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Institut für Heiße Chemie
Gesellschaft zur Wiederaufarbeitung von Kernbrennstoffen mbH

Fast Contactors in the WAK

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FAST CONTACTORS IN THE WAK[x)]

by

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1 Introduction

The first German reprocessing plant [1] - WAK - with a nominal throughput of 40 t/a has an intermediate character between test facility and industrial plant. The plant is presently under construction and scheduled to start operation in fall 1970. The plant has been designed to process slightly enriched uranium oxide fuels with burnup of about 20,000 MWd/t.

Recent achievements in the reactor fuel technology provide the possibility of higher burnup of fuels in thermal reactors, advanced converters and fast breeder reactors. The technical problems inherent to the aqueous reprocessing of such fuels have to be accounted for.

High burnup together with economically acceptable cooling times leads to intense radiation of the spent fuel due to the high content of fission products and actinide isotopes. One of the main problems in aqueous reprocessing of such fuel is to minimize radiation effects on solvents and aqueous process solutions to prevent the decrease of separation efficiency. Short residence times in the extraction equipment essentially contribute to the decrease of radiation damage.

2 Flow-sheet Considerations

Conventional mixer-settler batteries have been designed and will be used in the WAK for the extraction steps. The residence time of the organic phase in the high active region is about 15 minutes. The use of fast contactors is most advantageous in the co-extraction step (HA) of the first extraction cycle where the majority of the fission products is still present. At comparable throughput the residence time could be decreased by a factor of 20. A simplified flow-sheet of the WAK extraction cycles is given in fig. 1. The extraction cycles are succeeded by a silicagel tail-end treatment for uranium and an anion exchange step for final plutonium purification.

The use of fast contactors not only reduces the residence time but also permits higher throughputs. This is important for the processing of solutions with high content of fissile materials, which requires small critically safe equipment.

Another mode of application is an additional co-decontamination cycle preceding the first extraction cycle. Reconcentration of the dilute strip solutions would then become necessary before entering the first extraction cycle.

Although laboratory tests of the fast contactor type to be used (see below) gave satisfactory results, the reliability and stable performance of multistage batteries under rigorous plant conditions have not yet been proven. As there is ample time before the use of fast contactors becomes inevitable for the processing of high-burnup fuels, it has been decided to start with the installation of only two batteries of centrifugal contactors, which constitute a third extraction cycle for uranium. Due to the low activity level at this point the contactors remain easily accessible for adjustment, maintenance and test instrumentation. Only after extensive testing during plant operation a replacement of the conventional mixer-settlers by centrifugal extractors in the highly active section will be considered.

The chemical purex-type flow-sheet of the second uranium cycle cannot be used in the third cycle without change. The product of the second cycle is a low acid, dilute uranium solution. No intermediate evaporation is planned. Instead, the acid concentration is raised to 2.5 M resulting in more favorable distribution of uranium. This improves the ruthenium decontamination.

The extraction and strip batteries have 12 stages each. Fig. 2 shows the proposed flow-sheet of the third uranium cycle. This additional cycle will result in a further improved overall decontamination factor, thus reducing the need for rework operations.

3 Contactor Equipment

The extractors designed for WAK are similar to those developed at the Savannah River Laboratory in the United States.[2] The design principle of a compact WAK centrifugal contactor stage is shown in Fig. 3.

Its main components are a mixing chamber, a rotating bowl to separate the mixture in a centrifugal field, circular weirs to remove the phases separately and a driving motor for both bowl and mixer. The light and the heavy phase are sucked into the mixing compartment by an impeller pump through the two horizontal inlet tubes and are mixed intensely to yield an optimum mass transfer. The mixture is forced into the rotating bowl passing an anti-vortex baffle and steadying plates at the center of the axle. In the narrow, nozzle type inlet aperture the mixture is fed from the static mixing chamber into the rotating settling chamber (rotor) converting pressure into kinetic energy. The proper functioning of a stage depends on the correct choice of the free cross section of the nozzle for the respective speed and overall throughput.

A baffle plate or diversion baffle in the rotor on the mixer shaft radially diverts the entering mixture to the outside where it is subjected to the centrifugal field (about 400 g) over the entire length of the rotor and finally disengaged.

Separated by conveniently shaped circular weirs the two clean phases leave the rotor through radial bores at its top into collection chambers and further into the subsequent extractor stages via tangential outlet tubes.

The rotor shaft is supported by a plug and coupled to the motor which is located outside the radiation shield. The rotor must be balanced very carefully as the contactor has no bearing below the shielding. In the liquid stream there is thus no mechanically sensitive component which would require maintenance.

The extended motor shaft has an easily exchangeable rotary seal. Compressed air (0 - 300 mm Hg) is fed through the seal and the hollow motor and rotor shafts to the circular weir of the heavy phase to control the position of the interface so that both phases are completely disengaged [3].

The motor can easily be replaced. A special design feature allows also the removal of motor, plug, shaft, rotor and impeller as a unit for replacement.

4 Laboratory Tests

Hydraulic properties and the extraction efficiency of the centrifugal contactors were investigated using a 12-stage battery. Fig. 4 shows the stages arranged in a rectangle.

Characteristic data of a prototype contactor are summarized in Table 1. The weir dimensions were designed specially for the Purex system; however, they can be calculated for any other solvent system as well [4].

Hydraulic Capacity

The maximum total throughput of the centrifugal contactor depends on the

- size of the contactor
- mass flow ratio of aqueous and organic phase
- speed of mixer and rotor
- density ratio of the phases
- viscosity, temperature, and emulsibility of the phases

a) Without Interface Control

Fig. 5 shows the dependence of the maximum throughput on the mass flow ratio q_a/q_o at various impeller speeds and two different density ratios for a given degree of mutual entrainment of the two liquid phases (0,5 and 1 % resp.). The highest obtainable throughput (organic plus aqueous phase) was 600 l/h using the system 30 % TBP/Shellsol T - 1 M HNO_3 and a flow ratio of 1/1.

b) With Interface Control

Interface Control by means of compressed air requires an additional circular weir and a sealing baffle to prevent the escape of air through the aqueous outlet (Fig. 6).

The pressure range for the control air depends on the density of the aqueous phase, the rotor speed and the liquid head ($r_2 - r_a$). In the case of an air failure both phases leave together through the aqueous port into the collection chamber.

The maximum throughput as a function of the mass flow ratio for the system 15 % TBP/Shellsol T - 0.1 M HNO_3/ 1 M $NaNO_3$ is plotted in Fig. 7. The throughput curve holds for a constant rotor speed of 3.000 rpm and an optimum air control pressure p_a, i.e. that pressure at which the aqueous and the organic phase have a tolerated maximum of 1 % impurities. The maximum effective range of pressure control, $p_{h\ eff}$, was 266 mm Hg.

The maximum total throughput in the case of interface control is about 50 % higher than without control. Moreover, interface control allows with one type of contactor the handling of liquid systems with density ratios between 0.75 and 0.95 as is the case in the extraction cycles of the Purex process.

Mass Transfer Efficiency

The mass transfer efficiencies of the prototype contactor were determined for countercurrent extraction and re-extraction under the conditions of the third cycle of the WAK.

a) Uranium Extraction (Three Stages)

The uranium distribution is shown in Fig. 8.
Aqueous feed: 0.198 M U/2.5 M HNO_3
Organic feed: 30 Vol% TBP/Shellsol T
Slope of the operation line: 1.53
Contact time per stage : 14.1 sec.

After about six minutes steady state was established in the three stages with an overall throughput of 215 l/h.

b) Uranium Stripping (Six Stages)

The uranium distribution is plotted in a Mc Cabe-Thiele diagram (Fig. 9).
Aqueous feed: 0.22 M HNO_3
Organic feed: 30 Vol% TBP/Shellsol T - 0.325 M U
Slope of the operation line: 1.35
Contact time per stage : 15.1 sec.
Overall throughput : 200 l/h

Since the extraction was carried out at only about half of the maximum possible throughput, the contact time is relatively long. Steady state was established after about eight minutes.

Fig. 9 clearly indicates that the theoretical equilibrium was not obtained in the individual stages.
While the extraction yielded an overall efficiency (ratio of actual to theoretical number of stages) of nearly 100 %, only about 72 % were achieved in the stripping operation. This is possibly due to slow transfer kinetics[5]. The stripping temperature was 22 °C compared to 50 - 60 °C usually applied in plant operation.

5 Plant Installation

The two centrifugal contactor batteries together with pumps and small receiver vessels are placed in a concrete cell (3 m x 3 m, 2.1 m height) with a wall thickness of 0.5 m (Fig.10). The separation from other equipment and active areas makes the centrifugal contactors accessible and maintenance operations are conveniently done. The shielding efficiency would have to be raised by a factor of 10^4 for processing highly active solutions.

The product solution of the second cycle is collected in a pair of feed adjustment vessels and fed by air-lift to the first contactor. The TBP-solvent is recycled by canned motor pumps without intermediate cleaning.

All process vessels of the third cycle have critically safe dimensions with respect to 3 % U-235 fuel. The transfer of waste solutions to critically non-safe plant sections is controlled by an in-line uranium monitor.

The overall cost of the installation of the third uranium cycle including adaption to the already existing plant installation will be approximately $400.000. The scheduled date for the start-up of the WAK plant will not be postponed by this additional program.

References:

1) W. Schüller, P. Zühlke, WAK plant for reprocessing, Nuclear Engineering, 13, 768-769 (1968)

2) D.S. Webster et al., DP-MS-67-71 (1967)

3) A.A. Kishbaugh, DP-841 (1963)

4) B.F. Roth, KFK-862 (April 1969)

5) F. Baumgärtner, L. Finsterwalder, J.Phys.Chem. 1969 im Druck.

Table 1 Characteristics of the Prototype of Centrifugal Contactor

rotor (diameter)	80 mm
rotor (length)	130 mm
r_{o*} (organic weir)	16 mm
r_{a*} (aqueous weir)	19 mm
r'_{a*} (aqueous weir with air control)	26 mm
hold up, mixer	0.12 liters
hold up, settler	0.72 "
hold up, overall	0.84 "
centrifugal field	400 g
power requirement per stage	500 W
rotor speed	3000 rpm
material (SS)	DIN 14541

Fig. 1 - Schematic flow sheet of WAK extraction cycles

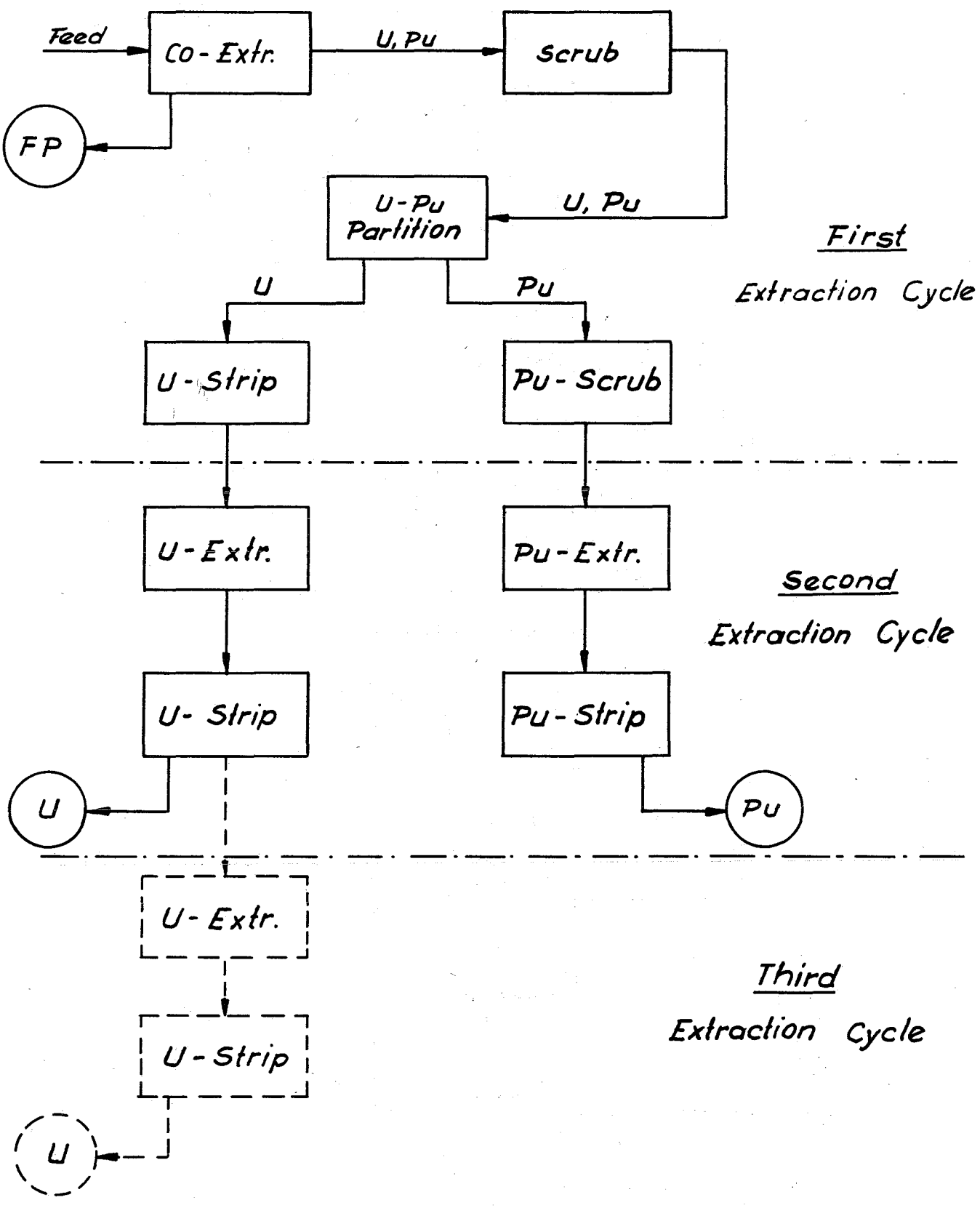
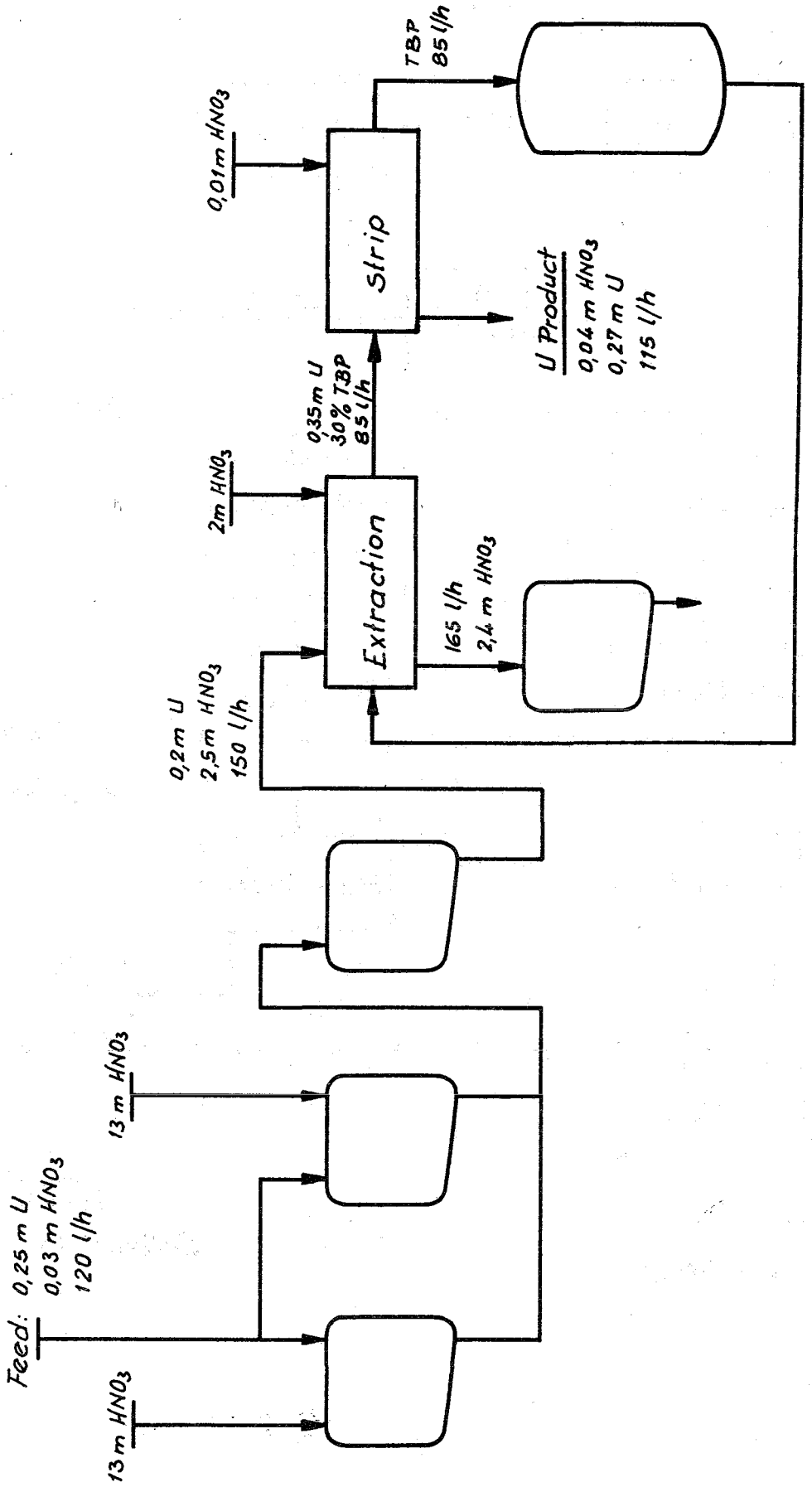


Fig. 2 - Third uranium extraction cycle with centrifugal extractor test batteries



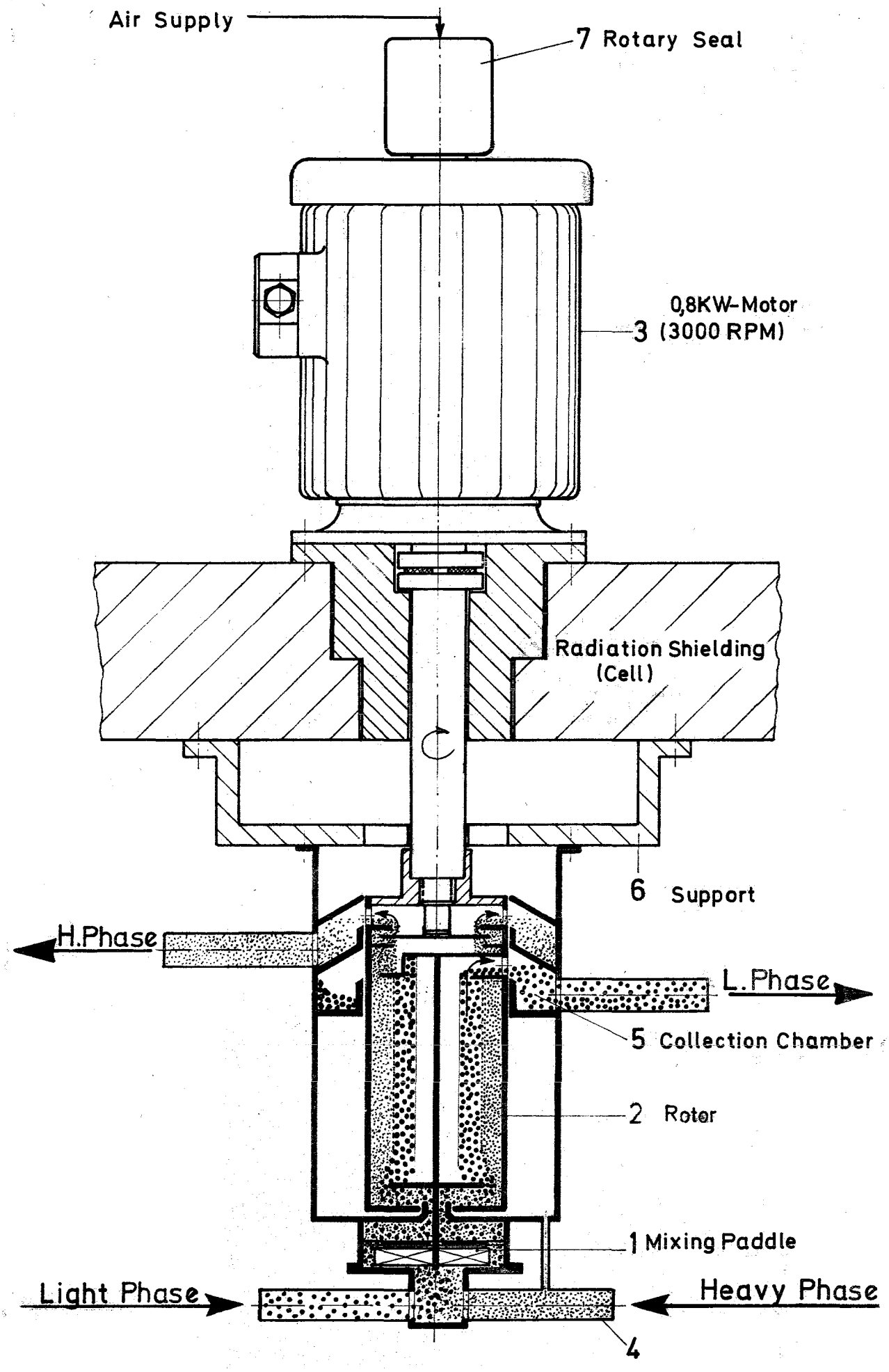
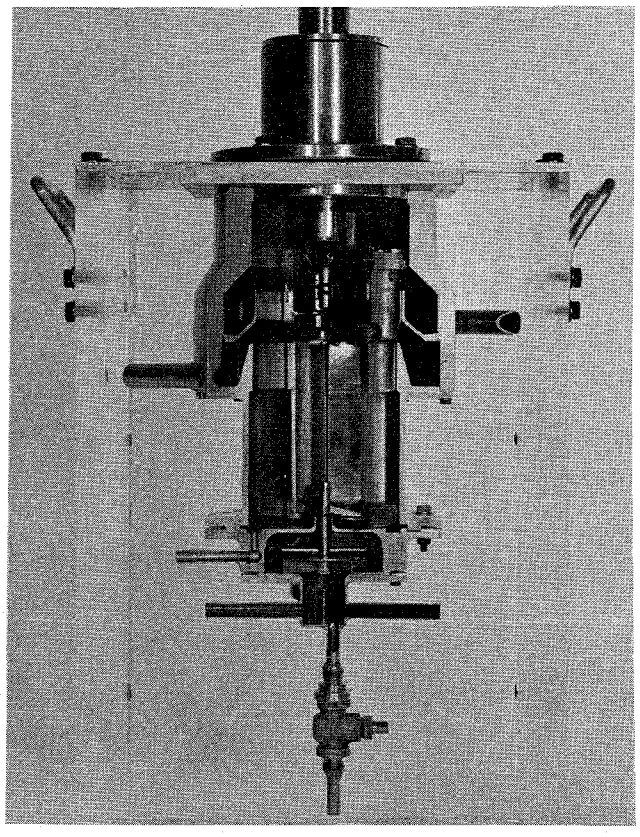
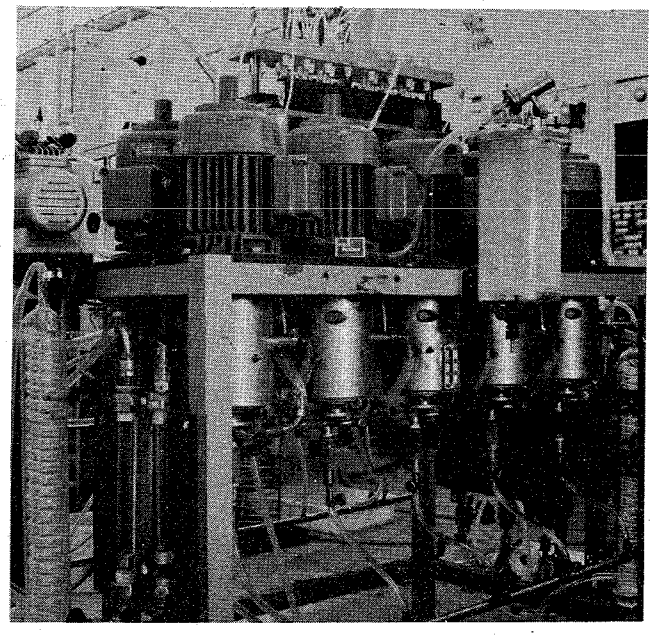
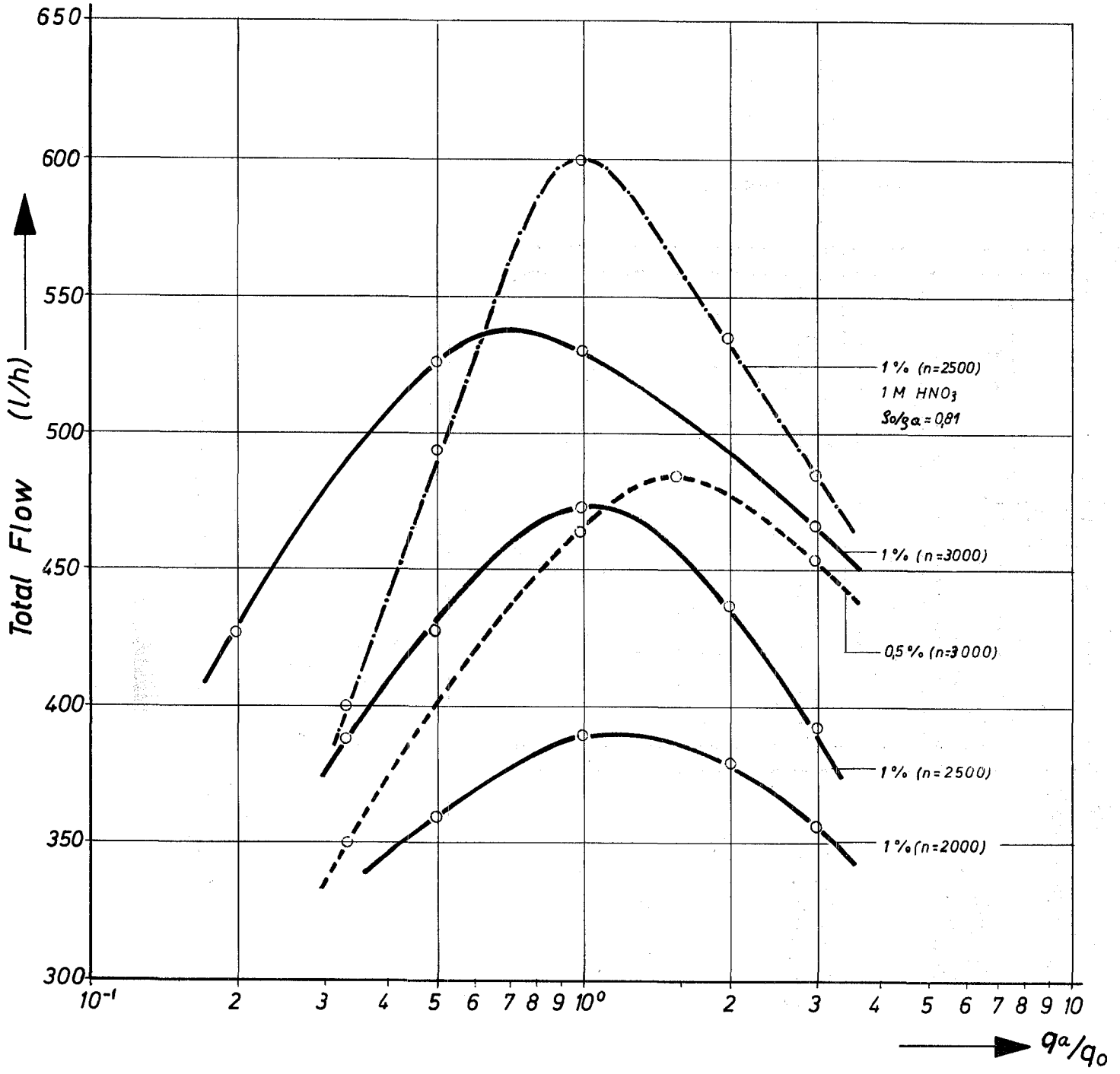


Fig. 3 - Centrifugal extractor stage

Fig. 4 - Centrifugal extractor model and
Laboratory test battery







Max. Hydraulic Capacity for 0,5% + .1% Entrainment of the Org. Phase at 2000/ 2500/3000 RPM

30% TBP/SST
0,075 M HNO_3
S_o/S_α = 0,85
Rotordiameter = 80 mm
r_o^* = 16 mm
r_α^* = 19 mm
t = 25 °C

Fig. 5 - Max. throughput at 2000, 2500 and 3000 rpm. 0.5 and 1 % entrainment

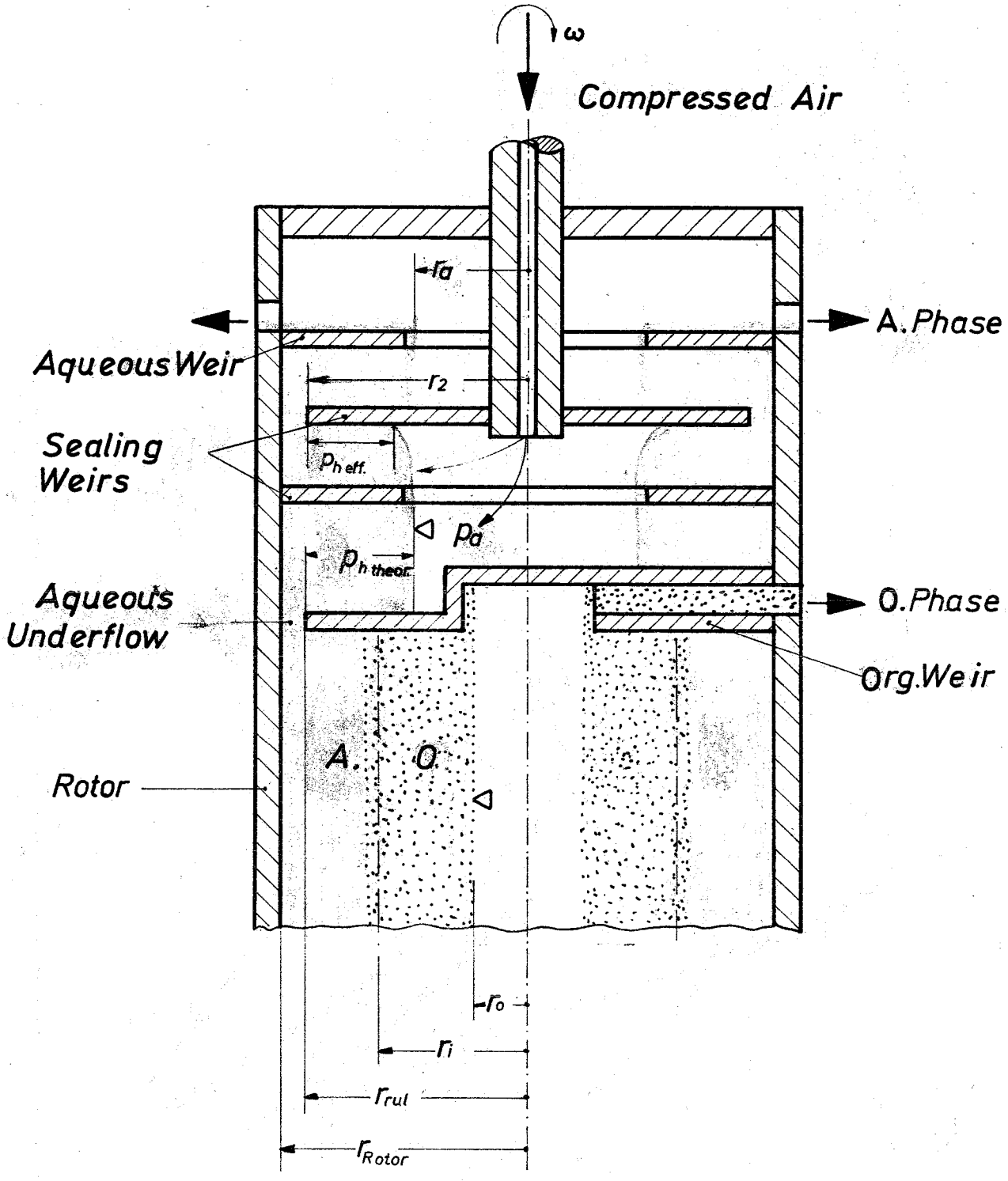


Fig. 6 - Interface control by air pressure

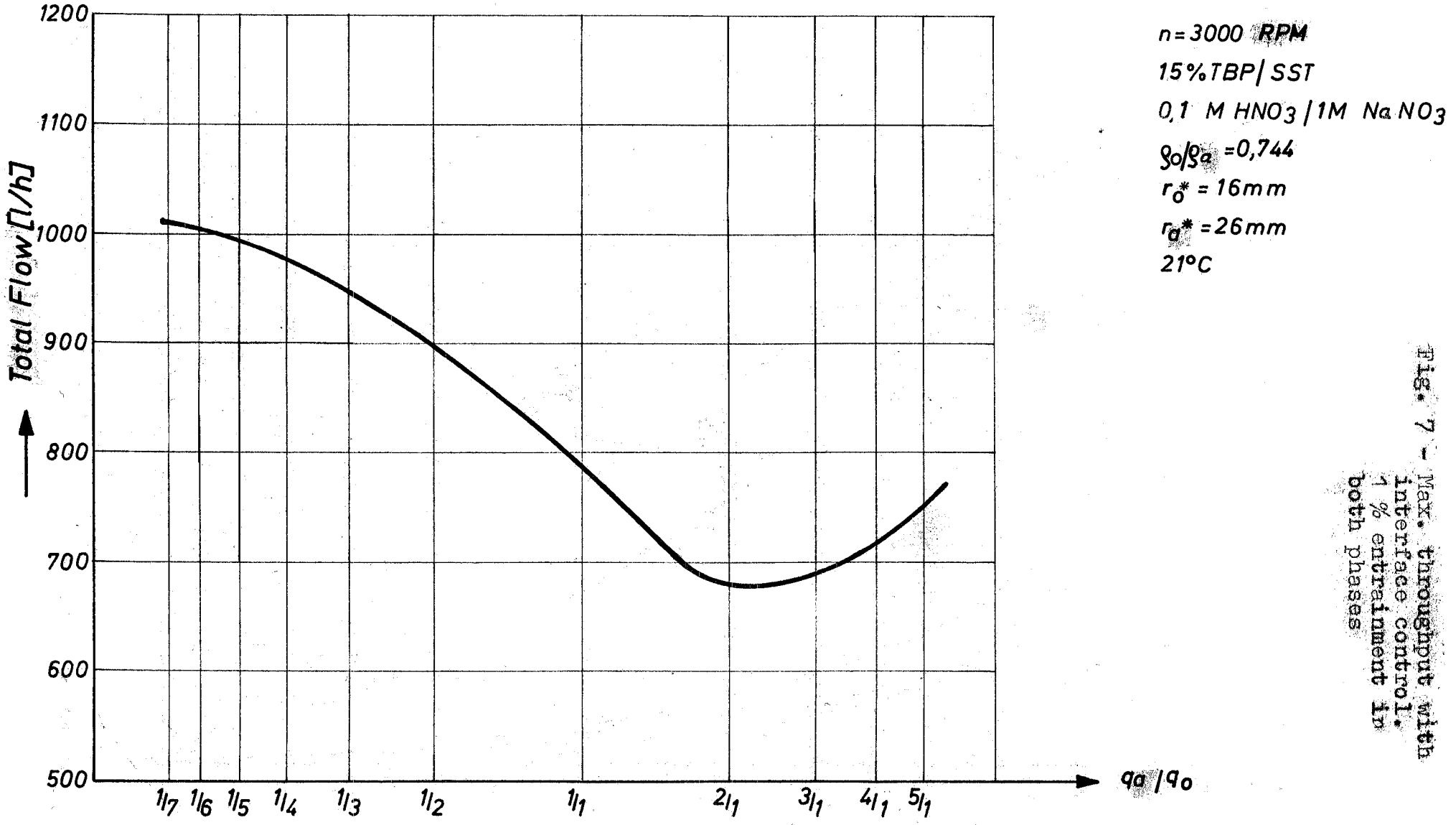
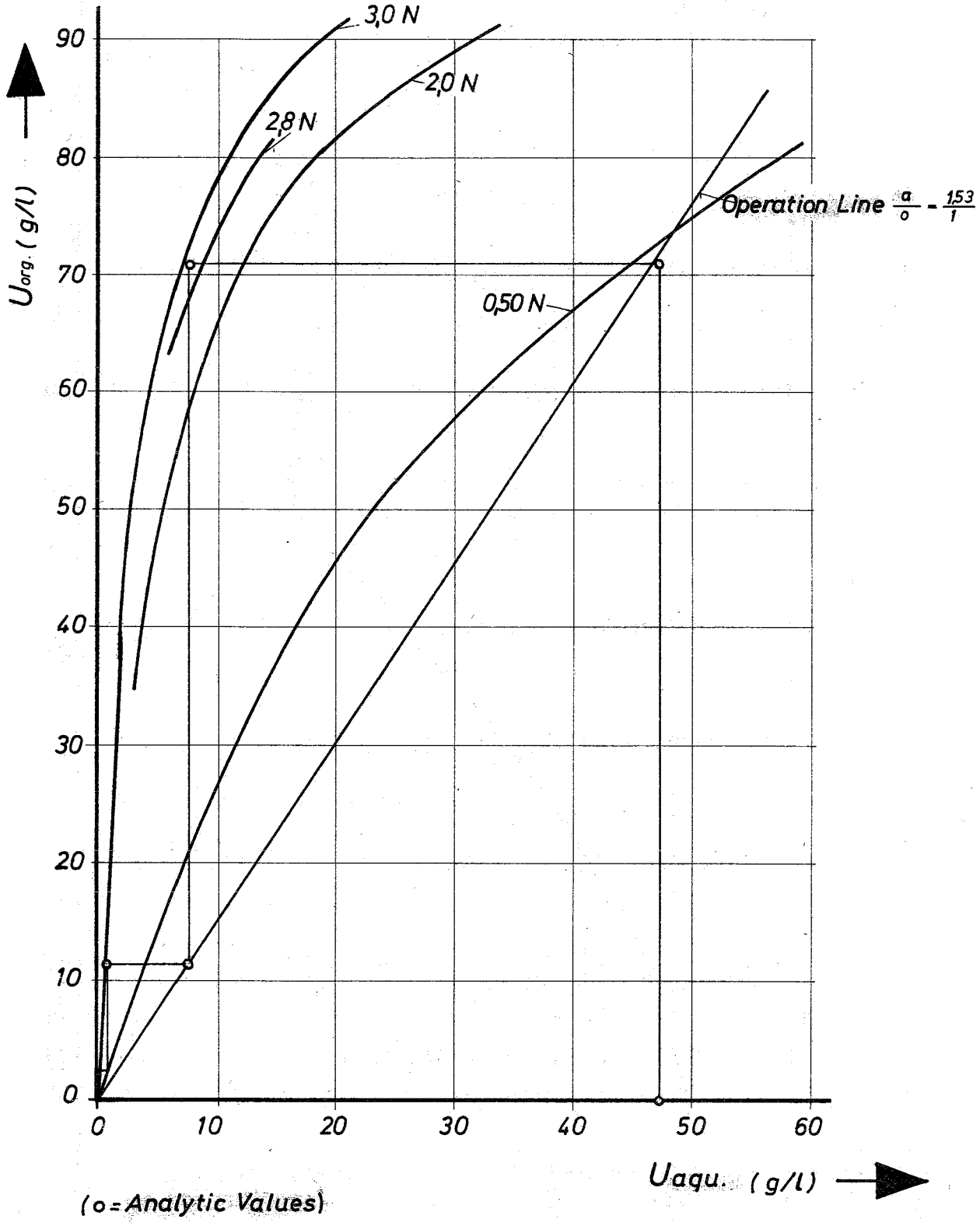


Fig. 7 - Max. throughput with interface control. 1 % entrainment in both phases

30 % TBP/SST

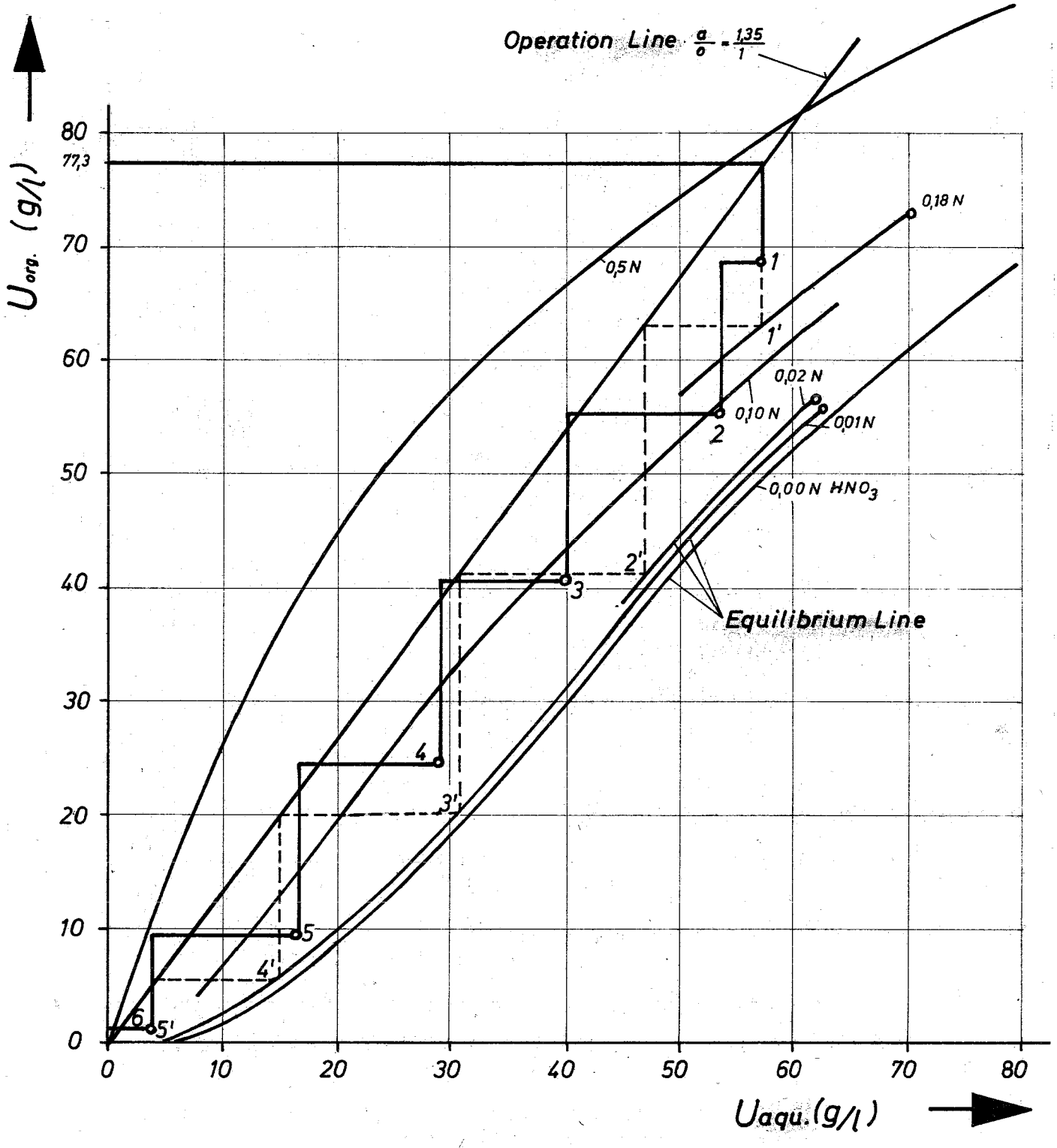
2,5 M HNO_3/0.198 M U

Fig. 8 - Uranium extraction operating diagram



30% TBP/SST - 0,325 M U
0,22 M HNO_3

Fig. 9 - Uranium stripping operating diagram



(o = Analytic Values)

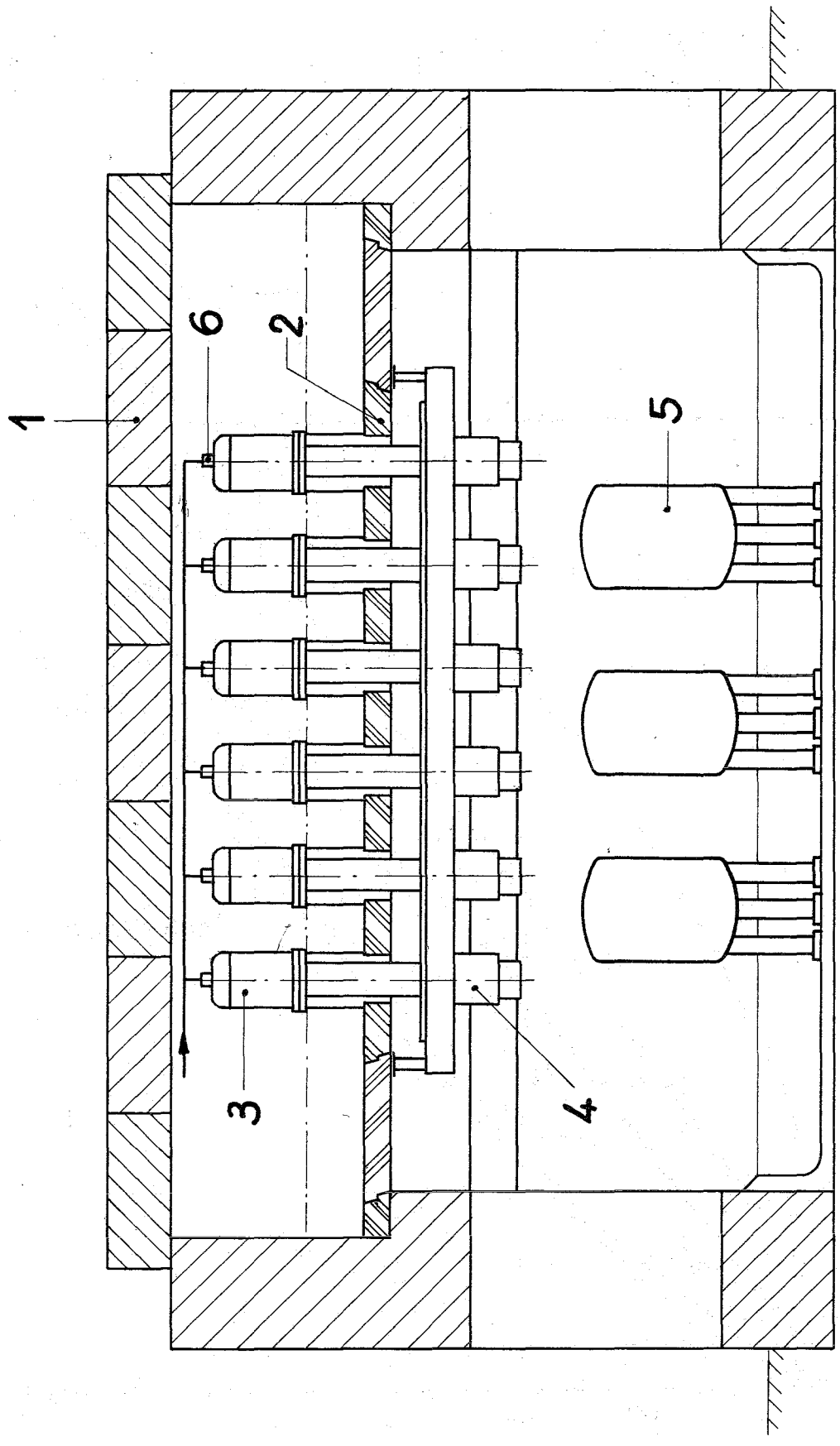


Fig. 10 - Sectional view of cell with centrifugal extraction battery.
(1) Removable shielding, (2) steel shielding, (3) motor, (4) extractor, (5) recipient, (6) rotary seal