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Apparatus for Measuring the Release of Fission Gases and other Fission Products by Degassing

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ABSTRACT

In gas-cooled high-temperature reactors the fuel is in general inserted in the fuel elements in the form of small particles, which are for example coated with pyrolytic carbon. The purpose of this coating is to keep the fission products separate from the coolant gas. The further development of these coated particles makes it necessary to check the retention capacity. One possible method of doing this is the degassing test after irradiation in the reactor. An apparatus is described below, which was developed and installed in order to measure to a higher degree of sensitivity and in serial measurements the release of fission gases and sparingly volatile fission products.

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1. INTRODUCTION

It has been found in the development of modern gas-cooled high-temperature reactors that graphite can not only be used for static reactor components but that it is of considerable importance in particular for the production of new types of fuel elements. On the one hand, fuel manufacturers have developed processes for the cladding of small fuel particles with pyrolytic carbon and, on the other hand, reactor fuel elements are being manufactured by means of graphite. The technology of graphite fuel elements is well advanced and in course of further development, in particular in the case of the pebble bed reactor.

When considering fuel elements, intended to be inserted in an existing reactor or proposed for use in a reactor still under development, the reactor engineer requires in particular information on fission product release during operation. This is of particular importance with modern reactor concepts, where energy conversion is planned to take place directly in the primary circuit by means of a gas turbine or magnetohydrodynamic generator.

The formula

$$\frac{R}{B} = \frac{\text{Release rate in the reactor}}{\text{Formation rate in the reactor}}$$

has been introduced here as the characteristic quantity for the release of a fission product. It indicates for the case where the formation and decay rates of the nuclide in question are in equilibrium, what fraction of the permanently present quantity is passing into the coolant gas.

A possible method for direct measurement of the release rate is an experimental irradiation in the reactor with sweeping of the capsule. The sweep gas transports the fission gases out of the capsule and they can then be measured as a function of time outside the reactor. An experiment of this type is, however, very laborious and for technical reasons may not always be possible. A procedure has therefore been introduced whereby the parameter R/B can be obtained from a post-irradiation investigation of the fuel element. The method consists in heating the irradiated element, followed by measurement of the fission products thus released. The basic assumption here is that the continuous release in the reactor ceases virtually immediately on shutdown of the latter, since the fuel element temperature falls rapidly at the same time. Since the release of fission products is very temperaturedependent, it decreases with a fall in temperature and is in general no longer measurable below 100°C. Furthermore, the release rates, acceptable to the reactor constructor on safety grounds, are very low, so that there is no question of an exhaustion of the amount available for release. The fuel element for investigation is exposed during the heating test to temperatures which either correspond to or exceed the irradiation temperature. The only reason for heating at low temperatures is to bring out experimentally the

marked temperature-dependence of the release of fission gases. While the fuel with its cladding are at the set temperature, fission products pass to the external medium continuously. These are then removed and, shielded from the intensively radiating the fuel element, collected in a trap for quantitative determination.

There has been general agreement in the conventional heating test to the determination of released xenon-133, which has played an important part in safety assessments for high-temperature reactors and is easy to handle from the point of view of the measurement technique. The test was introduced in order to provide information on ruptured or superficially contaminated coated fuel particles, since these were the principal initial difficulties encountered in the production of this fuel.

The measurement of fission product release was subsequently extended to coated particles which had neither highly contaminated clads nor visible cracks. On the assumption that this release was due only to diffusion, the test could be restricted to measurement of the xenon-133 and the release of other nuclides estimated from their diffusion constants. The xenon release would then provide a true criterion for the "leak-proof" properties of the cladding.

However, since these assumptions could not be maintained and it is also doubtful whether the release of fission products is due only to the cladding of the fuel particle, the attempt is made today to

measure expressly at least the nuclides, which are of critical importance for safety assessments, or better still to cover as many nuclides as possible. A specially modified apparatus was developed in order to achieve this aim, to the extent possible in a post-irr diation investigation due to the restricted half-lives. The degassing of Xe^{133} naturally retained its importance here also as a general behavior test for fuel elements, since this measurement is regarded as standard.

The development and installation of the apparatus took place within the framework of the THTR (Thorium High-Temperature Reactor) project at the Nuclear Research Plant Jülich GmbH.

1.1 Layout of apparatus

The layout of the apparatus was governed primarily by the measurement requirements and then by the factors of space and technical availability. The measurement requirements will be given in the subsequent section and their implications for planning described in predominantly qualitative terms.

The degassing experiments were to be carried out mainly on whole spheres, namely spherical graphite fuel elements, such as are used in the AVR and are envisaged for subsequent use in the THTR. They have a diameter of 60 mm and contain a charge of 11.41 g, consisting of 1.11 g uranium and 10.30 g thorium. It was also intended that experiments should be carried out on loose, coated fuel particles in small

quantities obtained from special irradiation experiments. The maximum quantity of 4 to 5 g, up to 50 % uranium burnup, taken after as short as possible a decay time, signified a maximum activity of ~1000 Ci during the experiment. The furnace was therefore installed in one of the cells of the "BZ III", a laboratory specially erected for the THTR subsequent investigations. The maximum possible measurement sensitivity is required, however, for the investigation of different sorts of loose coated particles, since in many cases the available particle quantities are small owing to the wide range of varieties in an irradiation experiment. This sensitivity could only be obtained by operating outside the cell in traps, which accumulate the xenon-133, and in detectors which determine the content. A special lead shielding was provided round the traps and detectors, in order to exclude the high and also variable background activity in front of the cells. In order to measure as much as possible of the accumulated xenon-133, the window in the trap was made as large and as thin as possible and sodium iodide crystals of exceptional quality were used for the measurement. The pipe connection from the furnace to the measurement apparatus was ~12 m in length. The maximum anticipated xenon-133 release was at a rough estimate 0.1 mm^3 in 10 h. The extraordinarily small quantities, which must therefore reach the measurement trap during the experiment, are most simply transported in a sweep gas. The released inert gases become frozen on to activated charcoal in the measurement trap at the temperature of liquid nitrogen or liquid air, so that only helium can be considered as a sweep gas in this case. The activated charcoal content in the should be kept as low as possible, in order to retain

favorable and constant conditions for the gamma-radiation yield during the activity measurement. The helium, which flows through the trap in large quantities during the course of the experiment ($\sim$ 50-100 l/h) should be thoroughly purified, since otherwise the charcoal becomes coated with impurities and loses its adsorption capacity. A further reason for this was the fear expressed by the particle manufacturer regarding the possible corrosion of the cladding, if the helium was not as pure and in particular water-free as in the reactor, i.e., a total contamination of 20-40 vpm. Provision was therefore made in the degassing apparatus for all the sweep gas, which is pumped through a membrane compressor, to be passed through a purification system before and during each experiment.

The degassing test was carried out at various temperatures. For determination of the behavior at a specific temperature, the latter must be held as constant as possible not only over a long period, but also over the full geometric extent of the object, in order to measure the release under equilibrium conditions. The temperature must also be reached quickly, however, if other processes of limited duration precede the steady state. This is in fact the case with initial bursts, which will be described in greater detail below, lasting from a few minutes to one hour. The power density, required to raise the temperature in a furnace from room temperature to 1200°C in a matter of minutes, can only be obtained with high-frequency induction heating.

It must not be possible for any fission

products (or activation products) apart from those carried by the inert gases to leave the cell and pass into the measurement trap during execution of the normal degassing test, since otherwise highly active substances would cause by their radiation an intensive background in the neighborhood of the line undergoing measurement, thus reducing considerably the measurement sensitivity. The overall activity obtaining outside the cell should be kept as small as possible. Furthermore, the nuclides, retained within the cell, must be reliably trapped, to permit their subsequent detection and, if necessary, quantitative measurement. The components, through which the released mixture in the sweep gas flows after leaving the heated zone, must be capable of easy replacement and removal after each experiment. These components are the pipe sections from the hot zone to the subsequent traps in the direction of flow of the sweep gas and the traps themselves. Apart from the inert gases, all fission products and impurities in dust form, which have not been previously condensed or become suspended during passage, are retained in these traps.

The anticipated sample size was found to be so great that it was necessary to arrange for an experiment to be carried out on all available working days, after setting aside 10-20 % of the possible working days were reserved for maintenance work. Since experience had shown that insertion of the sample in the furnace took several hours, if it was only carried out through a simple screwed cover with remote control by the cell manipulators, the fully automatic remote-controlled operation of the corresponding processes was incorporated

in the furnace design right from the start. The same applied to all valves inside and also outside the cell. A suitably designed control system sets automatically the preselected routine operating states, all processes being mutually interlocked in such a way as to exclude time-consuming or even dangerous operating errors.

1.2 Flow sheet

A diagrammatic picture of the whole apparatus has been derived from the above considerations. It is shown in Fig. 1 in the form of a flow sheet of the apparatus and is used below for a description of the principal aspects of the design.

A membrane compressor pumps the helium in the main circuit, which before reaching the furnace passes through the purification and a buffer vessel. The fission products are swept out of the furnace and the mixture passes through a so-called iodine trap followed by one of the two measurement traps, before again reaching the compressor. Helium for filling the apparatus is taken from a compressed helium cylinder. After initial purification it can be used at various different points : for charging the furnace, for charging the remainder of the apparatus and after heating, for desorption of the sorption traps.

The primary coolant line supplies first the generator and then a heat exchanger, both located outside the cell. The secondary water circuit is designed to cover all installations in the cell.

The electronic measurement apparatus and the high-frequency generator are located outside the cell. The oscillating circuit in the cell is excited via a high-frequency cable, the high frequency voltage being stepped down and passed through a duct into the box and into the furnace.

The liquid nitrogen is automatically distributed between four Dewar flasks.

The furnace temperature is continuously measured with an automatic pyrometer and registered by a recorder. A further high-precision reference pyrometer is installed outside the cell, to provide an automatic control of the measurements.

The inert gases released (Xe^{133} and possibly Kr^{85}) are accumulated in a measurement trap and measured by means of a NaI scintillation crystal.

Two single-channel discriminators are used for the measurement, one of which is adjusted on the desired line in the gamma-spectrum, while the other monitors the high-energy background in the measurement trap, in order to ensure that the measurement result is not falsified by the presence of further nuclides. These two measurements are also recorded on the same recorder as the temperature.

2. DESCRIPTION OF DESIGN AND STRUCTURE OF THE APPARATUS

The design and function of the individual

apparatus components are described in detail below.

Gamma-ray shielding is ensured in the BZ III laboratory by means of standard concrete, the working space in the interior of each concrete cell being surrounded by a gas-proof steel container. A special ventilation and air removal system operates via a filter, thus preventing the emergence of radioactive, in particular α-emitting dust.

2.1 Apparatus components in the main helium circuit

2.1.1 Furnace

The high-frequency oscillatory circuit is located outside this "alpha-ray-proof" box, but still inside the concrete cell. This is a parallel oscillatory circuit, which is excited by the generator outside the cell. It is tuned to a frequency of $\sim$ 300 kc/s. The maximum convertible HF energy is 35 kW and the HF voltage about 5 kV. The coil of this circuit has 20 turns and constitutes the primary winding of a high-frequency transformer. The secondary winding has a single turn and consists therefore of an almost completely closed shell. The transformer is cast in silicone rubber, the radiation resistance of which is sufficiently great to enable the oscillatory circuit to be operated in the cell for 1 to 2 years. The circuit capacitance consists of three ceramic capacitors. All individual components of the circuit are water-cooled and, in order to obtain at the non-earthed end the necessary electrical

interval to the earth potential of the coolant network, the circuit is fed with coolant water at that point through a 4 m hose. The latter is wound round a plastic insulator, so as to act at the same time as a high-frequency choke. The secondary shell of the transformer is attached directly, but in easily detachable fashion, to the HF duct, leading into the furnace. In order to penetrate the alpha-ray-proof box its doors must be opened and the whole oscillatory circuit housing is then disconnected and rolled aside. Hose and cable connections remain in position (Fig. 2).

The intensive gamma-radiation of the fuel element leads in the vicinity of its surface to an appreciable helium ionization, thus necessitating a number of compromise solutions in the power input. In the first place the original HF voltage is stepped down directly at the oscillatory circuit to 1/20 of its original value, so as to permit operation with a single-turn induction coil. The lead between the secondary shell and the induction coil must in this case, however, be of as low impedance as possible, i.e., must have as low as possible an ohm resistance and longitudinal inductance. If a determined minimum length has to be maintained for this lead, as is necessary in the present case due to the furnace geometry, two flat leads with high transverse capacitance are necessary, i.e., they must be a small distance apart and as wide as possible. This distance cannot, however, be less than 4-5 mm as a result of the low dielectric strength of the ionized helium, as was shown by preliminary experiments. The gap between the fuel element and the induction coil winding can be increased furthermore, by accepting

an inferior coupling but filling the gap with a solid insulator. The latter must, however, be highly radiation-resistant, thermally efficient - in contact with carbon - and highly resistant to temperature changes, since it extends over the range of most intensive gamma-radiation from the surface of the heated graphite susceptor to the wall of the water-cooled induction coil. With a coil height of 100 mm and a susceptor of equal length the heating will remain sufficiently constant over a susceptor length of 70 mm. Preliminary experiments in conjunction with the generator manufacturer, using this geometry and a mean workpiece temperature of 1000°C, gave deviations of ~ 20°C at the ends. The graphite susceptor radiated freely in this case at the ends of the coil, causing a rapid fall in the induced energy. These ends were therefore insulated on the final model of susceptor. Only boron nitride could be considered as an insulator for this purpose and for the gap between induction coil and susceptor. Although this ceramic material had a consistency and strength roughly equivalent to black-board chelk, it fulfilled all the above requirements perfectly. The following details are shown in Fig. 3; the cylindrical gap insulator is loosely suspended in the induction coil, a cover rests freely on it and a plate is held under the susceptor with a thin graphite nut. This ensures that the spherical fuel element is at least 20 mm distant from the induction coil and can nevertheless be uniformly heated to a maximum temperature of 2000°C, as was confirmed in preliminary experiments. The lead to the induction coil is as wide as the coil is high (i.e. 100 m) and about 500 mm long. It is also hollow, being used for the coolant water feed and thus itself being cooled at the same time. In order to ensure the required precision of the

coil bore and of the gap between the lead bands and at the same time to obtain an adequate compression strength, induction coil and lead were constructed of a single piece of solid electrolytic copper right up to the furnace duct. The coolant water pressure was 1.5-2 atm_g. The helium pressure in the furnace could be adjusted to between 1 and 5 atm_g, although the furnace could also be operated under vacuum. A mean copper wall thickness of 5 mm was required to take up this variable load. An inconsistency in the band shape of the lead is necessary at the gastight furnace duct. Attempts to obtain a hermetical seal by casting the conducting bands in epoxy resin were unsuccessful. Since the required seal could only be obtained with O-rings, the band cable was interrupted by 4 bolts, 2 for each conductor, which passed through a Super-pertinax cover. These copper bolts were sealed against the cover with Perbunan O-rings and through the bolts flowed coolant water (a hermetical seal better than 10^{-8} torr · l/s was obtained in this way over the whole duct). The bolts carried, outside the furnace area, the terminals for connection of the high-frequency transformer and the screw joints for the coolant water hoses. The insulating cover was 250 mm in diameter, since the dispersion of the magnetic field of the HF lead would have heated the metal parts in the neighborhood to such an extent that the losses would have been to great. The insulating plate is connected with a flanged ring by casting with epoxy resin. The latter is water-cooled and screwed into the furnace container with sealing by means of an O-ring. The induction coil together with insulator, lead and cover form a single unit, which can be replaced as such. Since these components become

severely contaminated from the furnace interior, replacement is effected by means of the plastic bag technique, which is normal in hot cell operation. The bag is secured by a crimpel flange surrounding the cover. The most important maintenance work on the furnace can be carried out in this way without opening the alpha-ray-proof box (Fig. 4).

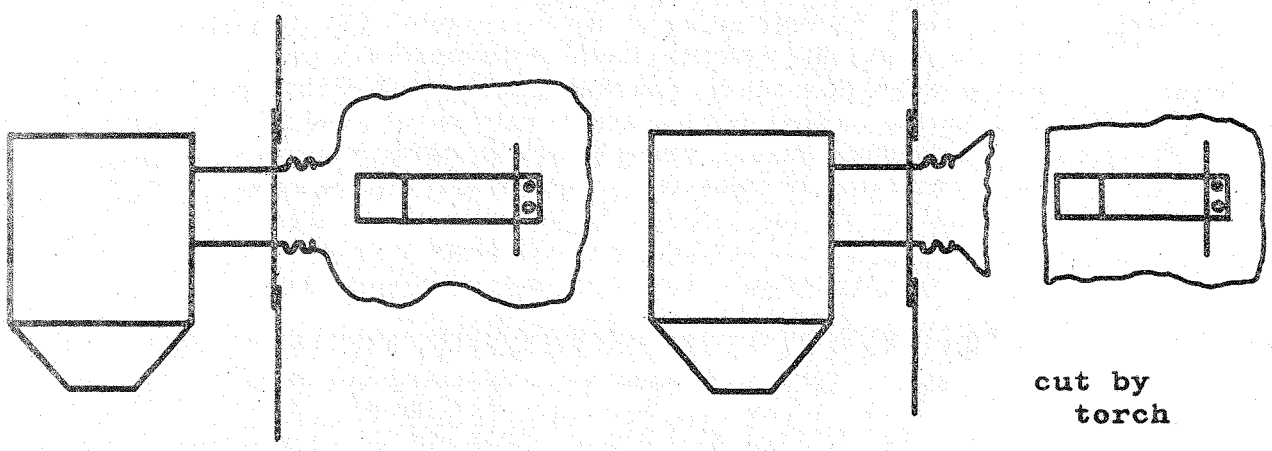

Fig. 4 Extraction of the induction coil

The helium-proof furnace container is constructed entirely of chrome-nickel steel (V2A), which has three properties absolutely essential for this purpose. It is paramagnetic, so that the electric losses due to the dispersion of the induction coil at a small distance from the furnace wall remain within acceptable limits. It is largely corrosion-resistant; the rust-free properties of the material are of particular importance for maintenance of the helium atmosphere, while the more general corrosion-resistance is a favorable

factor should any decontamination be required. It can be rolled to a pore-free state, is also capable of cold-working and can be relatively easily seal-welded, i.e., it is very suitable for the construction of a helium container of high hermetical properties.

Packings where necessary consist of Perbunan, which is relatively radiation-resistant but only tolerates a maximum working temperature of 80°C. All furnace components are water-cooled, in order to maintain a controlled temperature throughout. One branch of the coolant water circuit supplies the lower cover, while another flows through the upper cover, the casing of the furnace container and the casing of the pipe carrying the inspection window. This window consists of quartz glass and, in conjunction with two openings in the induction coil and the susceptor, permits direct observation of the heated object amd measurement of its temperature with the pyrometer. The axis of observation through the window of the hot cell is approximately at eye height, which permits also a favorable position for the reference pyrometer outside the cell. The quartz window is located 500 mm distant from the heated parts, in order to be certain that sparingly volatile substances do not strike against it, since even in very small quantities these would adversely affect the pyrometric measurement. The window is also screened by a plate diaphragm, which is folded aside electromagnetically only if so required, so that one can if necessary heat under vacuum. The window is slightly heated from the outside by a hot air stream, in order to avoid the formation of condensed water which in a humid atmosphere would be precipitated over the whole furnace. The window can

be replaced by remote control by means of the manipulator.

The purified sweep gas reaches the furnace at the upper cover (III). Since it can only leave the furnace via the lower cover (III), it flows through the pyrometer opening into the susceptor and further through its hollow stem into the pipe system at the lower cover. In this way it sweeps the released fission products out with it. The susceptor stem serves, therefore, not only as a mechanical carrier, but also as a means of conduction and a first condensation sector for sparingly volatile fission products, since it covers from the full annealing temperature down to the coolant water temperature ($\sim 30°C$) in the lower cover, at which point it fits tightly into a cone. The basic features of the furnace design are therefore directed at obtaining a complete separation of thermal insulation and hermetical sealing, all detachable sealing elements being arranged so as to permit cooling and separation from the hot points.

The lower cover of the furnace is adjustably secured to a bracket, which can be moved upwards and downwards by a pneumatically operated cylinder. In the upper position the susceptor is located in the induction coil and the cover with its O-ring is pressed flush against the furnace (this situation is shown in Fig. 3). In the lower position the upper end of the susceptor is about 150 mm below the lower furnace opening (Fig. 5). This position is used for charging or for complete replacement of the susceptor, if it is desired to determine the sparingly volatile fission

product content in its interior. After the charged sweep gas has passed the susceptor stem, it flows through a 4 mm diameter pipe into the iodine traps. This pipe section is also easily replaceable, to permit its examination for condensed fission products.

2.1.2 Iodine traps

After detailed preliminary experiments the following design was adopted for the iodine traps : the sweep gas enters from below (III, Fig. 3) and passes first of all through a sintered bronze filter ("Poral") and a copper scrap packing. The corresponding zone of the brass pipe is cooled by fixing the pipe at this point in a very accurately dimensioned water-cooled copper block (II, Fig. 3). The extremely fine (2 µ filter gap) Poral filter is firmly pressed in so as to make a good thermal contact with the brass pipe. This ensures that the sweep gas, which cannot flow in laminar fashion either through the filter or through the copper packing, is cooled at latest at this point to coolant water temperature, i.e., 25-30°C. All fission products, capable of being condensed on the metal surface, are retained. The sweep gas then passes through a further Poral filter (as above) into an activated charcoal packing, which is closed with a graphite felt plug and a third Poral filter. The graphite felt prevents the activated charcoal dust from the sweep gas being pressed on to and into the Poral filter and blocking it. The remaining iodine is retained in the activated charcoal packing. However, since the xenon would also be delayed here at room temperature for about 20 min, the corresponding part

of the brass tube, and thus the Poral filter and the charcoal packing contained in it, are heated to 100°C. The xenon is now delayed only approximately 1 minute and the retention capacity for iodine is virtually no worse, as was demonstrated in corresponding experiments in hot cells at Würenlingen. The packing was heated by fixing the brass pipe in an electrically heated copper block, again of exact dimensions (10, Fig. 3). The final element in the circuit is an absolute filter (Fiberfax) inserted in the gas flow, which is held between filter papers under the pressure of a spring. The last arrangement was found to be necessary in order to prevent even very small quantities of charcoal and graphite dust, charged with active substances, from entering the subsequent pipe system and measurement traps. Six of the iodine traps described above were arranged in succession, in order to be able to use them separately in experiments or in temperature stages during an experiment. The gas flow could be controlled as desired by a valve behind each trap. The traps could be replaced by means of the cell manipulator and were connected to the pipe system by the simple plugging in of a double O-ring packing into an easily accessible socket. They were held in position by the clamps of the cooled or heated block. The traps themselves could after replacement be removed from the cell and emptied by releasing the screw mechanisms on both sides and pressing out the filter packings with a suitable device. They were packed so as to fit the sample changer of the gamma-spectrometer. The carrier tube was decontaminated and refilled.

The whole furnace described above with all its component valves and pipes forms

from the point of view of apparatus design a single unit. It stands with its adjustable base frame on the bottom of the alpha-box, i.e., at about table height. With the oscillatory circuit disconnected and the box gate opened the furnace is freely accessible in the gate opening, but in such a way that the box itself is still accessible. All important parts are arranged in relation to the cell window in such a way that they are visible and can if necessary be reached by the manipulators (Fig. 5).

A diaphragm pressure manometer, showing the helium pressure in the furnace over a measurement range from -1 to +4 atm_g, is attached to the upper furnace cover. A Penning measurement tube was also attached at the same point, permitting pressure measurements down to 10^{-4} torr. To the side, on the left looking from the cell window are located pneumatically controlled valves for the helium circuit. They are designed so as to be operable with the manipulator in case of failure of the pneumatic system. These valves (nominal width 10) were specially developed for this apparatus.

The pipe from the furnace body to the vacuum pump on the cell roof is blocked off by a ball valve of nominal width 40, controlled by an electric motor, which is also pressure-tight. A vacuum filter prevents radioactive dust getting into the pump.

The sweep helium with the residual fission

gases xenon and krypton, after leaving the furnace unit, pass out of the cell into the measurement traps, which are reached about 1 minute after passage of the heated sample. The relevant pipe system and all further pipes, through which helium passes, have a nominal width 10 and are constructed of chrome-nickel steel (V2A).

2.1.3 Measurement traps

The design of the measurement traps is based entirely on the need for a good counting yield, i.e., on the sensitivity of the instrument as a whole. The xenon (and with it the krypton also) is condensed on activated charcoal at the temperature of liquid air. The gap between the activated charcoal packing and the measurement crystal must be kept as small as possible in order to retain a favorable counting geometry, and the material between charcoal and crystal must be of minimum density owing to the soft radiation of the Xe^{133} (81 keV). The charcoal layer between the point at which the xenon is adsorbed and the measurement crystal may not be too thick and the counting geometry should not be too greatly affected by the location of xenon condensation on the activated charcoal.

All the above conditions, cryotechnical considerations and production factors gave rise to the design described below (see also Fig. 7). The gas mixture reaches the trap through a 300 mm thin-walled (0.2 mm) chrome-nickel steel pipe (the outlet pipe being of identical construction).

The conduction of heat from outside the pipe system to the cooled components is kept as small as possible by this means. Inside the trap a Poral filter (similar to those in the iodine traps) is pressed into the pipe in front of the carbon layer, ensuring full cooling of the gas and diffuse distribution over the adsorbent layer. It provides at the same time the upper geometric demarcation of the activated charcoal. A few layers of ultrafine filter fleece prevent the dusty fractions of the charcoal penetrating the Poral filter (although the charcoal is carefully sieved into fractions and rendered dust-free prior to charging into the trap, small quantities of charcoal dust are produced during operation as a result of the temperature alternation). Adsorption of xenon-133 on activated charcoal particles inside the sintered bronze of the Poral filter would falsify the measurement result, since a proportion of the soft radiation of the xenon would be absorbed. The carbon layer is 3 mm thick in the direction of gas flow and has a diameter of 20 mm. This corresponds to a piled volume of approximately 1 cm^3, the particle size being about 0.2 mm. In consequence the sweep gas flow in the charcoal packing is not laminar at the given throughput. With an active surface of 1400 m^2/mol (Desorex F), the whole filling is capable of adsorbing a multiple of the expected quantity of xenon and krypton (the trap only becomes charged with small quantities of impurities due to the high degree of purity obtaining in the circuit). The lower boundary of the charcoal layer is a fine (0.05 x 0.05 mm mesh) V2A-gauze, through which the helium leaves the layer and flows radially out through the external housing of the trap. An ultrafine fleece paper filter in front of the gauze again ensures that no carbon dust escapes. The external

housing of the measurement trap is closed at the lower end with a welded cover, containing a finely machined window 0.25 mm thick and 35 mm in diameter. The whole trap, which is suspended in a metal Dewar flask of 3 l capacity, is concentric with the flask at the lower end and at a 3 mm distance from the base of the flask. The base of the double-walled brass flask contains also two finely machined (0.3 mm) points at a distance of 10 mm apart as windows for the gamma-radiation. The hermetical sealing of the Dewar flask is adversely affected by the presence of these windows and the marked alternation in the thermal stress to such an extent that a minimum of 10^{-4} torr must be continuously pumped out of it, in order to maintain an adequate vacuum. If the vacuum were not maintained, an ice layer would in course of time form on the vessel and also on the windows, which would both increase the absorption of the gamma-radiation for measurement and at the same time destroy by excessive cooling the measurement crystal, attached in the immediate vicinity (3 mm). As a further precaution dried compressed air can be continuously blown between the crystal and the external vessel window, to form a "heat cushion". The detector (referred to up to now as the "measurement crystal") is a complete "integral-line" detector, consisting of a sodium iodine crystal $1\frac{1}{4}$ inch in diameter and 1 inch thick, permanently connected with a photomultiplier; the whole unit can be plugged in to a preamplifier. The complete detector is clamped in a plastic mounting, which is again coaxially connected with the Dewar flask in easily detachable fashion, by screwing into a plastic intermediate piece, secured to the Dewar flask. This means that in conjunction with the centering part of the replaceable xenon trap a properly reproducible geometry is obtained for the whole unit.

A slit is provided in the plastic intermediate piece between Dewar flask and detector to permit the insertion of an also geometrically designed preparation, in order to check the functioning of the electronic measurement system before and during each experiment. Preliminary experiments have shown that the exact point on the activated charcoal layer, at which the xenon is adsorbed, is unimportant in the case of the present overall geometry. This has been demonstrated by measurements with suitable flat- and point-shaped preparations, which were positioned on the axis and at the boundaries of possible adsorption zones.

The arrangement described above and shown in the figure is full duplicated, so as to have a fresh trap available after a burst. The two traps with the Dewar flasks, etc., are surrounded with a 5 cm lead shielding, in order to maintain the background at a constant low level ($\sim$0.2 pulses/sec). The common front wall of the lead shielding of the two traps can be pushed aside on rollers for insertion of the test preparation referred to above or for maintenance of the electronics. Desorption of the measurement traps is in general carried out with hot helium, the liquid air being removed from the Dewar flask for this purpose (with an ejectro pump) and helium passed via a heater through the trap into the waste gas pipes (see flow sheet, Fig. 1; flow path through valves 14, 48 or 13, 49). The trap is in this case heated to $\sim$ 90-100°C and the temperature maintained for some 10 min. The adsorbed inert gases undergo desorption without residue, as can be observed directly by means of the measurement system. In order to ensure that the activated charcoal has retained its full sorption capacity, after a few

experiments and before starting a new series after a long period out of action the easily exchangeable traps also undergo desorption at 250°C under vacuum, thus ensuring removal of even very firmly bound water and hydrocarbon residues.

2.1.4 Compressor, helium purification, etc.

The sweep gas passes directly from the measurement traps to the compressor which drives the helium circuit. The components which impede flow in the helium circuit - such as the helium purification system, measurement traps and in particular the absolute filters after the iodine traps - cause a considerable pressure drop, amounting with a throughput of 100 l He/h to about two atmospheres. At this throughput the compressor requires 1 atm_g inlet pressure, to ensure that 3 atm_g pressure obtains in the furnace. A small membrane compressor was found satisfactory for this purpose and for the purity requirements of the apparatus. The membrane consisted of chrome-nickel steel plate 250 mm in diameter. The deflection of $\pm$ 1.5 mm is so small that a high service life is obtained. The membrane is operated by an oil hydraulic system and is insulated against the surrounding atmosphere with a triple O-ring packing, so that the compressor leak rate is less than 10^{-8} torr · l/s. The compressor was specially produced by the manufacturer to meet the required conditions.

The next stage after the compressor is helium purification. A zeolite trap

is used first of all, for adsorption of coarser impurities - in particular water and hydrocarbons. Adsorption is carried out at room temperature and desorption at 200°C, the latter by passing hot purified helium through the trap into the waste gas pipe with constant monitoring of the temperature in the trap. (The design of the zeolite bed corresponds in all details, with the exception of the precooler, to the activated charcoal bed, which will be described in greater detail below.)

Further purification is obtained in an activated charcoal trap, in which adsorption occurs at the temperature of liquid nitrogen and desorption at ~100°C. The design may be seen in Fig. 8. The helium flows initially through a coolant coil in liquid nitrogen, in order to ensure that it is fully cooled before reaching the activated charcoal bed. This avoids the possibility that only parts of the activated charcoal should have reached the required temperature owing to the poor thermal conduction of such a loose filling (this applies in principle also to the desorption process). As in the case of the measurement trap, a sintered bronze filter (Poral) is pressed into place under the filling for cooling and diffusion of the gas stream. The activated charcoal bed consists of Desorex F 12 with a particle size 0.5-1 mm. After leaving the activated charcoal, the helium passes through a further sintered bronze filter which retains the dust. All pipes, extending into the liquid nitrogen, have a finely machined sector in order to maintain the thermal conduction at a low level. The Dewar flask is constructed of chrome-nickel steel and welded, so as to withstand higher temperatures (up to 250°C). It need therefore only be

emptied during desorption of the trap and does not require removal. The helium leaves the trap above the large screw joint (metal packing) through which the filter filling is introduced, and flows along a choice of two paths through a water-permeated heat exchanger. The purpose of this during normal operation (direct passage) is to protect the packings of the following valve (pneumatic ball-cock) from excessive cooling due to the cold helium and the thermal conduction of the pipe system. The heat exchanger warms the helium in this case by means of the "coolant water" (18-25°C). During cleaning of the trap, on the other hand, i.e., during desorption with hot helium, it acts as a cooler, preventing excessive heating of the electromagnetic valve packings (the movement path being via the coiled pipe in the cooler into the waste pipe). For purification of the trap the helium is heated in a heater (Fig. 8 upper left), consisting essentially of a narrow gap between an external pipe and the casing of the heat conductor. The whole arrangement is bent into a U-shape in view of the variation in the thermal expansion of the two pipes. The temperature at the end of the packing is measured with a resistance thermometer.

The purified helium flows out of the purification traps into a buffer vessel, which it again leaves via a needle valve, - used for adjusting the throughput. The throughput is again measured before reaching the furnace, immediately after the needle valve by means of a rotameter - for an immediate reading after adjustment of the throughput - and then electronically with a debimeter, in order to be able to record the sweep gas flow. All components of the sweep gas circuit outside

the cell are - like the furnace - combined in a single unit from the point of view of apparatus construction. They are mounted on a frame, enclosed in a gas-proof (beta-technique) shoe box. This is intended to facilitate maintenance work, even when the interior of the apparatus was contaminated, as can occur with overloading or incorrect operation of the iodine traps. The box is connected to the cell waste air system and is thus continuously maintained at a slight vacuum.

2.2 Ancillary devices

Various ancillary devices are required in addition for operation of the main helium circuit (see also Fig. 1).

2.2.1 Helium supply

The required quantity of helium is taken from a gas cylinder (purity : 99.995 %). In order to obtain the mean required 30 vpm impurities and thus as complete as possible an absence of oxygen, it is passed initially through two purification traps of similar design to those in the circuit (these traps themselves undergo desorption by the passage through them of a highly pure, heated helium from a special cylinder). After purification the helium is used for charging the plant and for purification of the other traps.

2.2.2 Vacuum

Air penetrates the furnace during charging.

To prevent this air being swept into the purification traps, the furnace is evacuated, refilled with purified helium and again evacuated, prior to connection in the main helium circuit. A two-stage rotary slide valve pump is used for this purpose and a large heatable zeolite trap is incorporated between the pump and the furnace to prevent access of all the pump vapors (water and hydrocarbons) to the furnace. A vacuum of 10^{-2} - 10^{-3} torr is thus obtained in the furnace. The pump and the zeolite trap are located on the cell roof and the waste gas from the pump is passed back into the cell.

2.2.3 Compressed air

The compressed air required for different purposes is taken from an existing network, sufficient compressed air being stored in a storage tank to permit at least one experiment to be concluded in case of network failure. The air for the heat cushion in the measurement trap is dried and that required for the pneumatic servosystems is filtered and oiled. The electromagnetic control valves for all pneumatic cylinders in the cell are located outside the cell on the cell roof. Waste air from these is also passed back into the cell, to prevent the emergence of any contamination via the piston rods into the pistons and thus into the pipe system.

2.2.4 Liquid nitrogen

The liquid nitrogen for a four Dewar flasks is taken from a 100 l storage container. It is pumped under slight

excess pressure (0.6 atm_g) via a pipe network (steel pipes with Armaflex foam insulation) and four pneumatic cryovalves to the site of use. The liquid level in the Dewar flasks is maintained automatically.

2.2.5 Coolant water

Virtually all the energy converted into heat in the apparatus, approximately 100 kW at full furnace power, is removed by water cooling. Two water cooling systems are used therefor. The generator and one heat exchanger are cooled from the water mains. The heat exchanger then serves as a cooler for a closed secondary circuit, serving all furnace components, the iodine traps and the small heat exchangers in the purification traps. The whole secondary circuit is constructed completely of stainless steel, in order to prevent corrosion damage. The water is circulated by a centrifugal pump. The individual branches of the circuit are separated on disconnection of the system at a point still outside the cell by electromagnetic valves in the inlet and return pipes. In the case of pipe rupture, i.e., with a rapid pressure loss, this disconnection occurs automatically, thus ensuring that only small quantities of liquid can escape into the cell. This is necessary, since the relevant cell series is not designed for direct contact with liquids.

2.3 Electrical equipment

2.3.1 Generator

The high-frequency current for the furnace

is generated by a single-stage thermionic generator. The (already described) oscillatory circuit is located directly adjacent to the furnace in the cell, while the generator, exciting the circuit, is mounted outside the cell. Oscillatory circuit and generator are therefore connected by a 10 m high-frequency cable. The positive bias potential is produced in the generator by a built-in high-frequency transformer with a variable coupling. In consequence the inductance of the oscillating circuit is not distributed between the generator and the oscillatory circuit unit and the high-frequency cable is therefore loaded with minimum power and does not require cooling. The generator power is regulated by its d.c. voltage supply, which is generated with a mercury vapor thyratron in three-phase full-wave rectifiers. The d.c. voltage is cut in with the thyratron and regulated by phase intersection control, thus stabilizing the d.c. voltage at the same time. The transmitting tube in the generator has direct water cooling, while all other components are cooled indirectly by means of an air cooler. The generator produces 35 kW high-frequency power at 300 kc/s.

2.3.2 Measurement electronics

The electronic system for determination of the activaty, accumulated in the measurement traps, is designed as follows : each of the two measurement traps is fitted with a detector, consisting of a scintillation crystal (NaI), photomultiplier with high-voltage current supply and preamplifier. If desired - depending on which measurement trap is used - the signal from the particular detector is connected to the main amplifier (see circuit diagram below).

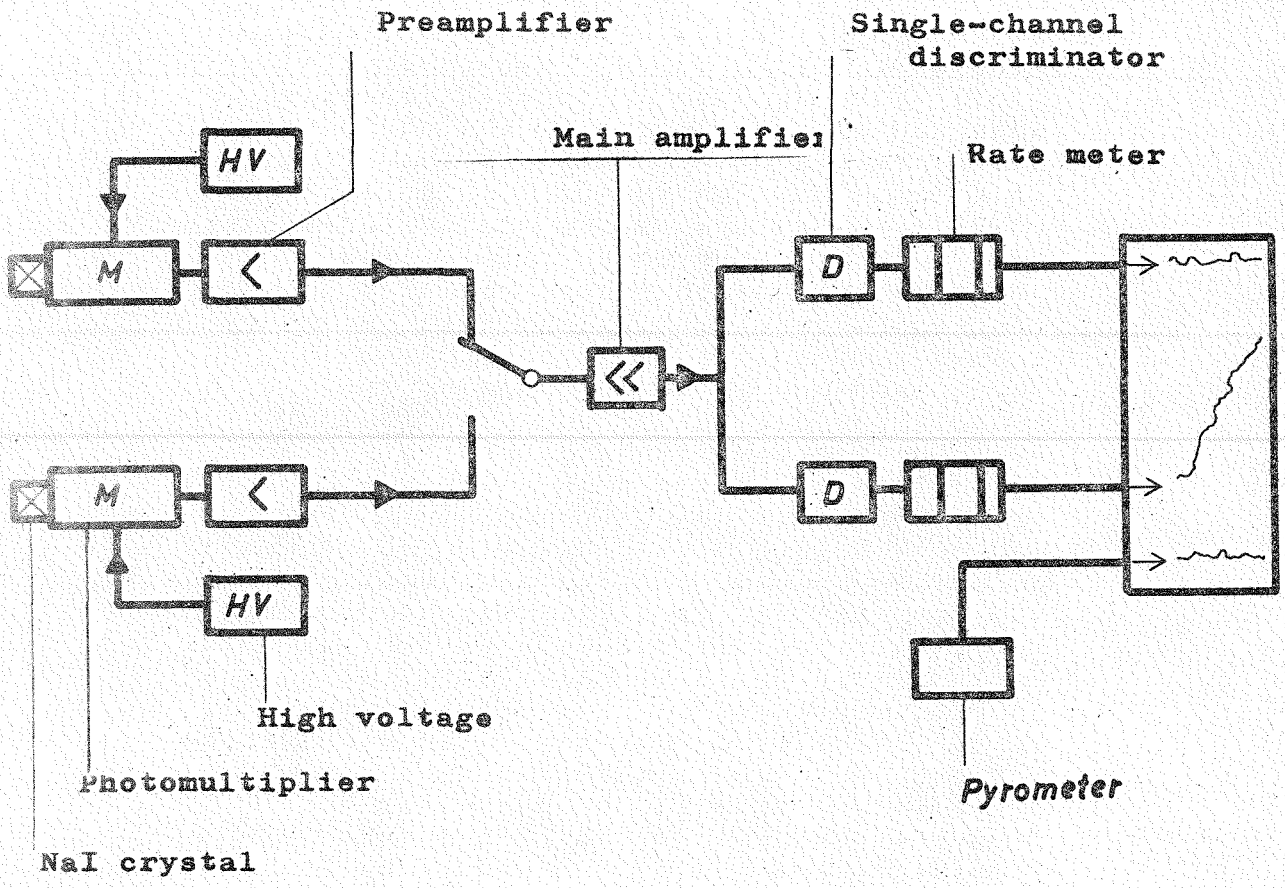

Fig. 10 Measurement electronics, diagram

This main amplifier is a linear, low-drift amplifier, adaptable to the impedance of the signal line, producing a 16-fold amplification. It divides the signal between two parallel single-channel discriminators, having windows which can be adjusted according to the position (energy) and in symmetry with this according to the width of the signal. These are followed by linear rate meters, the readings of which are recorded together with the temperature by a potentiometer recorder. The general practice is for one branch to monitor the intensity of the desired

line (xenon-133 or krypton-85), while the other monitors a wide range of the high-energy background (e.g. that containing the iodine-131 energy).

Serious difficulties occur if the sensitive measurement electronic system is operated in the immediate neighborhood of the running generator (35 kW at 300 kc/s). All control lines must be blocked at the generator and the control device with lead-in capacitors. All voltage supply units for the electronic system were provided with low-pass filters and the generator had an extremely low-inductance earthing system.

2.3.3 Control and operational monitoring

In order to permit rapid and routine operation of the apparatus, all operating states were controlled and monitored from a central control panel. The interlocking of individual states in this control panel was sufficient to prevent incorrect operation which might have serious consequences. The control panel was designed so as to show on its front side a circuit diagram of the essential elements of the apparatus, important functions being indicated by pilot lights and illuminated arrows (Fig. 11). It is thus possible to see at a glance the initiation of individual operating states, which are preselected by means of press buttons. A diagram of the interlocking system is shown in Fig. 12. The main switch disconnects the whole plant from the network (3 x 380 V three-phase current, 100 kW) and is only opened for purposes of repair. A key-operated switch on the control panel connects the electronic system and provides control voltages

for the power relays. All further operating states can only be obtained when the primary coolant water and secondary coolant circuits have been cut in.

The pressure in the primary system, which built up against a Pitot tube at the outlet, was monitored and amounted to at least 2.5 atm_g. This pressure monitoring ensures the correctness both of the through-flow and thus the coolant circuit and also the pressure gradient to the secondary circuit, which should be a maximum of 2 atm_g internal pressure. The pressure in the secondary circuit was maintained between this upper limit and a lower limit of about 1.5 atm_g and automatically monitored. Pressures in excess (penetration of primary water, steam formation or excessively high filling pressure) and also below these limits (pipe rupture or insufficient filling) causes immediate disconnection of the secondary circuit; all subsequent operating states in the sense of the interlocking plan are then disconnected and the secondary circuit itself is divided into several independent sections (see Fig. 1 and in text 2.2.5). The pressure gradient between the primary and secondary coolant water circuit ensures against the penetration of contamination into the primary coolant water pipe and thus into the outlet channel. These precautions obviate the need for monitoring or processing large quantities of water. When both coolant systems are running, it is possible after releasing the interlock by means of a key-operated button either to connect individual elements of the plant at will, which is a requirement for test and maintenance purposes, or to arrange mutual interlocking of the five routine operating states such as

charging, evacuating, etc., and their connection in delayed sequence.

The desired operating state is selected by the appropriately marked press button, the previous state being automatically disconnected. After a delay of approximately 15 sec (during which period the button must be held in the depressed position) to ensure synchronized closing of all valves and to avoid an uncontrolled connection, the desired state is engaged. The position "Heating" can only be engaged if an iodine trap in the helium circuit has been cut in. A measurement trap (xenon trap) can also be selected in this position and the high-frequency supply to the furnace can be cut in, provided that the generator has been preheated for 6 min (operating state III). The high-frequency supply can also be connected in the position "Evacuation" for short-term heating under vacuum. Failure of even one phase from the network supply causes disconnection of the whole apparatus, which will not cut in again automatically.

All valves are closed in the current-less state. The thermal capacity of the coolant circuits is in all cases adequate to prevent damage, should the coolant water supply be disconnected at the same time as the power supply. A number of operating data such as the helium throughput, the temperatures of the iodine traps and coolant water temperatures at several points were recorded by an operational data recorder during each experiment, so that the operator had available continuous information on the conditions at all technically critical points.

3. MEASUREMENTS

3.1 Systematic sequence of a measurement

The gamma-radiation measuring device was quantitatively calibrated for the nuclide requiring to be detected. Since xenon-133 is not available in quantities sufficient for calibration, thulium-170 was used, which at 84.5 keV has a peak in the immediate neighborhood of xenon-133 at 81 keV. A weighed quantity of the calibrated solution was evaporated on to the window for one measurement trap. The window was inserted, like a complete measurement trap, in a centered position on the base of the Dewar flask. The gamma-radiation now passes through all three windows (in one case a measurement trap, in two cases a Dewar flask and falls on the sodium iodide crystal. The calibration is based on the ratio of the calculated (actual) and measured activities. It compensates not only for the window absorption and geometry but also for the characteristics of the detector. This calibration is combined with a calibration using another preparation, which can be inserted in the slit between detector and Dewar flask, the geometry being in both cases reproducible. The calibration can thus be checked during the measurements also.

The apparatus is in general held ready calibrated and with the electronic system in operation. In the "Charge" position the furnace is opened and the spherical fuel element or a crucible with coated particles positioned on the lower part of the susceptor, which is then closed. After closing of the furnace the system is switched to "Evacuation" and the Dewar flasks for helium purification are filled

with liquid nitrogen. After 10 min a further helium sweep is carried out (filling, evacuation) and after a second, longer "Evacuation", the system is switched to "Charging". The furnace is now slowly (30 min) charged with helium which has been adequately purified by the slow passage through the inlet purification system. During this period the main helium circuit is already running on helium purification with furnace bypass. In the "Heating" position the whole helium content in the plant is pumped at a higher throughflow rate past the measurement traps (bypass) via the helium purification system. Circulation of the complete charge lasts about 10 min and purification is continued for several hours. The Dewar flasks of the measurement traps are then filled with liquid nitrogen and the gas stream passed through the measurement traps at 50-100 l/h, so that the zero effect is recorded in the measurement traps for 1 h. The high-frequency supply is then cut in and heating begins. The required temperature is reached after a few minutes (approximately 5 min from room temperature to 1250°C).

Fig. 13 shows a diagramatic representation of the curve recorded. The activity in the trap rose steeply almost immediately after the start of heating and then rapidly flattened out. This has led to the phenomenon being known as a burst. It lasts for a few minutes, less frequently for up to 1 h. The temperature set was maintained until the release rate reaches equilibrium, i.e., a linear rise in the curve. The latter was maintained sufficiently long for the mean slope of this activity increase to be accurately determined and the next temperature required in the particle specification was then set and the procedure

repeated. Fig. 14 shows two curves from actual tests. Heating lasted up to 20 h at two to three temperatures and on conclusion of the measurement the susceptor together with the sample was cooled for approximately 20 min. The helium was then released from the furnace - "Emptying" - and the furnace opened in the "Charging" position.

The sample is removed and the susceptor packed with minimum contamination in a hermetically sealed container, using a clean insert in the claw of the cell manipulator. If sparingly volatile nuclides are to be determined, the pipe connection from furnace to iodine traps and the iodine traps themselves are replaced and hermetically packed in the same way. All these components are transported to another cell for gamma-spectrometric investigation or removed from the cell.

The measurement trap undergoes desorption, while the apparatus is receiving a new charge.

3.2 Evaluation

The following procedure was used for evaluation of the measurements. The release of fission gases was defined as the ratio

$$\frac{R}{B} = \frac{\text{Release rate in the reactor}}{\text{Formation rate in the reactor}}$$

R is the variation in the number of atoms of a nuclide present in the cladding

as a function of time, this variation being due to a proportion of the nuclides leaving the cladding. Formation and decay rates are regarded in this case as being in equilibrium

$$R = \frac{dN(t)}{dt}$$

in which R is the release rate,

N the number of nuclide atoms

and t the time.

The formula in the case of a radioactive nuclide is

$$a = N \lambda$$

$$R = \frac{1}{\lambda} \frac{da}{dt}$$

in which α is the activity in decay incidents/sec and

λ the decay constant of the nuclide.

The activity is the parameter, which is accessible to measurement; since the released atoms accumulate in the measurement trap, by recording the counting rate I as a function of time the measurement yields the direct quantity

$$\frac{dI}{dt} \quad \text{with} \quad I = \alpha \times f$$

in which I is the counting rate in pulses/sec

and f is the calibration factor of the measurement system and the gamma-ray yield of the nuclide.

With a constant release rate, therefore, such as is reached after a short experimental period, the following formula applies

$$R = \frac{\Delta I}{\Delta t} \cdot \frac{1}{\lambda f} \cdot$$

In order to use the result of the heating test to predict the release rate of fission gases during reactor operation, the number of atoms of the nuclide in question is related to the time of reactor shutdown. If a time t_1 has elapsed prior to execution of the annealing test, the following formula applies

$$R = \frac{\Delta I}{\Delta t} \cdot \frac{e^{\lambda t_1}}{\lambda f} \cdot$$

where B is the number of atoms of the nuclide in question, produced during irradiation in a unit of time. It is obtained from the number of uranium-235 atoms present at a particular point in time.

$$B = N_{U\text{-}235} \cdot \sigma_f \cdot \phi_{th} \cdot \varepsilon$$

in which B is the production rate,

$N_{U}235$ is the number of uranium-235 atoms,

σ_f is the fission cross-section of the nuclide,

ϕ_{th} is the thermal flux

and ε is the fission yield of the nuclide.

With a uranium-235 batch of determined weight we obtain

$$B = \frac{N_L}{A_{U-235}} g_{U-235} \cdot \sigma_f \cdot \phi_{th} \cdot \varepsilon$$

in which NL is the Loschmidt constant,

A is the atomic weight

and g the weight in grams.

The weight of the U^{235} batch is corrected from the burnup obtained at the time of reactor shutdown.

The value R/B is meaningful, if at the time relevant to the degassing test, i.e., at the time of reactor shutdown, an equilibrium obtains for the nuclide in question between its formation rate B and its decay rate. The particle concentration within the cladding is then constant as a function of time and the release rate R is a function only of the temperature and the quality of the coating.

The result of the degassing test is normally given in the form of the value of R/B. To permit easier comparison of the release phenomena occurring with different samples, we determine for striking points on the recorded curve at corresponding times t the value

$$F(t) = \frac{N_{F(t)}}{N_{ges}}$$

in which

$$N_{F(t)} = \frac{I}{f \cdot \lambda}$$

and

$$N_{ges} = B \frac{e^{-\lambda t_1}}{\lambda}$$

$N_{F(t)}$ is therefore the number of atoms

of the nuclide in question, released during the experiment at the time t of the measurement and N_{ges} is the total number of atoms of the nuclide present at a time t_1 after reactor shutdown (normally a few weeks) which is assumed to be constant for the measurement period (a few hours).

3.2.1 Fission gases

The evaluation of the measurement will be described on the example of xenon-133, since this fission gas is normally used for measurement in the heating test. The required formation/decay equilibrium of its precursor I^{133} (half-life : 21 h) is reached after 3-4 days. The equilibrium for Xe^{133} itself (half-life: 5.2 days) is reached after 20-25 days irradiation.

Xenon is therefore well suited for the degassing test in view of the irradiation periods of reactors, normally of the order of months.

The calculations required in order to obtain R/B and the F(t) values are carried out on a special calculation form, in which the following symbols have a slightly modified significance :

R_I = the recorded pulses per second

f_E = the calibration factor.

Fig. 14 shows the original curves from two tests. The relevant calculating forms and the corresponding F(t) curves are shown on the following pages.

3.2.2 Sparingly volatile fission products

After leaving the heated zone of the graphite susceptor, the sweep gas becomes cooled. The lower furnace cover, in which the susceptor stem is inserted, is water-cooled. The subsequent connecting pipe to the iodine traps is at room temperature and the first filter zone in the iodine traps is again water-wooled. Passing through the above zones with the sweep gas (in the same sequence), the sparingly volatile fission products are condensed (a common term, namely "solid fission products", is not used here, since neither iodine nor cesium are solids at room temperature). All elements, apart from the inert gases xenon and krypton and the sweep gas helium, are trapped at latest in the iodine trap. In order to determine the total quantity of a sparingly volatile nuclide released, we need only determine the overall quantity, irrespective of where it is condensed. It is therefore necessary to examine the susceptor stem, the subsequent pipe system and the iodine trap packings. This is done by means of a gamma-spectrometer. The stem is stripped down, the interior of the pipe system is subjected to chemical decontamination (by surface etching of the metal or by an ion exchange process) and the solution obtained is also analyzed. In emptying the iodine traps the contents of the cooled and heated zones are kept separate. A number of samples are thus obtained with suitable geometries for the gamma-spectrometer, which are then packed individually in plastic bags for insertion by the sample changer. The sum of their activities yields - taking into account the calibration of the spectrometer - the total activity. A modern high-resolution spectrometer with a semiconductor detector permits the simultaneous determination of all the nuclides with which we are concerned

(fission products, e.g. cesium, and activation products, e.g. silver). Fig. 20 shows by way of example the spectra of two fillings of an iodine trap. The disadvantage of this measurement procedure is that only the total quantity of a nuclide, released during an experiment, can be determined. An attempt is therefore being made at the present time to show that the ratio of the quantities condensing in the iodine trap to those condensing at any other fixed point during an experiment under constant conditions (gas flow, temperature) will also remain constant. It is reasonable to expect this to be so, since the quantities condensed are so small that no overloading with condensation products is to be expected at any particular point. If experimental confirmation of this hypothesis is obtained, it will be possible to draw conclusions for the total quantity released from the amount collected in the iodine traps. Since six iodine traps are incorporated in the apparatus and can be used in succession during a single experiment, it is at least possible to give a maximum of six values for R/B at a single heating temperature.

Experiments have also been started to determine the local distribution of condensation along the sweep gas stream. The susceptor stem is of particular interest in this connection, since it extends from the susceptor itself, which is maintained at the degassing temperature, to the lower water-cooled furnace cover. The temperature drop with the present stem geometry (a long, thin hollow cylinder) is roughly exponential. Susceptor stems from several experiments have been examined by position-dependent gamma-spectrometry (gamma-scan), the stem being in this case displaced along its main axis by

a slit collimator. Fig. 19 shows the spectra obtained from one such measurement, while Fig. 18 shows the position-dependence of three lines. The stem is shown at the bottom of the figure in diagrammatic form, but to scale and in relation to the graph; the hot end and the sweep gas flow inlet is on the left-hand side.

In further experiments, which are now in preparation, the temperature gradient will be varied by suitable modification of the stem shape, the aim being to obtain a linear temperature drop. Further related possibilities of obtaining information on nuclide condensation will be described on the final pages under the heading "Future prospects".

4. FUTURE PROSPECTS

The routine execution of the heating test on coated particles and spherical fuel elements required the development and installation of an apparatus which had to satisfy strict conditions. The technical design had to ensure a high availability, simplicity of operation and a high reliability during operation. The measurement system required a particularly high sensitivity and the ability to detect sparingly volatile fission products. No such apparatus had been produced to date. A large measure of automation has made it possible to meet the technical operating requirements. More than 50 degassing tests (corresponding to some 1500 operating hours) have been carried out up to now (1969). The sensitivity is superior by a factor of 5-10 to that of other comparable equipments and this

high sensitivity enables us to investigate samples even a relatively long period after reactor shutdown. Either the remaining Xe^{133} residue (5.2 days) is sufficient for this purpose or it is possible - as was demonstrated in the most recent test - to measure Kr^{85} instead of Xe^{133}. The latter has a half-life of 10.6 years and accordingly a very low activity, but no formation/decay equilibrium is reached during irradiation, so that no results were obtained, which were fully comparable with the conventional Xe^{133} test. Nevertheless it was possible to recognize spherical fuel elements with damaged particles.

It is possible as a result of the short heating-up time to record as a function of time the course of the fission gas burst, occurring after heating-up and on the adjustment of each subsequent temperature stage. Analysis of this burst is of particular interest at the present time, since it can forseeably be used as a technique, permitting a sufficiently rapid division of the fuel elements, leaving a pebble-bed reactor, into the categories of damaged and re-usable elements.

The further development of coated particles to withstand higher fuel temperatures will always be a matter of general interest, in particular for the development of high-temperature reactors with a gas turbine in the primary circuit. The release of fission gases, which leave the reactor through the sum total of all the leaks present and are emitted,

obviously remains an important problem. However, the release of sparingly volatile fission products, which not only remain in the primary circuit but are condensed at specific points, is of very particular significance. It was pointed out in the last chapter that the total release of these products can be determined. Experiments are in course of preparation for measuring the deposition of these products as a function of temperature and sweep gas flow. In these experiments the temperature gradient along the susceptor stem will be given in advance and the flow rate of the sweep gas inside the stem will be varied by the insertion of graphite rods of different thicknesses in the bore of the stem, which cannot for reasons of production technology be reduced below a determined size (3-4 mm).

It has in general been found that the apparatus described above, in addition to being sufficiently flexible for the prescribed routine tasks, is capable of contributing to the solution of new problems, arising during the thermal treatment of highly active samples.

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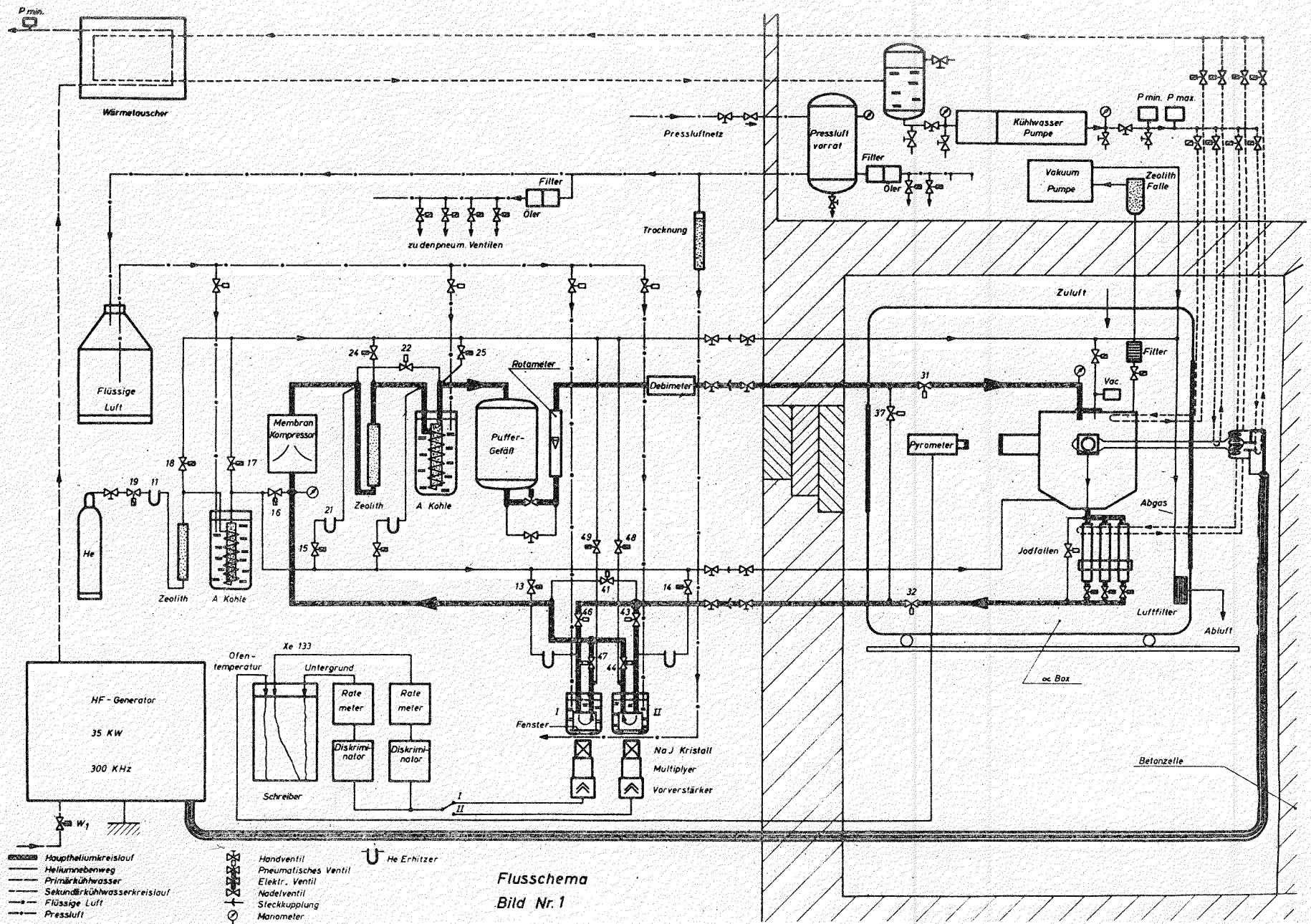

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Bild Nr. 1

52

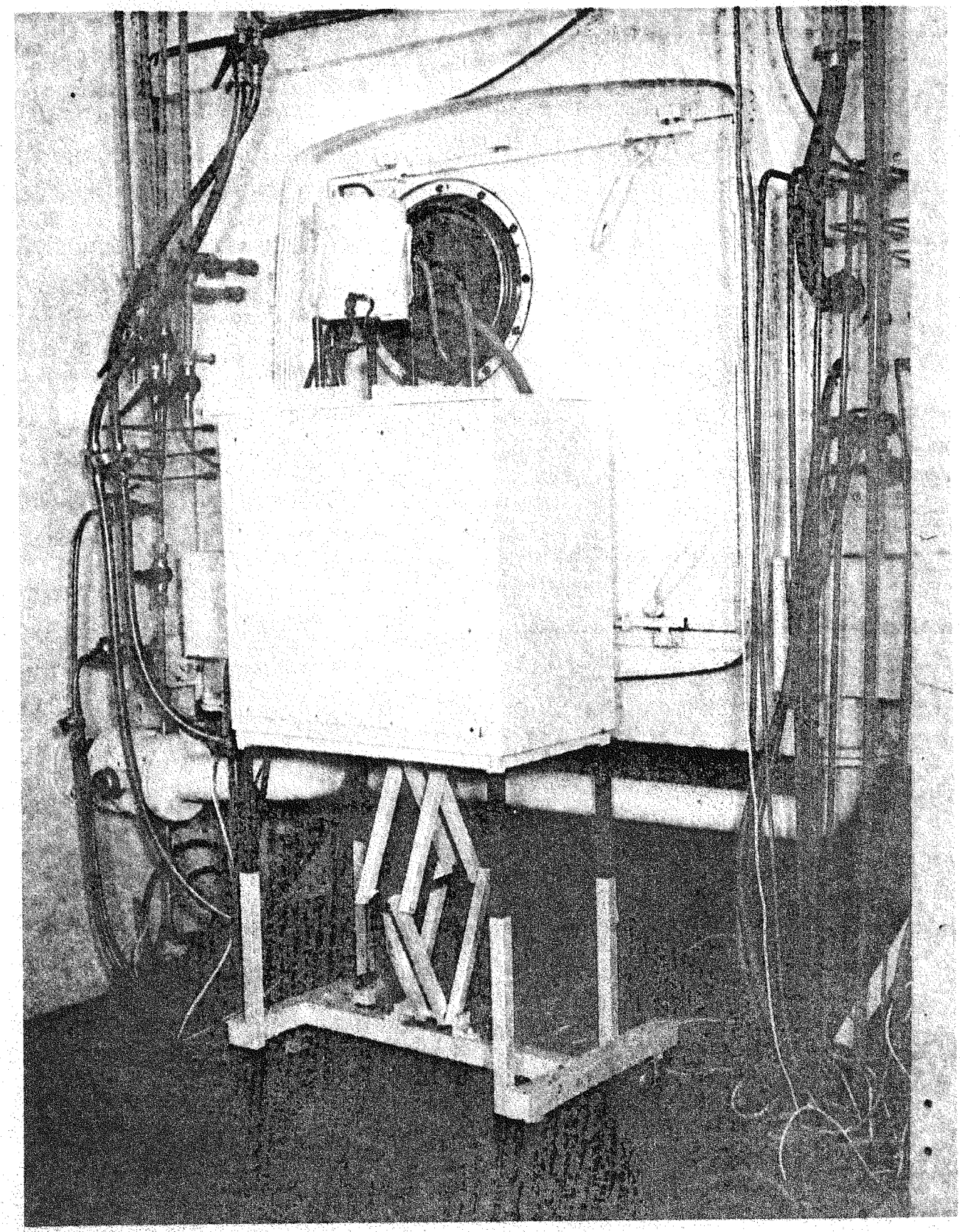
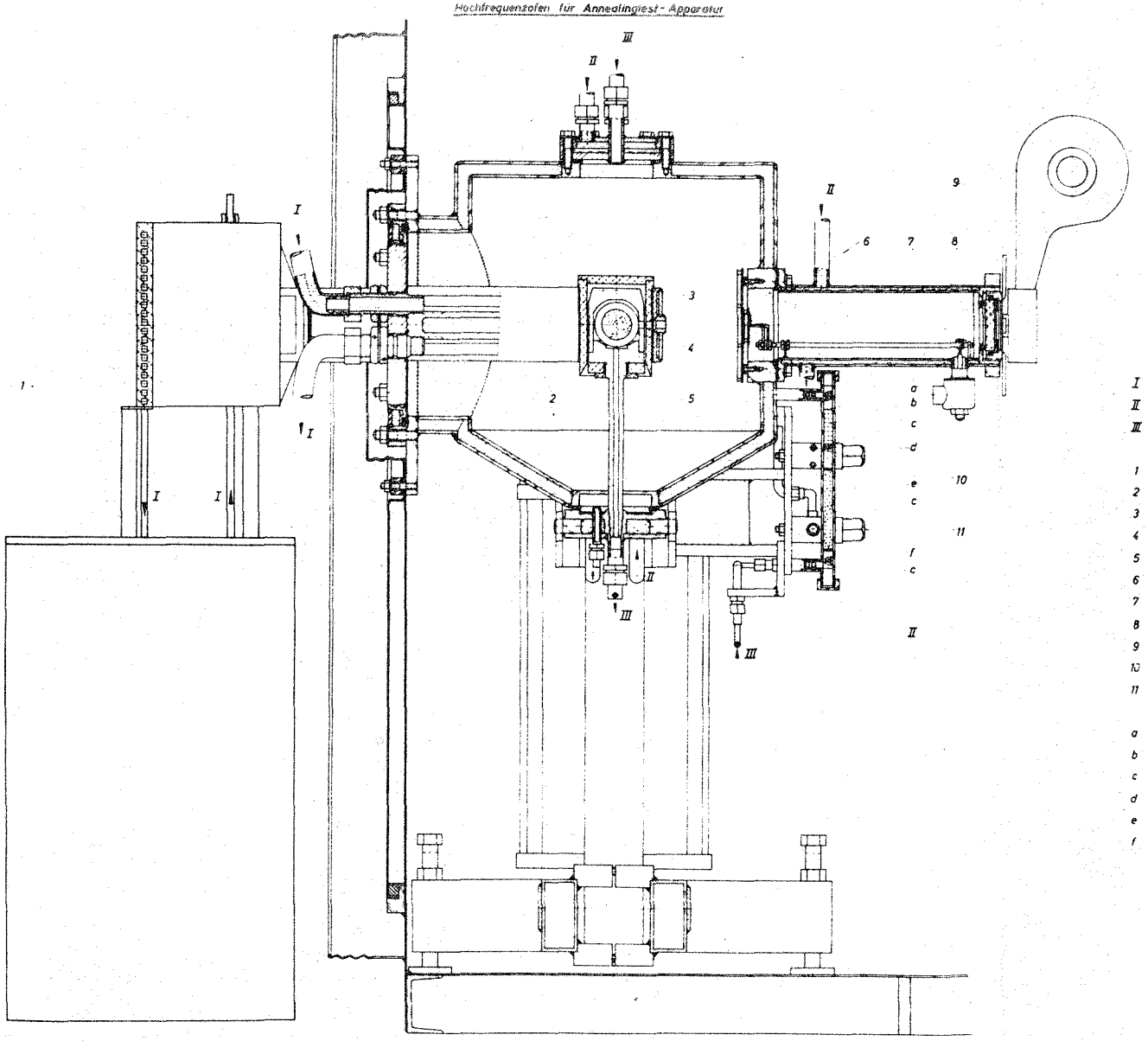


Bild Nr 2 Alpha-Box + Schwingkreis

Hochfrequenzofen für Annealingtest - Apparatur



I Kühlwasserkreislauf Induktor und Schwingkreis
II Kühlwasserkreislauf Ofen
III He - Kreislauf

1 Schwingkreis
2 Induktor
3 Bornitrit - Tiegel
4 Brennelement
5 Grafit - Tiegel
6 Wolfram - Hitzeschilde
7 Elektromagnet
8 Quarzglas - Fenster
9 Warmluftgebläse
10 Heizstäbe
11 Jodfalle

a Filterpapier
b Absolutfilter
c Poral - Filter
d Grafitfilz
e Aktiv - Kohle
f Cu - Schrot

Bild Nr 3 Schnitt - Ofen

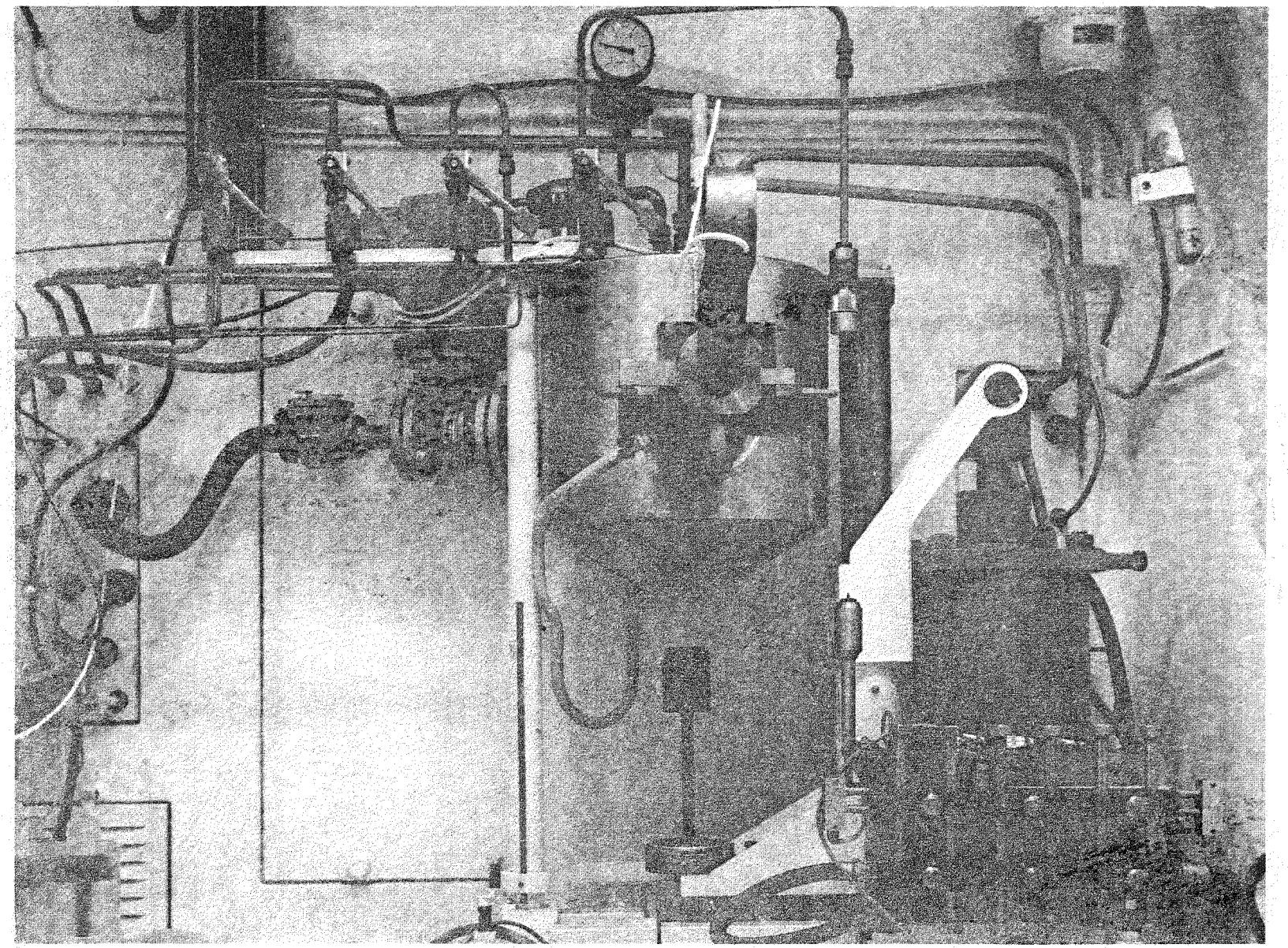
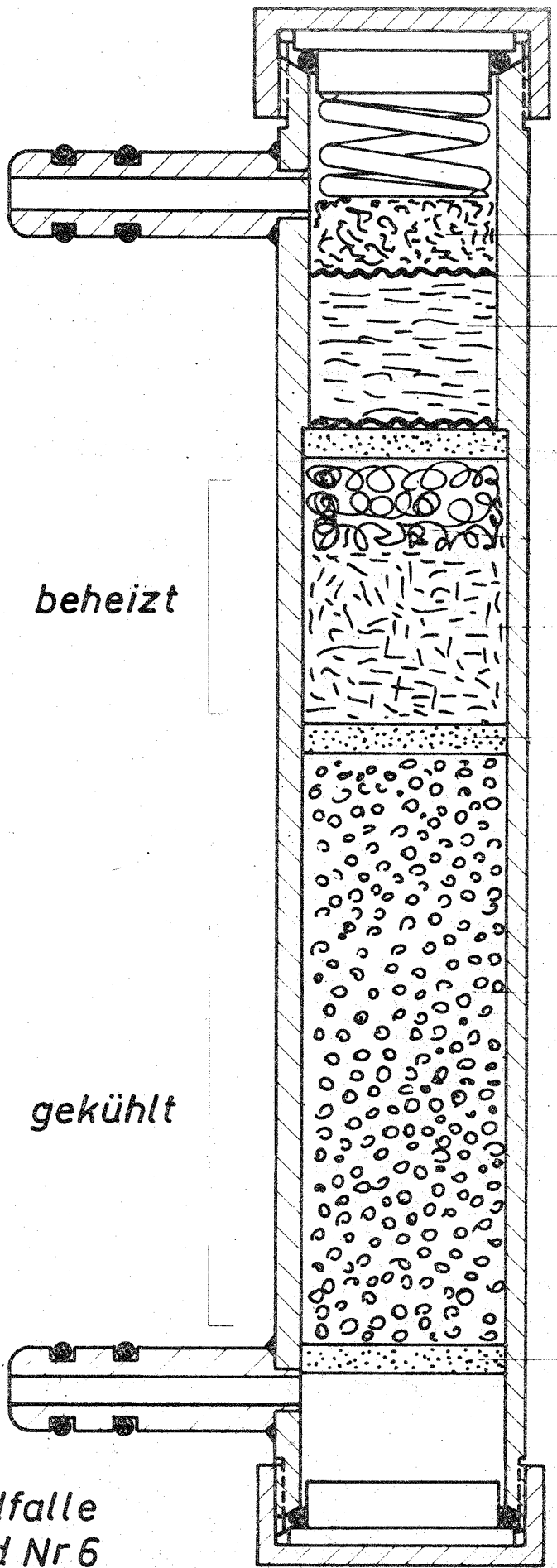
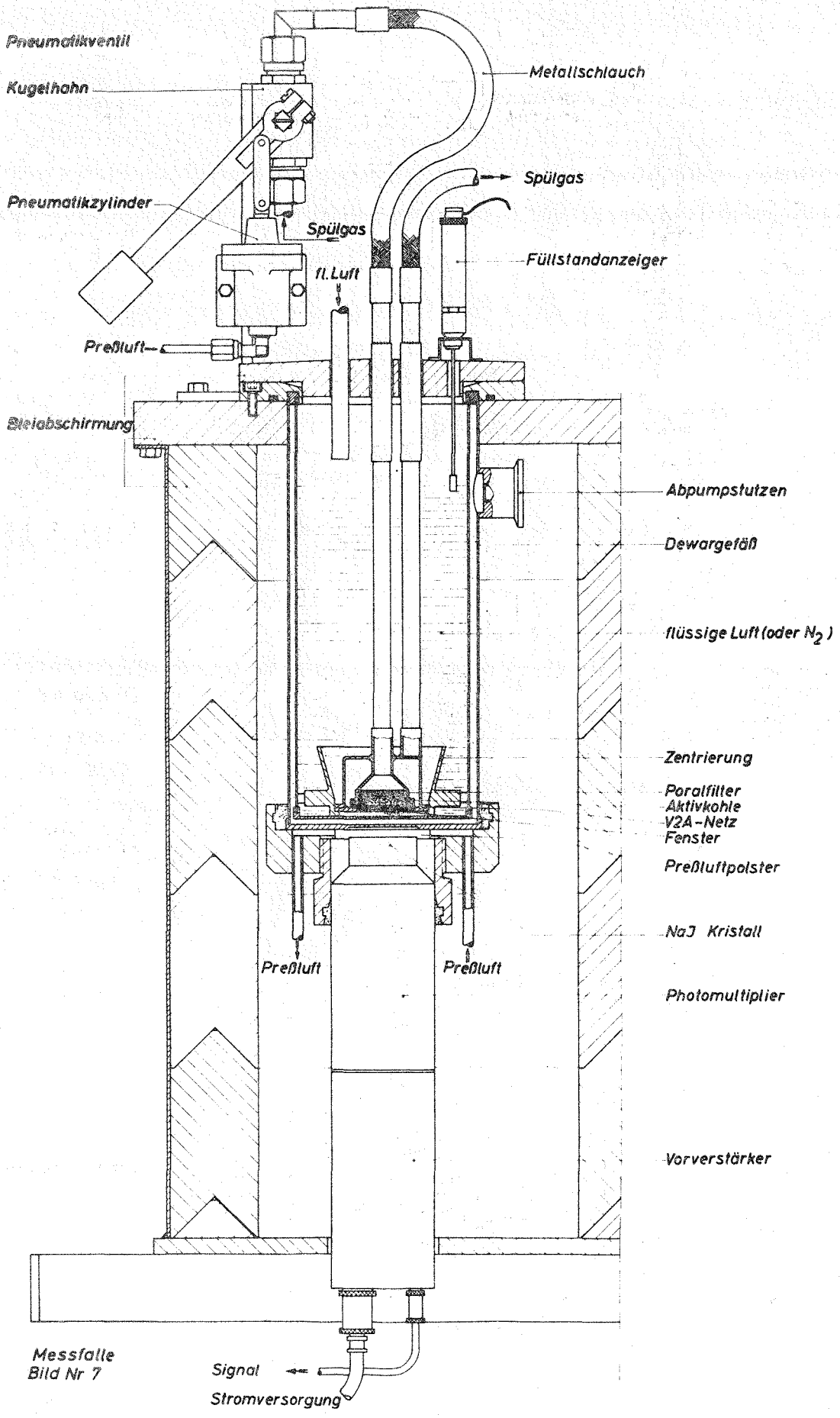


Bild Nr 5 Ofenansicht durch das Zellenfenster

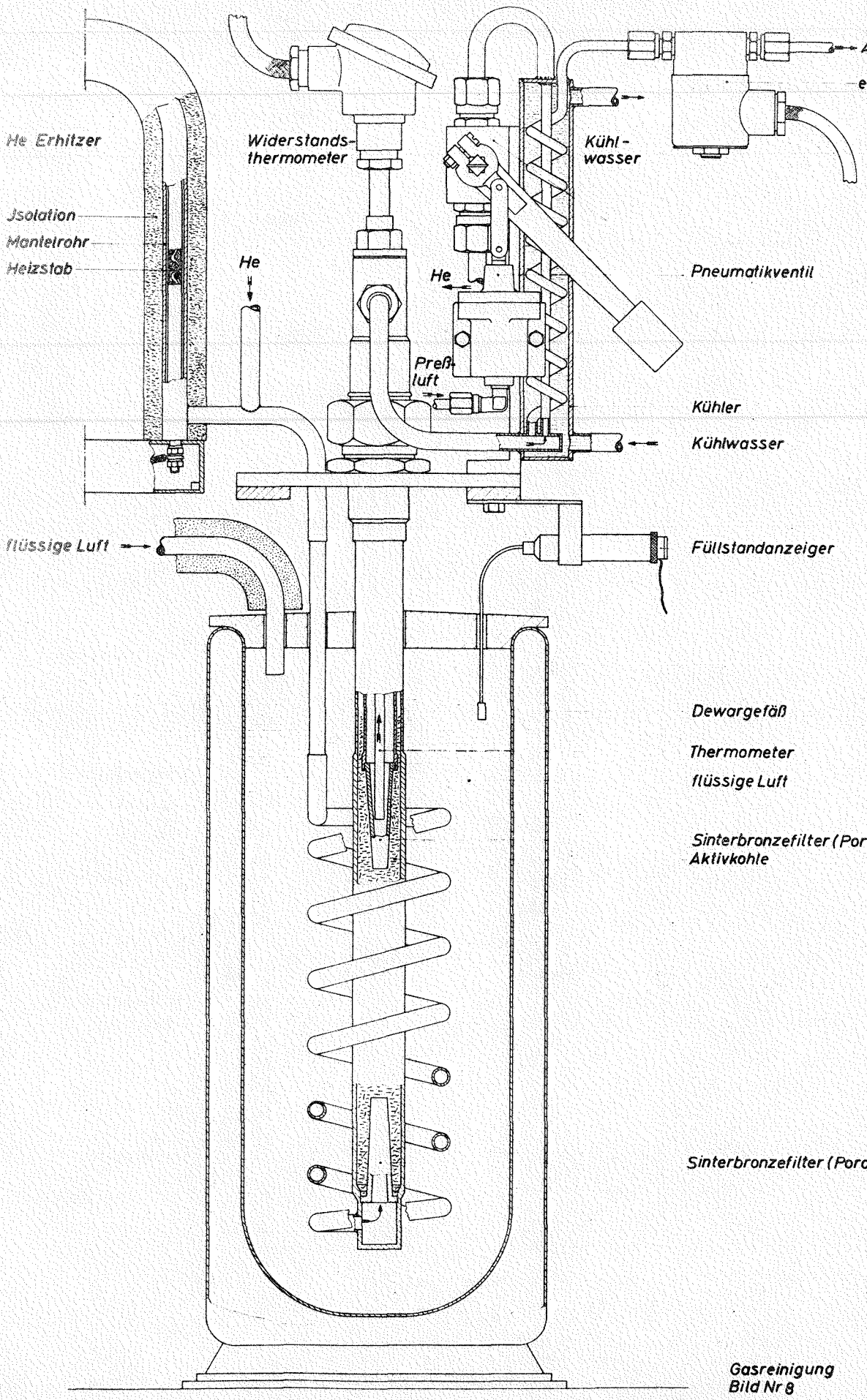


Jodfalle
Bild Nr 6

nicht maßstäblich
innsbesondere Länge und
Durchmesser



Messfalle
Bild Nr 7



Gasreinigung
Bild Nr 8

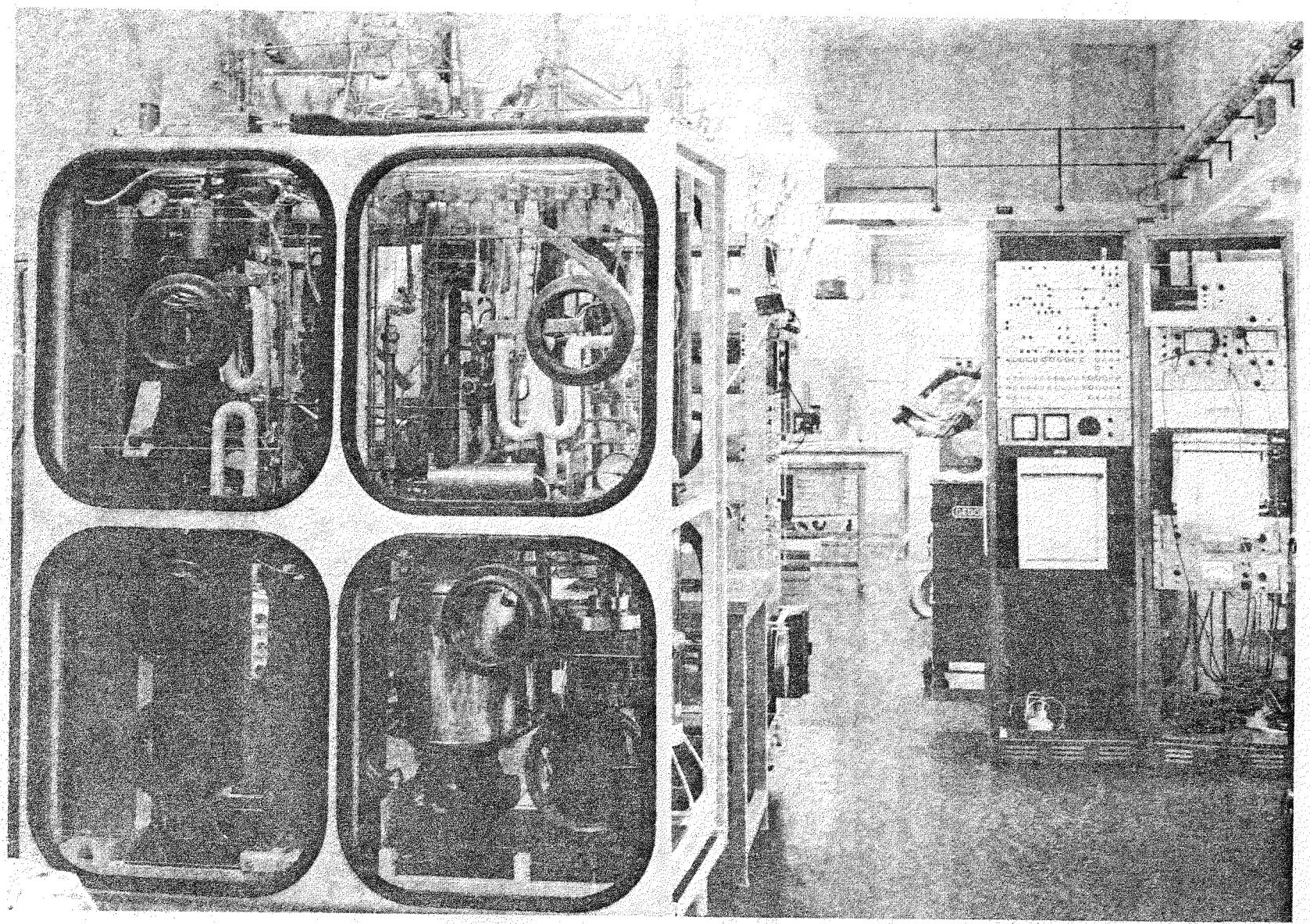


Bild Nr 9 Handschuhbox mit Meßfallen, He-Reinigung und Kompressor

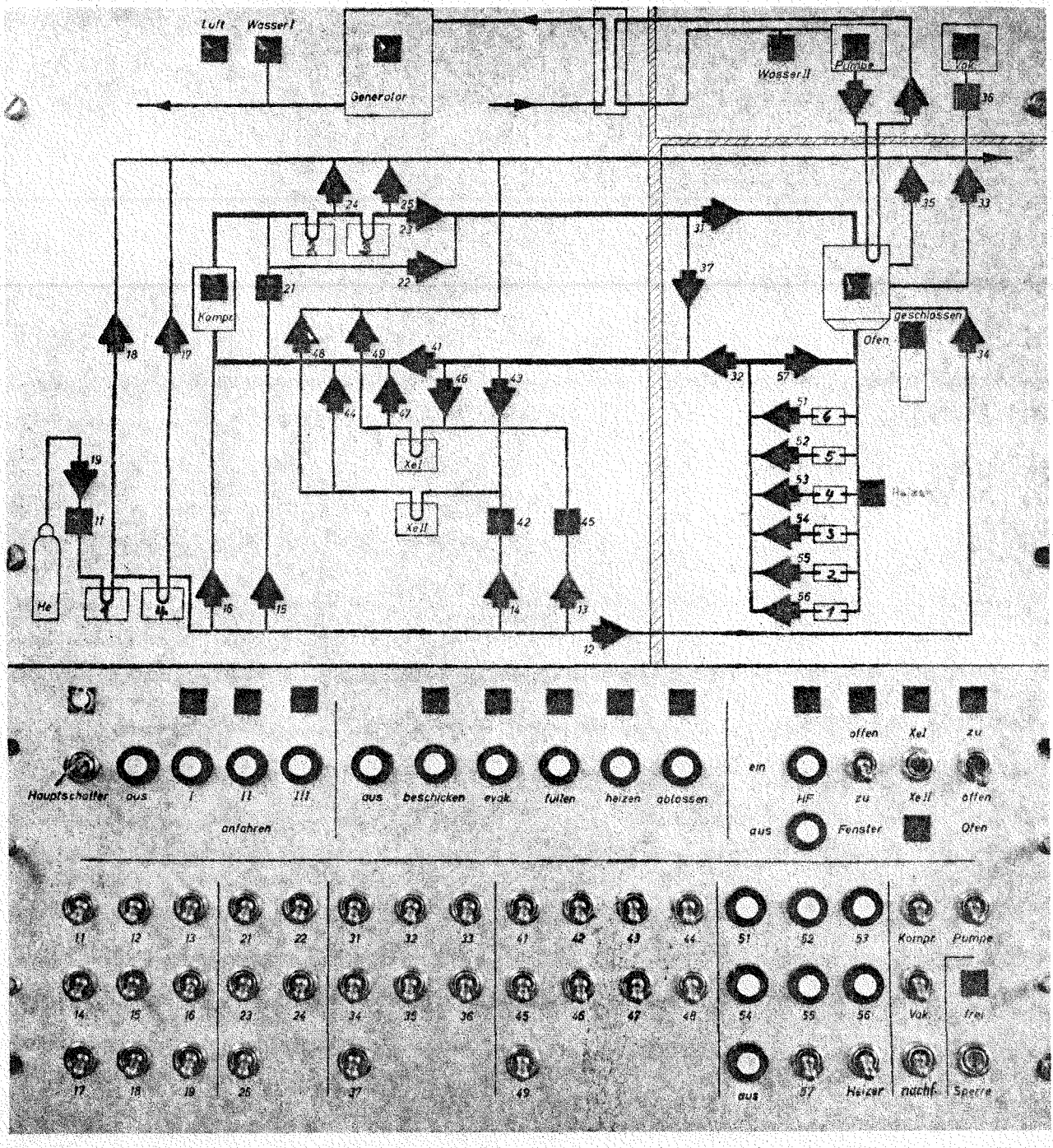


Bild Nr 11 Steuerteil

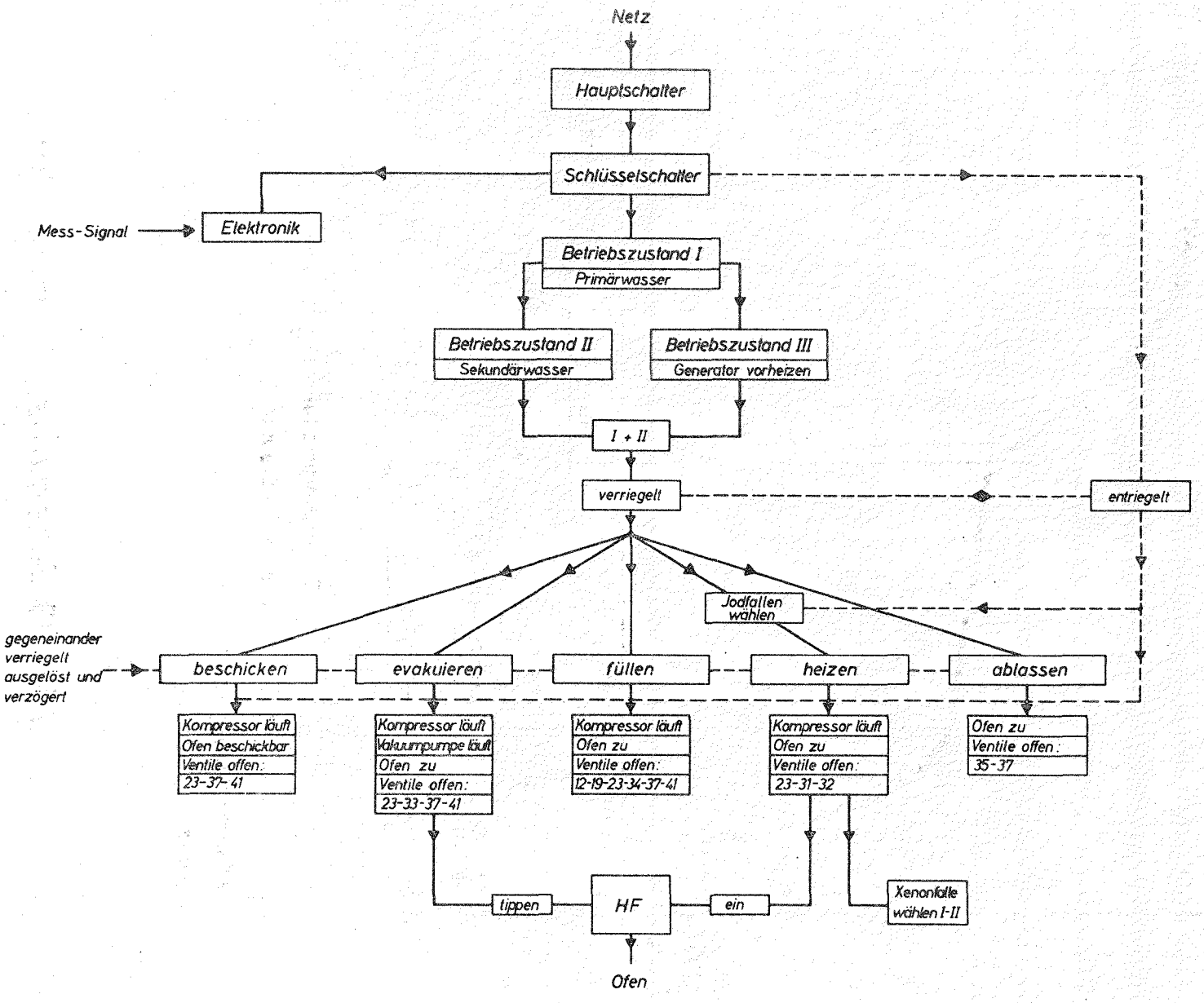


Bild Nr 12 Verriegelungsschema

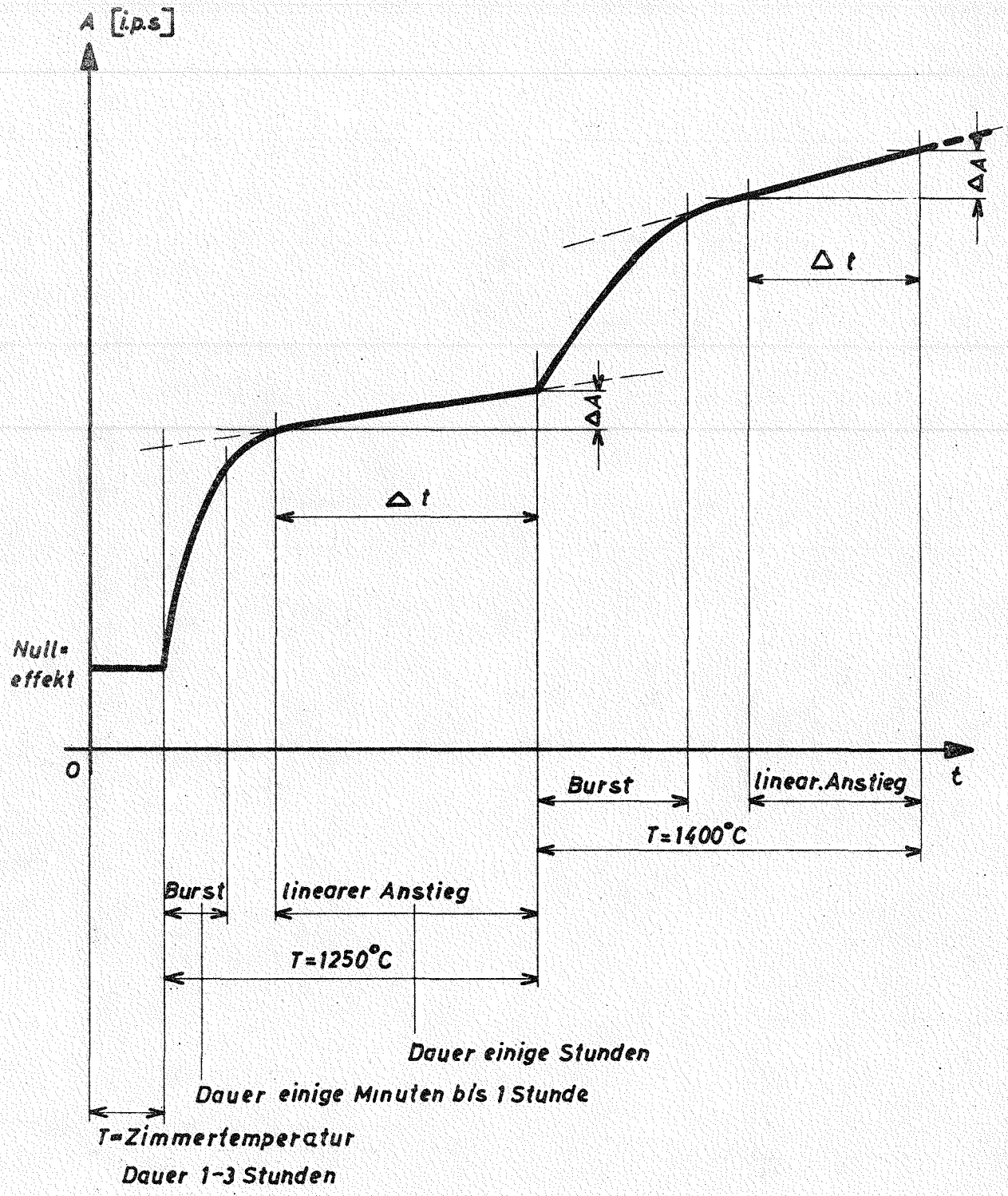
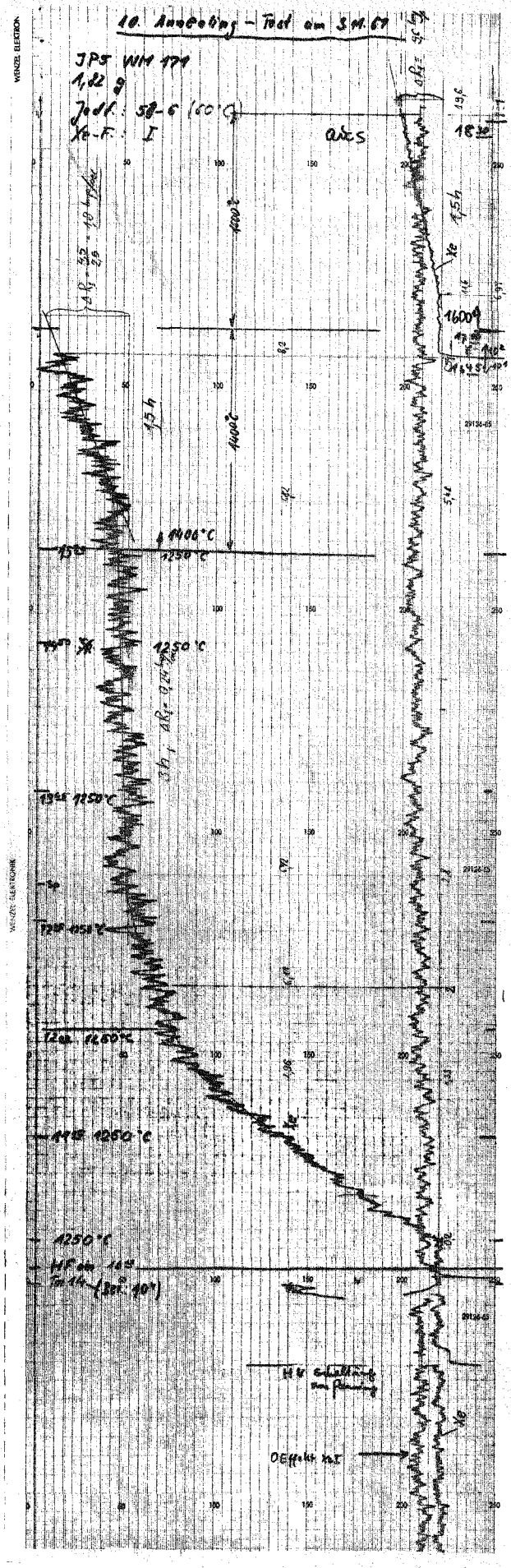
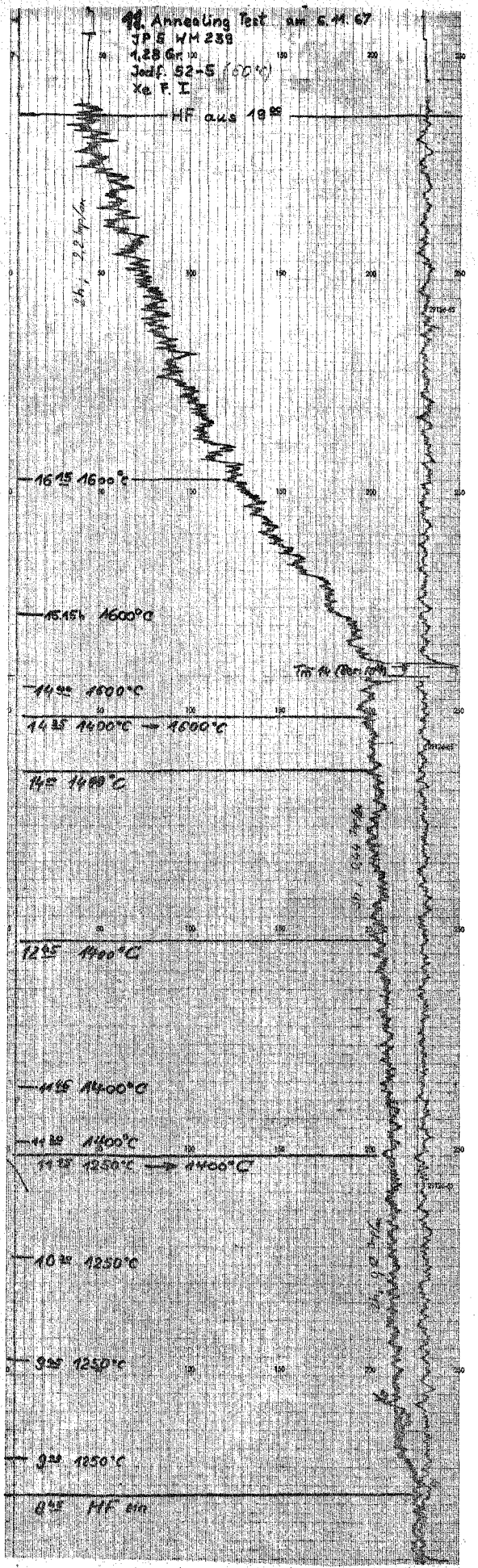


Bild Nr 13 Schematische Darstellung der Spaltgasfreisetzung



10 A-Test, JPS - WM 171 am 3.11.67



11 A-Test, JPS - WM 239 am 6.11.67

Bild: 14

Annealing - Test ..10... am 3.11.67

Experiment: JP5... Kapsel: ..48.. Part. Sorte: WM171
Gew.%U-235: .....[%]

Bestrahlung vom 26.1.67 bis 24.9.67..
therm. Fluß - ϕ_{th} 1,1·10^{14} [n·cm^{-2}·sec^{-1}]
Abbrand: .49...[%fifa] ≙ .....[%fima]

Bestrahlungstemp.: 1390..[°C]
schnelle Dosis: .....[n/t]
Einfang querschnitt - σ_f 613·10^{-24} [cm^2]

Part. Einwaage: 1,82...[g]
Abklingzeit - t_1: ..40....[d]

$\lambda_{Xe\text{-}133} \cdot t_1 = 0{,}131\ [d^{-1}] \cdot 40$
$e^{\lambda_{Xe\text{-}133} \cdot t_1} = 188$

$$g_{U\text{-}235} = g_{Einwaage} \frac{\text{Gew.\% U-235}}{100} \left(1 - \frac{\text{\% fifa}}{100}\right)$$

$$g_{U\text{-}235} = 1{,}82 \cdot 7{,}5 \cdot 10^{-2} \cdot 0{,}51 = 0{,}696 \cdot 10^{-1}\ [g]$$

$$B = 1{,}68 \cdot 10^{20} \cdot \sigma\,[cm^2] \cdot \phi_{th} \cdot g_{U\text{-}235}$$

$$B = 1{,}68 \cdot 10^{20} \cdot 0{,}613 \cdot 10^{-24} \cdot 1{,}1 \cdot 10^{14} \cdot 0{,}696 \cdot 10^{-1} = 0{,}789 \cdot 10^{13}\ [n \cdot sec^{-1}]$$

$$R = \frac{\Delta R_I \cdot e^{\lambda t_1}}{\Delta t\,[h] \cdot f_E \cdot 5{,}47 \cdot 10^{-3}} = \frac{\Delta R_I}{\Delta t\,[h] \cdot f_E} \cdot 3{,}44 \cdot 10^4$$

Berechnung R/B:

Temp. [°C]	f_E [Imp/Verf.]	ΔR_I [Imp·sec^{-1}]	Δt [h]	R [Imp·sec^{-1}]	R/B
1250	4,69·10^{-3}	0,24	3	5,86·10^5	7,44·10^{-7}
1400	4,69·10^{-3}	1,8	1,5	4,4·10^6	5,47·10^{-6}
1600	6,45·10^{-3}	9,6	1,5	3,41·10^7	4,32·10^{-5}

Berechnung F(t):

$$F(t) = \frac{N_f(t)}{N_{ges}}$$

T [°C]	t [h]	$R_I(t)$ [Imp/sec]	$\lambda \cdot f_E$	$N_f(t)$	N_{ges}	F(t)	F(t)/RI
1256	1,33	4,96	7,13·10^{-9}	6,96·10^8	2,76·10^{15}	2,5·10^{-7}	
1256	2	6,11	7,13·10^{-9}	8,57·10^8	2,76·10^{15}	3,1·10^{-7}	5,1·10^{-8}
1266	2,8	6,72	7,13·10^{-9}	9,43·10^8	2,76·10^{15}	3,4·10^{-7}	
1400	5,48	6,82	7,13·10^{-9}	9,57·10^8	2,76·10^{15}	3,5·10^{-7}	
1400	6,54	8,2	7,13·10^{-9}	1,15·10^9	2,76·10^{15}	4,15·10^{-7}	
1600	6,97	11,6	9,8·10^{-9}	1,183·10^9	2,76·10^{15}	4,3·10^{-7}	3,7·10^{-8}
1600	8,27	19,6	9,8·10^{-9}	2,0·10^9	2,76·10^{15}	7,25·10^{-7}	
1250	0,5	1,44	7,13·10^{-9}	2,02·10^8	2,76·10^{15}	7,3·10^{-8}	
1250	1,0	3,6	7,13·10^{-9}	5,05·10^8	2,76·10^{15}	1,83·10^{-7}	5,1·10^{-8}

Annealing-Test 11 ... am 6.11.67

Experiment: JP.5. Kapsel: J.A... Part. Sorte: W.M.239
Gew.% U-235: 7.26 [%]

Bestrahlung vom 25.1.67 bis 24.9.67	Bestrahlungstemp.: 1390 [°C]
therm. Fluß – ϕ_{th} 1.12.10^{14} [n · cm^{-2} · sec^{-1}]	schnelle Dosis: .....[nvt]
Abbrand: 5,15 [%fifa] ≙ .....[%fimx]	Einfangquerschnitt: 6,6.13.10^{-24} [cm^2]

Part. Einwaage: 1,28 ... [g]	$\lambda_{Xe\text{-}133} \cdot t_1 = 0{,}131\ [d^{-1}] \cdot 43$
Abklingzeit – t_1: 43 ... [d]	$e^{\lambda_{Xe\text{-}133} \cdot t_1} = 2{,}81 \cdot 10^2$

$$g_{U\text{-}235} = g_{Einwaage} \frac{\text{Gew.\% U-235}}{100} \left(1 - \frac{\text{\% fifa}}{100}\right)$$

$$g_{U\text{-}235} = 1{,}28 \cdot 7{,}26 \cdot 10^{-2} \cdot 0{,}485 = 4{,}51 \cdot 10^{-2}\ [g]$$

$$B = 1{,}68 \cdot 10^{20} \cdot \sigma\,[cm^2] \cdot \phi_{th} \cdot g_{U\text{-}235}$$

$$B = 1{,}03 \cdot 10^{-1} \cdot 1{,}12 \cdot 10^{14} \cdot 4{,}51 \cdot 10^{-2} = 0{,}52 \cdot 10^{12}\ [n \cdot sec^{-1}]$$

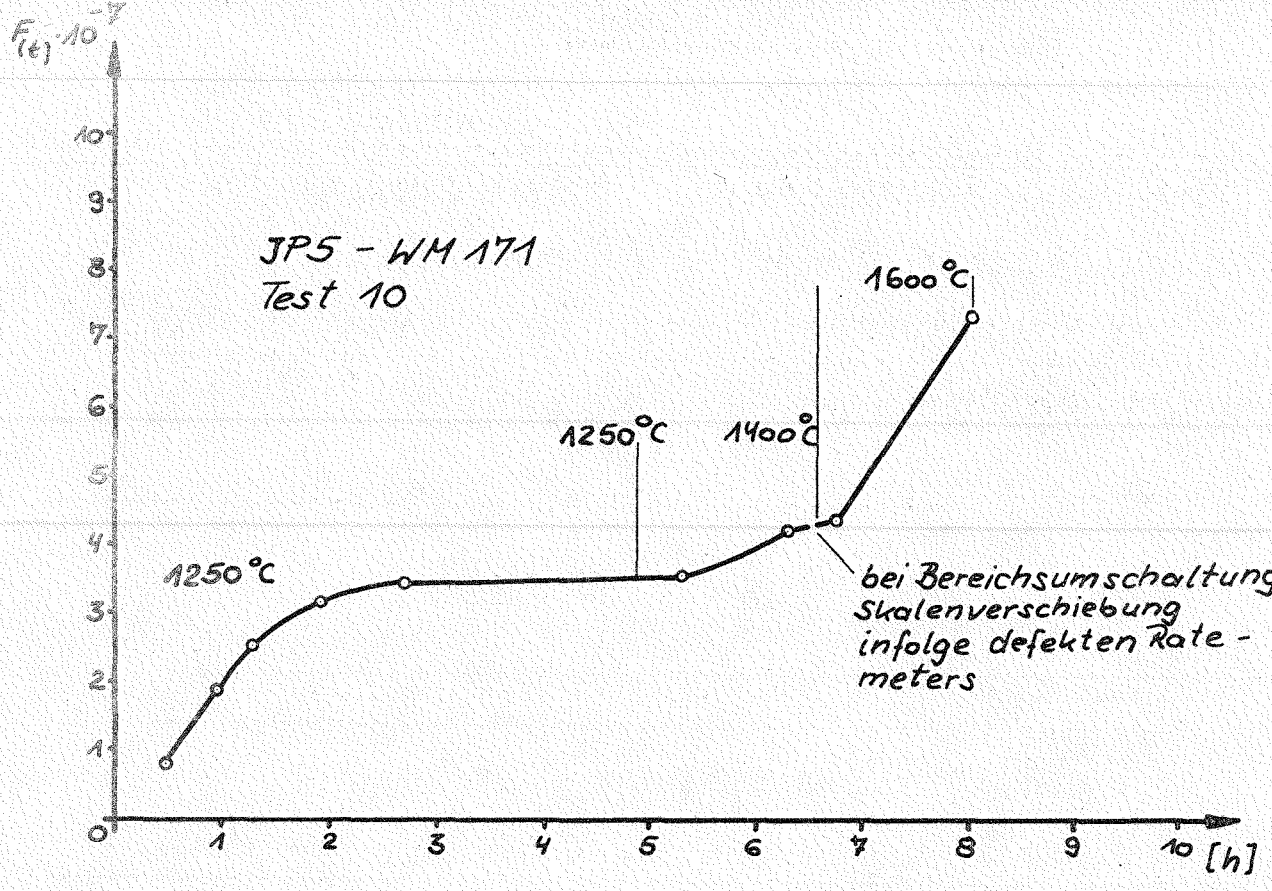
$$R = \frac{\Delta R_I \cdot e^{\lambda t_1}}{\Delta t\,[h] \cdot f_E \cdot 5{,}47 \cdot 10^{-3}} = \frac{\Delta R_I}{\Delta t\,[h] \cdot f_E} \cdot 5{,}14 \cdot 10^4$$

Berechnung R/B:

Temp. [°C]	f_E [Imp/Zerf.]	ΔR_I [Imp.sec^{-1}]	Δt [h]	R [Imp.sec^{-1}]	R/B
1250°	$4{,}69 \cdot 10^{-3}$	0,12	2 h	$6{,}57 \cdot 10^6$	$1{,}263 \cdot 10^{-6}$
1400°	$4{,}69 \cdot 10^{-3}$	0,44	3 h	$1{,}61 \cdot 10^6$	$3{,}1 \cdot 10^{-6}$
1600°	$4{,}69 \cdot 10^{-3}$	2,2	2 h	$1{,}204 \cdot 10^7$	$2{,}32 \cdot 10^{-6}$

Berechnung F(t): $F(t) = \frac{Nr(t)}{Nges.}$

T [°C]	t [h]	$R_I(t)$ [Imp/sec]	$\lambda \cdot f_E$	Nr(t)	Nges.	F(t)	F(f)/RI
1250	0,5	0,48	$7{,}13 \cdot 10^{-9}$	$6{,}74 \cdot 10^7$	$1{,}22 \cdot 10^{15}$	$55 \cdot 10^{-8}$	
1250	2,86	0,6	$7{,}13 \cdot 10^{-9}$	$8{,}4 \cdot 10^7$	$1{,}22 \cdot 10^{15}$	$6{,}9 \cdot 10^{-8}$	
1600	6,1	1,08	$7{,}13 \cdot 10^{-9}$	$1{,}51 \cdot 10^8$	$1{,}22 \cdot 10^{15}$	$1{,}24 \cdot 10^{-7}$	$1{,}15 \cdot 10^{-7}$
1600	8	5,1	$7{,}13 \cdot 10^{-9}$	$7{,}15 \cdot 10^8$	$1{,}22 \cdot 10^{15}$	$5{,}85 \cdot 10^{-7}$	
1600	10,1	7,4	$7{,}13 \cdot 10^{-9}$	$1{,}04 \cdot 10^9$		$8{,}5 \cdot 10^{-7}$	



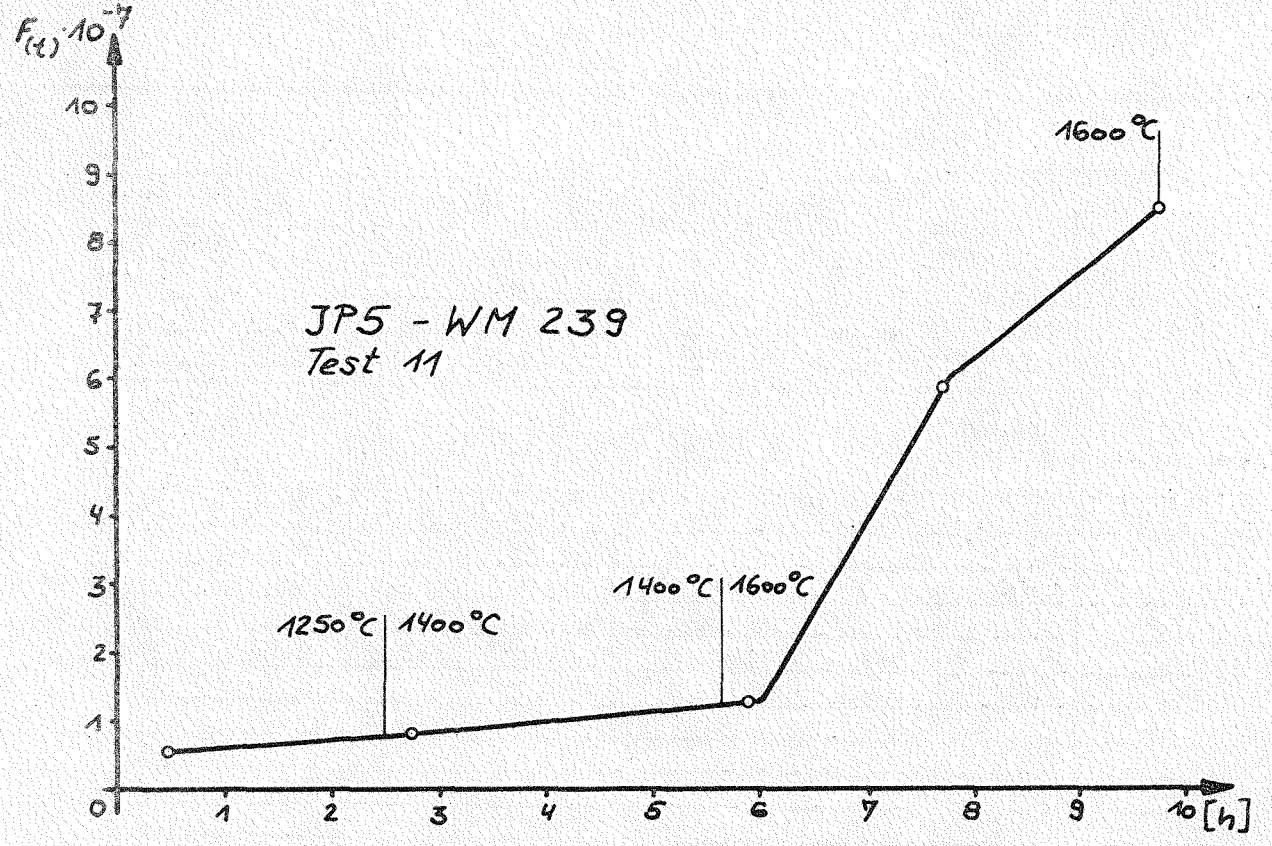


Bild Nr 17 F-Wert-Kurven

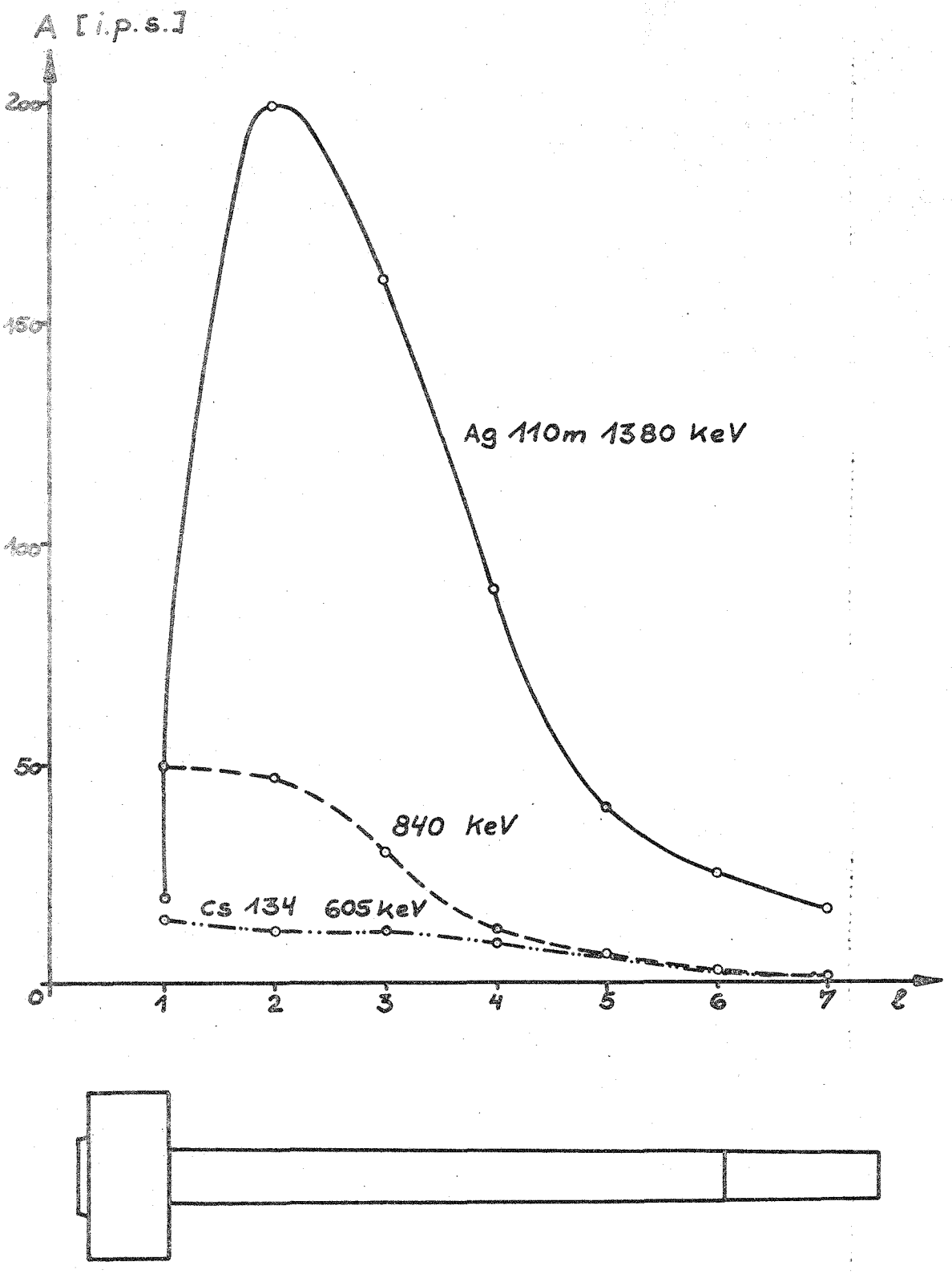


Bild Nr 18 Nuklide im Suszeptorstiel

Akkumulator
70
80
Energie
605-Cs134
660-Cs137+Ag110m
800-Cs134
840
880-Ag110m
1170
Co60(Untergrund)
1330 Co 60 (Untergrund)
1380-Ag110m
1450
1470 Ag110m
1490

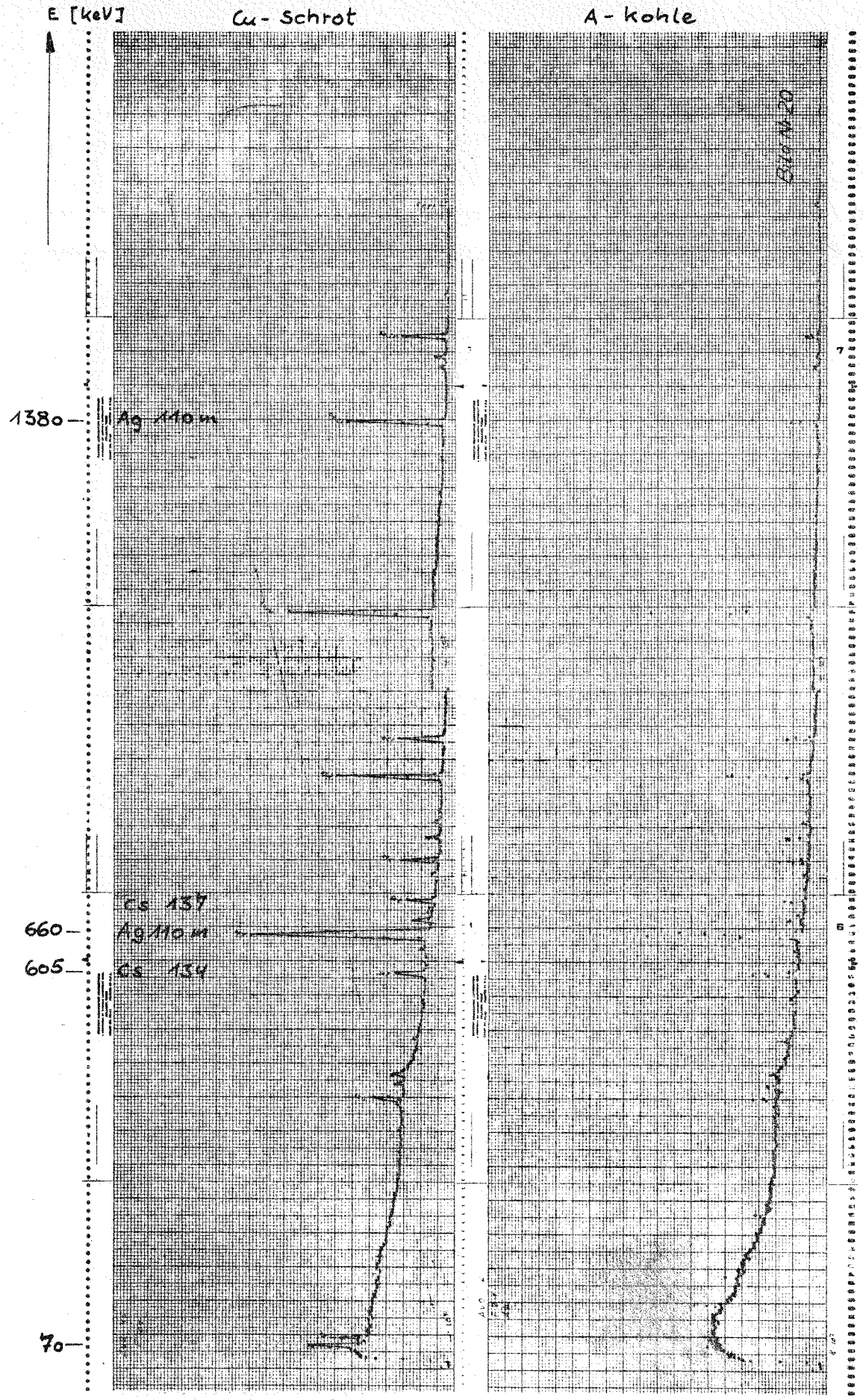


Bild Nr 20 Gamma - Spektren der Jodfallenfüllung