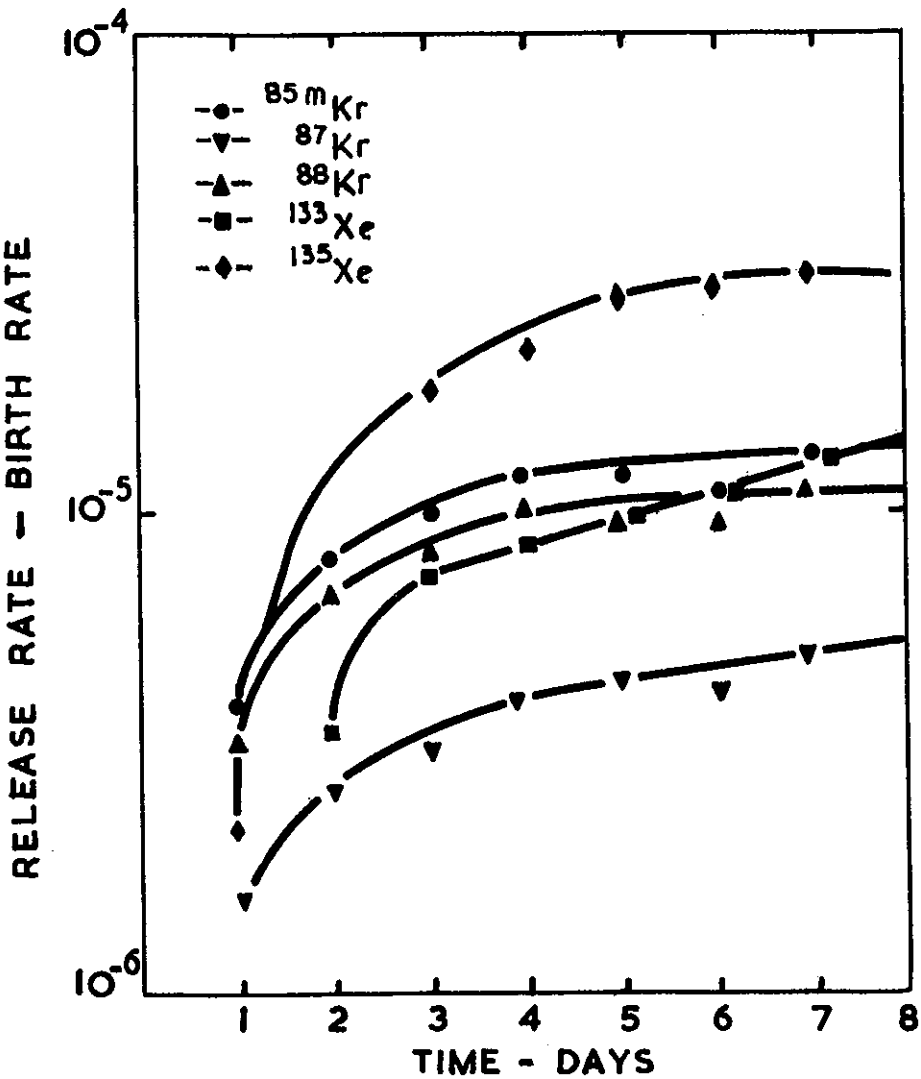

FIGURE 10. FISSION GAS RELEASE RESULTS FOR X-132-6

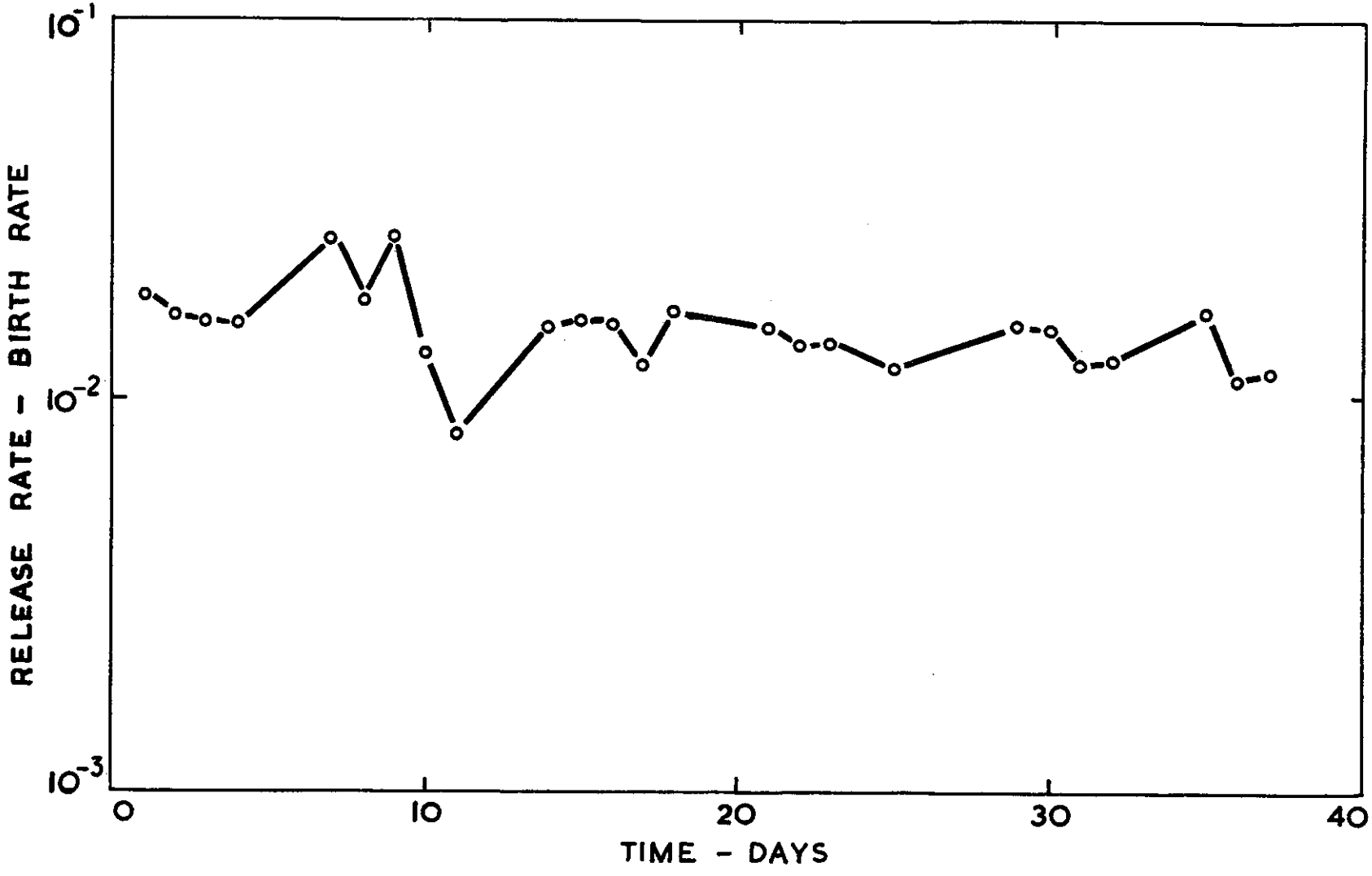

FIGURE 11. ^{131m}Xe RELEASE RATE FROM ^{131}I IN OUT-OF-PILE EXPERIMENT

APPENDIX

THERMAL CYCLING TESTS ON SMALL ABORIGINE FUEL ELEMENTS

1. INTRODUCTION

The early failures in experiments X-132-4 and X-132-5 raised doubts about the advisability of continuing with the sixth experiment in the series, particularly since this specimen would be taken to a higher fission burnup and would be subject to a fission heat rating and thermal stress about six times those in earlier experiments.

Four specimens were available from the same fabrication batch as the X-132-6 specimen and it was decided to subject these to a series of thermal cycles to test their resistance to thermal stresses.

2. THERMAL CYCLING PROCEDURE

Experience with in-pile sweep capsule experiments had shown that fission gas release is a very sensitive method of detecting cladding failure. Accordingly, specimens were irradiated at pile temperature to a fission density of 3×10^{15} fissions cm^{-3} of $(U,Th)O_2$ and thermally cycled under flowing helium with continuous monitoring of the gamma activity of the outlet gas. The apparatus, shown schematically in Figure A1, consisted of a helium-swept stainless steel capsule which was moved in and out of the hot zone of the furnace. On completion of each series of cycles, specimens were examined for macrocracks using a fluorescent dye penetrant.

Specimens were cycled as summarised in Table A1 from a nominal maximum surface temperature of 1000°C (the irradiation temperature used in the X-132 experiments) to a minimum of 500°C or 280°C. Temperatures of less than 280°C were not possible with the equipment used. Cycling times were 40 minutes in the hot zone and two hours and 60 minutes in the cold zone for temperature minima of 280°C and 500°C respectively. The cooling rate from 1000°C to 500°C was approximately 80°C per minute and the surface temperature reached 280°C after about 35 minutes in the cold zone. The profile of the heating cycle was very close to the inverse of that of the cooling profile.

During the first run (specimen LH65) the helium sweep-gas was passed over refrigerated charcoal for two periods of two hours each. Both

(cont...)

Appendix (cont.)

charcoal traps were counted for radioactivity on a gamma spectrometer but only background levels were detected.

When all specimens had been cycled and examined for macrocracks, each was re-irradiated to a further 3 x 10^{15} fissions per cm^3 and tested for fission gas retention at 1400°C by the post-activation diffusion (PAD) technique in the manner described by Roman, Randall and Hanna (1969). After specimen LH65 had been annealed it was deliberately cracked (confirmed by dye penetration) by quenching from about 400°C in a water spray. It was then re-annealed at 1400°C and the gas release compared with that observed before spray quenching.

3. RESULTS

All the tests done during and after thermal cycling indicated that the four specimens survived the cycling without fracturing. The results are summarised in Table A2.

The sweep-gas activity during cycling did not rise above background and the refrigerated charcoal samples showed no ^{133}Xe (the expected species if release did occur). Dye penetration did not show any cracks in the as-cycled specimens but clearly revealed numerous cracks in the spray-quenched sample LH65. In post-irradiation annealing of the as-cycled specimens the release of ^{133}Xe was undetectable or barely detectable, indicating a release rate parameter (D') of less than 10^{-17} sec^{-1} which is characteristic of spheres with satisfactory gas retention (Roman, Randall and Hanna 1969).

4. DISCUSSION

No adequate check of the sensitivity of the sweep-gas activity measurements during thermocycling was possible as the rig had to be dismantled before a cracked sphere could be tested. However, the dye penetration and PAD test on the cracked LH65 sphere indicated that these two tests were suitable crack detection techniques. It is concluded therefore, that none of the four specimens fractured during thermal cycling.

The cooling rates achieved in these tests compare favourably with those encountered in irradiation experiments during a normal reactor shutdown. Hence, the tests indicate that the spheres should survive the temperature cycling likely to occur during the X-132-6 experiment. Whatham (private communication) has studied the cooling rate obtained

(cont...)

Appendix (cont.)

during the thermal cycling tests and has estimated that the stress induced in a pure BeO sphere of uniform density would be about 2700 p.s.i. He believes that the stress would be less in a shell-and-kernel sphere where the porous buffer zone would reduce the rate of heat transfer to the inner surface of the BeO shell. This value is much lower than the fracture stress of BeO and is consistent with the survival of all balls on thermal cycling.

The stress generated during steady state irradiation at 1000°C of a sphere of uniform composition and density (assuming the same overall composition as the X-132-6 sphere) is calculated to be 12,100 p.s.i. However, the stress calculated for a hollow sphere of the same dimensions and density as the cladding and having the same ΔT as during the irradiation, is only 16 p.s.i. Measurements of the crushing strength at 600°C (Reeve, private communication) and stress relaxation rates in porous BeO (Rotsey and Wood, private communication) indicate that the buffer zone will deform very easily at 1000°C to accommodate expansion of the kernel and will stress the shell only slightly. It would seem justifiable then, to assume that the stress at the sphere surface during steady state irradiation will be comparable to that calculated for the hollow sphere.

5. CONCLUSION

The thermal cycling tests have adequately simulated the thermal stress conditions likely to be met in the X-132-6 irradiation and specimen failure due to thermal shock should not occur during that experiment.

TABLE A1

THERMAL CYCLING HISTORIES OF ABORGINE FUEL SPHERES

Specimen No.	No. of Temperature Cycles	
	1000-500°C	1000-280°C
LH-65	36*	18
LH-61	Nil	28
LH-64	Nil	30
LH-67	11	68*

* First series of cycles; dye penetration crack detection done before second series of cycles.

TABLE A2

SUMMARY OF RESULTS OF THERMAL CYCLING TESTS

Specimen	Sweep-Gas Activity	No. of cracks, Dye-penetration	Xe-133 activity after 100 min. PAD anneal
LH-65 as cycled	Background	Nil	Nil
LH-65 water quenched	-	Many	1.2×10^5 c.p.m.
LH-61 as cycled	Background	Nil	Nil
LH-64 as cycled	Background	Nil	Nil
LH-67 as cycled	Background	Nil	Nil

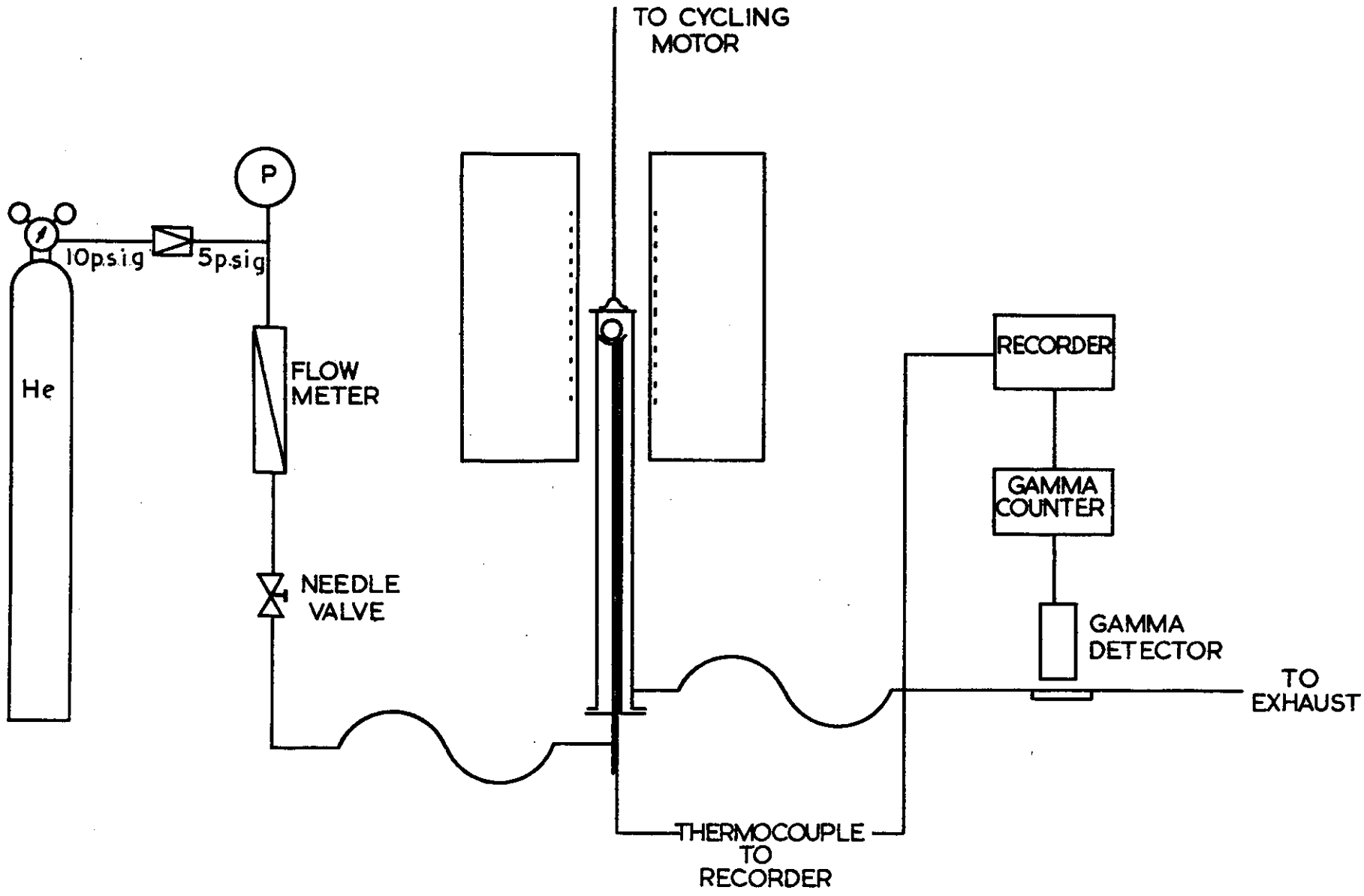

FIGURE A1. FLOW DIAGRAM FOR THERMOCYCLING RUNS USING A HELIUM SWEEP-GAS