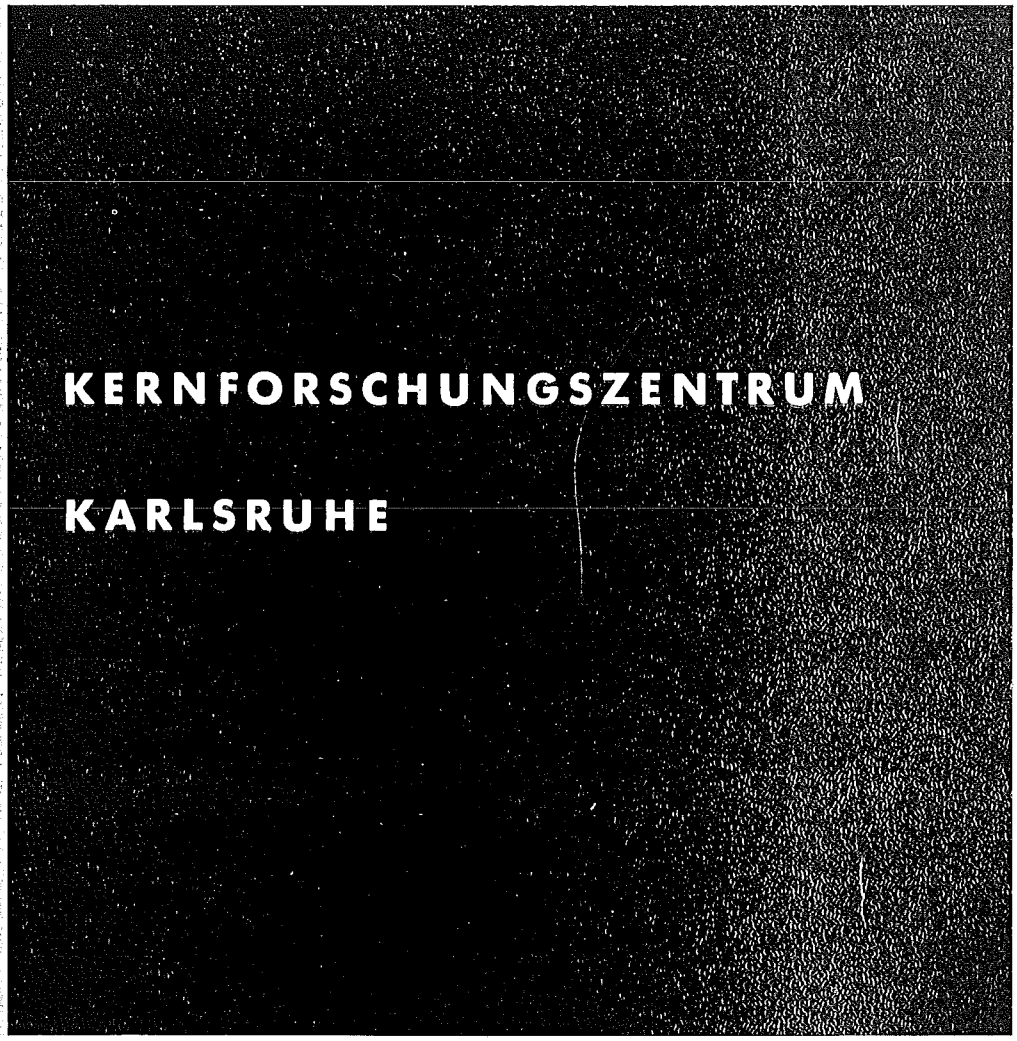

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Institut für Kernverfahrenstechnik

Present State and Development Potential of Separation Nozzle Process

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Present State and Development Potential of Separation Nozzle Process[+)]

by

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+) Revised version of a report presented by W. Ehrfeld at the Gordon Research Conference on Physics and Chemistry of Isotopes, Asilomar, Pacific Grove, Calif., July 1-5, 1974.

Summary

The separation nozzle process developed at the Karlsruhe Nuclear Research Center utilizes the mass dependence of the centrifugal force in a fast curved jet of gaseous uranium hexafluoride to effect a separation of the uranium isotopes. A light auxiliary gas is admixed to the UF_6 feed in a large molar excess in order to increase the velocity of the jet. In addition, the auxiliary gas prevents the UF_6-isotopes from concentrating rapidly in a small layer of the centrifugal field, which also has a favourable effect upon the separation of the isotopes.

The development work on process technology is carried out in cooperation with the STEAG AG, Essen. Two prototype separative stages of different size have been installed which allow to construct separation nozzle plants with a separative capacity of 5×10^5 to 1×10^6 kg USW/year without any connection of stages in parallel. Life and performance tests of commercially fabricated separation elements used in these stages show that the separative work capacity of these elements up to now has not changed over a period of 22,000 hours and that uniform separation characteristics are ensured. At the present state of development separative capacities up to 50 kg USW/year can be achieved with one commercial separation element, which has a diameter of 15 cm and a length of 2 m. Theoretical and experimental studies on separation nozzle cascades indicate that inherently stable operating characteristics exist.

Based on this engineering work and on complete design studies for a demonstration plant, the economics of the separation nozzle process has been analysed. It is shown by these studies that separative work costs of less than $ 50 per kg USW can be expected on the basis of the 1977 technology, if power costs of 9 mills/kWh and plant capacities of 2.5×10^6 kg USW/year are assumed.

Besides the development work on process engineering extensive studies have been carried out on the physics of the separation nozzle method. As a result of systematic optimization of the nozzle parameters the power utilization index of the process has been improved by a factor of 2 in the past few years. Studies now under way show a potential for significant further increase in separation factor through delaying UF_6 sedimentation in the centrifugal field of the flow. It is expected that the separation nozzle process will result in considerably lower separative work costs within a few years as compared to the other methods for the separation of uranium isotopes.

Zusammenfassung

Bei dem am Kernforschungszentrum in Karlsruhe entwickelten Trenndüsenverfahren beruht die Entmischung der Uranisotope auf der Massenabhängigkeit der Zentrifugalkraft in einer schnellen gekrümmten Strömung aus gasförmigem UF_6. Dem UF_6 ist ein leichtes Zusatzgas in hohem molarem Überschuß beigefügt, durch das die Strömungsgeschwindigkeit des UF_6 erhöht wird. Das Zusatzgas verzögert außerdem die Sedimentation der UF_6-Isotope im Zentrifugalfeld der Strömung, was sich für die Trennung der Isotope ebenfalls günstig auswirkt.

Die technische Entwicklung des Trenndüsenverfahrens wird gemeinsam mit der STEAG AG, Essen, durchgeführt. Im Rahmen dieser Entwicklungsarbeiten wurden zwei Trennstufen unterschiedlicher Baugröße installiert, welche die Erstellung von Trenndüsenanlagen mit Trennleistungen von $5 \cdot 10^5$ bis $1 \cdot 10^6$ kg UTA/Jahr ohne Parallelschaltung von Stufen erlauben. Die in den Stufen eingesetzten technischen Trennelemente zeichnen sich durch einheitliche Trenneigenschaften aus. Beim Dauertest der Trennelemente wurde in einem Zeitraum von 22.000 Stunden bisher kein Abfall der Trennleistung registriert. Beim gegenwärtigen Entwicklungsstand kann mit einem einzelnen Trennelement bei einem Durchmesser von 15 cm und einer Länge von 2 m eine Trennleistung von bis zu 50 kg UTA/Jahr erreicht werden. Bei theoretischen und experimentellen Untersuchungen zum Kaskadenverhalten von Trenndüsenanlagen wurde nachgewiesen, daß ein inhärent stabiles Betriebsverhalten erzielt werden kann.

Auf der Grundlage der technischen Entwicklungsarbeiten und auf der Grundlage detaillierter Planungsarbeiten für eine Demonstrationsanlage wurde eine Wirtschaftlichkeitsanalyse zum Trenndüsenverfahren durchgeführt. Hiernach sollten bei dem für 1977 zu erwartenden Entwicklungsstand des Verfahrens Trennarbeitskosten unter 50 $/kg UTA erzielt werden, wenn von einem Strompreis von 9 mills/kWh und einer Anlagenkapazität von $2.5 \cdot 10^6$ kg UTA/Jahr ausgegangen wird.

Neben den technischen Entwicklungsarbeiten wurden ausgedehnte Untersuchungen zur Physik des Trenndüsenverfahrens durchgeführt. Dabei konnte der Energienutzungsgrad des Verfahrens durch eine systematische Optimierung der Betriebsparameter der Trenndüse in den letzten Jahren um einen Faktor 2 verbessert werden. Zur Zeit laufende Untersuchungen weisen darauf hin, daß der Trenneffekt durch eine Verzögerung der UF_6-Sedimentation im Zentrifugalfeld der Strömung noch wesentlich gesteigert werden kann. Es ist zu erwarten, daß bereits in wenigen Jahren mit dem Trenndüsenverfahren die Trennarbeitskosten der anderen Verfahren zur Uranisotopentrennung beträchtlich unterschritten werden können.

Introduction

The separation nozzle method for the enrichment of the light uranium isotope U-235 has been developed at the Karlsruhe Nuclear Research Center as an alternative to the gaseous diffusion and centrifuge processes[1)2)]. Since March 1970 a German industrial company, STEAG AG of Essen, has been financially involved in the commercial implementation of the separation nozzle method.

In the separation nozzle method isotope separation, as in the centrifuge, is brought about by the mass dependence of the centrifugal force in a fast, curved flow. Fig. 1 shows the principle of the separation nozzle system used to date in the commercial implementation of the method[3)]. Gaseous uranium hexafluoride mixed with a light auxiliary gas - helium or hydrogen - expands along a curved fixed wall. At the end of the deflection section the flow is split up into a lighter and a heavier fraction by means of a skimmer. The light auxiliary gas, which is present in a large molar excess increases the flow velocity of the UF_6 and, hence, it increases the centrifugal force determining the separation. In addition, the light gas delays the joint sedimentation of the UF_6 isotopes in the centrifugal field, which also has a favorable effect upon separation of the isotopes[4)]. Under optimum operating conditions the maximum flow velocity of UF_6 in the separation nozzle arrangement is approximately the same as the peripheral velocity of a gas centrifuge.

Technical Development Status

In the separation nozzle method the optimum gas pressure is inversely proportional to the characteristic dimensions of the separation element. Since, for reasons of economy, the gas pressure should be made as high as possible, the characteristic dimensions of the separation element are selected as small as possible. Fig. 2 shows the design of the separation elements presently used in technical development: An extruded aluminum tube which is subdivided by baffle walls into ten compartments has ten slitshaped separation nozzle systems on its surface. Feed gas is introduced through every other chamber and the heavy fraction is removed through each adjacent chamber. The light fraction flows radially outward into a tank containing the separation elements.

The separation nozzle system proper is constituted by the deflection groove in the tube and by specially designed aluminum strips. The deflection groove has a radius of curvature of approximately 1/10 mm. The aluminum strips are fixed in the dovetail grooves of the tube by balls forced in between them. This special method of fabricating commercial separation elements, which lends itself particularly well to a mass production of separation elements, has been developed by Messerschmitt-Bölkow-Blohm, München.

Three years ago life tests of a section of a separation element manufactured in a commercial fabrication process were started in a special life test rig. As is evident from Fig. 3, in which the isotope separation effect ε_A [+)] is plotted over

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+) Separation effect $\varepsilon_A = \frac{n_L(1-n_H)}{n_H(1-n_L)} - 1 = A-1$

A = separation factor between enriched and depleted streams
n_L = mole fraction of the light uranium isotope in the light fraction
n_H = mole fraction of the light uranium isotope in the heavy fraction.

the operating period, the separative work capacity of this separation element has not changed within the error limits over a period of 22,000 hours.

Meanwhile, the fabrication technique has been continuously improved, manufacturing tolerances have been greatly reduced. As a result, variations formerly observed in the separation characteristics of various tubes have largely been eliminated. Fig. 4 shows the results of separation experiments carried out with a mixture of He/UF_6 with an expansion ratio p_o/p_M = 3.5 in five separation element tubes of 2 m length with 20 m slitlength each. The plot shows the separation effect ε_A, and the UF_6-cut as a function of the nozzle inlet pressure p_o. The different symbols are assigned to the individual tubes. It is seen that all five tubes show only minor deviations in the separation effect and in the cut. The separative work production of the tube when operated on a He/UF_6 mixture with an expansion ratio of 4 is approximately 12 kg USW/year. The use of a mixture of H_2/UF_6 would result in a separative work production of approximately 15 kg USW/year.

Fig. 5 shows the cross section of a prototype separative stage containing about 80 of these separation element tubes[3]. The stage consists of the tank containing the separation elements, the cross piece for gas distribution, the two-stage gas cooler, the two-stage centrifugal compressor, and the motor firmly coupled with the compressor. Because of the high gas flow the stage has been tested in closed-circuit operation. The heavy fraction is recycled to the upper closed-circuit loop and mixed with the light fraction coming from the separation elements.

Fig. 6 shows a photograph of this prototype separative stage. The stage was installed in July 1970. First of all, the compressor was tested. In September 1972 the separation element tubes were inserted into the stage. The separation experiments performed on a mixture of He/UF_6 with 5 mole-% of UF_6 were successful from the outset.

Fig. 7 shows another type of separative stage needed for a commercial plant in addition of the stage shown first. This separative stage, which was developed by STEAG and installed early this year, is an advanced version of the stage previously shown.

By an extented experimental program, the operating characteristics of separation nozzle cascades have been investigated using a 10-stage pilot plant. At present, the operation of industrial plants can be simulated by digital computer programs, whose accuracy has been checked in the previous experiments[5]. These investigations have shown smooth and stable operation of separation nozzle cascades even in the case of severe perturbations[6]. Also for more advanced process design an inherent stable cascade operation will be assured[7]. This means, that the operation of separation nozzle cascade will present no special control problems.

The stages shown in Fig. 6 and 7 allow of the construction of separation plants with a separative capacity of 500 to 1000 t USW/year without requiring any connection of stages in parallel. Since separation plants with a separative capacity of several thousand metric tons per year are more attractive for practical application, a technological program for the development of components of larger plants is planned presently. This technology program centers around the development of a high capacity separative stage with a separative work production of 6 to 10 t USW/year.

Economics of the Separation Nozzle Process

The development work performed to date constitutes the basis of an analysis of the economics of the separation nozzle method.

In order to obtain reliable data about the investment costs associated with the separation nozzle method, STEAG worked out the complete design of a separation nozzle demonstration plant with a separative capacity of 600 t USW/year. In addition, a number of industries were commissioned to develop design and fabrication methods for the most important plant components under the assumption of mass production. These orders, among other items, concerned the separation elements, compressors, the cooling system, the piping, tanks, and valves. On the basis of the costs determined in this way, detailed optimization studies on the design and operation of larger separation nozzle plants were performed in cooperation with Linde AG, München.

From these carefully confirmed capital costs the separative work costs were calculated for the separation nozzle process based on the development status attained by 1973. Table 1 gives the cost breakdown for a separation nozzle plant with a separative capacity of 2500 t USW/year determined in this way. The estimate was based on power costs of 9 mills/kWh, an interest rate on the capital of 10 %, depreciation over 20 years and a construction period of 5 years. The prices of plant components correspond to the cost status as of 1973, the rate of exchange was assumed to be $1 = DM 2.50. Under these conditions, specific capital costs turned out to be approximately $30 per kg USW, and specific power costs were found to be approximately $36 per kg USW. The other costs contribute only little to the costs of the separative work, which amount to $72 per kg USW.

TYPE OF COST	Technology 1973	1977
	$/kg USW	$/kg USW
Interest and Depreciation	30.2	22.6
Power Cost (9 mills/kWh)	35.6	21.2
Maintenance and Operating Costs	2.2	1.6
Stockpile	.8	.7
Salaries and Wages	2.8	2.8
Separative Work Cost	71.6	48.9

Depreciation: 20 years Interest: 10%/year

Price Level 1973 1$ ≙ 2.5 DM

Plant Separative Capacity: 2500 t USW/year

Table 1: Estimated cost breakdowns for separation nozzle plants using 1973 technology and 1977 technology, respectively.

Table 1 also indicates the cost breakdown resulting for the development status of the separation nozzle method anticipated for 1977. The cost extrapolation is based on current and planned work with regard to the scientific and engineering improvement of the simple separation nozzle system (Fig. 1). So-called advanced separation nozzle systems, which will be described in the third part of this paper, were not included in these cost assessments. On the whole, a separative work price of approximately $49 per kg USW is found for the technology of 1977, if a separation nozzle plant with a separative capacity of 2500 t USW/year and power costs of 9 mills/kWh are assumed.

Technology 1977	Power Cost 6 mills/kWh	9 mills/kWh	12 mills/kWh
SEPARATIVE WORK COST ($/kg USW) Plant Separative Capacity 3500 t USW/year	39	46	54
SEPARATIVE WORK COST ($/kg USW) Plant Separative Capacity 9000 t USW/year	30	37	45

Table 2: Estimated costs of separative work versus power costs and plant size (1977 technology).

Table 2 is a compilation of the separative work costs corresponding to the technological status of 1977 for separation nozzle plants of various sizes and for various power costs. Plant capacities of 3500 and 9000 t USW/year, respectively, were assumed. The power costs were assumed to be 6, 9 and 12 mills/kWh. Power costs of 6 mills/kWh are probably realistic for some countries with favourable natural resources. It is seen that the separative work cost will exceed $50 per kg USW only in the most adverse case. For the rest, it may be assumed that separative work costs around $40 per kg USW can be realized.

Development work on the physics of the separation nozzle method

The development work concerning the physics of the separation nozzle method is concentrated, above all, upon the optimization of the operating conditions of the separation nozzle system shown in Fig. 1. One of the major goals is the reduction of the specific energy consumption of the process. These studies must be conducted on the basis of extensive separation experiments with UF_6 because the complicated coupled flow and diffusion phenomena in the separation nozzle can be assessed theoretically only in a qualitative way.

Fig. 8 shows one of the experimental setups for UF_6 separation experiments of the type recently used for optimization studies. This apparatus has been greatly improved over earlier equipment with respect to its speed in performing measurements. Now, even an extensive experimental program can be handled within a reasonable period of time. It is possible during the separation experiment to directly measure and adjust the cut of the separation nozzle and to vary geometric parameters of the separation nozzle during the test. Adjusting the cut, which changes markedly in a fixed separation nozzle geometry when the operating conditions are varied, is a particularly important aspect. Only those experiments, in which the cut assumes the value required by the cascade circuit, can be used for optimization purposes. All the data produced by the equipment are evaluated in computer programs immediately after the experiment and analyzed in the light of technical and economic aspects. These data are used to set the conditions for the further experiments in finding the optimum operating conditions.

Despite the greatly improved experimental technique these optimization studies require a considerable amount of time because a multitude of parameters must be varied, and these parameters cannot be optimized independently of each other. Even in the simple separation nozzle system (Fig. 1) there are more than 10 such parameters; these are the most important geometric dimensions of the nozzle, the inlet pressure, the suction pressures, the cut, and the UF_6-concentration.

The success so far achieved in optimizing the parameters of the separation nozzle is shown in the improvement achieved in the power utilization index of the separation nozzle. In Fig. 9 the power utilization index in kg USW per megawatt day is plotted over the development time; all the losses and efficiencies of a separation nozzle plant, such as the cascade efficiency, the electric efficiency and the compressor efficiency, are fully taken into account. It is seen that the efficiency in the utilization of power has been improved by a factor of 2 in the past few years. The results of studies now under way seem to indicate that this development trend in the power utilization index will be continued for some years to come.

The greatly improved measurement technique of the new separation equipment made it possible to carry out systematic separation experiments with complicated advanced separation nozzle systems[8]. The advanced separation nozzle systems originated from extensive theoretical and experimental studies of the physics of the separation nozzle method. In this work, possibilities of reducing the friction losses in the separation nozzle flow and of using the kinetic energy of the flow in a second nozzle were indicated[9]. One particularly interesting possibility is the chance of greatly increasing the isotope separation effect by delaying the UF_6 sedimentation in the centrifugal field of the flow[4].

This delay effect will be explained on the basis of Fig. 10. First of all, the relation between the elementary effect of isotope separation and the UF_6 cut as determined experimentally is shown. In the experiments a mixture of 1.6 mole % UF_6 and 98.4 mole % H_2 was used with an expansion ratio of 8. At the lowest cut investigated in this case the separation effect is 4.2 %, i.e., it is approximately 10 times higher than in the gaseous diffusion process. Under practical operating conditions, in which mixtures with a higher UF_6-content are used at expansion ratios of about 2, the separation effect is about 4 to 5 times higher than in the case of gaseous diffusion. The experimental values greatly exceed those values which would result theoretically in the case of equilibrium separation even for an infinitely high flow velocity. Equilibrium separation in this case characterizes the condition in which each component of the mixture has assumed a density distribution corresponding to the centrifugal forces. This is the condition in which the separating pressure diffusion is just in an equilibrium with the remixing ordinary diffusion caused by the concentration gradients. So the equilibrium distribution is analogous to the barometric altitude distribution of components of different weights in a gravitational field.

The reason why in the case of equilibrium separation the isotope separation effect cannot exceed a specific maximum value even for infinitely high centrifugal forces or infinitely high flow velocities in the separation nozzle is the fact that UF_6 is concentrated into an increasingly narrower layer as the flow velocity increases. Although the concentration gradients in this layer increase as the square of velocity, the thickness of the layer at the same time decreases as the square of velocity so that the separation effect moves towards a finite limiting value at a given cut.

The upper limit of equilibrium separation was exceeded in the experiments because the additional gas delays the formation of a narrow UF_6 layer with steep concentration gradients. However, this delay effect, which tends to increase the separation factor, only has a small effect in the separation nozzle used so far. This is caused by the fact that due to the large difference in the molecular weight, UF_6 greatly separates from the auxiliary gas and concentrates at the deflection wall of the separation nozzle. In order to make better use of the delay effect, the separation between the UF_6 and the auxiliary gas must be counteracted. One basic possibility of making use of this concept is shown in the separation nozzle system as outlined in Fig. 11.

This separation nozzle system consists of two series connected separation units. The first separation unit is a conventional separation nozzle in which the flow is deflected at a fixed wall. In the second separation unit the flow is deflected by a gas stream. The separation process in the first nozzle develops in the familiar way: The heavy UF_6 is accelerated by the light auxiliary gas, there is a separation of the uranium isotopes in the fast curved flow which is accompanied by a large separation between UF_6 and the auxiliary gas. The light fraction enriched in U-235, which contains most of the auxiliary gas, is carried on to a higher stage of the cascade. The heavy fraction depleted in U-235, which contains the bulk of the UF_6, enters the second separation unit with a high kinetic energy.

Here the heavy fraction is deflected again and further split up. However, in this case, the deflection is not produced at a fixed wall but by a sweep gas which is taken from a lower stage of the cascade as a fraction rich in auxiliary gas. In this way, a more uniform UF_6 distribution is achieved in the centrifugal field, and the UF_6 is not concentrated in a narrow layer at the boundary of the centrifugal field, as

would be the case if a solid wall were used for deflection. Essentially, mixing of the fractions with high and low UF_6 content is done not in the pipe-lines of the cascade but within the separation element proper.

Fig. 12 shows results of a few preliminary separation experiments in the second nozzle, in which the flow is deflected by a sweep gas. The plot shows the dependence of the separation effect ε_A on the ratio between the sweep gas pressure and the suction pressure. In this case the sweep gas flow rate was set to 10 % of the overall gas flow, the cut being one third. If the suction pressure is reduced relative to the sweep gas pressure, i.e., the ratio between the sweep gas pressure and the suction pressure is increased, the velocity of the flow will increase as one result. However, there is another result also: If this pressure ratio is increased, the UF_6 jet will be swept by the auxiliary gas more and more intensively, i.e., the UF_6 flow lines are spread farther and farther apart and the UF_6 is distributed more uniformly in the centrifugal field. It is seen that already an increase in this pressure ratio from approximately 1.3 to 2.1 will cause the elementary effect of isotope separation to quickly rise to values of approximately 1 %. These values are comparable with the separation effect of 1.5 % existing in the first nozzle. In more recent experiments now under way, higher values than 1 % have already been achieved.

For the time being the studies conducted on such advanced separation nozzle systems still primarily serve the purpose of deepening the understanding of the phenomena involved. As soon as the current preliminary investigations have been finished work will be concentrated upon the arrangement appearing to be most promising, and an optimization based on technical and economical aspects will be performed as in the simple separation nozzle arrangement.

Even if an advanced separation nozzle arrangement were to be a much more complicated separation element, it has been demonstrated that it can be manufactured by a method developed by the Siemens AG, München. This method of fabrication is outlined in Fig. 13: The separation nozzle structure together with the gas ducts is photoetched into a metal foil. In the specimen shown here the structures of simple separation nozzles have been etched. Approximately 100 such metal foils are stacked one above the other, covered with cover plates and clamped together.

The separation element strips produced in this way are inserted in a tube which again serves as the basic unit of commercial separation elements. One such separation element tube is shown in Fig. 14. Approximately 80 strips are arranged in grooves parallel to each other between two tube halves. The tube halves are reinforced by a center web for stability reasons. To illustrate the details of this arrangement the upper half of the tube is shortened in this picture. The two tube halves serve for introducing the feed gas and removing the heavy fraction. The light fraction enriched in U-235 leaves the separation element tube between the two halves. A tube of this kind, which may be 2 m long and have a diameter of approximately 15 cm, can produce a separative work production of approximately 50 kg USW/year, if the simple separation nozzle geometry (Fig. 1) is used.

The success so far achieved in this development makes it likely that the separation nozzle method will henceforth establish itself side by side with the centrifuge method and the gaseous diffusion method. In the light of the relatively low capital and maintenance costs, and on the basis of the high development potential the separation nozzle process should result in considerably lower separative work costs within few years as compared to the other methods.

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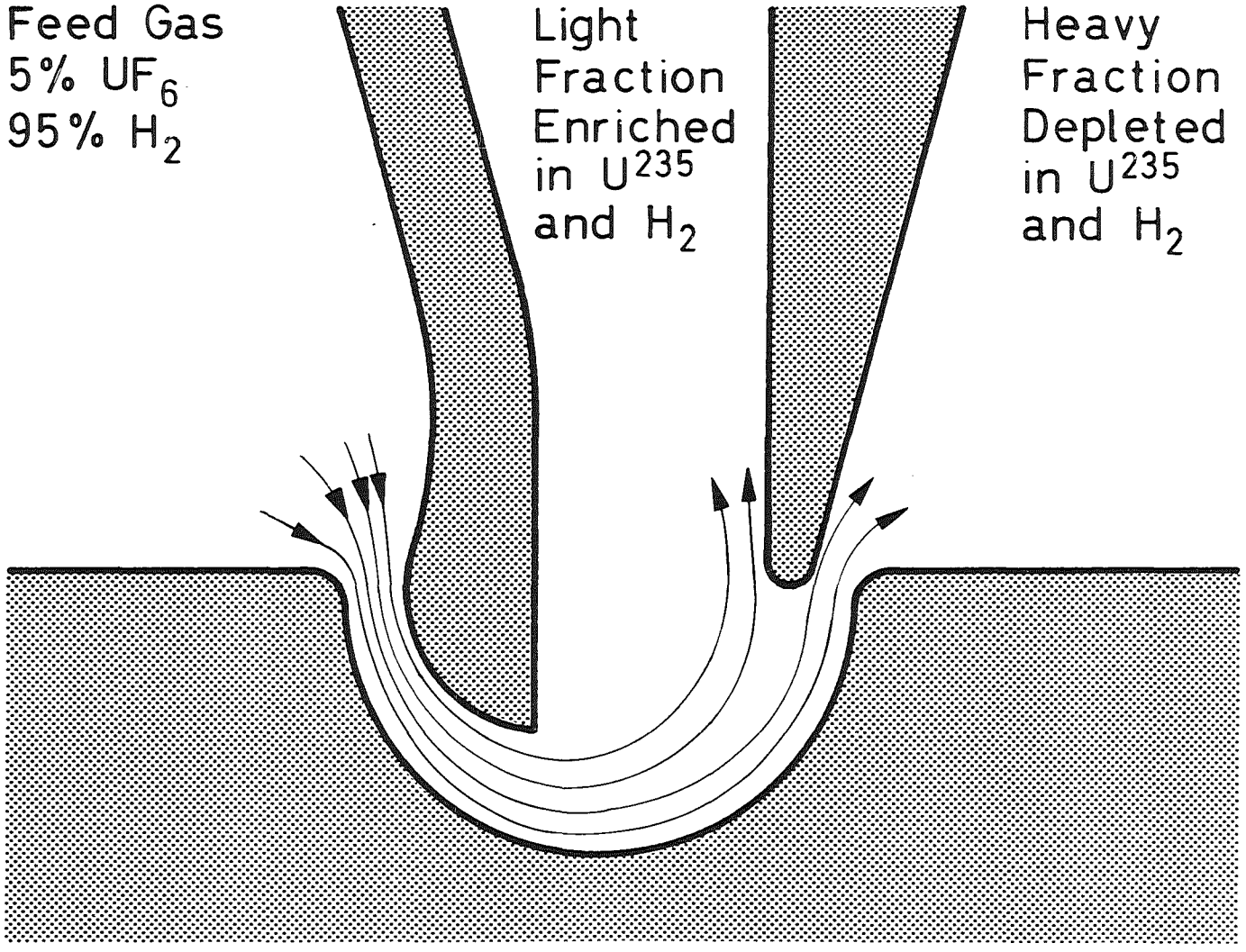

Fig. 1: Cross-section of the separation nozzle system used to date in the commercial implementation of the separation nozzle process.

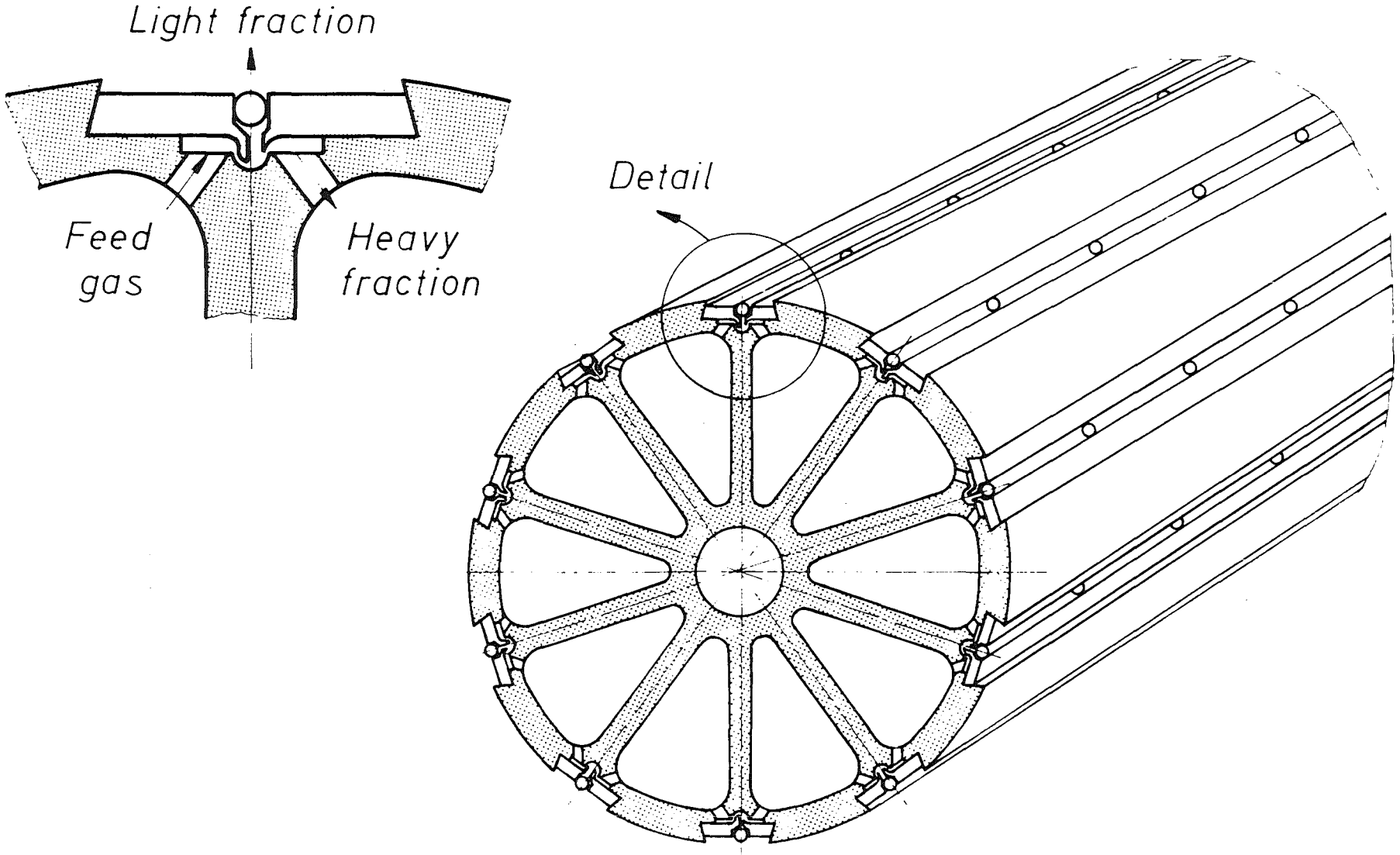

Fig. 2: Separation element tube with 10 separating slits manufactured in a commercial fabrication process.

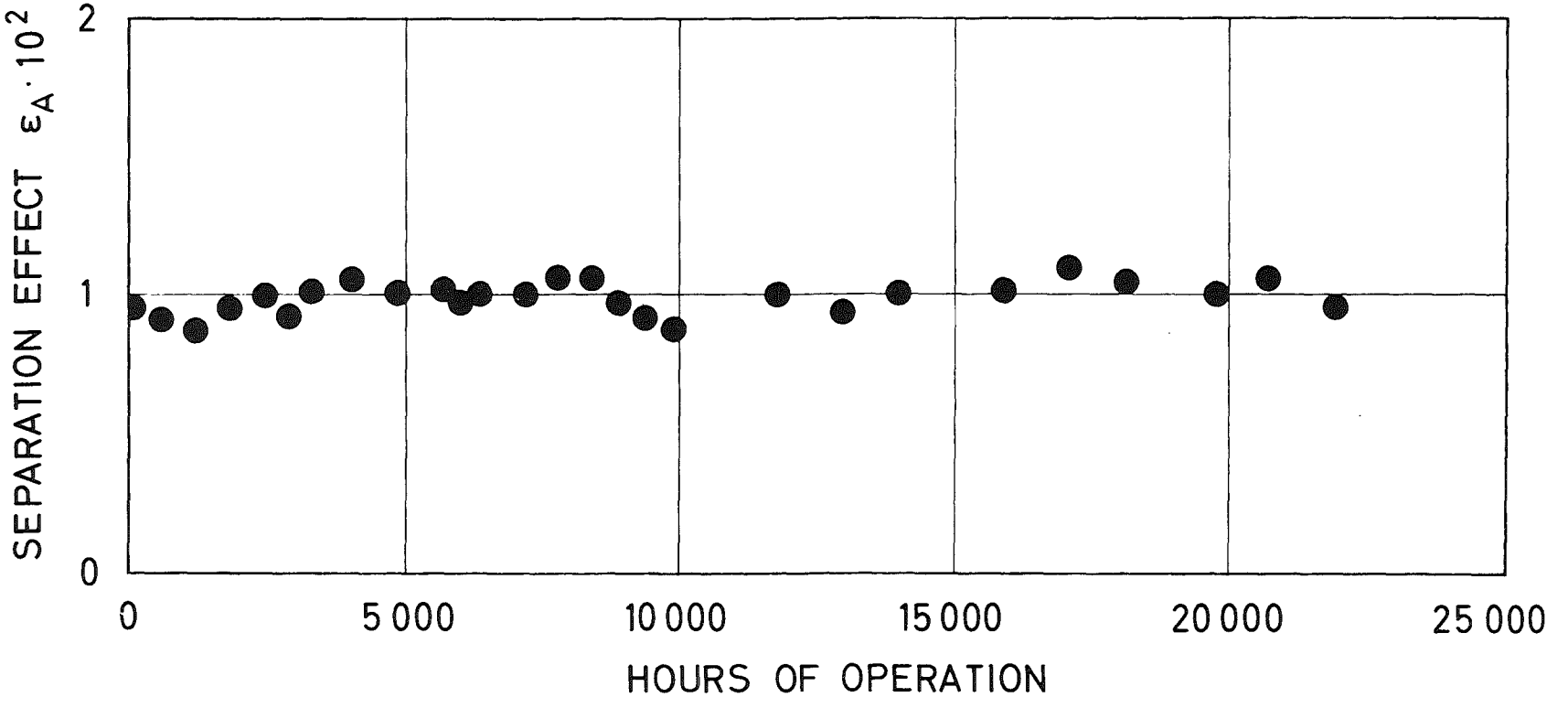

Fig. 3: Life test of a commercial separation element.

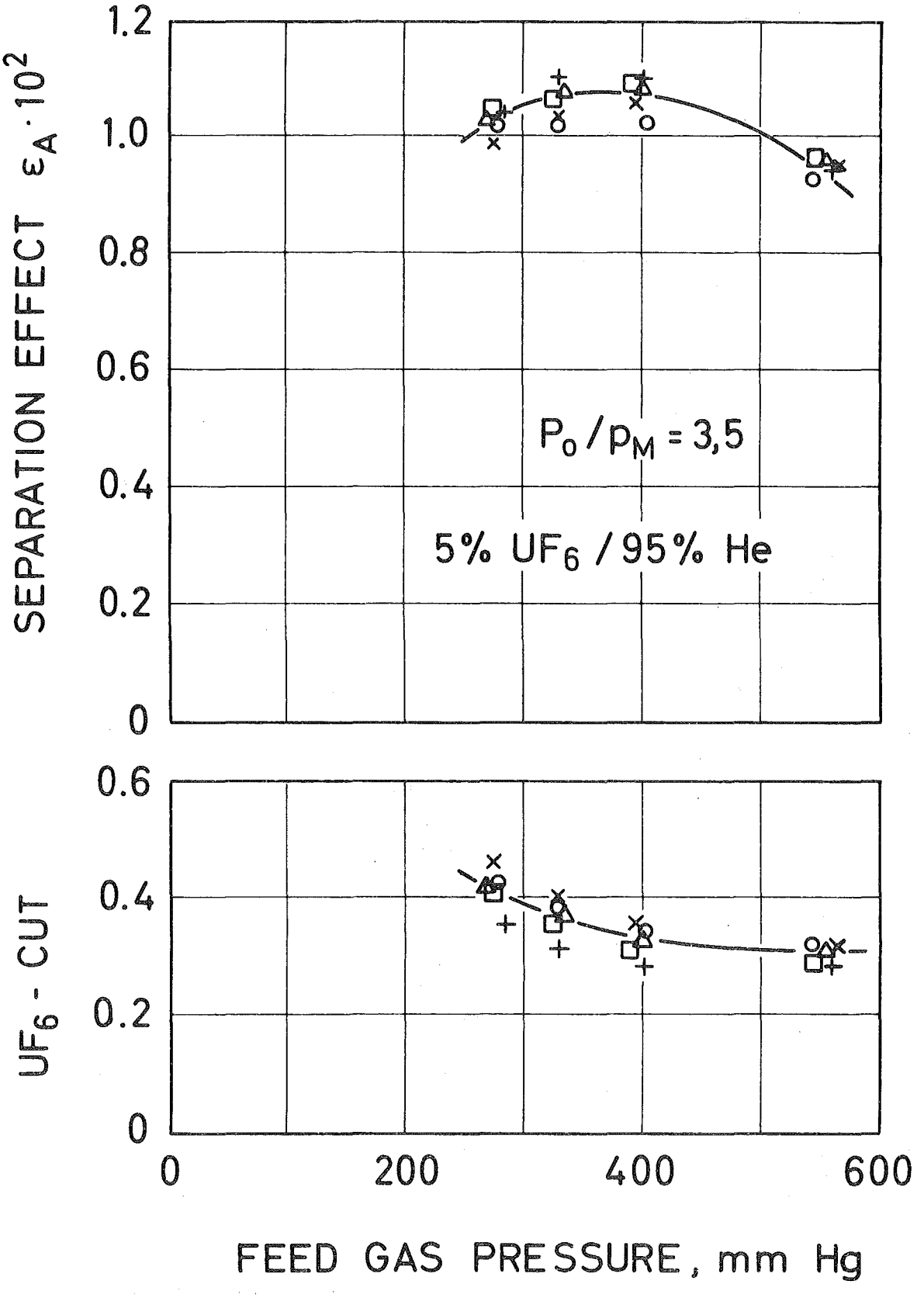

Fig. 4: Results of separation experiments in 5 separation element tubes of 2 m length with 20 m slit-length each.

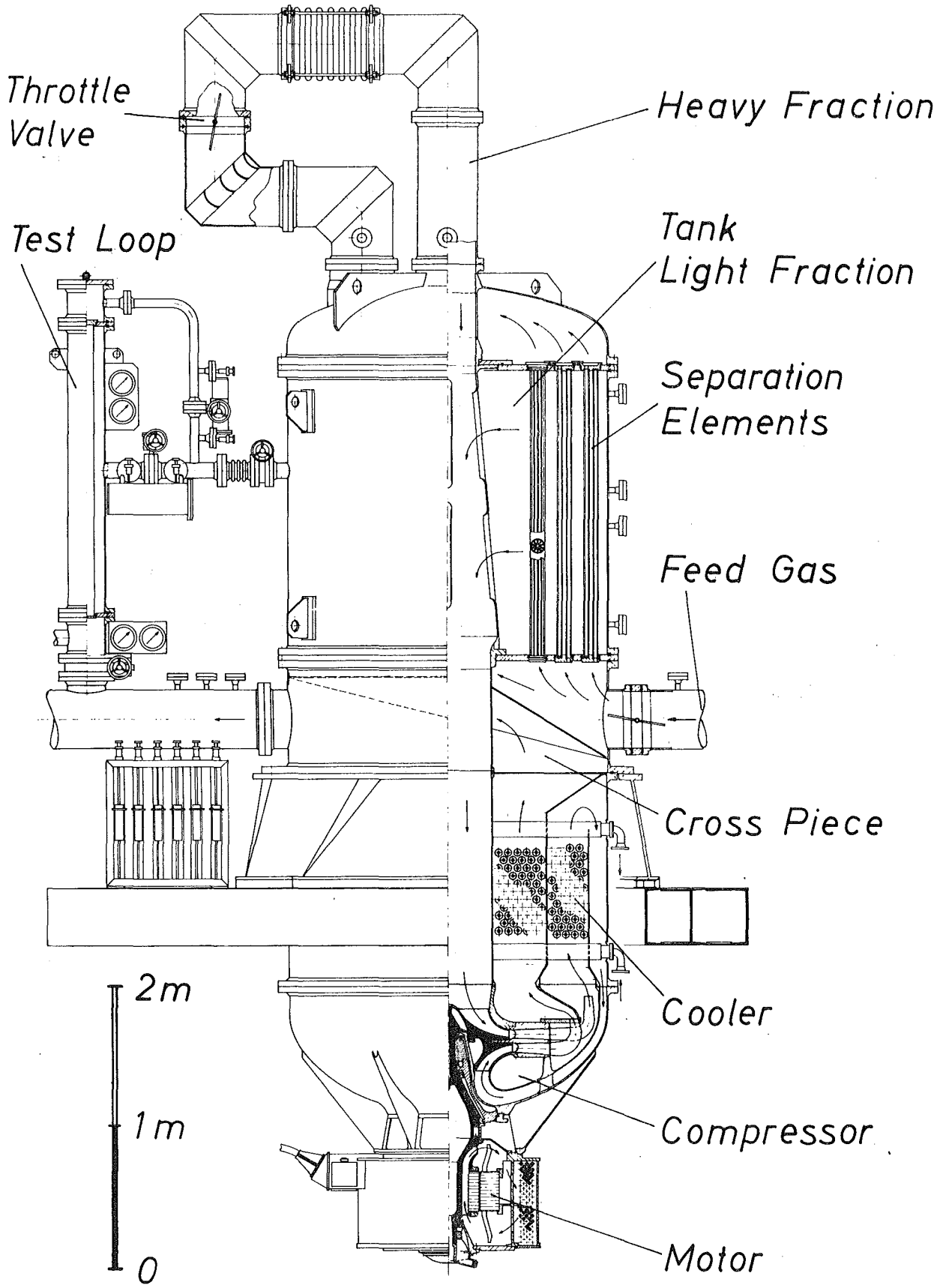

Fig. 5: Cross-section of the prototype separative stage.

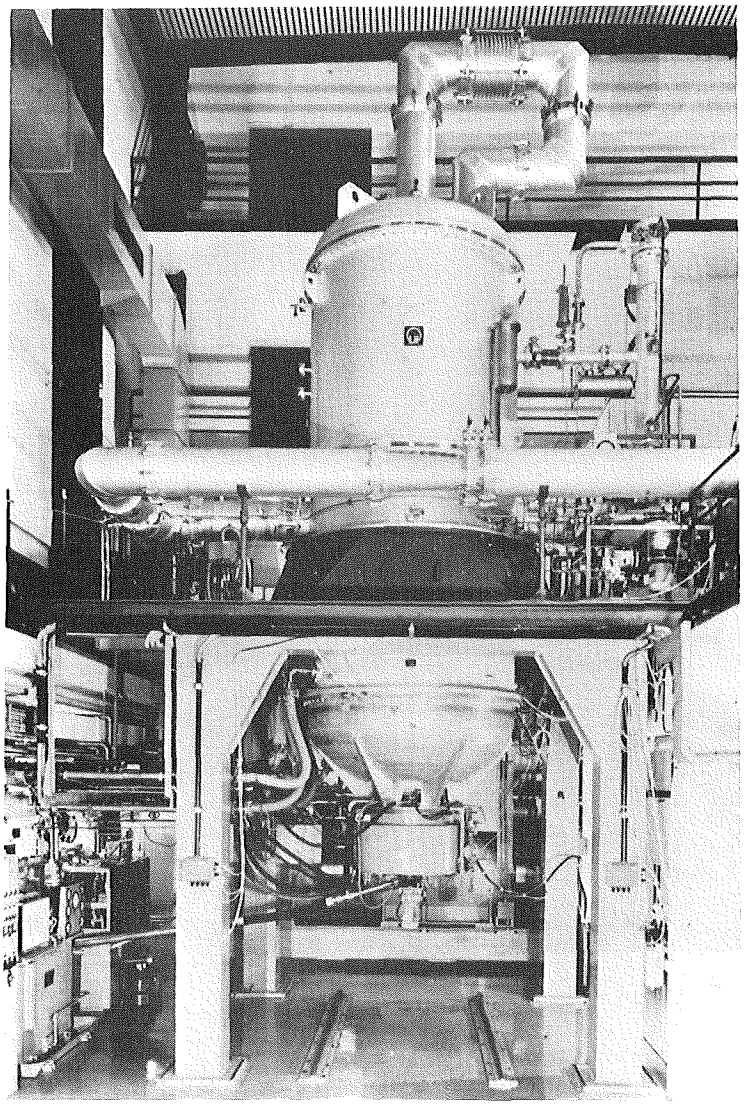

Fig. 6: View of the prototype separative stage.

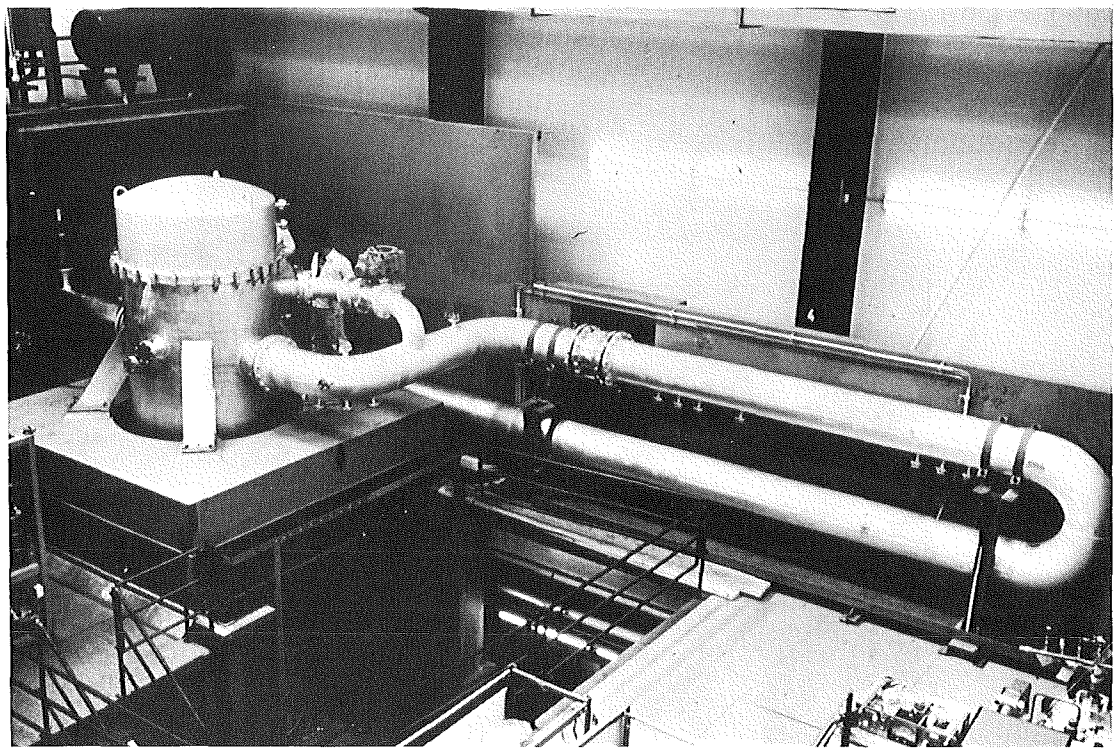

Fig. 7: View of the separative stage developed by STEAG AG, Essen.

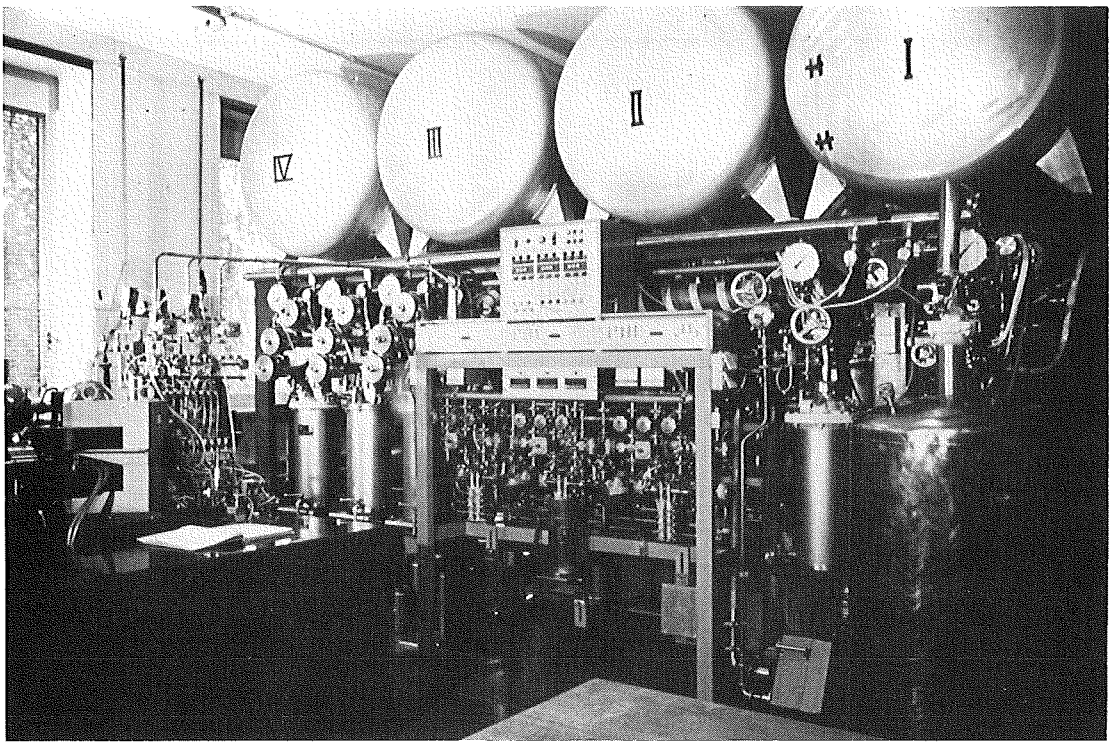

Fig. 8: One of the experimental setups for optimization studies.

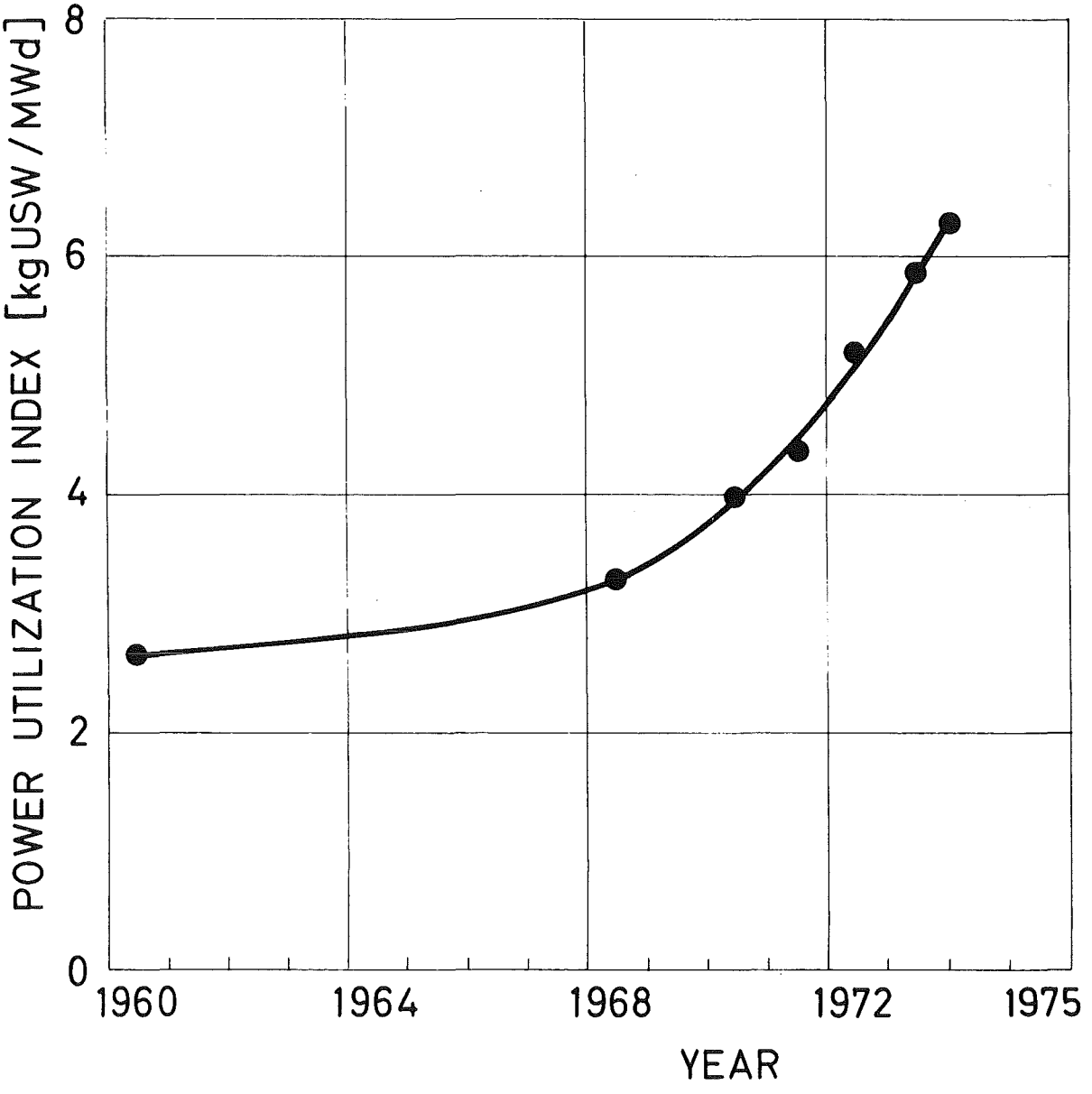

Fig. 9: Power utilization index versus development time.

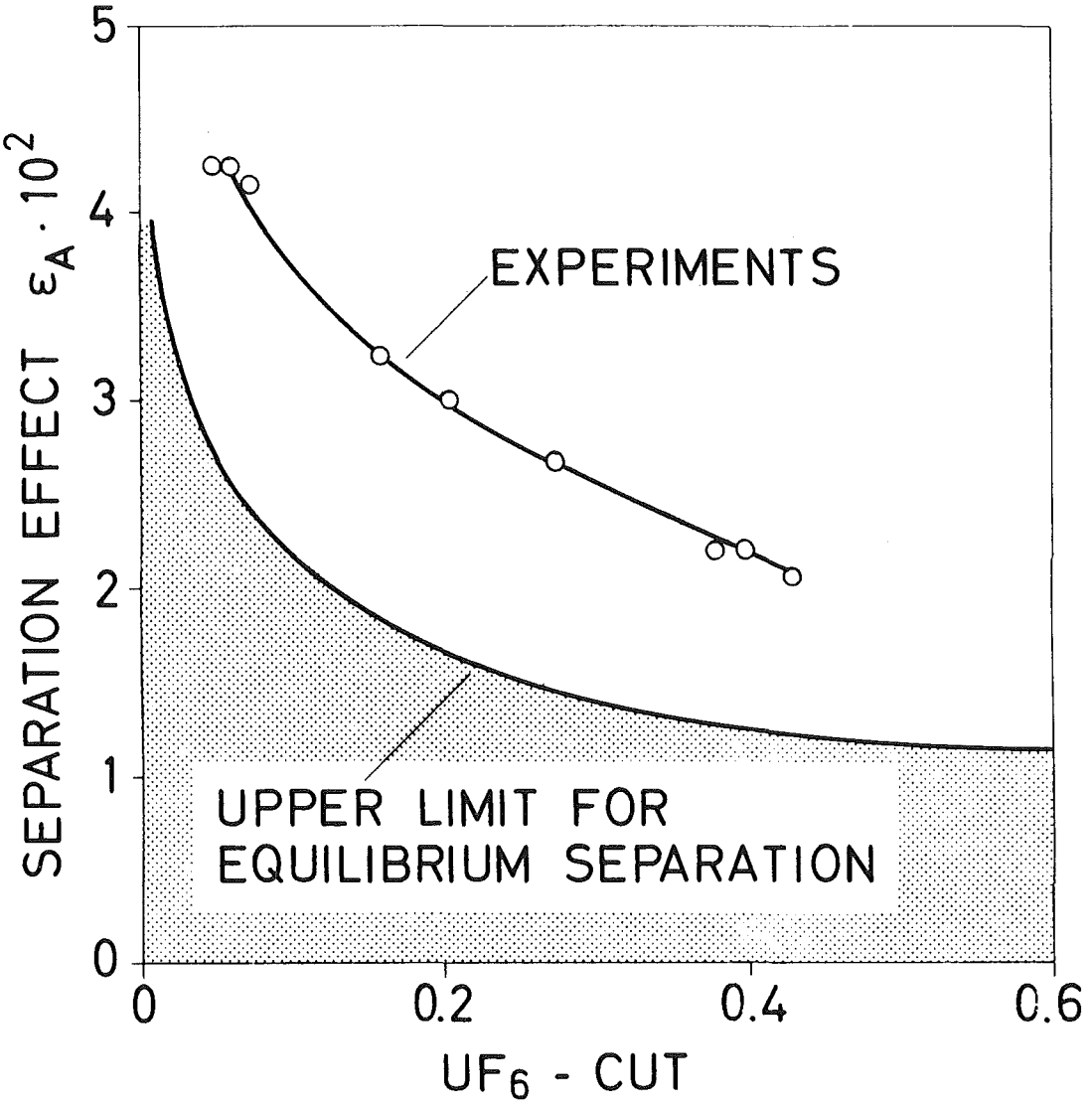

Fig. 10: The upper curves indicates the experimentally determined relation between the separation effect ε_A and the UF_6-cut ϑ (mixture of UF_6/H_2 with 1.6 mole-% UF_6; expansion ratio 8). The lower curve gives the upper limit for equilibrium separation calculated for infinite flow velocity.

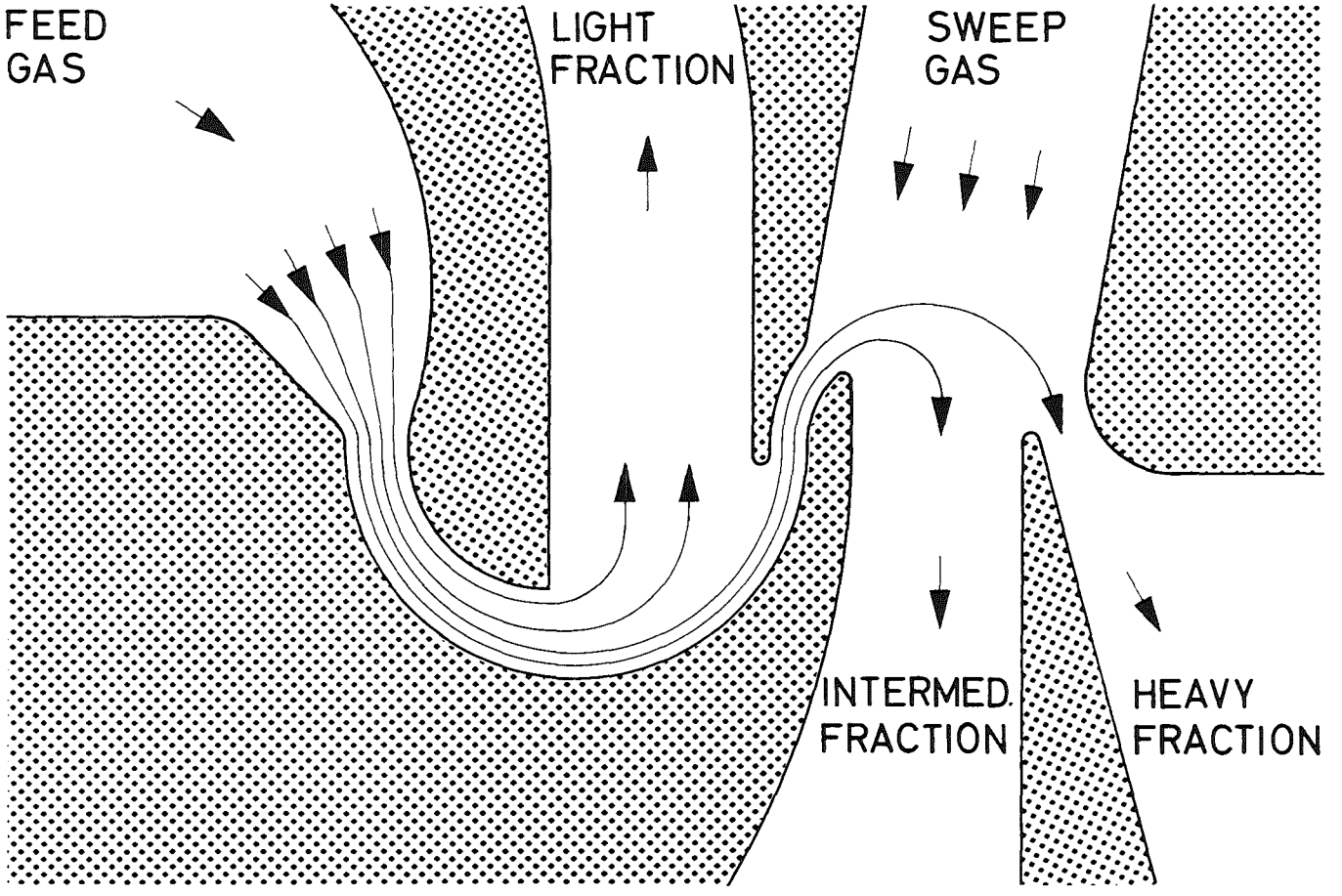

Fig. 11: Advanced separation nozzle system. The heavy fraction of the first nozzle is deflected in the second nozzle by a sweep gas.

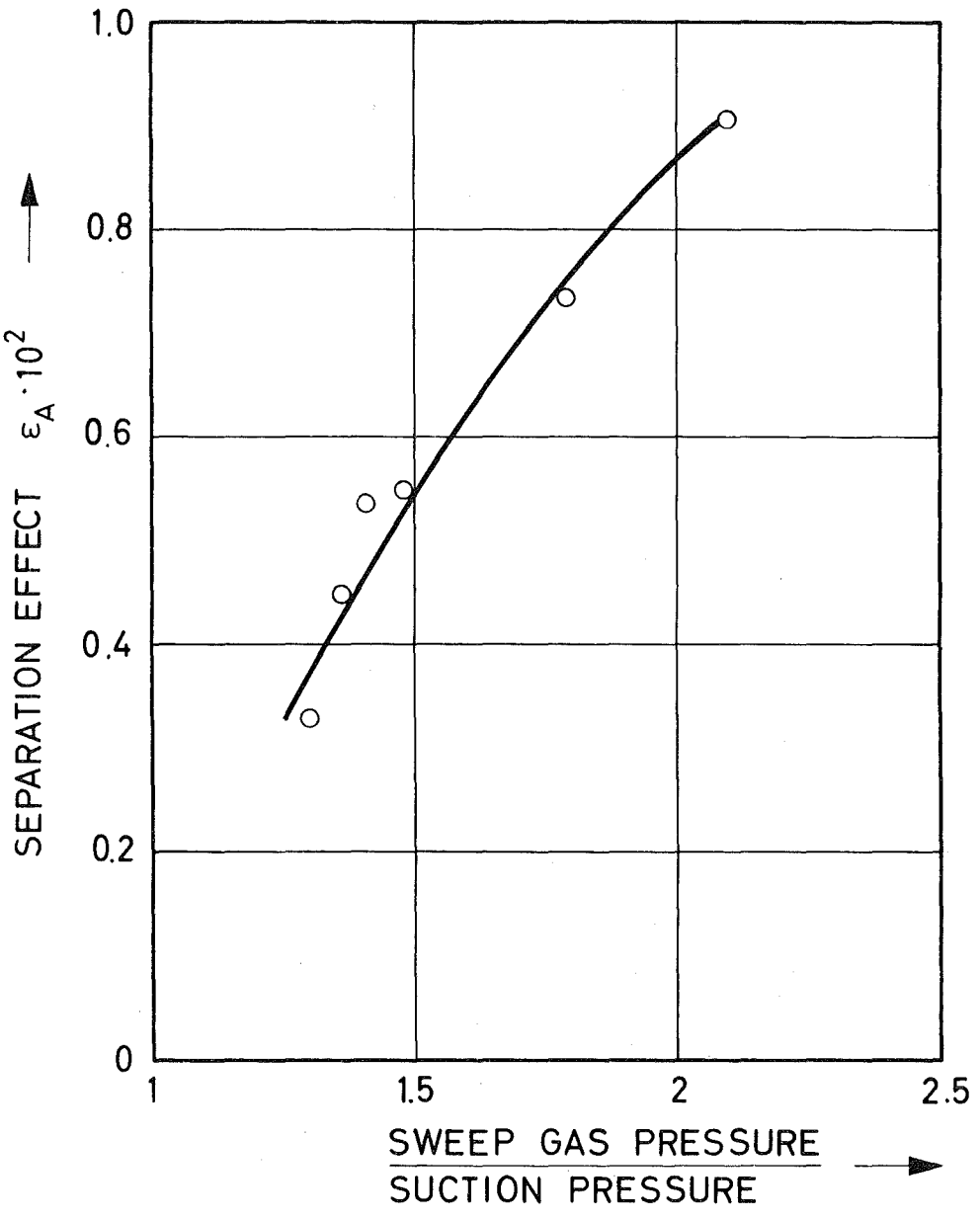

Fig. 12: Results of separation experiments in the case of flow deflection by a sweep gas.

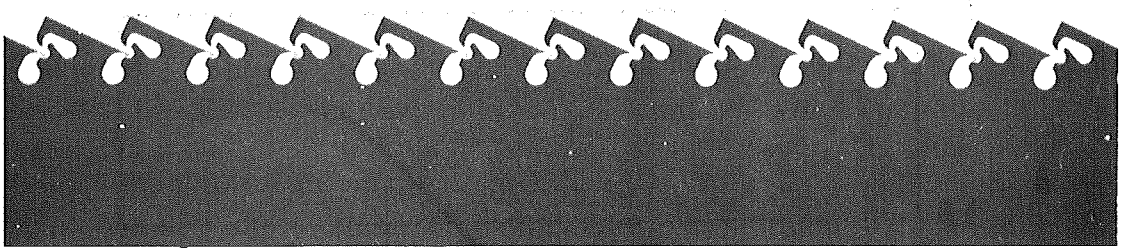

Fig. 13a: Metal foil with separation nozzle structures applied by means of photo-etching.

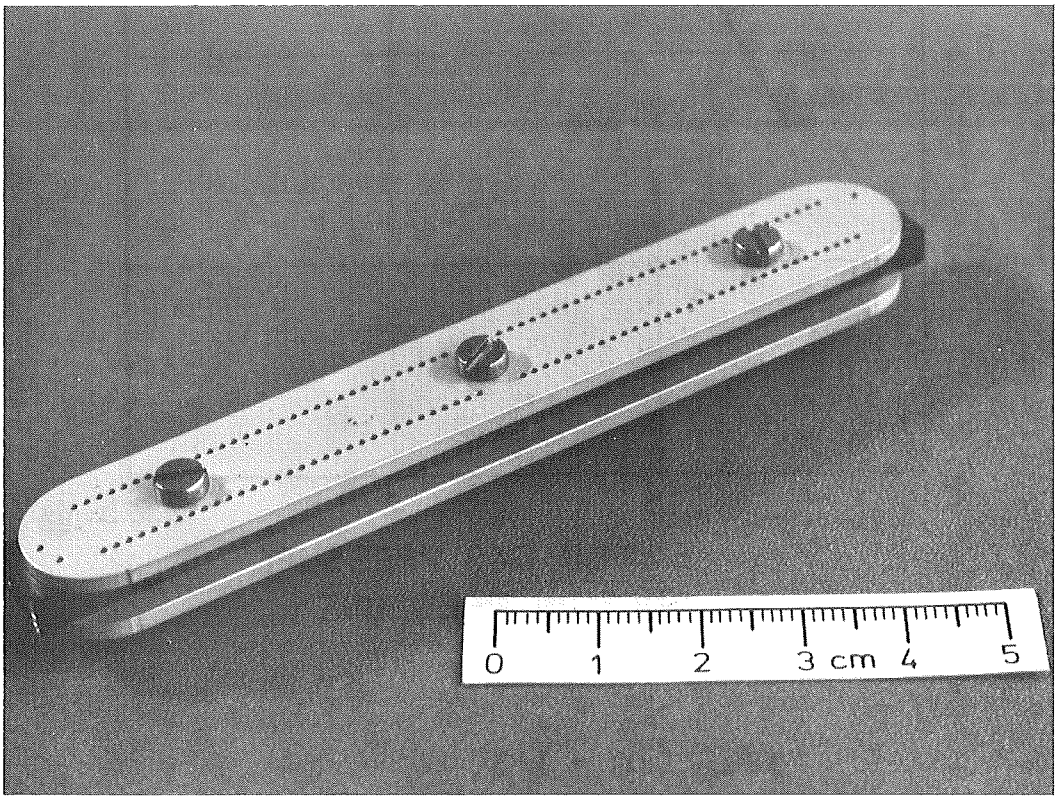

Fig. 13b: Separation element strip with cover plates.

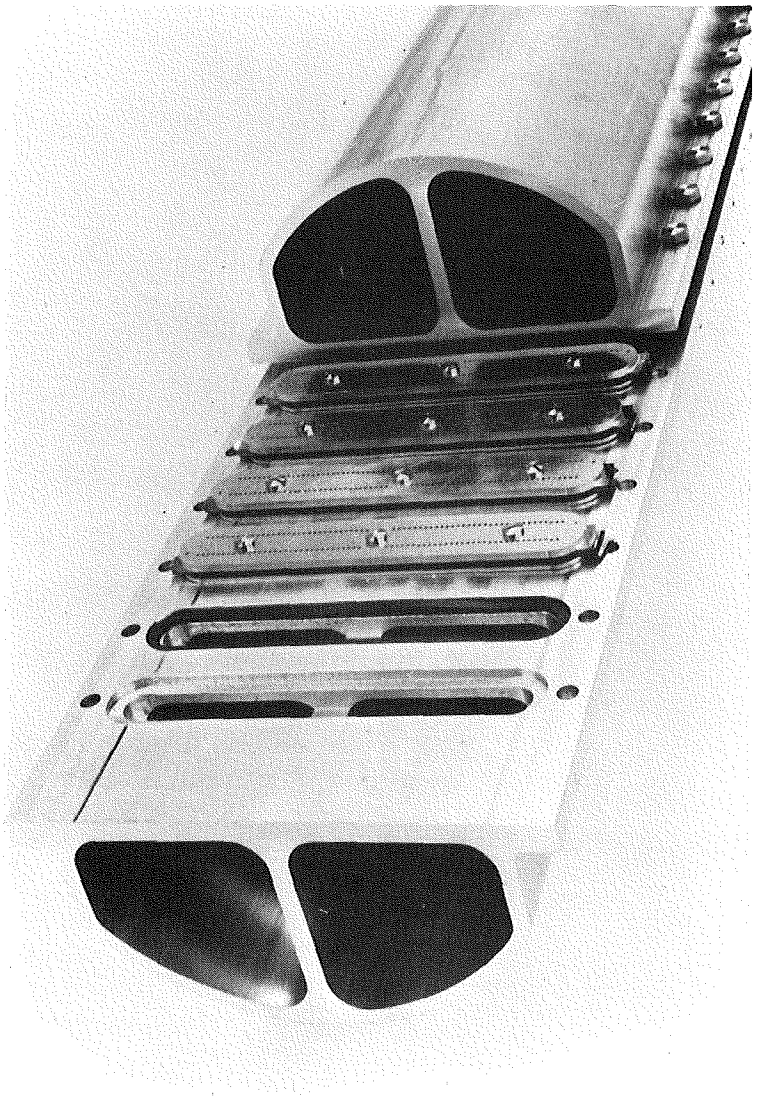

Fig. 14: Separation element tube with photo-etched separation nozzles.