Production of Separation-Nozzle Systems for Uranium Enrichment by a Combination of X-Ray Lithography and Galvanoplastics

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X-ray lithography using synchrotron radiation has been applied in a multi-step process for the production of plastic moulds to be used in the fabrication of separation nozzles by electrodeposition. For characteristic dimensions of a few microns a total height of the nozzle structure of about 400 μm has been achieved. Structural details of about 0.1 μm are being reproduced across the total thickness of the polymer layer. The surface finish of metallic separation nozzles produced by electrodeposition was equivalent to the high quality of the polymer surface. The separation-nozzle systems fabricated by the described method allow an increase by a factor three of the gas pressure in separation-nozzle plants as compared to the present standard. This results in considerable savings in the enrichment of ^{235}U for nuclear power production.

For the production of fuel elements for light-water reactors the uranium isotope ^{235}U has to be enriched from its natural abundance of 0.7% to about 3%. Besides the gaseous diffusion and centrifuge processes, the separation-nozzle method, developed by the Karlsruhe Nuclear Research Center, has qualified for this task [1]. It is based on partial separation of the uranium isotopes in a curved jet consisting of a mixture of UF_6 and a light auxiliary gas. The method will be applied on a technical scale under the Nuclear Power Agreement concluded between the Federal Republic of Germany and Brazil with the consent of the International Atomic Energy Agency [2].
Figure 1 shows a schematic representation of a separation-nozzle stage with double deflecting separation-nozzle systems, which is planned to be used in the commercial application of the process. In the first curved nozzle of the double deflecting system, the flow is split into two fractions. The light fraction is extracted from the separation stage while the remainder flows into the second curved nozzle. There it is split into an intermediate and a heavy fraction. The intermediate fraction is recycled to the intake of the compressor while the heavy fraction is transferred to a lower stage of the separation cascade.

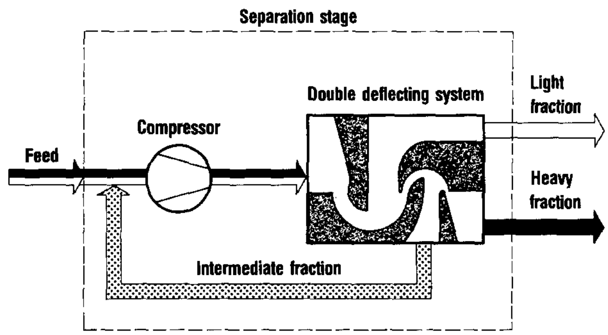

Fig. 1. Principle of a separation-nozzle stage with double deflecting separation-nozzle systems

The optimum operating pressure of a separation nozzle is inversely proportional to its characteristic dimensions. For reasons of economy, the operating pressure should be as high as possible. Therefore, the characteristic dimensions are chosen as small as permissible in regard of the tolerances of the manufacturing method. The methods presently available for the mass production of separation-nozzle systems rely either on machining with diamond tools (Messerschmitt-Bölkow-Blohm GmbH, Munich) or on stacking of photo-etched metal foils (Siemens AG, Munich). For these methods the mean radius of the deflection grooves is about 50 μm. The corresponding inlet pressure of the nozzle system is about 0.5 bar.

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Since the UF_6 is highly diluted by the light auxiliary gas, the separation nozzle can be operated at an inlet pressure of about 1.5 bar without the risk of UF_6 condensation. At this pressure level, the optimal radii of the first and second deflection groove are about 20 and 10 µm, respectively. The distance of the wedge-shaped skimmer from the deflection groove is about 3 µm.

Separation-nozzle systems with such extremely small characteristic dimensions can hardly be fabricated by the methods available at present. Therefore, a new fabrication method has been developed. It relies on *li*thographic generation of separation-nozzle structures in polymer layers which serve as moulds for *ga*lvanoplastic fabrication of metallic separation nozzles ("LIGA" method). Most favourable results have been achieved by X-ray lithography using synchrotron radiation which is superior by its high intensity and small divergence*.

The status of the development work on the LIGA method will be briefly described below.

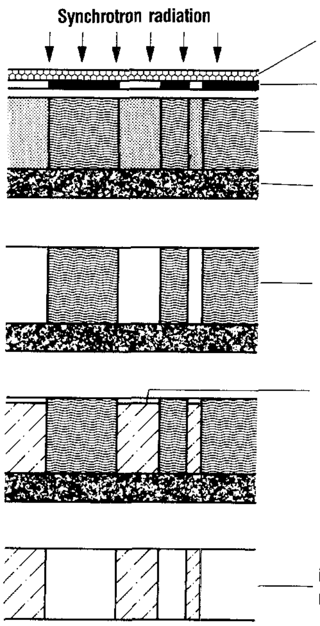

Fig. 2. Schematic representation of the LIGA process for production of separation-nozzle systems

Single-Step Processing of X-Ray Resist

First experiments on the various manufacturing steps were performed according to the schematic representation shown in Fig. 2.

The X-ray resist was a calandered foil consisting of polymethyl methacrylate (PMMA). The foil had a thickness of 100 µm and was laminated upon a metallic substrate using cyanoacrylate as an adhesive. The mask used in the lithographic process was made from polyimide with a 4 µm thick gold absorbing pattern. The characteristic wavelength of the synchrotron radiation was 0.2 nm. The irradiation dose was some 1000 J/cm^3. The resist was developed by means of a multicomponent developing agent which dissolutes the irradiated zones about 1,000 times faster than resist material not exposed to X-rays.

A sectional view of a PMMA mould produced in this way is shown in Fig. 3a. Such a mould comprises a large number of identical separation nozzle structures. A corresponding sectional view of a me-

* X-ray lithography has been developed primarily for producing microelectronic devices (see e.g. [3]). In this application the aspect ratio, i.e., the ratio of the thickness of the layer to the smallest characteristic dimension, is usually smaller by more than an order of magnitude as compared to the application described in this paper.

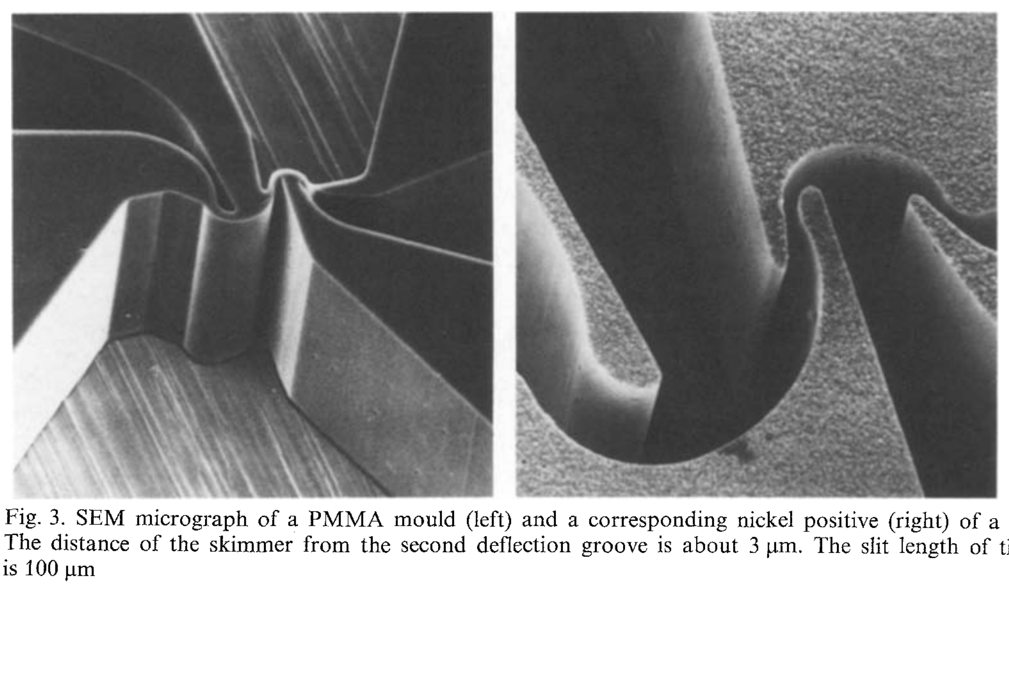
Fig. 3. SEM micrograph of a PMMA mould (left) and a corresponding nickel positive (right) of a separation-nozzle structure. The distance of the skimmer from the second deflection groove is about 3 µm. The slit length of the separation-nozzle mould is 100 µm

tallic separation-nozzle structure which was generated by electrodeposition of nickel is shown in Fig. 3b. The quality of the curved surfaces of the nozzle corresponds to that of the PMMA mould.

Multi-Step Processing of X-Ray Resist

In order to keep the influences of viscous losses at the ends of a slit-shaped nozzle below a tolerable limit, the ratio of the slit length to the distance of the skimmer from the deflection groove, i.e., the aspect ratio of the separation nozzle, should exceed a value of 150. Thus the minimum slit length of each nozzle structure should be greater than 400 μm, if an operating pressure of 1.5 bar is assumed.

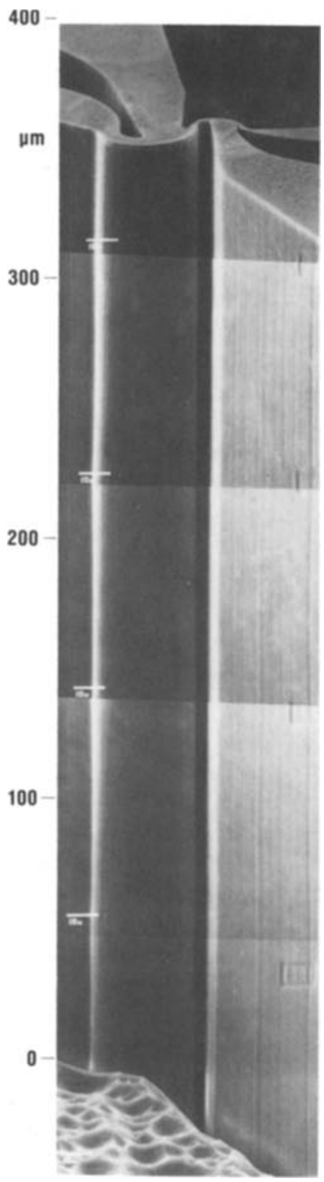

Fig. 4. PMMA mould of a separation-nozzle structure with a skimmer distance of 3 μm and a slit length of 400 μm. The picture has been composed of five single SEM micrographs

Lithographic moulds in sufficiently thick resist layers were generated by multi-step processing with alternating exposure and development. The potential of this method is illustrated by Fig. 4 where a PMMA mould of a separation-nozzle system is shown with a skimmer distance of 3 μm and a slit length of 400 μm. This mould was generated by three alternating exposure and development steps. The characteristic wavelength of the synchrotron radiation was 0.7 nm. The small vertical grooves with a typical size of some 0.1 μm result from corresponding grooves at the edges of the absorbing structure of the mask. They may be regarded as a demonstration of the high accuracy of the method.

Installation of Separation-Nozzle Systems in a Separation Stage

As illustrated by Fig. 5 the nozzles together with the ducts for the feed gas and the heavy fractions are arranged alternately on a line, so that each duct is in connection with two nozzle systems. The nozzle lines are arranged in a zigzag manner between two cover plates with openings for the feed gas and the heavy fractions, respectively. The light and intermediate fractions are collected alternately in the V-shaped ducts formed by the zigzag line. A nozzle arrangement together with the cover plates is called a separation-nozzle chip.
A great number of such chips are arranged in parallel as a chip assembly so that gas ducts are formed between adjacent chips alternately for the feed gas and the heavy fractions. The light and intermediate fractions are exhausted from the V-shaped ducts in opposite directions perpendicular to the large surfaces of the chip assembly.
About 50 chip assemblies, each of them comprising about 40 chips, are mounted as a star-shaped arrangement between two flanges (Fig. 6). This configuration allows a space-saving installation of the chip assemblies as well as a simple gas distribution in a separation stage.

Outlook

With the results available now we count on technical-scale application of LIGA technique as from 1985.
Although the costs of irradiation are acceptable, one may consider whether the dependence on a large-scale accelerator could not be eliminated by modification of the lithographic step. A possible

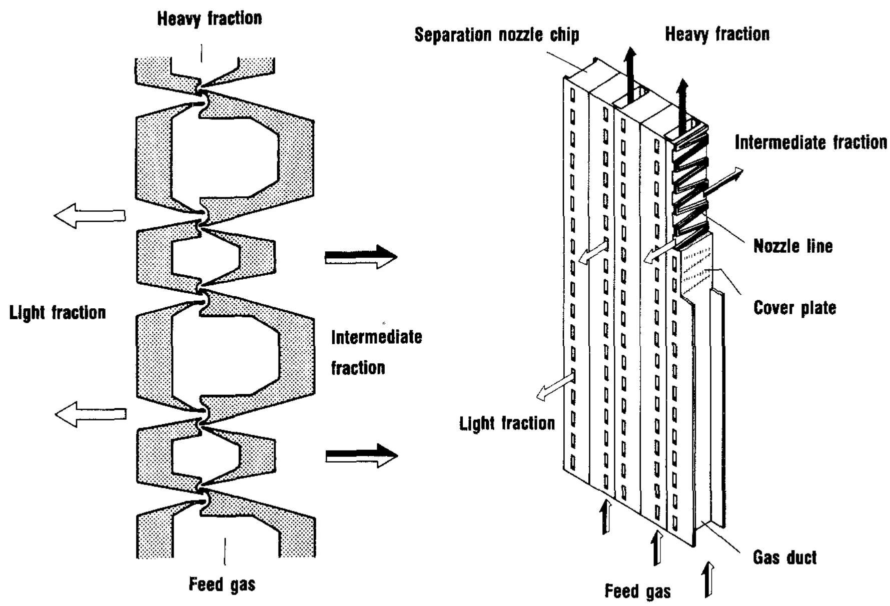

Fig. 5. Part of a separation-nozzle chip assembly (right). The nozzles together with the ducts for the feed gas and the heavy fractions are arranged alternately on a line as shown at the left

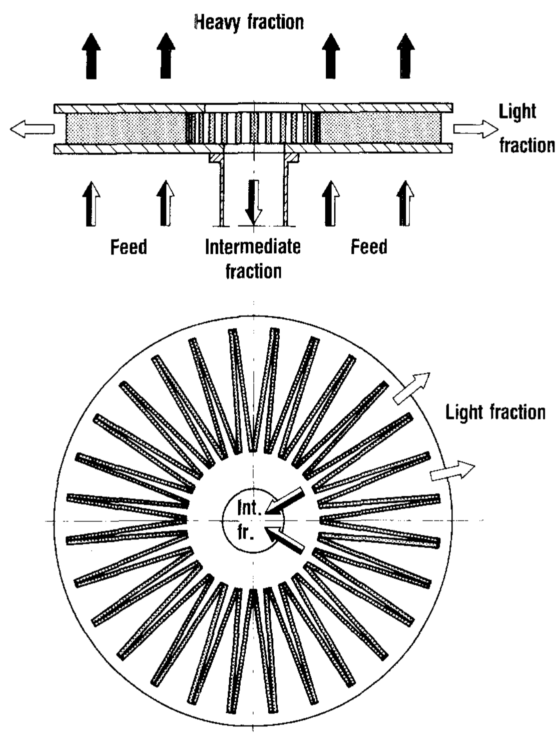

Fig. 6. Arrangement of separation-nozzle chip assemblies in a separation stage

solution would be to use for example ion beams instead of X-rays. In this case liquid-phase development might be replaced by continuous vapor-phase development. Furthermore, reactive ion etching could be taken into consideration which technique has already been used successfully for producing structures with relatively high aspect ratios [4]. However, to our knowledge, none of those techniques has achieved, up to now, results which are comparable to the combination of high aspect ratio and precise image formation as described in this paper.

On the other hand, the possibility is indicated of using the metal structure formed by combined lithography and microgalvanic deposition as a tool for producing separation-nozzle negatives by simple moulding of plastics. In this way, mass production of the separation-nozzle structures would become independent of the relative costly radiation sources.

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