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DEVELOPMENT AND TECHNICAL IMPLEMENTATION OF THE SEPARATION NOZZLE PROCESS FOR ENRICHMENT OF URANIUM 235

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Entwicklung und technische Realisierung des Trenndüsenverfahrens für die Anreicherung von Uran-235

Zusammenfassung:

Das Trenndüsenverfahren für die Anreicherung von Uran-235 wurde im Kernforschungszentrum Karlsruhe als Alternative zum Diffusions- und Zentrifugenverfahren entwickelt. Die Trennung der Uranisotope wird durch Ablenkung eines Strahls aus einem Uranhexafluorid/Wasserstoff-Gemisch erreicht. Seit 1970 ist die deutsche Gesellschaft STEAG an der technologischen Weiterentwicklung und der praktischen Anwendung des Verfahrens beteiligt. Ab 1975 wurde die Aktivität durch Beteiligung der brasilianischen Gesellschaft Nuclebras und der deutschen Gesellschaft Interatom weiter verstärkt. Hauptziele der gemeinsamen Arbeit sind zunächst der Bau einer Trenndüsendemonstrationsanlage mit einer jährlichen Kapazität von etwa 200 000 kg UTA und die Entwicklung von Komponenten für eine kommerzielle Anlage. Der Bericht beschreibt die wichtigsten Schritte bei der Entwicklung und technischen Anwendung des Verfahrens.

Summary:

The separation nozzle process for the enrichment of uranium-235 has been developed at the Karlsruhe Nuclear Research Center as an alternative to the gaseous diffusion and centrifuge process. The separation of uranium isotopes is achieved by the deflection of a jet of uranium hexafluoride mixed with hydrogen. Since 1970, the German company of STEAG, has been involved in the technological development and commercial implementation of the nozzle process. In 1975, the Brazilian company of NUCLEBRAS, and the German company of Interatom, joined the effort. The primary objective of the common activity is the construction of a separation nozzle demonstration plant with an annual capacity of about 200 000 SWU and the development of components of a commercial plant. The paper covers the most important steps in the development and the technical implementation of the process.

1. Introduction

The separation nozzle process for the enrichment of U-235 has been developed at the Karlsruhe Nuclear Research Center as an alternative to the gaseous diffusion and centrifuge processes (1 - 5). Isotope separation is achieved by the same basic mechanism as in the centrifuge method. However, the serious mechanical problems of highly stressed rotating machines are avoided, since, in the nozzle method, the separating centrifugal forces are generated by the deflection of a high speed jet of uranium hexafluoride and a light auxiliary gas.

Since 1970, the German company of STEAG, has been involved in the technological development and commercial implementation of the nozzle process (6). In 1975, the Brazilian company of NUCLEBRAS, and the German company of Interatom, joined the effort. The primary objective of the common activity is the construction of a separation nozzle demonstration plant with an annual capacity of about 200 000 SWU and the development of components of a commercial plant.

2. Basic Features of the Separation Nozzle Method

The basic features of the separation nozzle method can be understood from Fig. 1, which shows a cross section of the nozzle system used in the commercial implementation of the process. A jet of gaseous uranium hexafluoride mixed with hydrogen expands along a curved fixed wall. At the end of the deflection the flow is split into a lighter and a heavier fraction by means of a skimmer.

The hydrogen, which is present in a large molar excess, increases the flow velocity of the UF_6 and, hence, adds to the centrifugal force determining the separation.

This effect is illustrated in Fig 2, which shows the dependence of the centrifugal force on the hydrogen mole fraction. E.g. with an H_2/UF_6 mixture containing 5 mol-% UF_6 the centrifugal force acting upon the UF_6 molecules is nearly 20 times as high as with pure UF_6. A further positive influence on separation, exerted by the light gas, stems from the differing migration rates of the isotopes during establishment of the hypsometric density distribution (7). It appeared that the additional expenditure required to integrate the light auxiliary gas in the process is more than compensated by the advantages attainable in this way.

3. Optimum Operating Conditions

In order to achieve the enrichment required for reactor operation, the elementary uranium isotope separation effect supplied by the separating elements must be multiplied in a separation cascade. The gas expanded in the individual separation nozzle stages is recompressed before being fed to the separation nozzle systems of the next stages. The geometric parameters and the operating conditions of the separation nozzle systems, of course, must be set so that the separative work produced by the whole plant is as inexpensive as possible. A study of the cost structure of the method shows that, at the present state of development, the price per separative work unit is determined mainly by power costs and the capital cost fraction, which is proportional to the power consumption. Accordingly, the optimization can be carried out in a first approximation by finding the minimum of the specific ideal isothermal compression energy*): Fig. 3 shows the dependence of this figure on the expansion ratio and on the UF_6 mole fraction respectively. The minimum value of the compression energy accordingly will be reached at an expansion ratio of 2.1 and a UF_6 mole fraction of approximately 4%.

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*) The specific ideal isothermal compression energy is the ratio between the ideal isothermal compression energy to recompress the gas expanded in the separation nozzle and the separative work produced. See also (1).

The operating condidtions planned for the demonstration plant and the performance data they produce are summarized in Table 1:

UF_6/H_2 mixture with 4.2 mole % UF_6	
Inlet pressure p_o	0.26 bar
Expansion ratio π	2.1
UF_6 cut ϑ_U	0.25
Separation effect ε_A	1.48×10^{-2}

Table 1: Optimum operating conditions and performance data of the separation nozzle demonstration plant.

The results described in the table apply to a constant uranium cut ϑ_U = 25% which has turned out to be the economic optimum in a series of experiments. The corresponding cascade diagram is shown in Fig. 4. Fine adjustment of the uranium cut is possible in the individual separation stages by means of throttle valves installed in the suction lines of the heavy fractions.

4. Production of Separation Elements

To operate separation nozzles of the design shown in Fig. 1 at an inlet pressure of 0.26 bar (Tab. 1) the diameter of the deflection groove must be as small as 0.2 mm.

Several methods of inexpensive mass production of separation elements of such small characteristic dimensions have been worked out in cooperation with industry.

One method of producing separation nozzle elements by mechanical means has been developed by the German company of Messerschmitt-Bölkow-Blohm, Munich (4). Another method based on the stacking of photo-etched metal foils, introduced by the German company of Siemens, will be described in more detail:

Fig. 5a shows a metal foil along the edges of which a large number of separation nozzles are etched together with ducts for the feed gas and for the heavy fraction. Stacking such foils produces separation nozzle assemblies ("chips") with considerable packing densities (Fig. 5b). As shown in Fig. 5c, they are installed in tubes to one half of which feed gas is supplied while the other half is used to remove the heavy fraction. The light fraction (enriched in U-235) escapes to the outside of the tubes. Fig. 6 shows a separation nozzle structure produced by the etching technique. Considering that the radius of curvature of the deflection wall is only 1/10 mm, the picture demonstrates the accuracy of the fabrication technique.

The Siemens technique is particularly suited for the industrial scale production of separation nozzle configurations which are even more complicated than the version shown in Fig. 1 and which probably have to be realized in the course of long term process development (3). A largely automated production line for manufacturing photo-etched separation elements has been installed by Siemens (Fig. 7). At present its capacity is about 2500 m of slit length per annum.

5. Test Operation of Industrial Equipment

Tests of large batches of mass produced separation nozzle elements and of the equipment needed for commercial implementation of the process are performed in two prototype separation stages operated by Gesellschaft für Kernforschung (8)

and the NUSTEP company (6). The main stage components are a separation element tank, a two-stage radial compressor and a gas cooler. Fig. 8 shows the separation elements being loaded into the so-called small stage. As can be seen, the separation elements are arranged as a compact unit for easy installation. Fig. 9 shows the small stage ready for operation.
One target of test operations has been to work out the most economical methods of assembling the components of a commercial separation nozzle plant. Up to now, no "clean conditions" have been applied. Instead, dust particles present in the process loops have been collected in a filter during test runs prior to insertion of the separation elements. The typical test operation of a stage is shown in Fig. 10: Obviously the separation capacity is constant within the limits of error.

6. Cascade Design and UF_6-Recycling

Fig. 11 shows a schematic representation of an industrial separation nozzle cascade with two types of separation stages. The use of two types of stages with a 1:3 ratio of flows makes the performance approach that of a corresponding ideal cascade with an efficiency of approximately 90%. A total of about 500 stages must be connected in series in order to produce enriched uranium containing 3% U-235 and to strip to some 0.3% the U-235 content in the waste.

A separation nozzle cascade produces a net upward transport of the light auxiliary gas, which is of the order of the stage throughput. To prevent enrichment of light gas in the cascade, the upward transport has to be extracted from the top and recycled to the bottom of a section.
For this purpose, the light fractions of the top stages are processed in the so-called UF_6-recycling facilities shown in Fig. 11. Here, the UF_6-content of the upward flow is stripped off with high efficiency and fed back to the top stage feed flows.

The flowsheet of such a facility includes a special separation nozzle stage backed by a low temperature freeze-out heat exchanger system (9). As shown in Fig. 12, the heat exchangers include additional passages for the coolant flow by which the temperature profiles can be controlled properly. The UF_6 desublimes in relatively large passages equipped with serrated fins of the multi-entry type.

Experiments performed on UF_6/H_2 mixtures under process conditions have demonstrated that even with the highest temperature differences achievable between the gas stream and the heat exchanger wall no undesired desublimation of UF_6 in the gas phase has been detectable. This made it possible to run the heat exchanger with a steep temperature gradient as a function of time, as is shown in Fig. 13. By proper shifting of the temperature profile along the heat exchanger, the UF_6 is deposited in a layer of constant thickness. In this way, a high freeze-out capacity is achieved with a minimum pressure drop. A UF_6-content of the light gas well below 1 ppm is easily achieved. Accordingly, only negligible losses of separative work are associated with recycling of the light gas from the top to the bottom of the cascade sections.

7. Stabilizing the Operating Conditions along the Cascade

In the separation nozzle method, a high degree of stability is reached for the pressure distribution along the cascade: Local differences in the nozzle inlet pressure give rise to major local changes of the relatively pronounced upward transport of the light auxiliary gas in the sense of stabilization. Things are more complex with the UF_6 concentration along the cascade:

A ten stage pilot plant was built to work on these problems (10) and served for experimental studies both of the steady state and the dynamic control behavior of separa-

tion nozzle cascades (11). The UF_6 distribution was found to assume an inherently stable state under all operating conditions studied in the plant (Fig. 14). Without any active control measures the full anticipated multiplication of the elementary effect of uranium isotope separation was found (Fig. 14 lower diagram).

A theoretical analysis of the stability behavior clearly indicates that the stabilization of the UF_6 concentration is determined primarily by the dependence of the UF_6 mole fraction on the UF_6-cut. The separation stage can be made to react to an increase in concentration by raising the UF_6-cut so that any excess UF_6 is transported in the direction of the UF_6 recycling system which acts as a UF_6 buffer (12). This can be reached by proper choice of the characteristics and operating points of the plant components.

8. Economics

To obtain reliable cost data on the separation nozzle process, a number of industries were commissioned to develop design and fabrication methods for the most important plant components under the requirements of mass production. These orders, among other items, concerned the separation elements, compressors, cooling system, piping, tanks, and valves. On the basis of the costs determined in this way, detailed optimization studies were performed on the design and operation of industrial-scale separation nozzle plants (6).

These studies as well as the development work performed to date on the separation nozzle technology have provided a safe basis for analyzing the economics of the process. Cost evaluations corresponding to the present development status result in specific investment costs which qualify the separation nozzle process as an economically attractive technique. At the present state of development, the specific power consumption of the separation nozzle process corresponds approximately to that of the existing U.S. gaseous diffusion plants.

9. Future Development

Fig. 15 shows the organization for the commercial implementation of the separation nozzle method. It appears from the chart that the demonstration plant to be erected in Brazil will be owned by the firm NUCLEI. It is composed of the Brazilian firm NUCLEBRAS and the German firms STEAG and INTERATOM, the industrial architect being the STEAG-INTERATOM consortium.

The German-Brazilian company NUSTEP will be responsible for granting licenses for the method as well as for preparatory work preceding the construction of a commercial plant in Brazil. It will perform a so-called Technology Program which will involve the development and testing of components to be used in the commercial plant.

The future development of the method will be the task of the Karlsruhe Nuclear Research Center where the separation nozzle method has had its origin.

It is planned to start construction work with the demonstration plant next year. The last figure (Fig. 16) shows the construction site of the so-called technology building where the components for the commercial plant will be developed by NUSTEP. The decision for the construction of the commercial facility is anticipated for 1982. According to the provisional planning for this facility a capacity of 2 mill. SWU/a will be provided.

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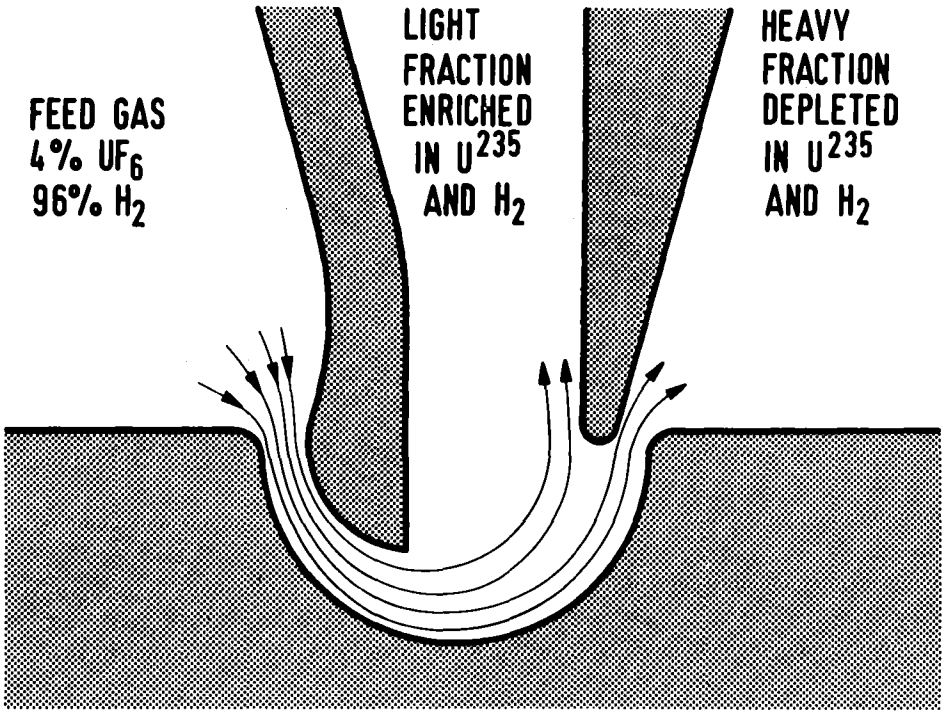

Fig. 1 Cross section of the separation nozzle system used in the commercial implementation of the process

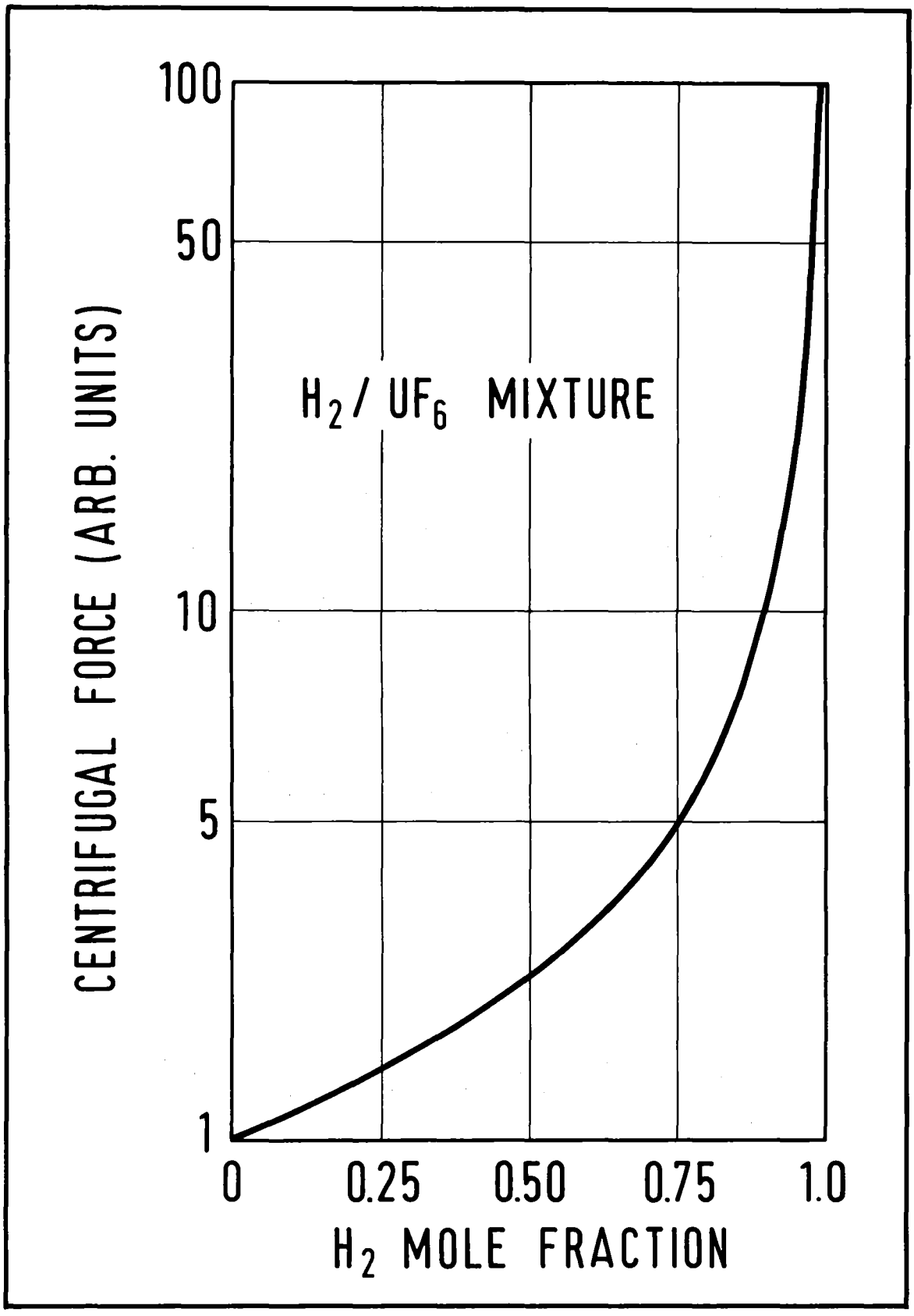

Fig. 2 Dependence of the centrifugal force acting upon the UF_6 molecules on the H_2 mole fraction

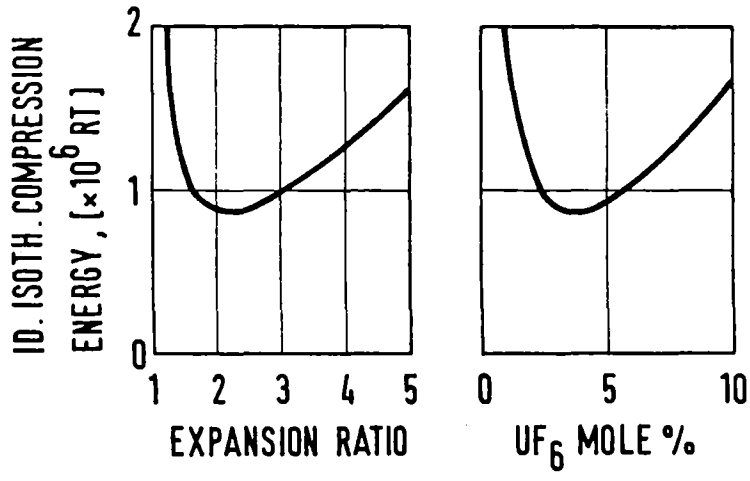

Fig. 3 Dependence of the specific ideal isothermal compression energy on the expansion ratio (left side) and on the UF_6 mole fraction (right side). Mixture of UF_6/H_2; UF_6-cut = 0.25

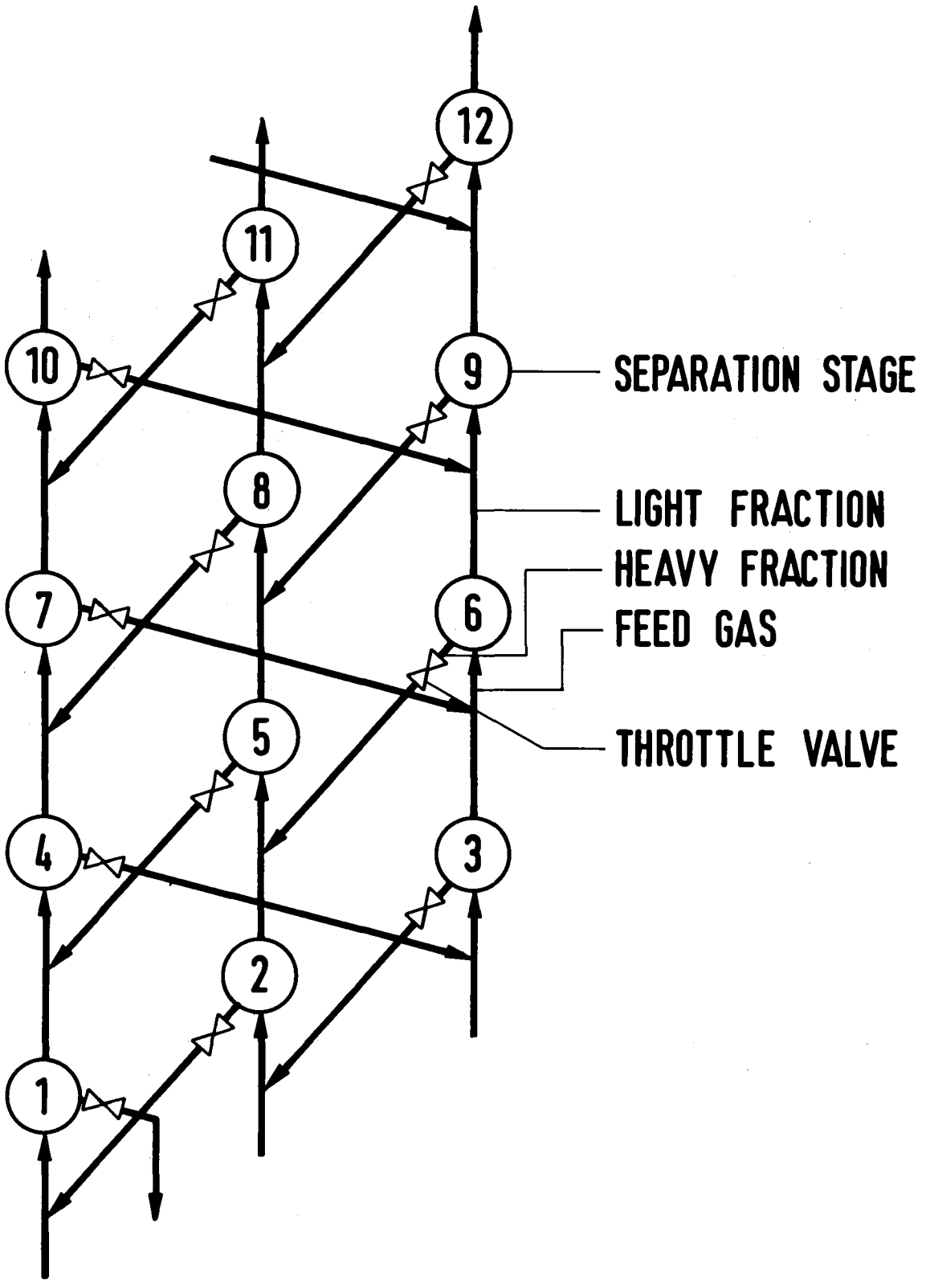

Fig. 4 Cascade flowsheed for a UF_6-cut of 0.25

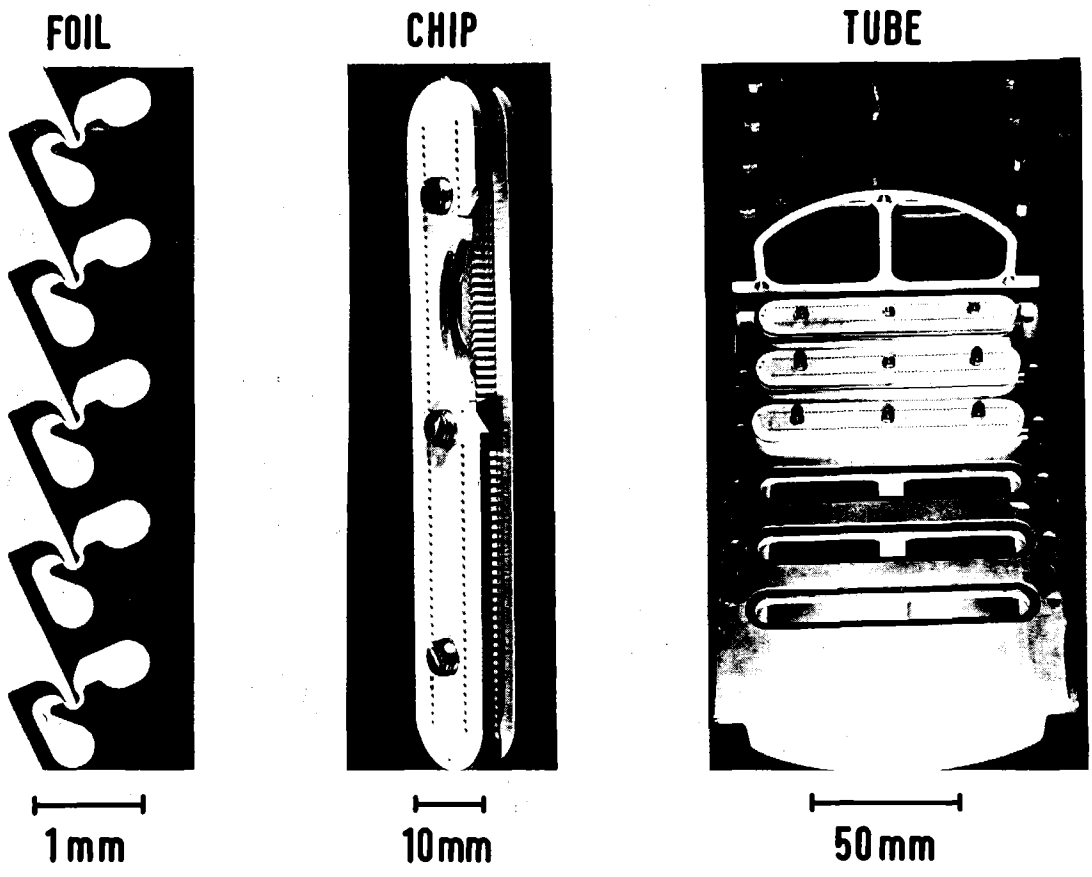

Fig. 5 The assembly of a commercial separation element manufactured by means of photo-etching (Siemens company, Munich)

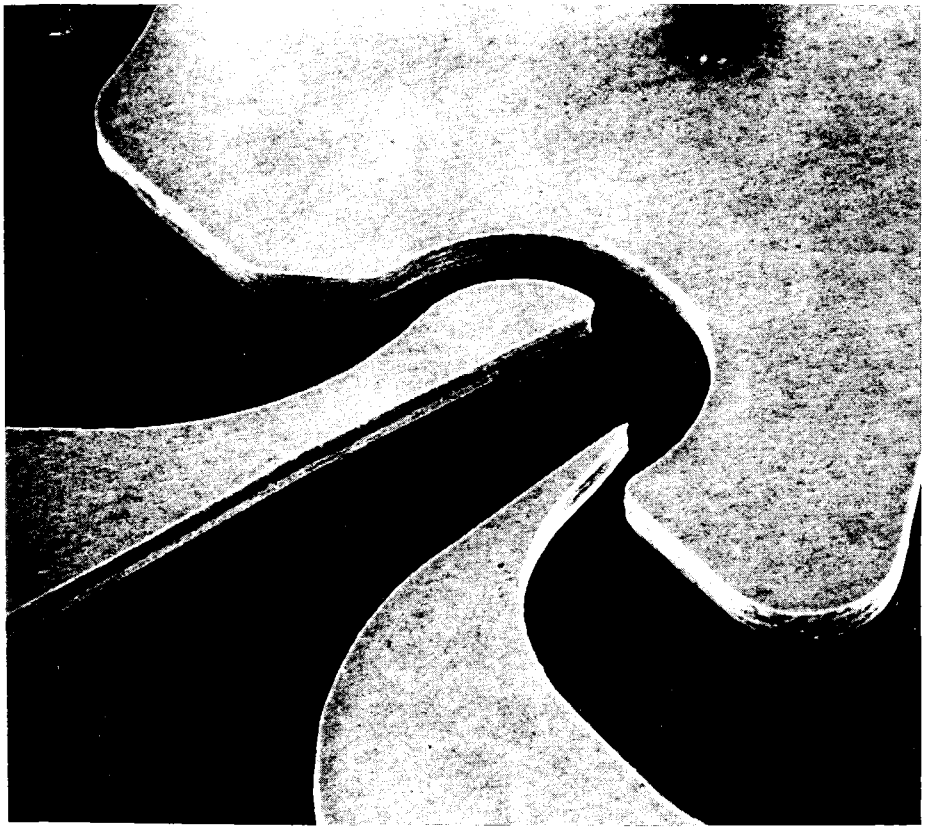

Fig. 6 Separation nozzle structure produced by the photo-etching technique (Siemens company, Munich)

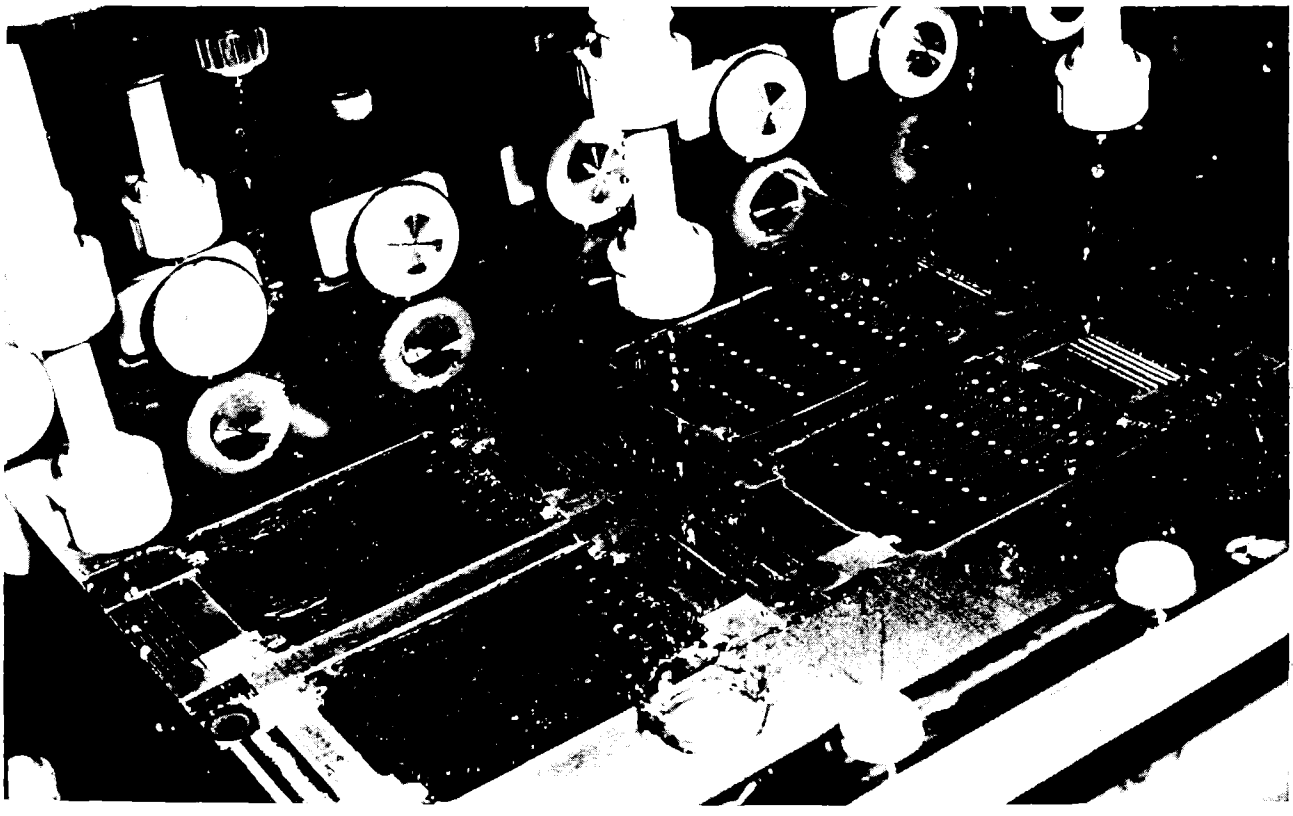

Fig. 7 Photo-etching device as part of the automated production line for manufacturing separation nozzle elements (Siemens company, Munich)

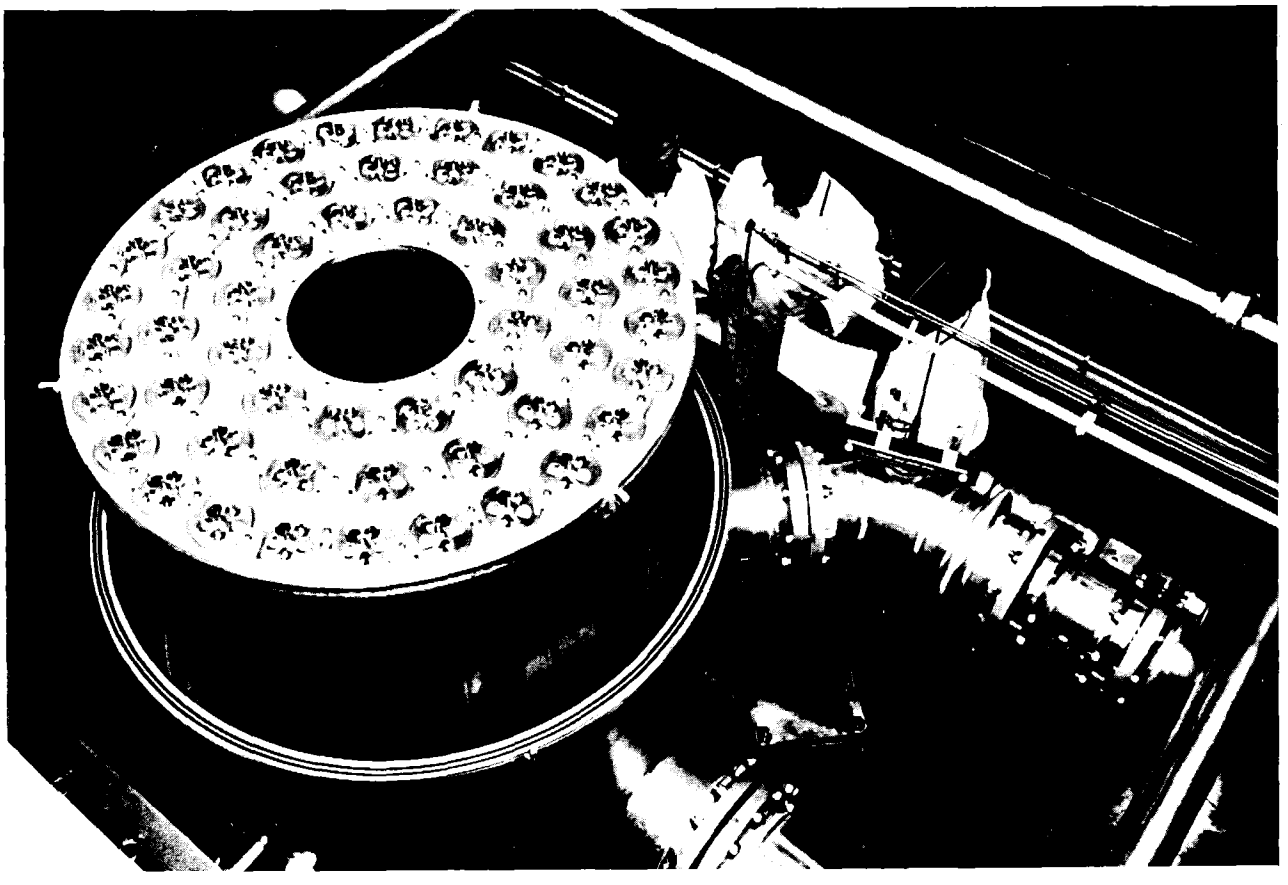

Fig. 8 Loading of the separation nozzle elements into the small separation stage. The separation elements are arranged as a compact unit to allow easy installation

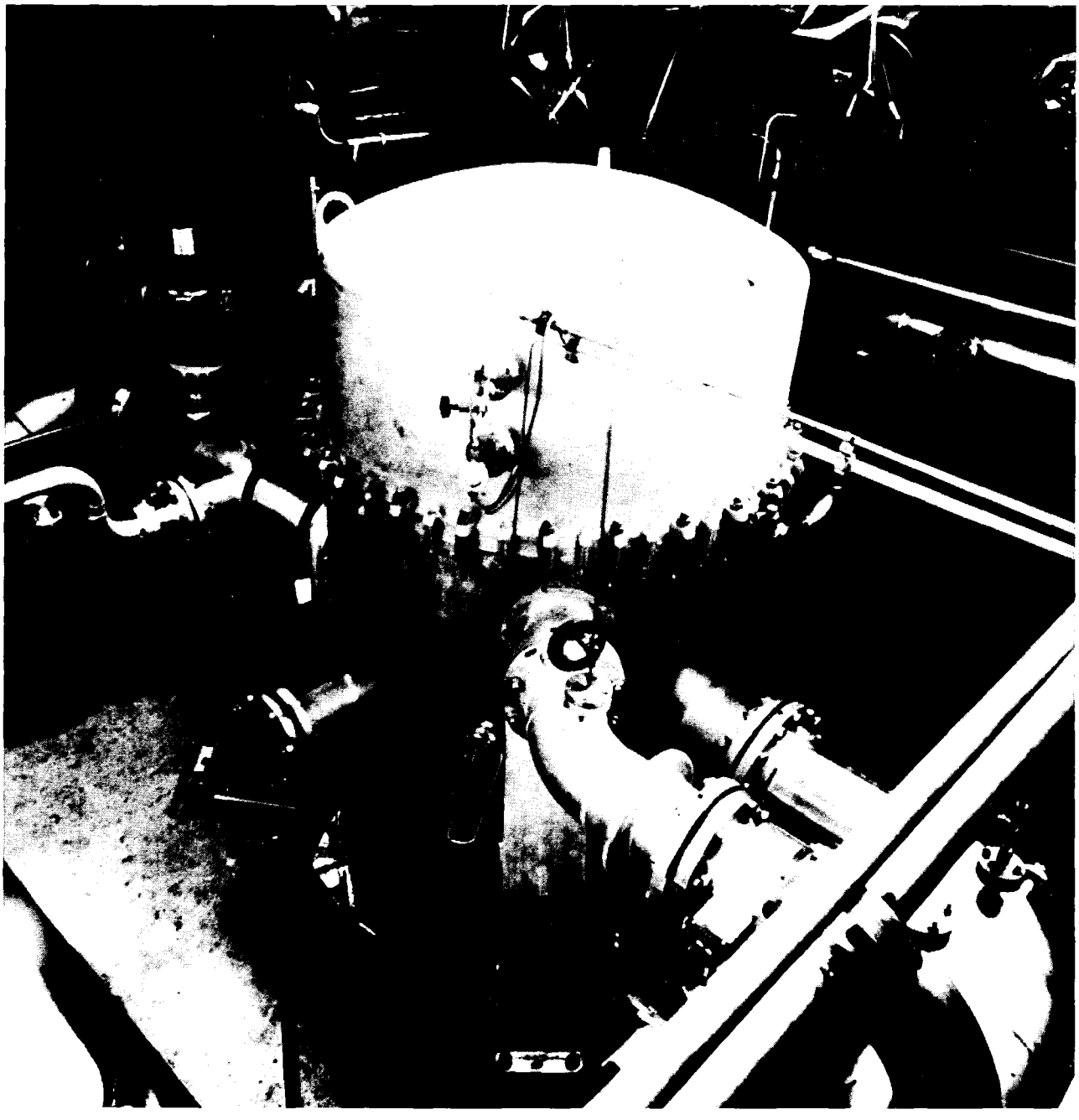

Fig. 9 Small separation nozzle stage ready for operation

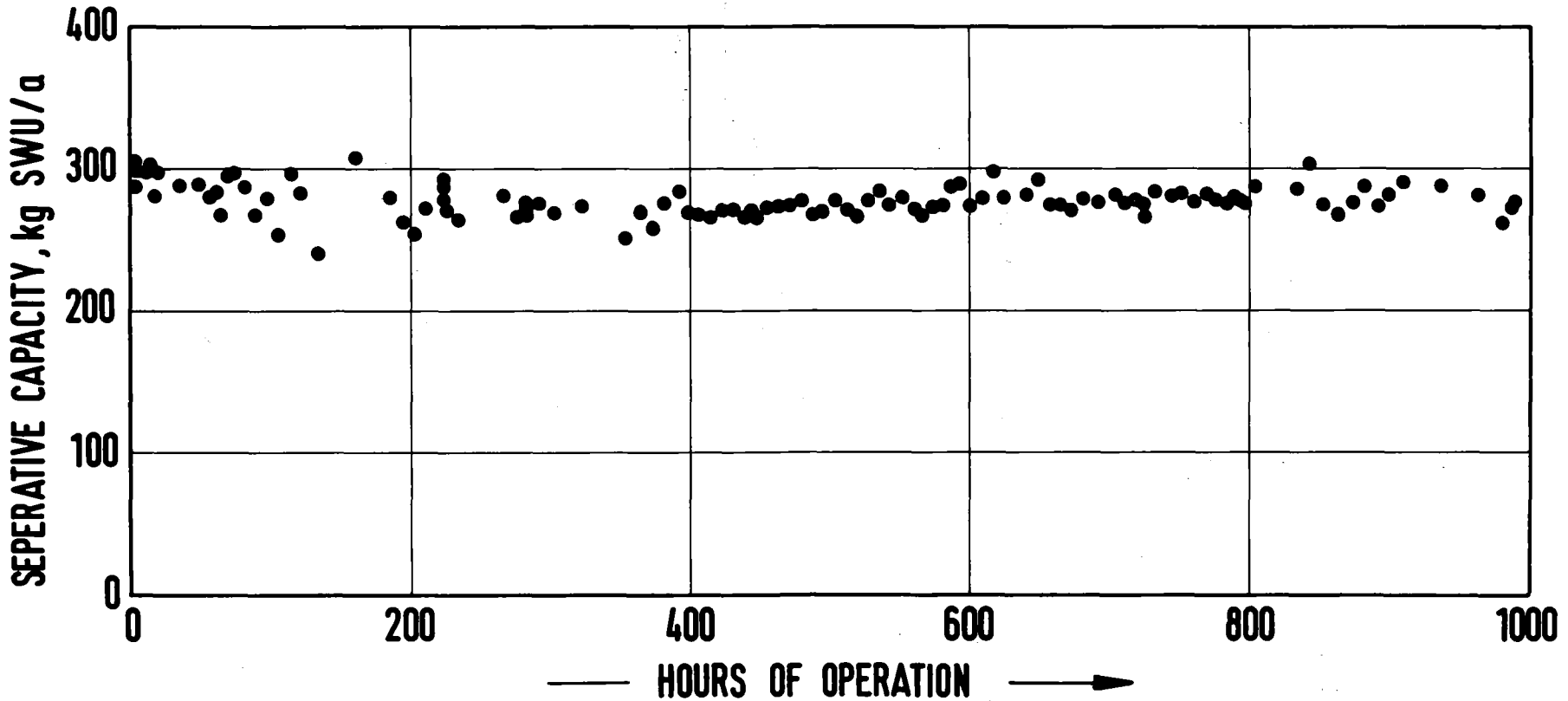

Fig. 10 Separation capacity versus time of the small separation nozzle stage

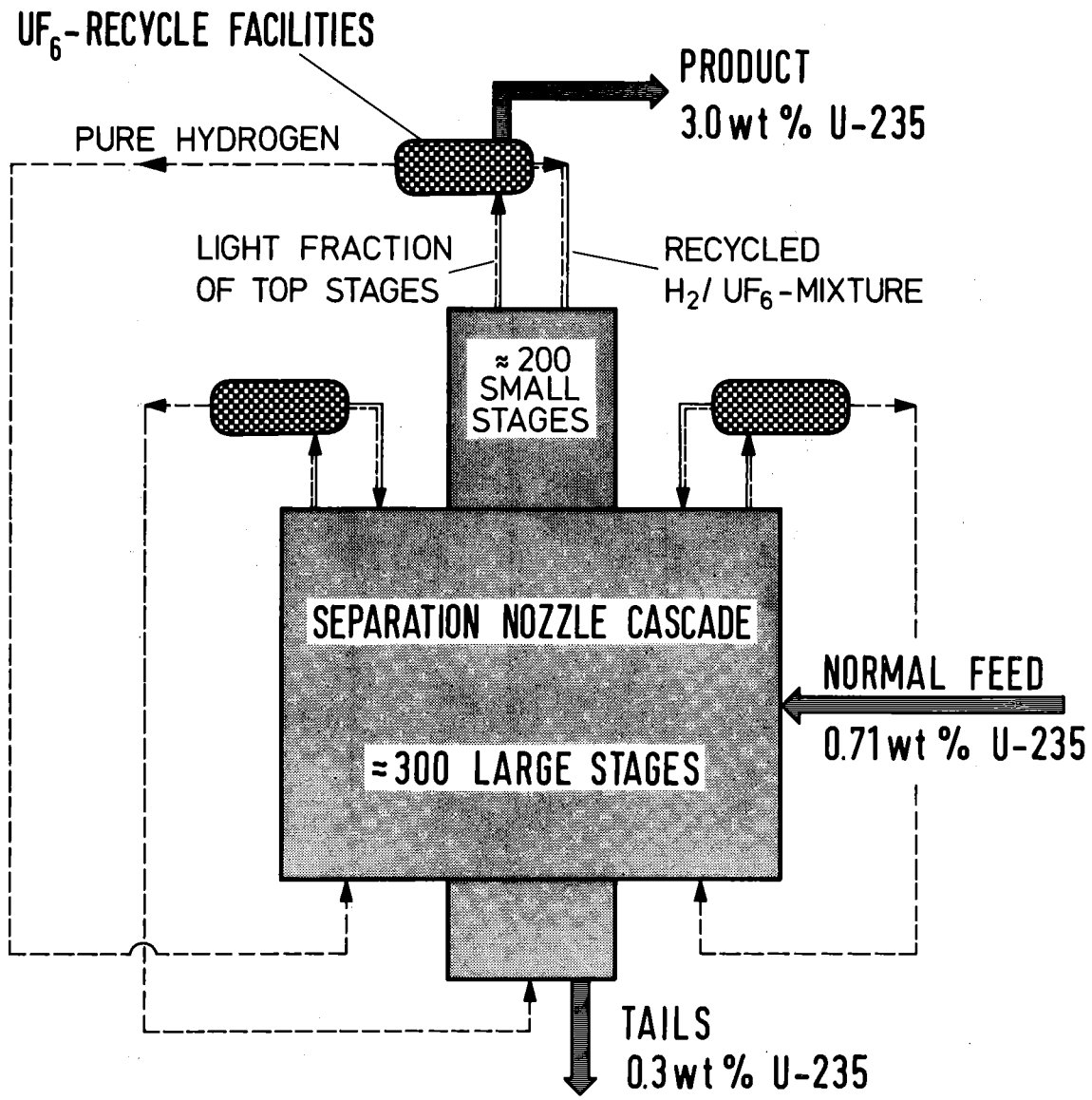

Fig. 11 Schematic representation of a separation nozzle cascade with UF_6 recycle facilities

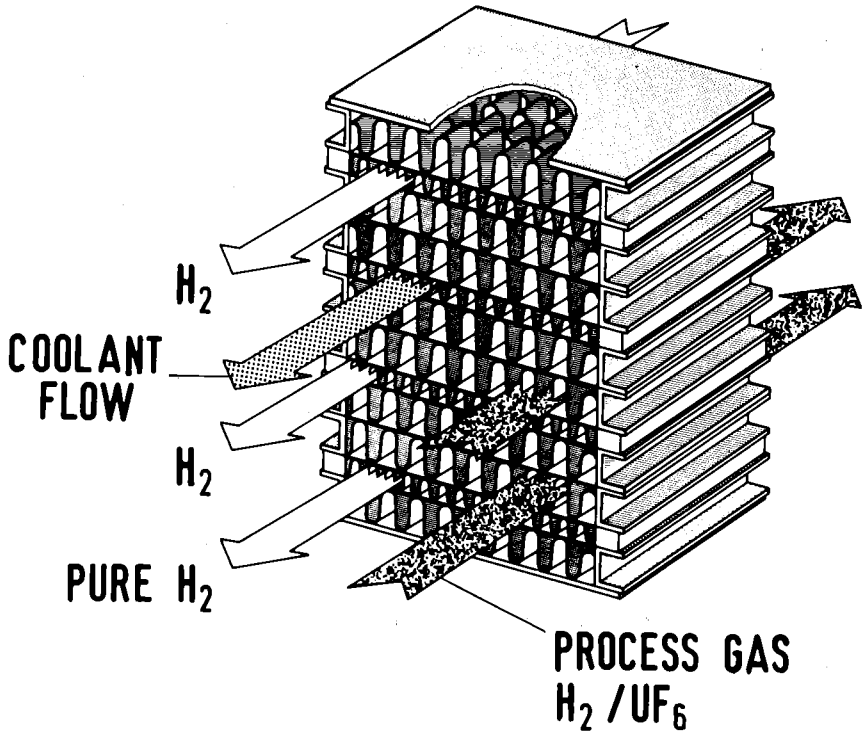

Fig. 12 Compact freeze out heat exchanger with an additional passage for the coolant flow

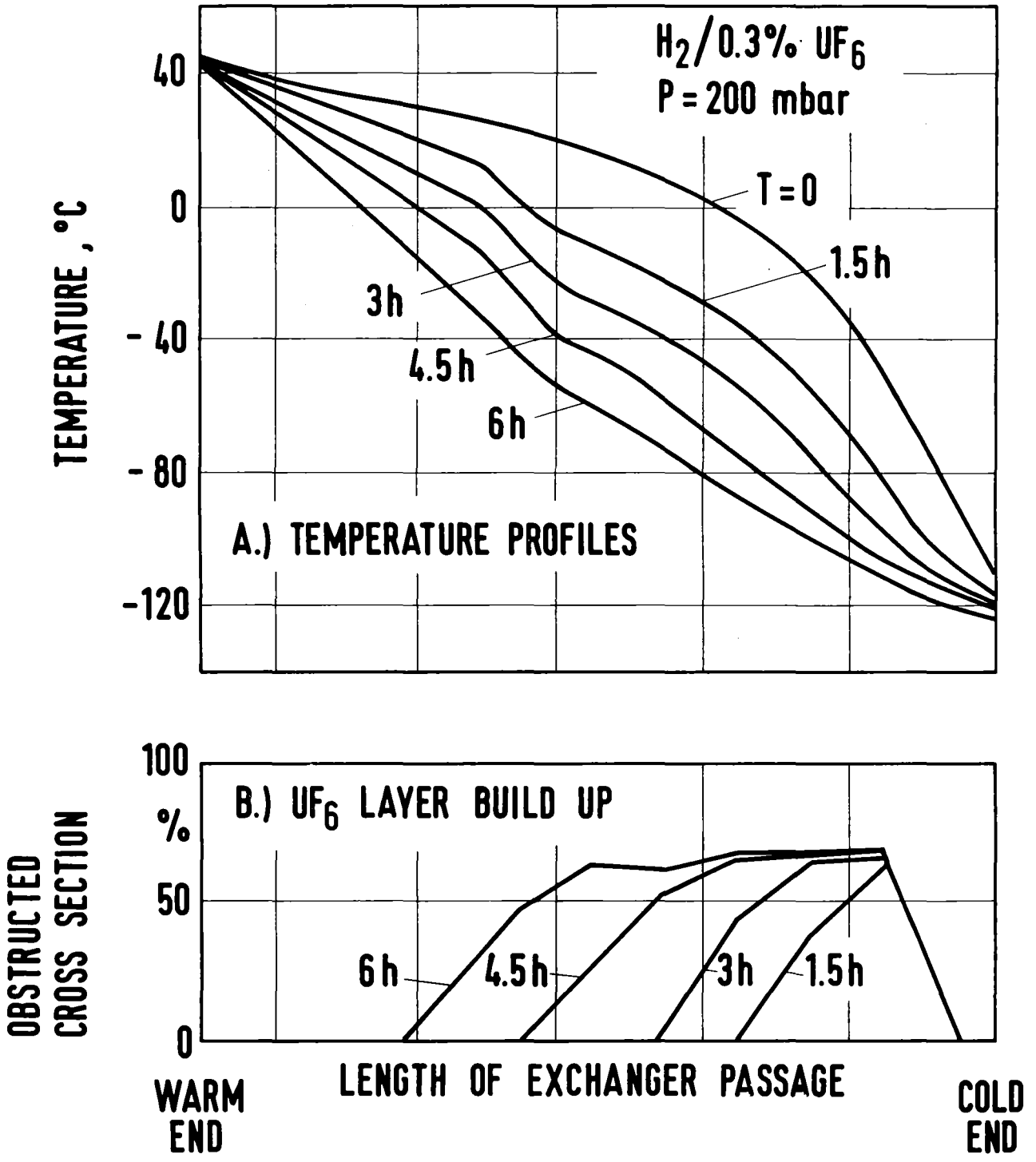

Fig. 13 Time dependence of the temperature profile (A) along the heat exchanger of a UF_6 recycle facility and resulting UF_6 layer (B)

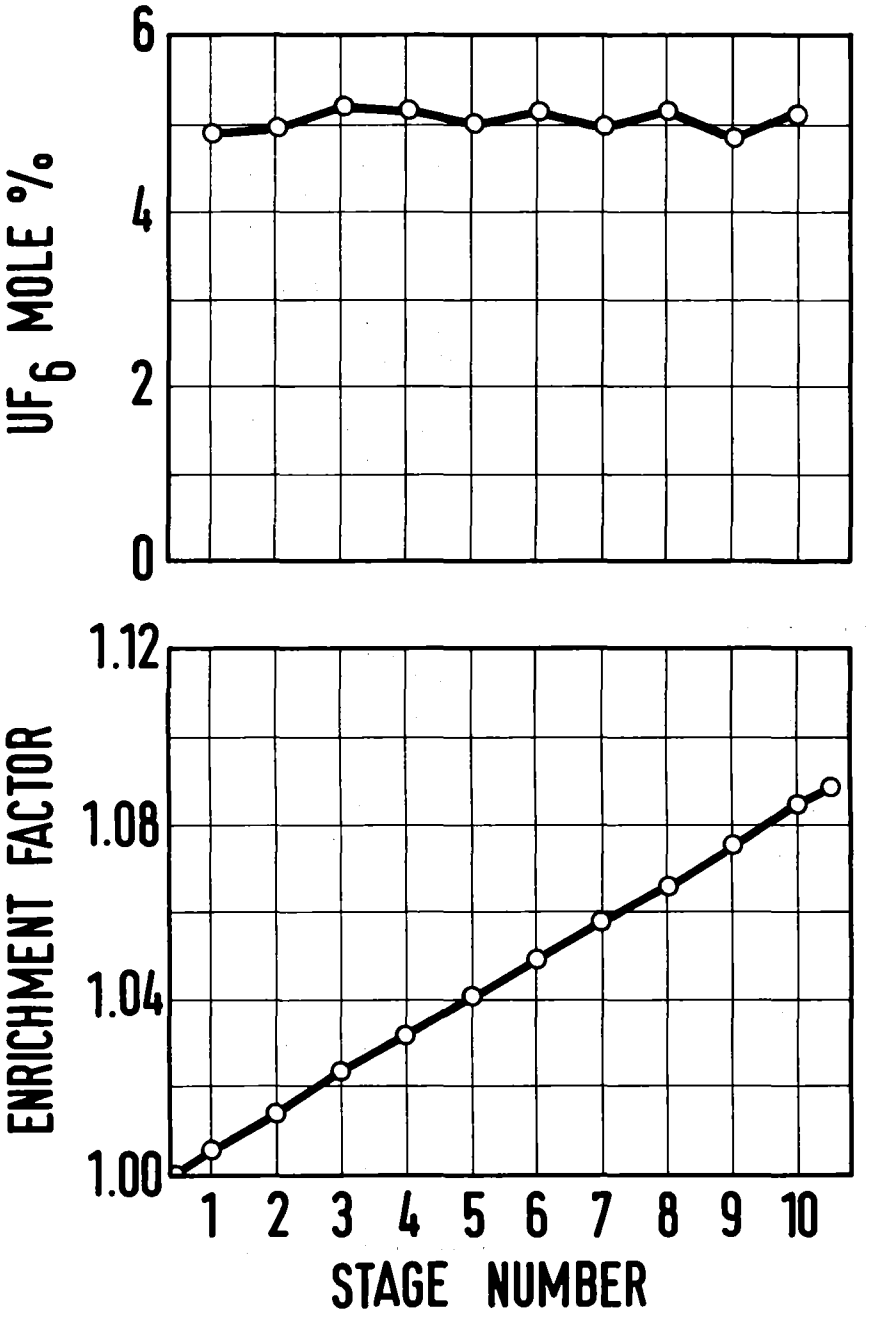

Fig. 14 Upper diagram: UF_6 mole fraction of the stages of the pilot plant, showing nearly uniform UF_6 content along the cascade
Lower diagram: Experimental enrichment factor of Uranium 235 relative to the heavy fraction of stage 1

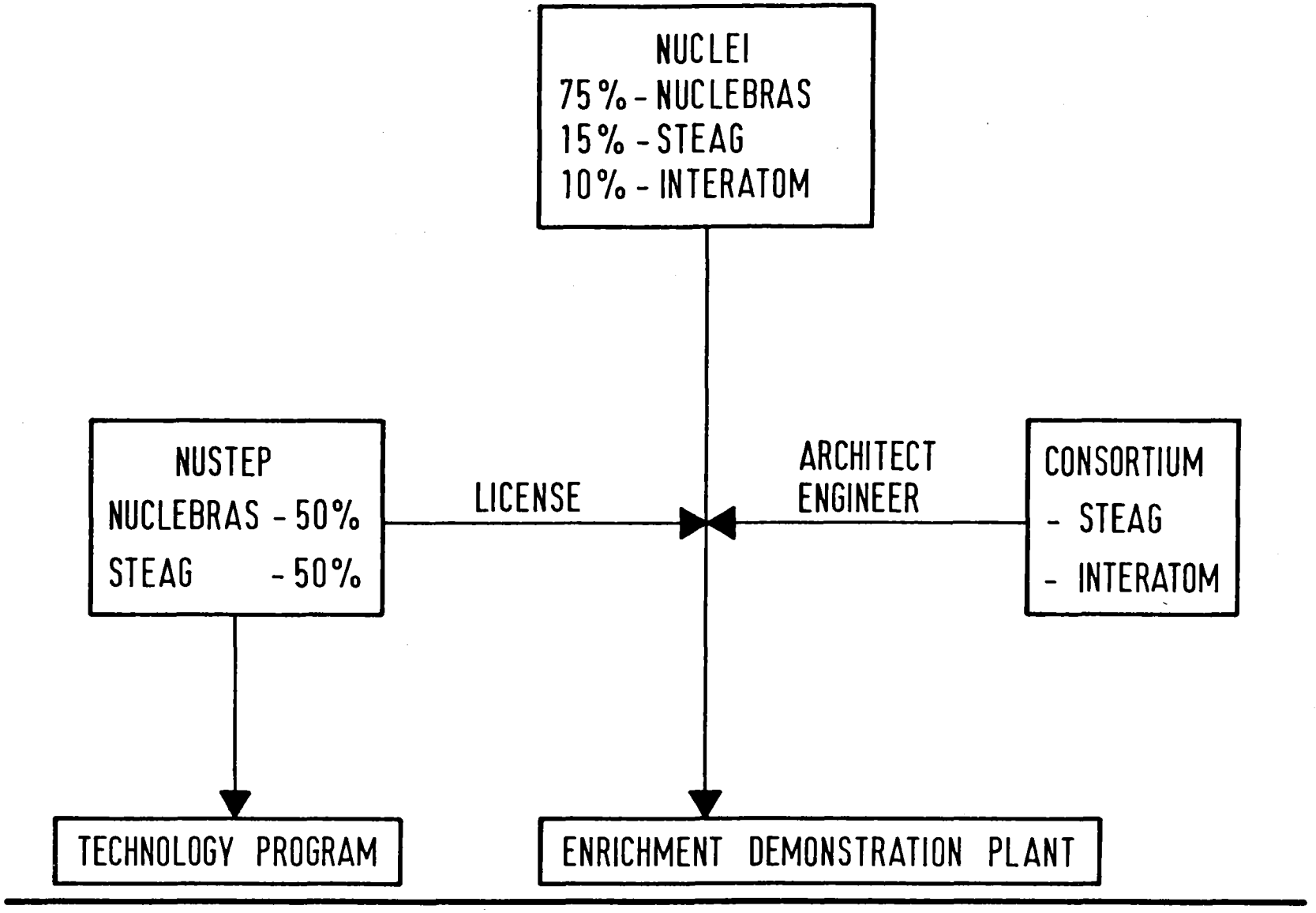

Fig. 15 Organization for the commercial Implementation of the separation nozzle process

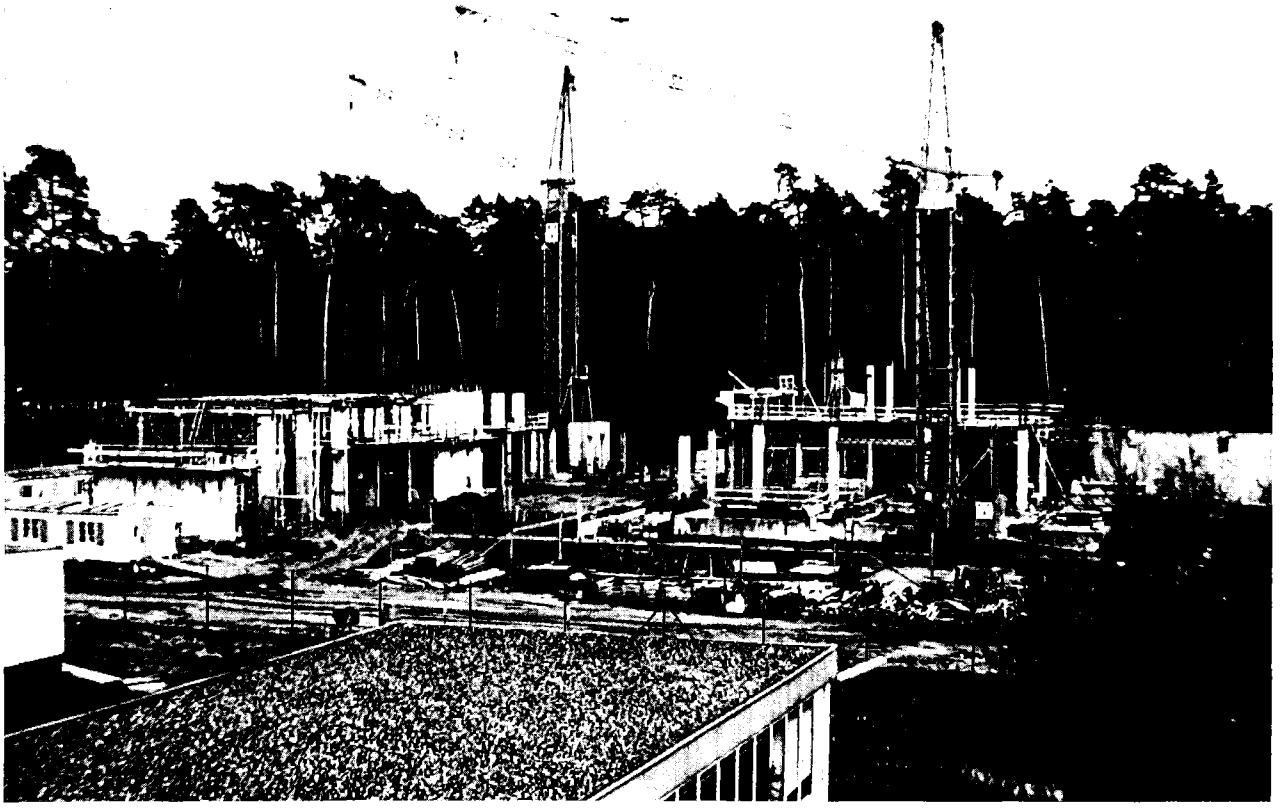

Fig. 16 Construction site of the building for the Technology Program