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R+D-WORK ON SAFEGUARDING THE INPUT TO A FUEL REPROCESSING PLANT (ACCOUNTABILITY TANK)

by

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This research has been carried out in the framework of a contract between the International Atomic Energy Agency (IAEA) and the Gesellschaft für Kernforschung m.b.H., Institut für Angewandte Systemtechnik und Reaktorphysik, Karlsruhe, Federal Republic of Germany.
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Gesellschaft für Kernforschung m.b.H., Karlsruhe

Zusammenfassung

Der vorliegende Bericht wurde im Rahmen eines Forschungsvertrages mit der International Atomic Energy Agency (IAEA) angefertigt. Untersucht wurden spezielle safeguards-relevante Probleme an einem inaktiven 1:1 Modell des Eingangsbehälters der Wiederaufarbeitungsanlage Karlsruhe (WAK). Zur Erfassung unkontrollierter Lösungsmengenbewegungen wurde ein Meßverfahren auf der Basis der Ultraschallmessung entwickelt, welches Flüssigkeit durch die geschlossenen Behälteranschlußrohre nachweist. Verschiedene Einflußparameter, wie Temperatur, Dichte, Inhomogenität der nachzuweisenden Flüssigkeit, wurden untersucht. Fernerhin wurden erste Schritte zur Entwicklung eines Systems zur Spaltmaterialerfassung mittels Tracer unternommen. Als Tracer wurden Zink und Rhenium verwendet. Im Zusammenhang mit den Tracerversuchen wurden angrenzende Gebiete behandelt wie Untersuchungen zur Homogenitätsverteilung des Tracers in der Lösung, Effektivität des Luftmischers, Einführmöglichkeit des Tracers in den Tank, usw. Zur Erstellung von Vergleichsmöglichkeiten wurden sorgfältige Behälterkalibrierungen unternommen. Ausblicke und Möglichkeiten für weiterführende Arbeiten werden aufgezeigt.

Abstract

The present report was prepared in the framework of a research contract with the International Atomic Energy Agency in Vienna. Special problems relevant to safeguards were investigated on an inactive 1:1 scale mock-up of the input accountability tank of the reprocessing plant at Karlsruhe (WAK). In order to monitor uncontrolled movement of input solution an ultra-sonic measuring device was developed which gives evidence of liquid present in the intact tank tubing. Different parameters influencing its performance were studied, like temperature, density, inhomogeneity, etc, of the liquid to be monitored. Furthermore, first steps were undertaken to develope an input accountability system with the help of tracers. Zinc and Rhenium were used as tracer. In connexion with the tracer works a series of related problems were investigated, like inhomogeneity distribution of the tracer in the solution, effectivity of the air-sparger, possibilities of introducing the tracer to the tank, etc. For comparison reasons, careful tank calibrations were performed. Conclusions are drawn and recommendations for further work are given.

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1.0 Introduction

It was recognized at an early date that the input stream to a reprocessing plant would require concentrated attention in the course of developing methods and techniques for safeguards. The earlier activities on this field are summarized in /1/. The main reason of interest in this point lies in the fact that the fuel elements irradiated in reactors, loose identity for the first time in the dissolver tank and a direct measurement of contained fissile material is possible there. Besides, the accountability tank connections are made in such a way that uncontrolled withdrawal of fissile material solutions into the process area is possible. Also, the information system at this point is normally such that some additional efforts may be required to ensure the credibility of all the relevant information required for safeguards.

A fairly large amount of work in this particular field was carried out at the Karlsruhe Research Center in the framework of the safeguards project. In this connection it was planned to erect an experimental set-up of the accountability section of a reprocessing plant of the WAK type (Wiederaufarbeitungsanlage, Karlsruhe). The systems analytical group of the safeguards project drew up a detailed program for experimental investigations of different problems, for testing of different instruments and techniques and for working out solutions.

The IAEA suggested to the systems analytical group of the project to investigate the following three problem areas in detail in the course of the present contract:

a) Development of a reliable instrument to indicate the presence or absence of liquid in the pipe lines connected to the input accountability tank in order to detect the by-passing of dissolver solution.

b) Investigation of the problems of inhomogeneity and recalibration with the help of tracers.

c) Investigation of the possibilities of tampering and protection of the measurement data system.

The work carried out in these areas is presented in this report. In chapter 2, a detailed description of the experimental set-up has been given. The set-up consists mainly of two fully instrumented accountability tanks, one with a plexiglass outer shell to permit visual observations, and the other with a stainless steel shell - both having approximately the same dimensions as that of the accountability tank at the WAK.

Chapter 3 deals with the problem of liquid indicating devices. After setting up the criteria for the choice and characteristics of a liquid sensing device, seven different methods have been analysed with regard to their suitability. The system chosen for further analysis is a commercially available liquid indicating device with ultra sonic sensors. The operating principle of this method as well as, the experimental set-up with the commercial unit used for testing, have been explained in detail. A parametric study was carried out for different liquid temperatures and densities as well as, for different ratios of air-liquid mixtures. The use of the unit for the purposes mentioned above has been demonstrated clearly in the experimental set-up. Its applicability has yet to be proved under radioactive operating surroundings in a commercial plant.

In chapter 4 the problem of the use of tracers has been analysed in detail. As in the case of liquid sensing devices, the reasons for the use of a tracer and the conditions to be fulfilled by it have been discussed first. Although such an analysis indicated a number of elements like, Li, Ge and Zn etc. as suitable for tracer use, actual experiments were restricted to zinc and rhenium (Re). The results indicate that zinc may be used as a tracer for the verification of volume calibrations. However, further tests are essential under plant operating conditions before more definite recommendations on its use can be made.

The possibility of use of Re as a tracer requires further investigations. The neutron activation analysis suggested in this report for the measurement of Re is still at an initial stage of development.

In chapter 5 the data generation, transmission and indication methods for the level and density measurement systems have been analysed for their sensitiveness to tampering. The analysis showed that it is technically not feasible with reasonable means, to monitor the data system at the input of a reprocessing plant in such a manner as to detect intentional tampering. A much better way of safeguarding fissile material at the entrance of a reprocessing plant appears to be the use of correlations or to a certain extent the use of suitable types of tracers.

The last chapter deals with some recommendations for further work in this area. It is found desirable to continue work in three areas namely:

a) on the liquid sensors in the experimental set-up to refine its use,

b) improve different analytical methods for tracers like zinc and rhenium in the laboratory scale and

c) testing of liquid sensors and the tracers under radio-active and operational environments.

2.0 Mock-up

The experimental set-up consists mainly of 2 vessels which are 1:1 scale replica of the input accountability tank of the fuel reprocessing facility at Karlsruhe (WAK). Fig. 1 shows the original drawing of the WAK tank no. 13.2.08, Fig. 2 shows a cross-section of Tank I which has been used previously for R+D work reported in /2/ and is slightly different from the original. Its main difference is the plexiglass outer wall which is of stainless steel in the original. In Fig. 3 a top view of the same tank with the pipe connections can be seen. Fig. 4 shows the work shop drawing of Tank II. It resembles exactly the original but for the fact that the cooling jacket has been omitted and that 2 additional sets of U-tubes to measure the density in various positions in the tank and 4 additional sets of sample taking lines also in various positions in the tank, have been installed.

The installation of a second tank made entirely of stainless steel became necessary because it is foreseen to use uranyl nitrate for the tracer experiments, and thereby simulate, as far as practicable, operating conditions in a plant. Furthermore, the calibration with a liquid of a density of about 1.5 g/ml (see chapter 4.3.1.2) will give more realistic statements on the geometrical deformation on the stainless steel tank than on the plexiglas one. Besides, with two tanks plant conditions can be simulated for the solution transfer from one tank to the other.

2.1 Description of the experimental set-up

The experimental set-up is assembled in Room 109 of the Institute of Applied Reactor Physics at the Nuclear Research Center Karlsruhe. The room is two-storied which makes tank calibration and sample taking easier. Fig. 5 shows schematically the experimental set-up. The two tanks are placed in catch tanks to comply with the safety regulations. The tanks are connected with each other as shown in Fig. 6. The individual transfer routes can be pre-set and called off automatically, thus preventing an unintended switching error. They can also be switched manually to allow for a greater flexibility for tests on transfer simulation and its tampering. The electrical connection diagramme is shown in Fig. 7. Transfers are done with the help of a commercially available pump model ROVEX 50 E by Klein, Schanzlin & Becker. This does not ecactly represent plant conditions but as the transfer system itself was not within the scope of the Contract it seemed legitimate to do this substitution, particularly because of the fact that steam for steam-jet operation is not available in the laboratory.

Tank I is equipped with a level and density instrumentation as shown schematically in Fig. 8. The transmitters are CMR-transmitters type TDE 240 from Schoppe and Faeser with a range from 0 to 2500 mm H_2O. The special advantage of these transmitters lies in the fact that the beginn of the measuring range can be shifted between 0 and 90 % of the maxinum range and the range can be adjusted on the ratio 1:10 electronically, i.e. without mechanical alteration of the transmission system. This opportunity was used on the density transmitter: Supposing that the maximum pressure difference in the density dip-tubes would be achieved with a liquid of 1.5 g/ml, this pressure would be put equivalent to a 100 % indication on the transmitter. The lowest density which can normally be expected in an input accountability tank will be somewhere around 1, which then would be equivalent to a 67 % indication on the transmitter. This means that the range between 0 and 67 % is lost and that the significant range between 1.0 and 1.5 g/ml is restricted to the 67 % to 100 % range on the transmitter. By shifting the zero point and spreading the remaining range electronically it was possible to equate 1.0 g/ml to 10 % and 1.5 g/ml to 90 % indication. Such a flexibility makes these transmitters very valuable for laboratory experiments.

The recorder is a twinned line recorder from Hartmann and Braun, type ERLN 2/144 K. The measuring range is 2 x 0 to 20 mA. The level and density transmitter outlets are coupled to the respective scales on the recorder.

The instrumentation of Tank II is virtually the same as of Tank I. Additionally, 2 dip-tube systems to measure the density at various positions in the tank are installed. The additional instrumentation needed - air flow-rate meters and recorder - is installed on the panel, whereas the transmitters have been left out for cost reasons and those from Tank I will be taken over for the inhomogeneity tests (see chapter 4.3.2.2). Fig. 9 shows schematically the level and density instrumentation for Tank II.

Tank I is equipped with a sampling device. The needle-block is fixed on the upper floor of the room. For Tank II 5 such sampling devices are installed. The needle-blocks are also mounted, together with the one from Tank I, on the upper floor.

Both tanks are equipped with air-spargers for solution homogenization purposes with corresponding air supply. The instrumentation and connection is shown schematically in Fig. 10.

Furthermore, to measure the temperature of the solution in the tank, a temperature measurement device is installed on each tank. A resistance made of Pt-wire and embeded in a protecting tube of 1400 mm length is introduced into the tank. The temperature then can be read directly on a calibrated voltmeter installed on the panel. The measuring range of the device is from 0 to 40°C.

3.0 Liquid indicating sensor

3.1 Basic considerations

The range of problems related to the proper safeguarding of in-going fissile material to a reprocessing plant have been dealt with in great detail in /3/. These problems are summarized below for ready reference.

Fig. 11 shows schematically the input accountability area of an EUROCHEMIC-type reprocessing plant. It consists principally of the dissolver, the accountability tanks for in-going solution, for recirculated recovered acid and for jacket dissolution waste as well as, of a monitoring device for solid wastes. The input accountability area of a WAK-type plant is the same excepting the jacket dissolution waste tank as the WAK operates on the chop-and-leach basis. The problems there are of a similar nature.

Analyses of the plant design of the existing reprocessing plants of WAK type and the experience gathered during the three integral exercises at EUROCHEMIC, Mol, indicated at an early stage that several possibilities existed for the uncontrolled withdrawal of fissile material containing solutions at the head end of the existing facilities. Some of these possibilities are listed below.

1. During the filling of the input accountability tank, the solution could be pumped off simultaneously, thus withdrawing the unmeasured solution in an uncontrolled manner. If the pumping-off is done at a lower velocity than the filling, the recorder reading will continue to show an increasing slope. Also during the emptying of the accountability tank after having accounted a batch a filling from the dissolver is possible, thus also withdrawing unaccounted-for solution.

2. A short-circuitting of two adjacent needle-blocks can also be used to withdraw uncontrolled amounts of fissile material. In times of

necessity (break-down of the normal transfer systems) whole batches have been transfered from one tank to the other by this method at the EUROCHEMIC reprocessing plant.

Because of these possibilities it would seem desirable to know for safeguards purposes whether and at what time a batch of solution has actually been transfered. This could lead to the installation of an inter-lock system of the transfer mechanisms. However, such a system may hamper the normal operation of the plant and would probably be inadequate. For example in the case of two short-circuitted needle-blocks the switching-on of the circulation belongs to the normal practice.

A detailed analysis of the various possibilities has led us to the conclusion that a fairly simple solution exists for the highly complex problem, namely, to monitor the absence or presence of liquid in the tubes leading to or coming from the tanks in the input accountability area. With this, the indicated presence of liquid in two or more tubes of one tank would mean an abnormality in the operational process requiring further investigations. Besides, with this suggested method the sequence of presence, and/or absence of liquid in the pipes connecting the vessels in the input accountability area can also be monitored. This gives an inherent check on a properly performed input measurement and informs the inspector of any irregularity, e.g. of an ommited sample-taking.

3.2 Conditions to be fulfilled by a liquid indicating system

Before searching for an adequate measurement method considerations were given to the conditions which a liquid indicating system would have to fulfill:

a) The tubes to which the sensor is to be coupled should not be cut, firstly for safety reasons (leakproofness of welding) secondly because such an intervention would not be possible in working plants without unreasonable costs and intrusion into the running process.

b) The sensor should be able to be mounted on the tubes with the help of remote handling devices.

c) The sensor should require a minimum of maintenance, should have a high operational life-time and should be easily replaceable.

d) The sensor should be resistant against corrosion and radiation.

e) The sensor should be inexpensive.

f) The sensor should give unequivocal response to the presence or absence of liquids.

A number of liquid measuring methods were analysed and commercially available apparatus studied. These methods are shown in Table 1 together with the conditions mentioned before. This matrix helped in the choice of an adequate liquid measuring device.

A summarized review of the different measuring methods analysed is given below.

Conductivity probe: This method would mean the cutting of the tubes but would on the other hand be very economic and once the ceramic insulated probe has been properly welded into the tube it could be regarded as very safe against leackage. The afforded electronic needed is straightforward, simple and cheap. On the other hand, it should be well understood that also in the future reprocessing plants will be few and far between, so that the possibility of installing such a device in a new to build plant will be scarce. Nevertheless, it was suggested in the contract to investigate the applicability of this method to this measurement problem, but due to the very well known technique it was later agreed upon in discussions related to this work, that a study without any hardware effort would seem enough to outline the problem.

Inductivity coil: Commercially available apparates are used for through put measurements or even liquid density measurements. The tube length surrounded by the coil must be a material with a low relative permeability coefficient in order to be able to destinguish between the filled and the unfilled tube. For this reason this method is inadequate for this problem because of the high relative permeability coefficient of stainless steel.

Heated wire device: Here a heated wire is introduced into the tube which changes its resistance according to the different temperatures obtained through the different heat flux in air or liquid. This method also seems very unsatisfactory because of oxydation and corrosion effects expected on the wire.

Condensor probe: This apparatus is similar to the first device described. A higher potential level on the insulated probe causes this and the walls of the tube to act as a condensor. The electrical current will flow in the presence of liquid in the tube.

Inter-lock system: As already mentioned, this would mean the mechanical or electrical inter-locking of the transfer and sample-taking power supply (steam-jet, air-lift, etc.). This has the great advantage of being able to be performed on the cold side of the plant installation which however may mean that normal operation of the plant may be hampered. For the sampling lines additional efforts have to be foreseen. It could be that the necessary tamper-proof instrumentation needed in addition may also make this system uninteresting.

Activity measurement: By installing a γ-monitor to the tubes a signal on whether liquid is in the tube or not will certainly be obtained. A disadvantage could be the high back-ground radiation level when the vessel is filled.

Ultra sonic sensor: This apparatus is usually applied in material testing and is operated at fairly high sonic frequencies (in the range of some MHz). The sensor itself is uncomplicated, easy to mount, mechanicallly resistant and is expected to have a high life-time in γ-radiation surroundings. The number of measuring points to be supplied by one central unit can be chosen as high as needed, which makes this system financially attractive. All these positive aspects led us to the conclusion that this device should be investigated thoroughly for its use as a go-no-go liquid sensor.

3.3 Characteristics of the ultra-sonic measurement technique

3.3.1 Basic sound wave theory

The details of the ultra-sonic measurement technique are to be found in the open literature /4, 5/. A short survey is given below:

Basically one distinguishes between a series of fundamentally different kinds of waves, like longitudinal waves, transversal waves, Lamb waves etc., of which the first one is of main interest to our measurement problem.

In the following the behaviour of a plane longitudinal wave is described. In reality we will be confronted mainly with spherical waves, which behave similarly, but are far more elaborate to describe. The unit of greatest interest here is the sonic pressure P, which is defined as

$$P = \rho \cdot c \cdot f \cdot A \qquad [\mathrm{N/m^2}] \qquad (3.1)$$

where ρ = specific density of the material $[\mathrm{kg/m^3}]$
c = sound velocity $[\mathrm{m/s}]$
f = frequency $[\mathrm{s^{-1}}]$
A = amplitude of one particle, max. turn from zero position

$\rho \cdot c$ is often defined as W = specific sonic resistance. The value for steel is $W_{ST} = 45 \cdot 10^6 \ [\frac{\mathrm{kg}}{\mathrm{m^2 \cdot s}}]$, for water $W_W = 1.5 \cdot 10^6$.

A further unit is the wave intensity

$$I = \frac{1}{2} \frac{P^2}{W} \qquad [\mathrm{Watt/m^2}] \qquad (3.2)$$

The intensity is hereby proportional to the square of the amplitude of the sonic pressure.

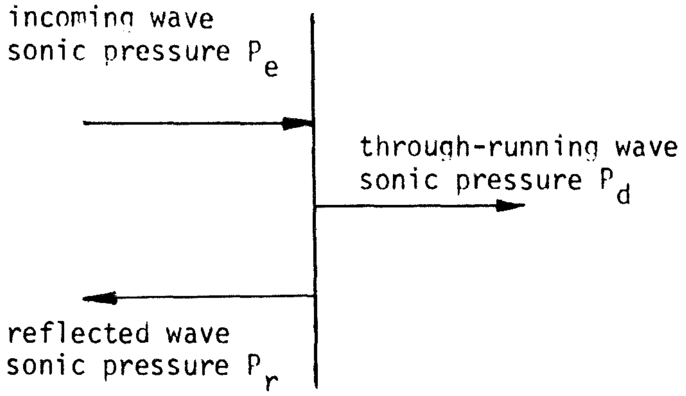
To have an idea of the order of magnitude of the absolute values of e.g. the particle amplitude it should be noted that in material testing wave intensities of 10 Watt/cm^2 are not exceeded. This leads to particle amplitudes of some 0.04 % of the wave length (in water).

In our special application in monitoring the presence or absence of liquid in a steel tube, we can consider the simplified case of the wave behaviour at the boundary surface steel/water on one hand and steel/air on the other hand.

We consider the case of a plane wave hitting the boundary surface at right angle. For symmetry reasons, only plane waves reaching the boundary surface at right angle, produce one reflected and one through-running wave. Their sonic pressure may be calculated in the following manner:

Material 1 Material 2

$W_1 = \rho_1 \cdot c_1$ $W_2 = \rho_2 \cdot c_2$



We refer the sonic pressure of the reflected and through-running wave to that of the incoming and form their ratio:

$$\frac{P_r}{P_e} = R \quad \text{and} \quad \frac{P_d}{P_e} = D$$

R is called the reflexion factor, D the permeability factor.

The derivation leads to

$$R = \frac{W_2 - W_1}{W_2 + W_1} \; , \quad D = \frac{2\,W_2}{W_2 + W_1} \qquad (3.3)$$

For the boundary surface steel/water and with $W_{Steel} = 45.10^6$ kg/m^2.s and $W_{water} = 1.5.10^6$ kg/m^2.s

$$R = -0.935 \; , \quad D = 0.065$$

i.e., -93.5 % of the incoming wave is reflected (the minus sign means phase inversion, which is of no interest in our case) and 6.5 % runs through into the water.

This wave then hits the boundary surface water/steel. R and D is then calculated to

$$R = 0.935 \; , \quad D = 1.935.$$

A higher sonic pressure than 100 % seems at first glance paradox. But in accordance with equ. 3.2 the wave intensity (i.e. energy per time and surface unity) not only depends of the square of the sonic pressure, but also of the sound resistance, in every case the sum of the wave intensities is always zero:

$$I_e = I_r + I_d$$

For the sonic pressure one has

$$P_e + P_r = P_d \quad \text{or} \quad 1 + R = D.$$

As a next step, the wave behaviour at the boundary surface steel/air has to be considered. Here

$$W_{steel} = 45.10^6 \text{ kg/m}^2\text{.s}; \qquad W_{air} = 0.004.10^6 \text{ kg/m}^2\text{.s}$$

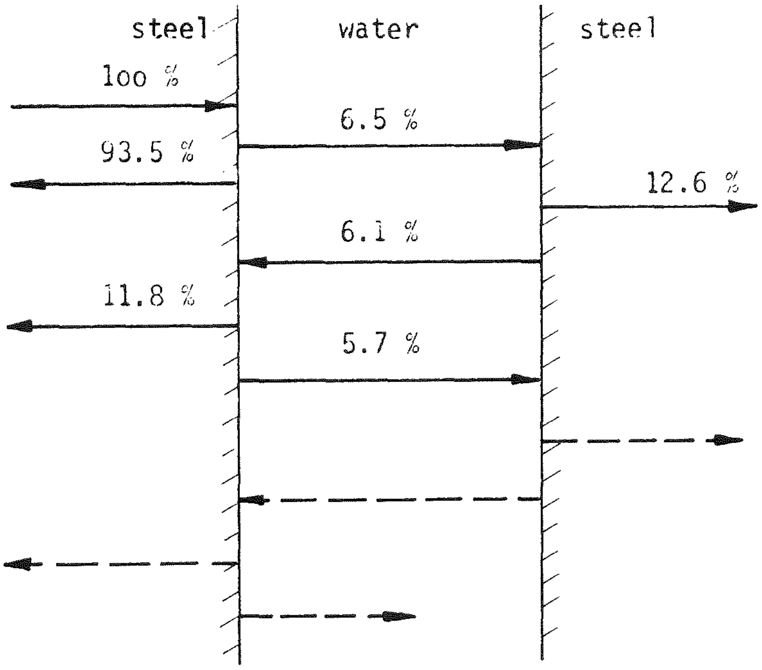
This leads to

$$R = 0.99998222 \; ; \qquad D = 0.00001778$$

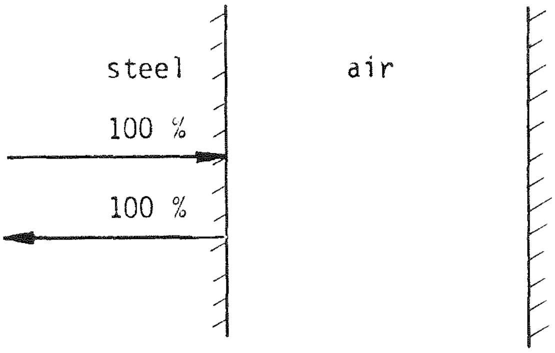
Practically, the totality of the incoming wave is reflected.

In principle we will have the two following cases (for ideal reflexion behaviour):

I.



II.



As can be demonstrated, only the tube filled with water (or any other liquid) will deliver a rear wall echo wave which can be used as a criteria for the presence of such a liquid in the tube. On the other hand, with air in the tube, no rear wall signal will be received (within the intensity range monitored in our case).

It should however, be noted, that the reality looks far more complicated. Wave behaviour, as described is only valid for absolute even boundary surfaces, which cannot be achieved. On the (microscopically) rough surfaces a series of scattering and interference of the sound waves will occur which will influence reflexion behaviour and lead to a reduction of the wave intensities. Furthermore pure longitudinal waves are restricted to liquids only, where only this kind of wave can be transmitted, whereas, in steel sound is also transmitted in form of e.g. transversal waves, which can obey slightly different laws. Moreover, wave inflexion, interference, absorption, scattering, etc. on small material inhomogeneities should be kept in mind.

3.3.2 Generation and reception of ultra-sonic waves

For the generation and reception of ultra-sonic waves a piezo-cristallic material is needed. This material reacts with the generation of an electric tension on its surface when submitted to mechanical deformation. On the other hand the same material reacts with deformation when an electric tension is layed out on its surfaces, which is then called the reciprocal piezo-electric effect.

To generate ultra-sonic waves, a piezo cristal is submitted to an alternating current of a defined frequency of e.g. 4 MHz. The cristal then expands and shrinks due to the changing of polarity on its surfaces in rhythm to the frequency. The cristal so generates a sound wave in any adjacent material.

Alternately, a sound wave hitting the piezo cristal will generate an alternating current: it acts as a microphone.

For an ideal X-quartz, which is presumed to change only in thickness, we can describe the two piezo-electric effects in the following elementary manner, for the static case only:

Having a known tension U_e (=wave emission) on the cristal, the change in thickness ΔX_e can be calculated to:

$$\Delta X_e = d.U_e \qquad [m] \qquad (3.4)$$

where d = piezo-electric modul

$(= 2.3.10^{-12} \frac{m}{V} \text{ or } \frac{A.s}{N})$

Having a known change in thickness ΔX_r caused from outside (= wave reception), the tension is calculated to:

$$U_r = h \ . \ \Delta X_r \qquad [V] \qquad (3.5)$$

where h = piezo-electric deformation constant

$(= 4.9 \ . \ 10^9 \ \frac{V}{m} \text{ or } \frac{N}{A.s})$

We can also calculate U_r by knowing the sonic pressure P which creates change of thickness. U_r will then depend on the cristal thickness d:

$$U_r = g.d.p \qquad [V] \qquad (3.6)$$

where g = piezo-electric pressure constant

$(= 0.057 \ \frac{Volt/m}{N/m^2} \text{ or } \frac{m^2}{A.s})$

3.3.3 Electronic supply unit

Under a variety of different ultra-sonic measurement methods we have chosen the impulse-echo method as the most promising for our measurement problem. In the following a very brief survey of the method is given, for a more detailed study, reference is made to /4, 5/.

In principle, the supply unit consists of an oszilloscope with the corresponding deflexion tension generation units. Furthermore, an emitter unit, power supply and an amplifier is needed. Fig. 12 shows schematically on the left the whole supply unit, on the right the corresponding time sequence of the different tension alterations. The electric impulse tension of the emitter is transported to the sensor head. From there the electric echo impulses are transported back to the amplifier and displayed on the CRT (Cathode Ray Tube).

The trigger unit generates the deflexion tension for the CRT and simultaneously the steering tension for the HF-emitter unit which then rhythmically gives a pulsed HF rate to the sensor head. Fig. 12 A shows as an example a highly attenuated HF tension. The pulsed signal together with the back-coming echo signals are amplified, rectified and after filtering the carrier frequency are displayed on the CRT. The cathode ray is cut off according to the trigger tension (Fig. 12 D). To make the run-back of the cathode ray invisible, the trigger unit steers the brightness of the ray with the help of a rectangular tension (Fig. 12 E).

3.4 Experimental set-up

Central part of the experimental set-up is the electronic supply unit ECHOSKOP MPT 10 from Dr. Lehfeldt & Co., Heppenheim/Bergstrasse, working on the impulse-echo basis as described in chapter 3.3.3. The echo signals are transfered to a monitoring point switch (block diagramme see Fig. 13), to which a maximum of 8 sensor heads can be coupled. In our set-up only 7 sensor heads were coupled, using the 8th step to reset the alarm triggering unit (see chapter 3.4.2). In the following, some technical and commercial details on the purchased apparatus are given:

3.4.1 Data of the purchased apparatus

The data of the purchased apparatus can be summarized as follows:

i. Central supply unit

Type : ECHOSKOP MPT 10
Manufacturer : Fa. Dr. Lehfeldt & Co., Heppenheim/Bergstrasse
Frequency : 0.2-1 MHz, 1-6 MHz, 4-15 MHz
Testing region : Continuous between 10 mm and 10^4 mm (for steel, other materials equivalent)
Impulse emitter : 3 steps, max. output 3 kVA
Impulse rate frequency : 30 - 3000 Hz
Amplifier : linear with switchable selective pre-amplifiers for the 3 frequency ranges. Total amplification 120 dB
Power supply : 220 V/60 VA. 50 Hz
Dimensions : 350 x 230 x 450 mm
Price : DM 11.000,-- + tax (July 1971)

ii. Sensor head

Type : NT 4/10 K
Manufacturer : Fa. Dr. Lehfeldt & Co., Heppenheim/Bergstrasse
Frequency : 4 MHz
Diameter : 10 mm
Price : DM 350,-- + tax (July 1971)

iii. Measuring point switch

Interrogation frequency : 1.5 sec per measuring point
Manufacturer : Fa. Spinner GmbH., Stuttgart
Price : DM 3150,-- + tax (July 1971)

iv. Memory and alarm unit

Manufactured in the electronics work-shop at the Institute.

2 cycles (1 signal storage + 1 interrogation) with bi-stable relays

2 triggered relays block the next storage cycle and "freeze in", which sensors reported liquid.

Estimated costs: ≈DM 1000,--

3.4.2 Adaption to the measurement problem

As already shown in Fig. 12, principally two techniques are possible for the reception of the echo signals, either with one sensor head, this functioning then both as emitter and receiver, or with two sensor heads, one functioning then as emitter, the other as receiver.

Although the two-sensor-technique has a number of advantages (sound wave must not hit rear wall at right angle; no careful centric sensor mounting needed; no double echo absorption; higher echo intensity, etc.) it was decided to use the one-sensor-technique because it would be clearly less elaborate and costly to mount one sensor once an adequate tube adaption device has been developed (see chapter 3.5.1). Nevertheless, the experiments carried out and the experience gathered so far call for thorough investigations on the two-sensor-technique which have not been carried out for lack of time. It should be left open for the moment, which of the two possibilities should finally be given preference.

The greatest difficulty which had to be overcome (and which is the same for both techniques) was the adaption of all signals from various sensor heads on different tube-diameters to one pre-set signal gate and amplitude threshold. A shifting of the gate would have meant a great effort in additional electronic apparatus. The problem was solved as described in chapter 3.5.5.

It was then decided to store the signals coming from the different sensors in a memory and alarm unit. This has a very simple and straightforward design. Parallel to the measurement point switch positions bistable relays are installed. In a 1st cycle of the measurement point switch, these relays are triggered or not, according to a yes or no echo signal. In a 2nd interrogation cycle 2 or more triggered relays short-circuit an alarm buzzer and lighten up their corresponding optic alarm signals (bulbs) and block the automatic re-set, so that the information is frozen-in until the re-set is done by hand. If only one relay is triggered, it will be re-set automatically and the next signal storage cycle begins. A detailed block-diagramme is given in Fig. 14.

3.4.3 Simulation of input accountancy measures

In accordance with the basic considerations mentioned in chapter 3.1, a variety of sensors is assembled as shown schematically in Fig. 15. As the supervised area cannot in reality be restricted to the accountability tank alone and as two tanks are available to simulate plant conditions, the input accountability area can be simulated to a great extent. A comparison with Fig. 11 shows that the main fissile material streams are monitored firstly in their timely sequence. which can give the plant operator a guarantee that the input data are valid for the input balance and secondly gives the inspector evidence of any uncontrolled transfer of solution to be accounted for. The tube connections needed to meet the conception expressed in Fig. 15 are shown in Fig. 16.

Also in Fig. 15 the time sequence of the sensor signals as received with a properly performed input measurement, is shown. This measurement starts with the supply of the accountability tank with recovered acid (RAR) coming from the chemical process carrying minor amounts of fissile material (sensor 4). The acid should be released to the dissolver (sensor 3) only after the inspector has accounted for the batch (sensor 6). During the delivery of the acid to the dissolver (sensor 3), no further acid should reach the RAR-tank (sensor 4). Sensor 3 also shows that the measured acid reaches the dissolver and is not substituted by acid coming from somewhere else, thus justifying the deduction of fissile material content in the recovered acid from the input amount of fissile material measured in the input accountability tank (AFU). The route of the solution bearing fissile material from the dissolved elements and from recovered acid is checked by sensor 1. During the filling period of the AFU tank sensors 2, 3, 5 and 7 should not give a signal, otherwise an uncontrolled withdrawal of the unmeasured fissile material has taken place.

Until the input is accounted for (sensor 5), no movement of solution should be monitored (sensors 1, 2, (6), 7). During release of the solution to the chemical process (sensor 2), no further solution should be pumped from the dissolver (sensor 1).

3.4.4 Parameter variation

In order to determine the limits within which the sensor heads are usable, it was found desirable to investigate the following parameters influencing specifically the performance of the liquid sensor system

i) the solution temperature

ii) the solution density and

iii) the solution inhomogeneity (e.g. mixed media water/air)

It was furthermore necessary to investigate means of coupling the sensor head to the tube to be monitored.

Results of these tests are given in the following chapter 3.5.

3.5 Experimental results

3.5.1 Investigation of different coupling techniques

As could be demonstrated in chapter 3.3, the sound transmission between two media is in our case only possible when these two are either solid or liquid. This means that in coupling the sensor head to the steel tube to be monitored an air gap between the two and herewith total sound reflexion must be avoided. In the commonly applied ultra-sonic nondestructive material testing technique a coupling media between sensor head and test sample is used, this being generally water, oil or grease. Testing is also performed by submerging both sensor head and test sample in water.

Due to this, and according to what is said in chapter 3.4.2 tests were made with liquid coupling and by glueing the sensor to the tube by resin. For the liquid coupling a special adaption device has been developed (see chapter 3.5.5). In the glueing technique the echo delay column is made of steel, glueing this to the tube and mounting the sensor on top. Both techniques have shown identical satisfying results. It will be a question of long-time testing under radiation as to whether the resin changes its sound transmission characteristics. By an adequate lay-out of the adapter, the resin will not be submitted to mechanical stress. The main advantage of the liquid coupling is its great flexibility in readjusting the sensor, whereas the handling of any liquid in the severe plant surroundings is probably problematic. With the glued coupling the sensor head has to be adjusted very carefully because a readjustment is not easily possible.

3.5.2 Temperature influence

The fuel element dissolution in a reprocessing plant is done with hot acid to accelerate the dissolution process. Of interest to our problem is the temperature of the solution when this is transfered from the dissolver to the accountability tank. On the basis of information supplied by different plant operators it seems that the normal range of temperature is between 35-60°C /6/.

The technical data delivered with the sensor head showed that these can resist working temperatures upto 50°C. The manufacturer stated that this is due to the resin with which swinger cristal and the absorber are glued together, which melts at temperatures above 60°C.

In a test performed, the sensor was mounted on a tube filled with water which was heated gradually with an immersion heater. The sensor showed normal performance upto 60°C. Above that the echo signal was deformed gradually and at 70°C disappeared completely.

Due to information received from the manufacturer, he is willing to deliver a special sensor head which will be resistant against temperatures upto 100°C. Also this will be submitted to a performance test. The costs, as could be estimated at this moment, will not exceed significantly those for the normal sensor head.

3.5.3 Density influence

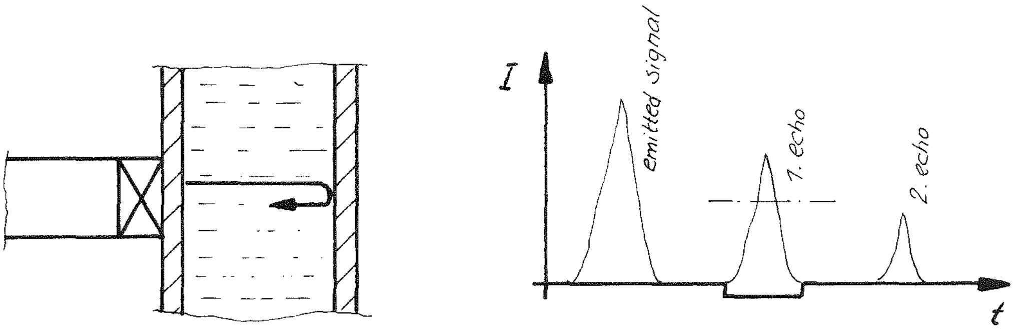
Equ. 3.1 shows that the sonic pressure P is amoungst others a function of the density of the material, through which the sound beam travels. To have an idea on the influence of this variable on our set-up, three different solutions of densities 1.0, 1.2 and 1.45 were monitored with the same adjustment. The results are shown in Fig. 17. The relative echo intensity changes between 80 % (density 1.0) and 110 % (density 1.45) and the relative echo time lapse between 65 % (density 1.0) and 50 % (density 1.45). This means that with increasing density we have a lower echo time lapse and a higher echo intensity. However, the responses are obtained with the same setting. The differences in the intensity and time may be utilised in differentiating among solutions of different densities e.g. with or without uranium.

3.5.4 Investigations of effects of mixed media (liquid/air) in the monitored tube on the echo signal

In practice the solution transfer is done with the help of steam jets or air lifts. Due to this the transfer tubes are not completely filled with liquid. The gas bubbles transported through the tube interrupt the echo signal when passing the sound beam. As these gas bubbles do not form a continuous interruption of the liquid column, the echo arrives in the form of pulsed signals.

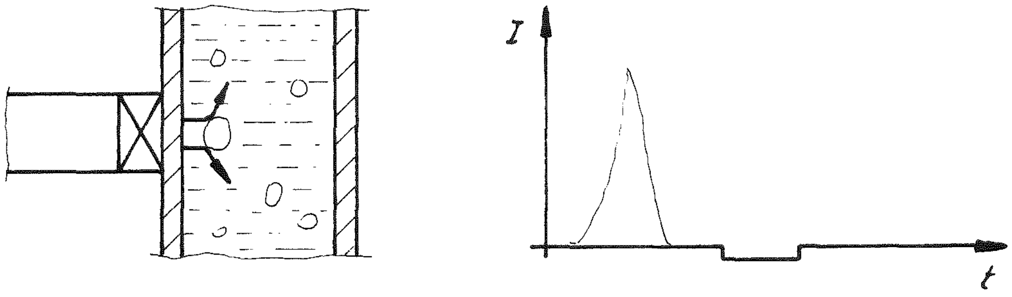
In a series of tests, these conditions were simulated by passing pressurized air through a tube filled with water. The results show that in very short time intervals the sound beam crosses the completely filled tube, creating then the desired echo. The minimum time needed for an echo to be registered as such by the electronic signal monitoring unit is very low compared with the frequency with which the air bubbles interrupt the echo signal. In other words, even with a ratio of air/liquid considerably higher than that ever to be encountered in a functioning air lift system, the monitoring sensor will give an unequivocal signal of presence of liquid in the tube. Fig. 18 gives a survey of the test array showing the echo signal at different air-throughputs. We have chosen a 30 mm diameter tube filled withwater and have taken the pictures demonstrating the CTR displayed echo signal at 12 l/h and 30 l/h air throughput respectively. On the left the signal is shown when the sound beam crosses the filled tube entirely, whereas on the right a moment is shown were the sound beam is interrupted by an air bubble. The very small peak visable in the gate is a small echo coming from the boundary surface water/air-bubble. The difference of height of the signals in the two left-hand pictures is due to the fact that the upper photo was taken in a moment when the sound beam crossed the tube diameter entirely, whereas in the photo below, the sound beam hit a small bubble passing near the rear wall (and is therefore not reflected with the same intensity).For a better demonstration the following schematic diagrammes are given:

1.



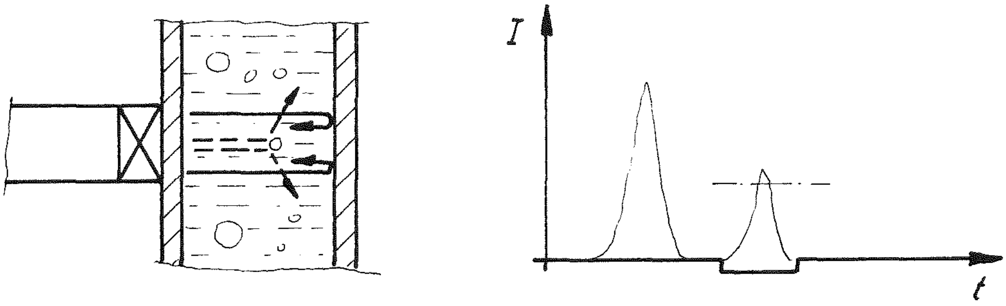
Entirely filled tube, alarm is triggered by the 1st echo signal

2.



Sound beam hits big bubble and is scattered, weak echo is not strong enough to trigger alarm.

3.

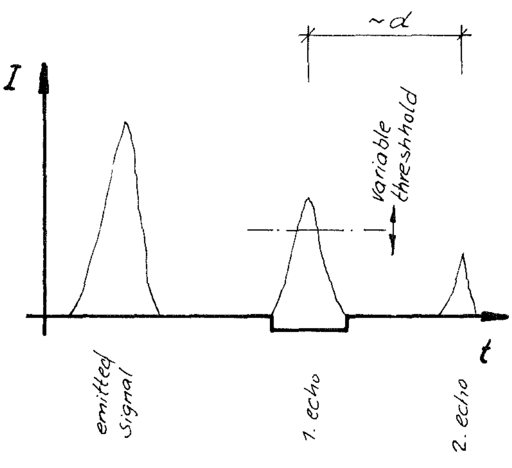


Sound beam hits (partly) small bubble, is scattered partly, the echo is weaker than in 1. but sufficient to trigger an alarm.

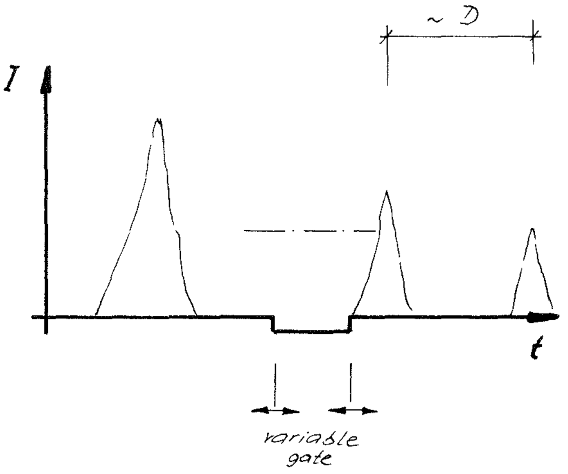
3.5.5 Investigations on monitoring different tube diameters

In the case of the WAK accountability tank the connected tubes carrying liquid range between ≈14 (= outer diameter) x 3 (= wall thickness) (sampling lines) to ≈34 x 4 mm (input feed and removal lines). In a series of tests it could be demonstrated that the tube diameter is of no influence on the quality of the echo signal.

On account of the fact that on the CRT a gate can be blended out so that the monitoring can be restricted to one defined echo signal, a way had to be found to shift the echo signal from different tube diameter to the same gate (see also chapter 3.4.2). The situation is shown schematically below:



Signal and positive registration



Signal, but no registration

As an electronical shifting of gate and amplitude threshold would have been far too costly, it was decided to create artificially a constant sound path irrespective of the tube diameter. The solution is shown schematically in Fig. 19. The gate is adjusted to the 1st echo of the signal coming from the tube with the biggest diameter (Fig.19A). At any smaller diameter the time lapse can be enlarged by a liquid foreleg (Fig.19C). In case that the smaller diameter is just one half of the biggest, the 2nd echo falls in the gate (Fig.19B).

To achieve this, an adaption device with great flexibility for laboratory needs was developed. Later on a prototype was developed which seems to be able to cope with the severe plant conditions. The device is shown in Fig. 20.

Furthermore, a series of pictures were taken to illustrate the situation, these are presented in Fig. 21.

3.5.6 Results on the simulation of transfers

The entire apparatus was assembled and the sensor heads coupled to the corresponding tubes as described in chapter 3.4.3. This instrument has proven to be adequate to a very large extent for the measurement problem considered. Over a long-time test run of 20 days without readjustment of either sensor heads or supply unit, there was no perceivable alteration of test results. Back-ground noises coming from pumps, electro-magnetic valves etc. were of no influence to the echo signals. It became evident, that inherently the tube monitoring could indeed serve as a cross-check for a properly performed input measurement. It was felt as a deficiency that a recording of the time sequence of the echo signals was not possible (preferable on the same read-out as the transfer record) with the given instrumentation.

4.0 Use of a tracer

4.1 Reasons for using the tracer method

The most common way of knowing the input to a reprocessing plant is by measuring level and density of the input solution in the input accountability tank with the help of a dip-tube system and determining the fissile material concentration of the solution. The corrected dip-tube readings are compared with a calibration curve established commonly by adding weighed increments of water to the tank and recording both the incremental amount and the respective readings on the level indicating instruments /7,8/. This method is generally reliable enough and its accuracy will meet safeguards requirements if measurement conditions are and can be carefully controlled.

However, the problems of safeguarding conventionally generated input measurement data are manifold: The safeguards inspector will have to be present when the tank is calibrated and when an input accountability measurement is performed in order to provide credibility to the data supplied by the plant operator.

The wish to assure the credibility of input measurement data will inherently lead either

1. to supply the present input measurement system with additional instrumentation so as to make it tamper-resistant, or

2. to apply an alternate and independent input measurement method which in turn could supply the safeguards authority with

   a) an independent mass determination of in-going fissile material,

   b) a verification of calibration data,

3. to use correlations.

A promising means to fullfill the condition a) to b) seems to be the use of the tracer technique as described in /9/. By this method, a known amount of an element used as a tracer is brought into the accountability tank, homogenized there and a sample from the homogenized solution is analyzed for the ratio of U to tracer or Pu to tracer if the amount of fissile material in the tank is required, or for the weight of tracer per volume unit of solution when verifying the calibration curve.

4.1.1 Independent mass determination of fissile material

To determine the total amount of fissile material in the tank only two data have to be generated:

a) the tracer to fissile material ratio and

b) the total amount of tracer added to the tank.

According to this, the total amount of fissile material can be calculated directly, without having to measure the solution volume:

$$F_W = W_T : r \qquad (4.1)$$

where:

F_W = weight of fissile material (U or Pu) in grams

W_T = weight of added tracer in grams

r = chemically analized fissile material to tracer ratio.

4.1.2 Verification of the calibration of the accountability tank

For the verification of the calibration curve, a known amount of tracer can be added to an amount of input solution in the tank present at any time. This amount corresponds to a particular point on the calibration curve. This point can then be determined by chemically measuring the weight of tracer per unit volume of input solution:

$$V_T = w_{TR_{tot}} \cdot \frac{V}{W_{TR}} \qquad (4.2)$$

where: V_T = total volume in the tank in liters

$w_{TR_{tot}}$ = total weight of tracer added to the tank in grams

$\frac{w_{TR}}{V}$ = the chemically analyzed tracer-volume fraction in grams/liter.

4.2 Relevant aspects to be considered for a tracer system under plant operating conditions

It is to be noted that in present day reprocessing plants normally no possibilities exist for the introduction of a tracer into the accountability tank. For the purposes of independent mass-determination the general trend is to keep the amount of tracer as low as possible ($<$ mg quantities). For the verification of calibration, there may not be any undue restriction on these amounts.

In principle, the tracer can be introduced either in the liquid or in the solid form. For small quantities of liquid, their introduction into the tank may be associated with some difficulties; it is quite possible that a part gets lost on its way or somewhere else in the tank and thereby causes a major error in the estimation. Although introduction in the solid form, particularly in a capsule, may not be associated with such difficulties, the use of a tracer may also be restricted because of prevailing plant conditions.

This short introduction is given mainly to draw attention to the fact that in connection with the use of tracer, the introduction system itself may pose a major problem. This has been found elsewhere also /9/.

A series of tests was undertaken to investigate the feasibility of using the sampling system to introduce the tracer in liquid form into the accountability tank.

About 0.5 g of methylene blue were introduced into a sample vial. This was then coupled to the needle block. After starting the circulation, the methylene blue was flushed out of the vial in about 10 min. time but it took about 2 h more to flush all of it down the sample return line. Due to the fact that the plant sampling system cannot be simulated to the last extent for space reasons it was felt useful to not further carry on with these tests, but to wait for results on finding a 'minor quantity' tracer and to repeat these tests if possible at the WAK plant.

It was then decided to perform the tracer experiment on two alternatives:

1. to find an adequate tracer which is used in larger quantities. Here losses during the introduction will have minor relative effects.

2. to find an adequate tracer which is used in minor quantities and which can be used to introduce in solid form to the tank (e.g. enclosed in a soluble capsule) provided however, that this is possible by the individual plant lay-out.

For the choice of an adequate tracer, further circumstances have to be considered and the tracer has to fulfill a number of conditions:

a) it should be easily introduceable to the tank,

b) for independent mass-determination, the tracer must be compatible with plant conditions, i.e. it should be extracted together with the fission products and it should not in any way alter or hamper the chemical process as such,

c) it should be measurable within the accuracy of the common volume-concentration determination,

d) it should be measurable by the same analytical methods used for the common volume-concentration determinations,

e) it should be inexpensive,

f) it should be easy to handle, should have good mixing characteristics (homogeneity) with the input solution and should not change its chemical properties during the process operations.

It would, however, by far exceed the scope of work set out in this Contract to investigate the different possibilities and aspects of using one or the other tracer and analyzing this by one or the other chemical method. This should be done on a laboratory scale in a chemical analytical laboratory.

The main interest of the tracer experiments carried out so far on the accountability tank mock-up was concentrated on the mixing and homogeneity behaviour of a tracer already in the tank. By the introduction plant conditions were simulated to the utmost possible extent.

After having pre-selected some 5 elements which seemed to comply to a large extent with the conditions mentioned before, it was finally decided to restrict the tests to using Zn as a "large quantity" tracer and Re as a "minor quantity" tracer.

4.3 Investigations carried out prior to tracer experiments

In combination with the actual tracer experiments, a series of related problems were investigated, like regional inhomogeneity, mixing effectivity of the air-sparger, effects of the recirculation time on the representativeness of samples taken and so on. Moreover, an accurate vessel calibration was done in order to compare the accuracies of both input determination methods. In the following, tests and results are described.

4.3.1 Calibration of the vessels

4.3.1.1 Calibration with water (density 1.0)

In order to be able to compare the accuracy of the tracer method with that of the volume-concentration method, careful calibrations of the two vessels were performed. In each tank 1 run with tap water and 3 runs with demineralized water were done. In Annex 1 the calibration procedures are described in detail and the results are given. Furthermore a unified calibration manual is proposed.

The results obtained show that the confidence region for the calibration process does not exceed ± 0.2 % from the measured value. For the process volume measurement after calibration the confidence region does not pass 0.7 % from the measured value.

4.3.1.2 Calibration with a solution of 1.5 g/ml density

Concern had been expressed as to whether it would be correct to calibrate the input accountability tank with water as the input solution thereafter measured has densities in the range of 1.5 g/ml. A higher deformation of the tank expected on account of the higher weight of the measured liquid might change the values in the calibration curve with water. In order to check this point, both the mock-up vessels were calibrated with a potassium carbonate solution of a density of about 1.5 g/ml. The corrected measured values (according to Annex 1) are shown in Table 2. The resulting slopes and intercepts of the calculated regression line are:

	Tank I		Tank II	
	slope [1/mm]	intercept [1]	slope [1/mm]	intercept [1]
total height	0.5247	7.66	0.5545	10.60
0 - 500 mm	0.5235	7.96	0.5551	10.47
500-1000 mm	0.5245	7.94	0.5547	10.37

A comparison with the data obtained from calibration with water shows no significant difference. All the calibration points in the range from 500 to 1000 mm of Tank II lie in the 95 % confidence range of the respective calibration values obtained with water (see Fig. 32).

These results would tend to show that a calibration of input vessels of the WAK-type with a solution of a higher density than water could be omitted. Nevertheless, this particular problem should be kept in mind, as these results depend strongly on the vessel dimensions and it might well be possible that vessels of other types could show load deformation influencing the calibration values.

4.3.2 Investigations on problems of solution homogeneity

4.3.2.1 Influence of sparger time on homogeneity

During the experiments carried out to introduce the tracer to the tank it became evident that the mixing and homogenization time was, at least visually, extremely short, so that a change in sparger design did not seem to bring any significant improvements. However, it was decided to mix two liquids with different specific densities and measure the mixing time to confirm the visual observations with the experimental results.

Originally it was planned to use the readings obtained from the two additionally installed density dip-tube systems to measure when homogeneity was reached, and to perform the entire tests in Tank II. During preliminary tests it was found more convenient, however, to perform the tests in Tank I so that the mixing procedure could be followed visually also. For this reason the dip-tubes were temporarily transfered to Tank I. Furthermore, the mixing process itself takes very little time, so that a follow-up of the change of 3 U-tube readings was hardly possible. On the other hand, the change of density down to values of 1 % is far below the possibility of detection by the dip-tube system. Moreover, the recorder print-out is unsatisfactory, being still more inaccurate and having too low a time axis to analyse the change of density with its help. It was therefore, decided to measure the change of electric conductivity of the solution during the mixing process and verify the homogeneity reached with the 3 dip-tube readings.

To this end, about 300 l of potassium carbonate solution with a specific density of about 1,45 g/ml were introduced into Tank I. Then about 300 l of water were added very carefully so that a pre-experimental mixing could be reduced to a minimum. The conductivity probe was submerged about 10 cm beneath the surface of the liquid and showed a slightly higher conductivity than tap-water. Moreover, the probe was introduced at a place into the tank where the air-bubbles are less frequent (due to the slope of the sparger), firstly because there the bubbles would produce less disturbances to the recorder readings and secondly because there the mixing time would be the highest.

The experiment was repeated with 200 l of potassium carbonate solution and 200 l of water in order to see how the mixing time depended on the quantity of solutions to be mixed. The data from the two experiments are compiled in Table 3 and the measured and calculated curves for the conductivity change [+)] are given in Figs. 22 and 23 whereas Fig. 24 shows the experimental set-up for the experiment.

The results show a dependency of the time constant on solution quantities. In the first case a time constant of T = 2.6 min was found, in the second T = 1.85. After some 7 time constants = 18 min (13 min) the calculated values differ 0.1 % from the asymptotic value. Actually the measured values seem to reach the asymptotic value somewhat sooner.

The U-tube readings were done after 10 min and 2 h after beginning of the mixing. No difference in the time sequence or in the reading between one of the 3 could be observed. One can say, that at least within the accuracy of the dip-tube system, no regional inhomogeneity of the density could be traced.

The results obtained indicate that with the existing sparger design mixing times in the range of 20 min. would be required to ensure proper homogenization. On the other hand, the test conditions have been - purposely - very extreme, so that in practice these mixing times may not be required unless specifically dictated by some operation reasons. Since in most of the plants of the type considered here, it is the normal practice to allow for about 20 minutes of sparging time, and this time does not form a bottle neck for the sequence of related operations, no further experimental activities were carried out on the design of a sparger in the framework of this contract. Some parameter study on this sparger design has been done in /2/.

[+)] The conductivity values are not absolute and show only the qualitative change in the solutions's conductivity. Furthermore the range has been standardized for time constant calculation.

4.3.2.2 Detection of spatial inhomogeneity with the help of tracer

Experiments to investigate the extent of inhomogeneities in the input tank solution carried out so far in working reprocessing plants could only be restricted to monitor inhomogeneities on a series of samples taken in timely sequence at one and the same sampling point in the input tank. The wish to know something about regional inhomogeneities distributed over the whole tank led us to install additionally 4 sampling devices, so that samples could be taken at 5 different points in the tank.

In total 2 runs were performed in which some 550 l of demineralized water were filled into the tank and some 550 g of Zn were added as a tracer. The solution was acidified to 0.1 M with nitric acid. Per run 5 samples were taken from the sampling lines positioned at various points A to E of the tank.

Chemical analysis gave the following results:

Samples from position	A	B	C	D	E
1st run	1.004	0.980	0.972	0.984	0.953
2nd run	1.078	1.036	1.036	1.025	1.048

In order to determine inhomogeneity the following statistical analysis was performed, following two steps:

a) Test of significance to find out whether the data reveal inhomogeneity or not.

b) In case of inhomogeneity estimation of the order of magnitude of the inhomogeneity.

a) Test of significance

The test of significance consists in the construction of a significance threshold x_c for the differences between the single measurement results as a function of the accuracy of the single measurement and the error of the first kind α. This relation is given by

$$\Phi\left(\frac{x_c}{\sigma_D}\right) = 1-\alpha$$

where σ_D is the standard deviation of the difference of the two single measurements and Φ is the Gaussian distribution.

For $\alpha = 0.05$ and $\sigma_D = \sqrt{2} \cdot 0.005$ (± 0.5 % was the standard deviation of the chemical analysis reported by the analyst) one obtains

$$x_c = 0.0117.$$

As the maximum differences of the single measurements for each test run are greater than this threshold value, inhomogeneity in the tank solution, errors in sampling or errors caused by geometrical restrictions etc. (expressed in general with the word 'inhomogeneity') have to be assumed.

As in the 2nd run the data for the single repeated measurements were available, a test of significance has been performed in the course of which all the data obtained in the 2nd run were used (not only the extreme values as above). The results from this run also confirmed that inhomogeneity or other sources of errors as in the previous case have to be assumed.

b) Estimation of the inhomogeneity

The standard deviation of the inhomogeneity is estimated by an analysis of the variance-type calculation. (As the repeated measurements of the single samples are not reported, only the reported value of the reproducibility can be used.) It should be well noted, however, that it is not allowed to calculate the mean value and the standard deviation because the measurement contains error and inhomogeneity. For this reason the estimated measurement error must be deducted.

The estimate of the variance of the inhomogeneity $\hat{\sigma}_I^2$ is given by

$$\hat{\sigma}_I^2 = \frac{1}{p - 1} \sum_{i=1}^{p} (y_i - \bar{y})^2 - \frac{1}{n} \sigma_r^2$$

where p is the number of samples (p = 5), y_i is the reported value of the concentration of the i-th sample, $\bar{y}$ is the average value of the p sample concentrations, n the number of repeated measurements per sample (n = 2) and $\frac{1}{n} \sigma_r^2$ the variance of the reported measurement per sample ($\frac{1}{n} \sigma_r^2 = 0.005^2$).

These analyses lead to the values of

1.	1.78 %
2.	1.97 %

as inhomogeneity in the tank.

As mentioned above in the second run the data for the single repeated measurements were available. With the help of these data estimates of the variance of the inhomogeneity (σ_I^2) and of the reproducibility of the chemical analysis (σ_r^2) as well as confidence intervals for these estimates could be established. The following estimates were obtained:

$$\sigma_I = 2.06 \cdot 10^{-2} \qquad \sigma_r = 0.42 \cdot 10^{-2}$$

The 95 % confidence intervals for σ_I and σ_r are

$$0.588 \cdot 10^{-2} \leq \sigma_I \leq 5.46 \cdot 10^{-2}$$

$$0.27 \cdot 10^{-2} \leq \sigma_r \leq 1.05 \cdot 10^{-2}$$

A survey of the analytical results and the statistical analysis is given in table 4. It is to be noted that these values of inhomogeneities depend on the value of the standard deviation of chemical analysis assumed. They will naturally be lower if the actual values of standard deviation were to be higher. Furthermore it should be noted that the samples taken may not be fully representative of the tank solution on account of local concentration inhomogeneities in the sampling lines. It is therefore, desirable to continue investigations on this problem.

4.3.2.3 Influence of recirculation time of sampling system

In order to determine the time needed to circulate the tank solution through the sampling system to obtain a representative sample from the tank, a series of samples were taken consecutively. The tank was filled with about 550 l of demineralized water, slightly acidified with HNO_3 (≈ 0.1 $\underline{M}$) and about 550 g of Zn were added as a tracer. Chemical analysis gave the following results:

Sample No.	Circulation time	Calculated tracer concentration [gZn/g sol.]	Analysed tracer concentration [g Zn/g sol.]
1	5	$\frac{536 \text{ g Zn}}{560.6 \text{ kg sol.}}$ = 0.957	0.647
2	10		0.889
3	15		0.961
4	30		0.972
5	45		0.953

Fig. 25 shows graphically the increase of Zn-concentration over the time.

Comparing these results, we can say that after 15 min. of circulation we get a representative sample. This value is, of course, restricted to our experimental set-up, depending mainly on the solution through-put through the sampling device.

Here again we can at first glance suspect inhomogeneity when looking at the results of the last 3 samples. In applying the equations from chapter 4.3.2.2

$$\sigma_I^2 = \frac{1}{p-1} \sum_{i=1}^{p} (y_i-\bar{y})^2 - \frac{1}{n} \sigma_r^2$$

we calculate an estimated inhomogeneity of

0.8 % .

This result, together with those obtained in chapter 4.3.2.2 show that significant inhomogeneity may exist in the tank solution. It seems to be of importance to investigate these phenomena in greater detail, as similar experience on routine input analyses has been reported elsewhere /10/.

4.4 Experimental results of tracer tests

4.4.1 Compiled data on the Zn-experiments

A series of tests were performed using Zn as a tracer. This was added in quantities to produce tracer concentrations of ≈1 g/l of tank solution, leading to the added quantities of max. 800 g Zn per input batch. With these fairly high quantities the amounts lost in the introduction to the tank may be considered to be relatively negligible.

It was intended to verify two points of the tank calibration curve namely, at about 350 l and 550 l respectively. The procedure was to fill the tank to approximately these volumes with demineralized water, which was then slightly acidified with HNO_3 (≈ 0.1 M). **Zn** was used in form of $Zn\,Cl_2$, because of its easy availability. The $Zn\,Cl_2$ was carefully weighed and analysed for Zn. It was then dissolved in diluted HNO_3 and drained down into the tank from the upper floor through a plastic tube, which was rinsed afterwards with water. This was the utmost which could be done to simulate plant conditions.

With the U-tube readings the volume was calculated according to the following equation (see Annex I):

$$V = \frac{\rho_M}{\rho_T} \left[(h_1+h_2)\cdot 0.5571+10.33 \right]$$

(The densities of the U-tube liquid (= demin. H_2O) and the tank solution being both 1, the term $\frac{\rho_M}{\rho_T}$ is neglected.)

The calculated volume is then corrected to mass by multiplying it with the respective density to eliminate the temperature influence. The weighed tracer amount is then divided by the analysed tracer concentration [in g Zn/kg solution] to obtain the mass content of the tank in this manner. This is then compared with the calculated content obtained through U-tube measurement.

For ready reference, the calculated mass content difference between the dip-tube and tracer method are given below. They were calculated according to

$$D = (1- \frac{G_{TR}}{G_U})\cdot 100 \quad [\%]$$

run	1	2	3	4
D [%]	+0.34	+0.32	1.)+2.28 2.)+0.62	+0.88

In the first two runs 1 sample for each was taken from position E.

In the 3rd run 5 samples were taken consecutively from position E after 5, 10, 15, 30 and 45 min. of circulation respectively. Furthermore, also after 45 min 1 sample was taken from each position A, B, C and D. The first value (2.28 %) given above for this run was calculated with the mean concentration value from samples A, B, C, D and E_{45} (= sample taken after 45 min. of circulation). This value is fairly high and reference is made to comments on the concentration value deviations in chapters 4.3.2.2 and 6.0. The second result (0.62 %) is obtained by taking the mean concentration value from samples E_{15}, E_{30} and E_{45}.

In the 4th run, an effort was made to verify the Zn-factor in $ZnCl_2$ supplied by the manufacturer by analysis at the chemical laboratory of the Research Center. The two values show a deviation of 0.13 %. Furthermore 5 samples were taken, 1 each from positions A, B, C, D and E. The difference shown above (0.88 %) was obtained by using the mean concentration value of the 5 samples taken.

The experiments done so far show encouraging results (apart from the 1st value of run 3), the difference between the two volume determination methods being less than 1 %. Nevertheless, runs 3 and 4 have clearly indicated, that there is a major source of error which may be caused by analytical error, spatial tank solution inhomogeneity, sampling error, geometrical or design insufficiencies, etc. Further efforts are required to clarify the causes of this source of error.

Furthermore, the results clearly indicate a positive bias either for the chemical analysis, by which always a higher tracer concentration is found, or for the volume determination by U-tube which always indicates a higher volume. Here also further investigation is necessary.

4.4.2 Error analysis

As could be demonstrated in Annex I, the confidence interval for a single vo-volume V measured with the U-tube method is given by

$$(V-\xi,\ V+\xi) \qquad \text{(AI.7)}$$

where

$$\xi = c\cdot\frac{s}{|b|}\sqrt{1+\frac{1}{n}+\frac{(V-\bar{V})^2}{\sum\limits_i^n (V_i-\bar{V})^2}} \qquad \text{(AI.8)}$$

For an estimation of the error of the tracer method, we use the following equation:

$$\delta_G^2 = \underbrace{\delta_{m_s}^2 + \delta_{m_r}^2}_{\text{weighing error}} + \underbrace{\delta_{c_s}^2 + \frac{\delta_{c_r}^2}{n_c\cdot n}}_{\text{concentration analysis error}} + \underbrace{\frac{\delta_c^2}{n}}_{\text{inhomogeneity}}$$

where δ_m and δ_{mr} are the standard deviations of the systematic and random weighing error (which can be neglected in the following because they are small compared to the other error components). δ_{c_s} and δ_{c_r} are the standard deviations of the systematic and random concentration analysis error, n_c is the number of analyses per sample and n is the number of total samples taken per batch. δ_V is then entered in the following equation

$$1.96\ \sigma_{TR} = \delta_G \cdot 1.96 \cdot G_{TR} \cdot \frac{1}{100}$$

In the following δ_{c_s} is estimated to be 0.25 %, δ_{c_r} is given with 0.5 %.

For the tracer runs performed so far, the following relative standard deviations δ_G and the half lengths of the confidence intervals 1.96 σ_{TR} have been calculated.

run	1	2	3A	3B	4
δ_G /‾%_7	1.8	1.8	0.93	1.06	0.63
1.96 σ_{TR} /‾kg_7	19.6	13.3	9.9	11.6	7.1

The half lengths of the confidence intervals for the corresponding U-tube volume determinations are given in Annex I.

In Fig. 28 the results obtained by the two volume determination methods are represented graphically. Presuming that both methods deliver exact values, they should all lie on the 45^0 line. The crosses formed by the confidence intervals and the regions covered by them give an idea on the accuracies of the two measurement methods: If the 45^0 line is touched or covered, the results obtained can be considered as satisfactory.

Again it is evident, that (with the exception of run 3A) the obtained values may be within the confidence interval.

4.4.3 Compiled data on the Re-experiments

In the search for a sensitive analytical method capable of analysing amounts down to some ng (= 10^{-9} g) it was felt that the neutron activation analysis could give satisfying results. Re was added to the tank to produce Re concentrations of some 25 ng/ml. Again the aim was to verify two points (350 and 550 l) of the calibration line. Test conditions were the same as with the Zn experiments.

In analysing the sample for Re, the sample is exposed to a neturon flux together with a sample of a known Re concentration. Through neutron capture, part of the ^{185}Re is built up to ^{186}Re which decays with a half life of 90 d, emitting a typical 0.14 MeV γ-line. A comparison of the peak intensities of the known with the unknown sample leads to the knowledge of the Re concentration.

In table 6 the obtained data from the 4 runs with Re are compiled. Again the deviation values for the two volume determination methods are given below for ready reference. They were again calculated, using the following equation to the values listed below:

$$D = (1 - \frac{G_{TR}}{G_U}) \cdot 100 \quad [\%]$$

run	1	2	3	4
D [%]	-7.19	+ 2.1	+ 5.32	+11.42

The variations in the difference D are still rather large and should be considered as preliminary as this is the first and only set of data available for the moment. The runs were performed in the time sequence as indicated in the above list, and although very careful rinsing was done in between, there is a clear indication of an accumulation of Re (perhaps on the tank walls?) deducted from the fact that the Re concentration in the samples becomes higher with each subsequent batch. Nevertheless, the data have been graphically shown in Fig. 29, giving the points measured by both the U-tube and the Re-tracer method. Also the confidence interval for the U-tube volume determination method is shown. An error analysis for the Re-tracer method has not been done on account of lack of sufficient data.

4.4.4 Remarks on the results obtained on the application of tracers

The results obtained on the application of Zn as a tracer for volume verification appear to be encouraging. The amount used, about one kg per calibration may be reasonable. The differences obtained lie in a range (0.6-1.8%) which may be considered to be useful for practical application. The accuracies obtained however, have to be tested under plant operating conditions. The use of Zn as a tracer for independent determination of uranium amount in the accountability tank appears to be doubtful at the present stage of development.

The use of Re as a tracer can be commented upon only after the method of analysis has been developed further. In view of the fact that this method is at an initial stage of development the relatively large standard deviations (-7 % +11%) obtained so far should not be considered to be a disadvantage for the method. A large number of factors not directly associated with the measurement method (absorption on the surface, accumulation, faulty sampling, irregular dissolution rate etc.) may influence the accuracy.

5.0 Tampering of level and density indicating system

The level and density measurement system can generally be divided in the following 3 categories (see Fig. 26):

a) The data generation: the dip-tubes in the tank and the air supply lines.

b) The data transmission: the pneumatic lines leading to the transmitters and to the U-tubes, the transmitters themselves and the electronic conduits leading to the recorders.

c) The data indication: the U-tubes and recorders.

A tampering at any of these three points will influence the material balance. A number of tests were performed to locate the sensitive points in the whole system:

a) The dip-tubes themselves being not easily accessible, are relatively immune to tampering. The air supply lines, however, together with the air flow rate meters are very sensitive to tampering. A test was performed by merely changing the air-throughput at the flow meters. The flow rate in the tubes I to III was altered consecutively, leaving the respective two others at a constant rate of 10 l/h. In Fig. 27 the values for the altered air flow rate in l/h are drawn up over the values for the change in U-tube readings in mm. The curves show that any level or density reading can be simulated. It is either possible to simulate entire transfers without that the solution has been moved, or to supress completely any indication of movement of solutions.

b) The data transmission lines are exposed to a large number of possibilities of tampering: cutting of the pneumatic lines to superimpose an altered signal, direct or inductive alteration of the electric signals, tampering of the power supply or transmission characteristics of the transmitter, etc. One example may illustrate this point.

As shown in Fig.8 for Tank I (and respectively in Fig. 9 for Tank III), a number of valves are foreseen in the dip-tube system which serve to close the system for a test of leakproofness (see Annex I). If the MV8, MV9 and MV10 are not opened properly, the back pressure leads to falsified level and density readings.

c) The U-tubes themselves have not been investigated for tampering as they may normally be read in presence of the inspector and an eventual tampering could have a chance of being detected. The recorders, however, were investigated. It was found for example that in diminishing the tension, the indication needle remains at its last position, whereas the paper transport motor still works down to relatively low tensions (≈ 70 % of max.). This gives the possibility to perform transfers that will not be registered on the recorder.

The examples given here illustrate mainly the point that it is technically not feasible with reasonable means to monitor the data system at the input in such a way as to detect an intentional tampering. A much better way of safeguarding the input material appears to be the use of correlations or suitable tracer techniques.

6.0 Conclusions and recommendations for further work

The present investigation was undertaken to analyse in detail three areas of interest at the input of a WAK type reprocessing plant. They were: a) Liquid sensing devices for the indication of the presence or absence of liquid in pipelines from and to the accountability tank; b) Tracer techniques for the independent measurement of fissionable materials in and verification of the volume calibration curve for the accountability tank and c) Sensitiveness to tampering of the information system for the accountability tank.

6.1 Conclusions

All the investigations have been carried out in the experimental set-up for the WAK-type accountability tank under inactive laboratory conditions. Before some definite recommendations or conclusions can be made for the use of one system or the other for safeguards purposes under industrial conditions, the feasibility of the application of such systems under ratio-active and operating conditions has to be checked. However, a number of conclusions of general interest can still be drawn subject to this restriction:

6.1.1 Liquid sensors

The commercially available ultra-sonic single head sensor of the type NT 4/10 K gives a clear-cut yes-no indication on the presence or absence of liquid in a pipe attached vertically to the accountability tank. The liquid phase has to be continuous from wall to wall inside the pipe (for vertical mounting) to obtain the echo signals through a preset gate. The gate setting can be kept unaltered (which is a great technical advantage) for different pipe diameters by attaching a compensating variable liquid leg to the sensing head. The echo intensity and the echo time lapse vary by more than 20 % for liquid densities between 1 and 1.45 g/ml, although the same gate setting can be used. These differences in intensity and time lapse could probably be used to differentiate between liquids with or without uranium. The liquid sensor system is simple to mount and maintain, is robust in nature and remains stable over a long period of operation. The signal to noise ratio is very good.

The unit used was found to be stable only up to 50 - 60 ^{0}C, above which no signal was obtained probably because of the disintegration of the resin used in the sensing head. The sensor has not been tested for its functions under irradiation. It has also not been checked for horizontal pipes since normally, all the pipelines leaving or entering the accountability tank of a WAK-type reprocessing plant go through a vertical position. Also the possibility of installing such sensors remotely in highly radio-active environment has yet to be investigated. No registration unit was used in the present set-up to register the echo signals but only an oscilloscope. This was found to be a disadvantage particularly during the analysis of the time sequence of solution transfers.

6.1.2 Tracer techniques

A number of additional investigations had to be carried out before the feasibility studies on the tracers could be started. They included a careful calibration of the tank with liquids of different densities, homogeneity studies with conductivity measurements, density measurements at different points inside the tank and with concentration measurements (with Zn) at different sampling points and finally studies on the recirculation time in the sampling system and the representativeness of samples taken.

The important point in this series of investigations was the fact that a certain inhomogeneity (1.5 - 2 %) in the tank solution could be concluded to exist on the basis of results obtained from zinc measurements, provided that a reproducibility of about 0.5 % was assumed for this measurement. Since the differences in the calculated values giving rise to inhomogeneity may be influenced among others, by the sampling errors, other values of reproducibility for the chemical analysis, geometrical registrations etc., further investigations are necessary before something more definite can be said about the inhomogeneity in the tank solution.

The U-tube calibration error of ± 0.7 % (2σ value) appears to be in the range of values commonly found in industrial practice. The verified values using Zn as tracer were found to be always higher (+ 0.34 % to + 2.91 %) than the corresponding values obtained by U-tube calibration. Further investigation on this point is necessary. The actual calibration error using zinc as a tracer was found to be 0.6 - 1.8 % (2σ), the major contribution coming from the error on account of inhomogeneity in the tank solution. Without the inhomogeneity error, this calibration error with Zn as a tracer reduced to ± 1.0 % (2σ). These ranges of errors, with or without inhomogeneity, are such that this tracer method may be considered to be suitable for the verification of the volume calibration error for the accountability tank. However, the feasibility of its application for safeguards purposes has to be checked carefully under plant operating conditions.

The apparent inhomogeneity in the tank solution as found on the basis of chemical analysis of Zn may be caused by a number of factors,as mentioned above. Since, inhomogeneity, if it does exist, will influence the overall accuracy of mass-determination at this point, it appears essential to continue investigation on this point in the present set-up.

The series of tests with Re is at its initial stage of development. The high range of measurement errors obtained (-7 % +11 %) may be due to a number of factors (inhomogeneity, absorption on the surface of the tank, incomplete dissolution, sampling errors etc.) which may not be associated directly with measurement errors. However, the analytical method itself may require further work for standardization for routine use. The main advantage of this method lies in the fact that very small amounts can be used and that the measurement accuracy is expected to be high.

6.1.3 Sensitiveness of the input information system to tampering

The analysis of the tamper resistance of all the three components (generation, transmission and indication) of the information system for the accountability tank indicate that with a few exceptions, all of them can be tampered with varying degrees of ease. However, it does not appear justifiable to investigate technical means of making them tamper resistant. This is because of the fact that sufficient redundant information in the form of isotopic and other correlations, proper use of tracers etc. are available and may be utilized to the fullest extent to improve the credibility of or verify the information obtained from the information system at the input of the accountability tank.

6.2 Recommendations for further work

The experience gained so far in the operation of the experimental set-up and analysis of the results indicate clearly that further work in this context should be continued under the following three conditions:

6.2.1 In the present experimental setup

6.2.2 In a radio-chemical laboratory

6.2.3 In a reprocessing facility

6.2.1 In the present experimental setup

a) Inactive work on liquid sensors

Although the use of the ultrasonic liquid sensor has been demonstrated in principle under inactive conditions, some further work is desirable to refine and extend the use of the sensors. They are among others:

i) Improvement and testing of the sensor head for operation under higher temperatures than 60^0 C in collaboration with the manufacturer of the sensor. It would be recalled that the resin used in the head was probably not stable above this temperature.

ii) Testing of the sensor system for horizontal pipelines. It is quite possible that a different signal system (e.g. time of flight instead of echo method) would be more advantageous for this purpose.

iii) Testing of a double-head sensor system which promises to offer some inherent advantages over the single head type.

iv) Investigations on whether the differences in the intensity and the time lapse for the echo signals for different densities could be utilized to distinguish clearly between liquids with or without uranium. This finer differentiation might even be useful for plant operation as in that case flows of acid, wash or dissolver solutions could be differentiated in a quantitative manner by registering the sequence of transfers with the help of the liquid sensing system.

b) Inactive work on tracers

Different types of investigations are still necessary in the present setup for further clarifications on the use of tracers for the purposes mentioned at the beginning of the report. They are for example:

i) Careful analysis of the inhomogeneity problem for large amounts of tracers (Zn) - on its cause, its components and possibilities of improvement. Also investigations on the attainable accuracies under the experimental setup conditions.

ii) Investigations on different introduction mechanisms for small amounts of tracers (Re) and the attainable measurement accuracies under the conditions of the experimental setup.

6.2.2 In a radio-chemical laboratory

a) The major parts of the future work is expected to involve development and standardization of various analytical methods for tracer elements like Zn and Re in a radio-chemical laboratory.

b) Investigations with the present sensor head under highly radio-active environment corresponding to that present in the input tank of a reprocessing plant (e.g. with a cobalt source in a hot cell) and if required, introduction of modifications in the sensor head in collaboration with the manufacturer to make the head useable under highly radio-active conditions.

6.2.3 In an operating reprocessing facility

Both the liquid sensor system and the proposed tracer methods have to be tested in an operating reprocessing facility. For this purpose all the investigations enumerated under 6.2.1 a) and b) as well as under 6.2.2 b), will have to be repeated in such a facility.

6.3 Concluding remarks

Because of the importance attached to the sphere of activities on and around an accountability tank, in the framework of the safeguards project Karlsruhe, the recommended activities will be carried out even after expiring of the present contract with the IAEO. As far as feasible, the results of such future activities will also be published in the usual way.

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Annex I

AI.0 Tank calibrations

In order to be able to compare mass or volume determination by the tracer technique with the commonly used input measurement (volume-sampling-chemical analysis) a very careful calibration was performed on both Tank I and Tank II. The technique used was the "incremental calibration" technique as described in /7/ and /8/.

AI.1 Calibration of the balance

The balance used is manufactured by TOLEDO, Cologne, model 0851, with a range from 0.4 to 30 kp in 1 step and a standard deviation of ± 10 p (= 1/2 scale division). The balance was adjusted prior to calibration by the manufacturer. The calibration was done with standard weights from 10 p to 5 kp in steps of 5 kp. There was no difference perceivable between standard weight and balance indication.

AI.2 Zero-point correction

For the level indication, a U-tube of 1.5 m length filled with demineralized water was used. Due to unequal temperature distribution in the two U-tube legs an indication different to zero can occur at balanced pressure conditions. All readings have then to be corrected to zero. In all calibrations done on both tanks no deviation from zero indication could be perceived.

AI.3 Leakage test

In order to detect eventual leakages which would falsify measurement results, a leakage test was performed on the system. Numbers and symbols used are shown in Fig. 8 for Tank I (corresponding values for Tank II are from Fig. 9 and are given in brackets).

1. Valves MV3, MV4, MV5 (MV11, MV12, MV13, MV14, MV15, MV16, MV17) were closed. With this the air supply was interrupted.

2. Valves MV8, MV9, MV10 (MV22, MV23, MV24, MV25, MV26, MV27, MV28) were closed. This means a closing of the dip-tube ends.

3. The by-pass valves MV6, MV7 (MV18, MV19, MV20, MV21) were closed.

4. Alternatively the valves MV4, MV5 (MV12, MV13, MV14, MV15, MV16, MV17) were opened to produce desequilibrium in the U-tubes.

5. The U-tube indications are noted.

6. After a certain time U-tube indications are compared with the ones taken before. In case of a difference, a leakage exists in the system.

7. All pneumatic connections are moistened with NEKAL (or similar) to locate the leakage.

8. The procedure is repeated in case of necessity.

AI.4 Calibration of the vessels

The set-up for the calibration can be seen schematically in Fig. 30. The vessels were filled in batches of some 24 kg. No special attention was given to the lower or upper part of the vessels as they are completely cylindrical and the dip-tubes do not reach down to the unlinear part. U-tube readings were taken after each added batch of water and the temperature noted.

AI.5 Evaluation of calibration data

AI.5.1 General comments

Each tank was calibrated with 1 run with normal water, 3 runs with demineralized water and 1 run with potassium carbonate solution of a density of about 1.5 g/ml (see chapter 4.3.1.2.).

First of all the corrected heights and volumes are calculated from the measured values:

$$h_i = \frac{\rho_M}{\rho_T} (h_{L_1} + h_{L_2})_i \qquad [\text{mm } H_2O] \qquad (AI.1)$$

$$V_i = \frac{1}{\rho_T} \sum_1^i \Delta G_j \qquad [1] \qquad (AI.2)$$

wherein ρ_T = density of the solution in the tank

ρ_M = density of U-tube liquid

h_{L_1}, h_{L_2} = U-tube readings in mm

In table 7 the corrected values for the run with tap water are given, for both Tank I and Tank II.

Table 8 shows the corrected values for Tank I, Table 9 those for Tank II, both for the 3 runs with demineralized water.

With the corrected values a linear regression according to the least square method was performed. Herein the error in the independent variable (calibration volumes, weighing error = 10 g) are negligible compared with the error in the dependent variable (height, reading error = 0.5 mm $\hat{=}$ 250 g).

AI.5.2 Linear regression

Linear regression was performed over all the measured points of each single run. Additionally, the height was divided into 3 regions, from 0 to 500, 500 to 1000 and 1000 to 1500 mm. A linear regression was then performed over each of the 3 regions. This was done in order to consider the effects of geometrical tank deformations and tank inserts on the linearity of the regression line. The calculated slopes in 1/mm and the intercepts in 1 are given in the following tables.

Tank I

	1st run		2nd run		3rd run	
	slope	intercept	slope	intercept	slope	intercept
total height	0.5251	7.2	0.5242	7.67	0.5261	6.81
0-500	0.5225	8.13	0.5236	8.28	0.5263	6.91
500-1000	0.5270	5.46	0.5254	6.13	0.5261	6.59
1000-1500	0.5305	2.73	0.5290	1.66	0.5305	1.02

Tank II

total height	0.5537	10.40	0.5538	10.13	0.5537	10.75
0-500	0.5551	10.07	0.5546	9.97	0.5545	10.59
500-1000	0.5534	10.42	0.5535	10.35	0.5544	10.12
1000-1500	0.5534	10.86	0.5538	10.12	0.5517	13.27

For the calculation work done for the linear regression, a small program was used on the DIEHL COMBITRON S 10, which reduced evaluation effort to a great extent. The program is shown in Table 10.

A significant difference cannot be observed in the values shown in the tables above, neither between the different runs nor between the different regions. Only in Tank I there seems to be a slight geometrical deformation in the upper region of the tank. In order to analyse the question whether it is legitimate to perform the regression over the entire run, the deviation of the measured points from the calculated regression values for the entire run as well as the values for each of the 3 regions have been entered into the graph shown in Fig. **31**. The values shown are from the 1st run with demineralized water in Tank II and are given here as an example. It can be seen that a significant difference does not exist, even though the values obtained from linear regression done over the single regions are somewhat closer to the regression line. Due to these results it seems legitimate to perform the regression over the entire run; this seems reasonable as no tank inserts exist excepting at the top or bottom of the tank and no geometrical deformation has been found.

AI.5.3 Error analysis

AI.5.3.1 Basic considerations

As it is expected to perform the tracer experiments in Tank II and there with tank fillings of about 400 l it was decided to do error analysis on the middle region of the regression line of Tank II. Firstly the mean values of the slopes and intercepts of the middle regions of all three tanks were calculated and compared with the values obtained from linear regression over all points of the middle regions of the 3 runs together. In the table below the values are given:

	mean values	calculated values
slope /̲l /mm̲/	0.5538	0.5571
intercept /̲ l̲/	10.30	10.33

As can be perceived, the difference is not significant but as in statistics it is more correct (in the sense of unbiased estimates) to use the calculated values, they have been used in the following.

AI.5.3.2 Analysis of the calibration line error

Under the assumption that the error of the dependent variable (h) is independent of the value of the independent variable (V) the estimate of the variance of the dependent variable is given by the following formula /11/.

$$s^2 = \frac{1}{n-2} \sum_{i=1}^{n} (h_i - H_i)^2 \qquad \text{(AI.3)}$$

Here, h_i is the measured value, corresponding to the calibration volume V_i; and H_i is the value lying on the regression line and corresponding to the volume V_i. H_i is determined by

$$H_i = \frac{V_i - V_o}{a} \qquad \text{(AI.4)}$$

where a and V_o are the slope and the intercept of the regression line.

One obtains (as stated before, for the middle region and the lumped values of all three runs)

$$s^2 = 0.8748 \qquad s = 0.9353$$

This variance is important for all kinds of error propagation (see the tracer experiments).

In the following, a confidence region for the calibration line shall be constructed.

With respect to a single calibration volume V the confidence interval for the height H is given by

$$(H-c\sqrt{S^2},\ H+c\sqrt{S^2}) \tag{AI.5}$$

where S^2 is given by

$$S^2 = s^2\left(\frac{1}{n} + \frac{(V-\overline{V})^2}{\sum_i (V_i-\overline{V})^2}\right) \tag{AI.6}$$

and s^2 is given by equ. AI-3 . The value of c depends on the confidence $1-\alpha$ and is determined by the t-distribution with n-2 degrees of freedom.

In table 11A for a given V, the half width $c\sqrt{S^2}$ of the confidence interval for the calibration process is shown. In Fig. 32 , the confidence region for this process is also shown. As can be seen, for the shown part of the calibration curve, the confidence region for the calibration process does not exceed ± 0.2 % from the measured value.

AI.5.3.3 Analysis of the error of the process: calibration plus measurement of an unknown volume

It is clear that the error of the process: calibration plus measurement of an unknown volume must be larger than the error of the calibration alone, as the uncertainty of the U-tube reading happens twice in this process. This is reflected by the confidence interval for this process which is given for a single volume V by

$$(V-\zeta,\ V+\zeta) \tag{AI.7}$$

where
$$\zeta = c \cdot \frac{s}{|b|}\sqrt{1+\frac{1}{n}+\frac{(V-\overline{V})^2}{\sum_i (V_i-\overline{V})^2}} \tag{AI.8}$$

As above, c is determined (for a given confidence $1-\alpha$) by the t-distribution with n-2 degrees of freedom. In table 11B for a given V, the half width ζ-V of the process: calibration plus measurement is given for different values of V. In

Fig. 32, the confidence region for this process is also shown. As can be seen, for the shown part of the calibration curve the confidence region for the process: calibration plus measurement of an unknown volume does not exceed ± 0.7 % from the measured value.

In principle, the once calculated confidence regions have not to be repeated when having performed another calibration. c is quite insensitive to a change in the number of degrees of freedom and n does not alter the value of ζ significantly. It is therefore allowed to transfer the confidence regions to another set of calibrations (performed under the same conditions).

AI.6 Recommendations for a standardized calibration program

To ensure a careful and thorough calibration, the recommended proceedings of a calibration run are given in the following:

1. Calibrate the balance to be used for the weighing of the calibration increments according to AI.1. If necessary, call for a re-adjustment of the balance. In case of indication deviating from calibration value, correct the measured increments according to

$$G_j = G_M \pm \Delta g$$

   wherein G_M is the measured value and Δg is the difference between the indication and calibration value.

2. Make the zero-point check on the U-tubes according to AI.2. In case of deviation all readings have to be corrected accordingly.

3. Perform the leakage test of the U-tube system according to AI.3.

4. Fill the weighed increments of water into the tank and note the U-tube readings for level and density and the temperature of the water in the tank. At least, 3 calibration runs should be performed.

5. Enter your readings into equ. AI.1 and equ. AI.2 and obtain your corrected values for height (h_i) and volume (V_i). Note that the density values are temperature dependent and have to be corrected according to the temperature measured in the tank and at the U-tubes, e.g.:

temp. [°C]	density [g/ml] for H_2O
20	0.99823
21	0.99802
22	0.99780
23	0.99756
24	0.99732
25	0.99708

6. Perform linear regression according to table 10.

7. Divide the tank height into 3 regions, perform linear regression over each of these regions and compare the results with those obtained from regression over the entire height. If there is no significant difference it is legitimate to lump all values obtained from the 3 runs and perform linear regression over the entire height and use the obtained values for process and error analysis purposes.

8. For an estimation of the variance of h, enter the calculated slope and intercept of the regression line into equ. AI.4 and obtain H_i. Enter this into equ. AI.3 and obtain s^2 and s.

9. The confidence region for the calibration is found by calculating S^2 according to equ. AI.6. The values for c can be looked up in any handbook on statistics, e.g./11/. Both values entered in equ. AI.5 give the confidence region for the calibration.

10. The confidence region for the process: calibration plus measurement is given by calculating ζ according to equ. AI.8. Equ. AI.7 gives then the confidence region.

Table 1   Properties of various liquid measuring methods

	Conductivity probe	Inductivity coil	Heated wire device	Condensor probe	Inter-lock system	Activity measurem.	Ultra-sonic sensor
a) no cutting of tubes		x			x	x	x
b) remote mounting		x			(x)	x	x
c) low maintenance	x	x	x	x	(x)	x	x
d) resistance against corrosion and radiation	x	(x)	x	x	x	(x)	x
e) inexpensive	x	x	x	x		(x)	x

( ) = with restrictions

Table 2 Data from calibration with potassium carbonate solution

Tank I		Tank II	
h [mm]	V [l]	h [mm]	V [l]
68.5	43.425	35.9	30.296
98.5	60.000	65.7	46.639
131.6	76.561	94.9	63.330
162.8	93.109	126.1	80.394
194.7	110.119	155.9	97.213
226.6	126.382	185.7	113.577
257.1	142.719	215.6	130.219
288.3	159.246	246.1	147.025
320.2	175.563	275.9	163.797
351.4	192.022	306.4	180.772
383.3	208.536	337.6	197.890
415.2	225.424	368.1	214.818
446.4	241.313	398.6	231.393
475.5	257.106	427.8	248.035
508.1	273.790	458.3	264.691
539.3	290.344	488.1	281.517
569.8	306.715	519.3	298.234
601.0	323.046	547.8	314.782
630.9	338.724	578.3	331.343
662.8	355.319	608.1	347.897
694.0	371.833	638.6	364.295
725.2	388.347	667.8	380.625
757.1	404.895	697.6	397.207
789.0	421.707	727.4	413.910
819.5	438.099	757.3	430.702
851.4	454.409	787.8	447.222
881.9	470.800	817.6	463.593
911.7	486.567	847.5	480.127
942.3	502.482	875.9	496.498
973.5	518.391	905.8	512.774
1004.0	534.416	935.6	529.593
1034.5	550.250	966.1	546.222
1064.4	566.118	998.6	564.636
1099.7	584.284		

Table 3 Conductivity measurements: Data compilation

Experiment I

	U-tube readings		Calculated Density	Calculated Volume	Air-throughput	Time Constant
	Level	Density	[g/ml]	[l]	[l/h]	[min]
Potassium carbonate solution	465, 359	214, 153	1,4596	296,5		
+ Water	762, 639	213, 152				
Mixed solution	764, 641	189, 127	1,2567	587,2	2300	2,5

Experiment II

Potassium carbonate solution	331, 221	207, 145	1,3999	207,1		
+ Water	540, 431	207, 145				
Mixed solution	542, 433	184, 122	1,2169	420,4	2300	1,85

Table 4:

Results and statistical analyses of inhomogeneity tests

sample from position	A	B	C	D	E
1st run	1.004	0.980	0.972	0.984	0.953
2nd run	1.078	1.036	1.036	1.025	1.048

[g Zn/g solution]

Test of significance:

$$\Phi\left(\frac{X_c}{\sigma_D}\right) = 1-\alpha$$

σ_D = standard deviation

Φ = Gaussian distribution

for $\alpha = 0.05$ and $\sigma_D = \sqrt{2}\cdot 0.005$

$X_c = 0.0117$ = threshold value

max. difference for single measurements

$$\left.\begin{array}{ll} 1. & 0.051 \\ 2. & 0.053 \end{array}\right\} = \text{both} > X_c,$$

this means: solution is inhomogeneous.

Estimation of inhomogeneity:

Estimation of the variance of inhomogeneity

$$\hat{\sigma}_I^2 = \frac{1}{p-1}\sum_{i=1}^{p}(y_i-\bar{y})^2 - \frac{1}{n}\sigma_T^2$$

p = number of samples ($p = 5$)

y_i = reported concentration of i-th sample

$\bar{y}$ = mean value of p sample concentrations

n = number of analyses per sample ($n = 2$)

$\frac{\sigma_r^2}{n}$ = reported variance of measurement per sample ($\frac{\sigma_r^2}{n} = 0.005^2$)

Inhomogeneity (estimated)

1. 1.78 %
2. 1.97 %

Table 5 Compiled data for the Zn tracer experiments

run no.	1	2	3	4
added Zn (W_T) [g]:	553.7 1)	351.4 1)	536.0 1)	602.43 2) 603.2 1) 602.8 3)
U-tube readings (h_1+h_2) [mm]:	982	660	989	1028
calculated volume in tank (V_U) [l]:	557.4	378.0	561.3	583
solution temperature [°C]:	19	19	17	18
calculated mass content in tank (G_U) [kg] 4):	556.5	337.4	560.6	582.2
sparging time [min]:	120	120	180	180
no. of samples taken:	1	1	9 5)	5 6)
anal.tracer concentr. 7) $[\frac{g\ Zn}{kg\ sol}]$:	0.9983	0.9342	A = 1.0037 B = 0.9799 C = 0.9718 D = 0.9839 E_5= 0.6470 E_{10}=0.8888 E_{15}=0.9610 E_{30}=0.9728 E_{45}=0.9527	A = 1.078 B = 1.036 C = 1.036 D = 1.025 E = 1.048
mean concentration value (C_A) $[\frac{g\ Zn}{kg\ sol}]$:	0.9983	0.9342	1.) 0.9784 8) 2.) 0.9622 9)	1.0446 10)
calculated tank content 11) (G_{TR}) [kg]:	554.6	376.2	1.) 547.8 2.) 557.1	577.1
deviation $(1-\frac{G_{TR}}{G_u}) \cdot 100$ [%]	+0.34	+0.32	1.) +2.28 2.) +0.62	+0.88

1) found by weighing Zn Cl_2 and consecutive analysis for Zn. Values reported by analytical laboratory

2) value reported by manufacturer of Zn Cl_2

3) mean value

4) = V_U x density of H_2O at given temperature

5) 1 each at sampling points A, B, C, D after 45 min of circulation, 1 sample each at point E after 5, 10, 15, 30 and 45 min of circulation resp.

6) 1 each at sampling point A, B, C, D, E

7) values are mean values of 2 single measurements per sample analysed by complexometric method. Values reported by analytical laboratory.

8) = mean value of samples A, B, C, D, E_{45}

9) = mean value of samples E_{15}, E_{30}, E_{45}

10) = mean value of samples A, B, C, D, E

11) calculated by $G_{TR} = \frac{WT}{C_A}$.

Table 6 Compiled data for the Re-tracer experiment

run no.	1	2	3	4
added tracer (W_T) [g] : 1)	$10.242\cdot10^{-3}$	$10.218\cdot10^{-3}$	$15.298\cdot10^{-3}$	$15.296\cdot10^{-3}$
U-tube readings (h_1+h_2) [mm]:	642	642	1004	967
calculated volume in tank (V_U) [l]:	368	368	569.7	549
solution temperature [°C]:	18	18	18	19
calculated mass content in tank (G_U) [kg]2):	367.5	367.5	568.9	548.2
sparging time [min]:	120	120	120	120
no. of samples taken:	1	1	1	1
anal. tracer concentr.(C_A) $\left[\frac{\text{g Re}}{\text{kg sol}}\right]$: 3)	$26.0\cdot10^{-6}$	$28.4\cdot10^{-6}$	$28.4\cdot10^{-6}$	$31.5\cdot10^{-6}$
calculated tank content (G_{TR}) [kg]4):	393.9	359.8	538.7	485.6
deviation $(1-\frac{G_{TR}}{G_U})\cdot100$ [%]:	-7.19	+2.1	+5.32	+11.42

1) weights reported by the analytical laboratory
2) = V_U x density of H_2O at given temperature
3) analysed by neutron activation
4) calculated by $G_{TR} = \frac{WT}{C_A}$

Table 7 Calibration of Tank I + II with tap water

Tank I		Tank II	
h [mm]	V [l]	h [mm]	V [l]
113	66.830	26	24.509
155	88.977	66	46.666
198	111.133	109	70.324
240	133.310	152	93.943
263	142.131	194	117.431
283	155.176	237	141.265
328	179.065	280	164.993
375	203.494	323	188.917
386	209.521	368	212.915
432	233.440	411	237.095
479	257.809	455	261.864
		497	285.693
526	282.529		
573	307.239	542	309.982
620	331.678	585	334.251
666	356.107	628	358.150
714	381.067	671	382.219
760	405.597	715	406.348
807	429.806	749	430.707
853	453.795	803	455.246
900	477.513	847	479.385
943	501.622	891	503.755
988	525.140	935	527.953
		979	552.283
1035	550.120		
1082	574.319	1023	576.832
1128	598.468	1067	601.081
1175	622.867	1112	625.831
1220	646.456	1156	650.110
1265	670.134	1200	674.609
1312	694.804	1244	698.899
1359	719.533	1288	723.498
1404	743.392	1332	747.697
1451	767.841	1375	772.147
1497	791.960	1419	796.596
1540	813.887	1466	821.225
		1509	845.445
		1552	869.944
		1596	894.303

Tank I	Tank II
$S_I = 0.5229$	$S_I = 0.5534$
$I_I = 7.42$	$I_I = 10.01$
$S_{II} = 0.5244$	$S_{II} = 0.5536$
$I_{II} = 6.72$	$I_{II} = 10.94$
$S_{III} = 0.5239$	$S_{III} = 0.5540$
$I_{III} = 7.65$	$I_{III} = 9.89$
$S_T = 0.5243$	$S_T = 0.5537$
$I_T = 6.94$	$I_T = 10.28$

Table 8 Tank I Calibration with demineralized water

1st run		2nd run		3rd run	
h [mm]	V [l]	h [mm]	V [l]	h [mm]	V [l]
32	24.823	147	84.729	102	60.377
78	48.826	193	109.393	149	85.181
125	73.339	240	134.087	195	110.235
171	97.612	287	158.881	244	135.199
217	121.976	335	183.815	291	160.123
264	146.288	383	208.649	338	184.787
313	171.163	429	233.363	386	209.400
360	195.987	477	258.176	432	234.365
407	220.700	525	283.000	479	259.059
454	245.604	572	307.444	526	283.863
501	270.008				
549	295.212	619	331.657	574	308.616
600	320.597	667	356.410	621	333.320
645	345.802	714	381.114	668	358.134
693	370.846	760	405.658	715	382.698
740	395.279	808	430.522	762	407.461
784	419.132	855	455.255	809	432.205
830	442.864	902	479.839	858	457.766
875	466.526	950	504.843	904	482.490
920	489.967	996	529.757	951	507.294
966	514.380	1043	554.321	999	532.057
1012	539.024			1046	556.821
				1093	581.665
1059	563.117	1092	579.047	1143	606.399
1108	588.041	1140	604.169	1188	631.243
1156	613.245	1184	628.171	1235	656.127
1201	637.989	1230	652.975	1282	680.871
1253	662.833	1278	677.679	1329	706.105
1296	687.998	1325	702.473	1375	731.040
1344	713.353	1370	726.996	1423	755.894
1390	737.715	1418	751.860	1470	780.667
1434	761.457	1465	776.574	1499	796.195
1481	785.941	1508	798.843		
1528	811.316				

Table 9 Tank II Calibration with demineralized water

1st run		2nd run		3rd run	
h [mm]	V [l]	h [mm]	V [l]	h [mm]	V [l]
96	62.691	53	38.929	4	13.400
140	87.395	96	63.192	50	37.566
184	112.329	139	87.404	95	62.390
224	136.632	182	111.477	136	86.844
273	161.195	228	136.241	181	111.207
318	186.119	272	160.424	226	136.091
362	210.753	315	184.697	270	160.524
406	235.306	358	208.699	315	185.298
451	260.070	402	232.972	361	209.911
495	284.954	446	257.115	404	234.876
539	309.408			449	259.379
				493	284.183
584	333.821	489	281.138	540	309.337
629	358.585	531	304.529	585	334.572
673	383.118	576	328.822	630	359.566
718	407.882	619	353.305	674	383.859
763	432.606	665	378.149	719	408.693
807	457.229	710	403.093	764	433.397
856	482.123	754	427.587	808	458.231
897	506.857	799	452.140	852	483.015
942	531.811	843	476.683	898	507.678
986	556.615	887	501.487	941	532.202
1031	581.369	932	526.602	987	557.046
		977	551.075		
1076	606.073	1021	575.719	1031	581.940
1120	630.736	1065	600.192	1077	606.784
1165	655.390	1110	624.575	1116	631.287
1209	680.194	1153	648.868	1165	655.530
1254	705.098	1199	673.582	1209	680.044
1300	729.912	1242	698.196	1255	705.218
1344	754.686	1287	722.799	1299	729.411
1389	779.299	1331	747.162	1344	754.333
1434	804.444	1375	771.836	1387	778.618
1479	829.228	1420	796.620	1431	803.031
1512	847.610	1463	820.442	1460	819.330
		1487	833.605		

Table 10 Program for calibration data evaluation on a DIEHL COMBITRON S 10

The regression is done according to the least square method:

$$y = n\,a + [x]\,b$$
$$[xy] = [x]\,a + [x^2]\,b$$

this gives

$$b = \frac{n\,[xy]-[y]\,[x]}{n[x^2]-[x]^2}$$

$$a = \frac{[x^2]\,[y]-[xy][x]}{n\,[x^2]-[x]^2}$$

wherein n = number of measured points
b = slope
a = intercept
$[\;] \mathrel{\hat{=}} \Sigma$

P0	(X_i)	#	⊻	5	+	X	S	⊽	8	+	A
P1	*	⊻	8	⊽	9	S	$\overset{*}{S}$	⊻	9	(y_i)	A
P2	#	+	X	⊽	5	S	⊽	7	S	$\overset{*}{S}$	A
P3	⊻	7	⊽	6	+	*	⊻	6	J	0	A
P4	(n)	#	+	X	⊽	9	S	⊽	8	X	A
P5	$\underline{S}$	$\overset{*}{S}$	⊻	5	⊽	6	X	⊽	8	$\underline{S}$	A
P6	⊽	7	X	*	S	$\overset{*}{S}$	:	⊽	5	=	A
P7	#	⊽	9	X	⊽	6	S	⊽	8	X	A
P8	⊽	7	$\underline{S}$	$\overset{*}{S}$	:	⊽	5	=	#	*	A
P9	⊻	6	⊻	7	⊻	8	⊻	9	J	0	A

Table 11

A. Half width of confidence region for calibration $(c\cdot\sqrt{s^2})$

V [l]	$2{,}04\sqrt{s^2}$ [mm]
300	0.6268
350	0.4693
400	0.3552
434, 83	0.3272
450	0.3327
500	0.4169
550	0.5618
600	0.7321

B. Half width of confidence of process: calibration plus measurement

V' [l]	ζ-V' [mm]
300	3.8779
350	3.7940
400	3.7475
434, 83	3.7380
450	3.7398
500	3.7712
550	3.8406
600	3.9461

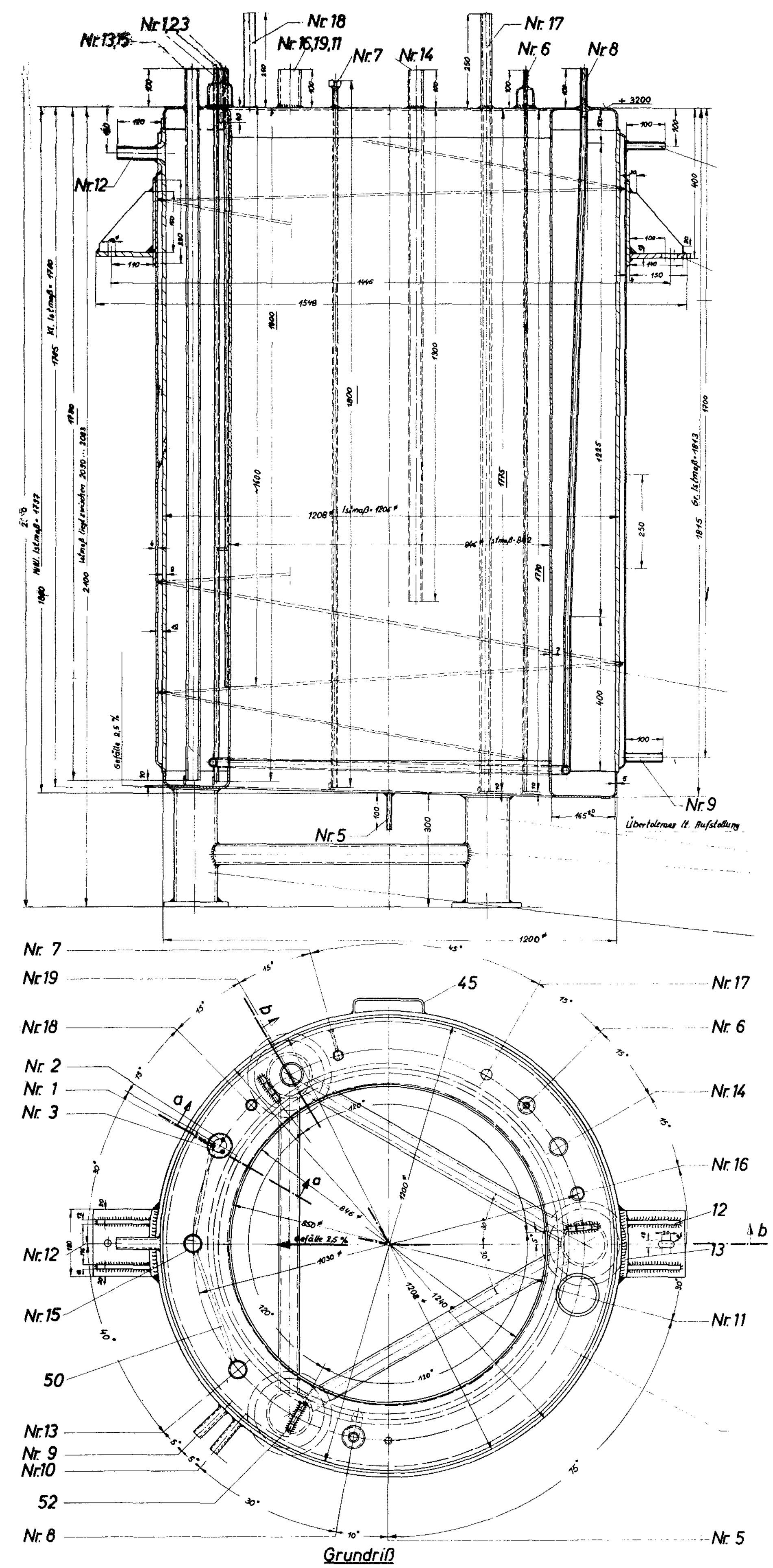


Fig.1 WAK accountability tank

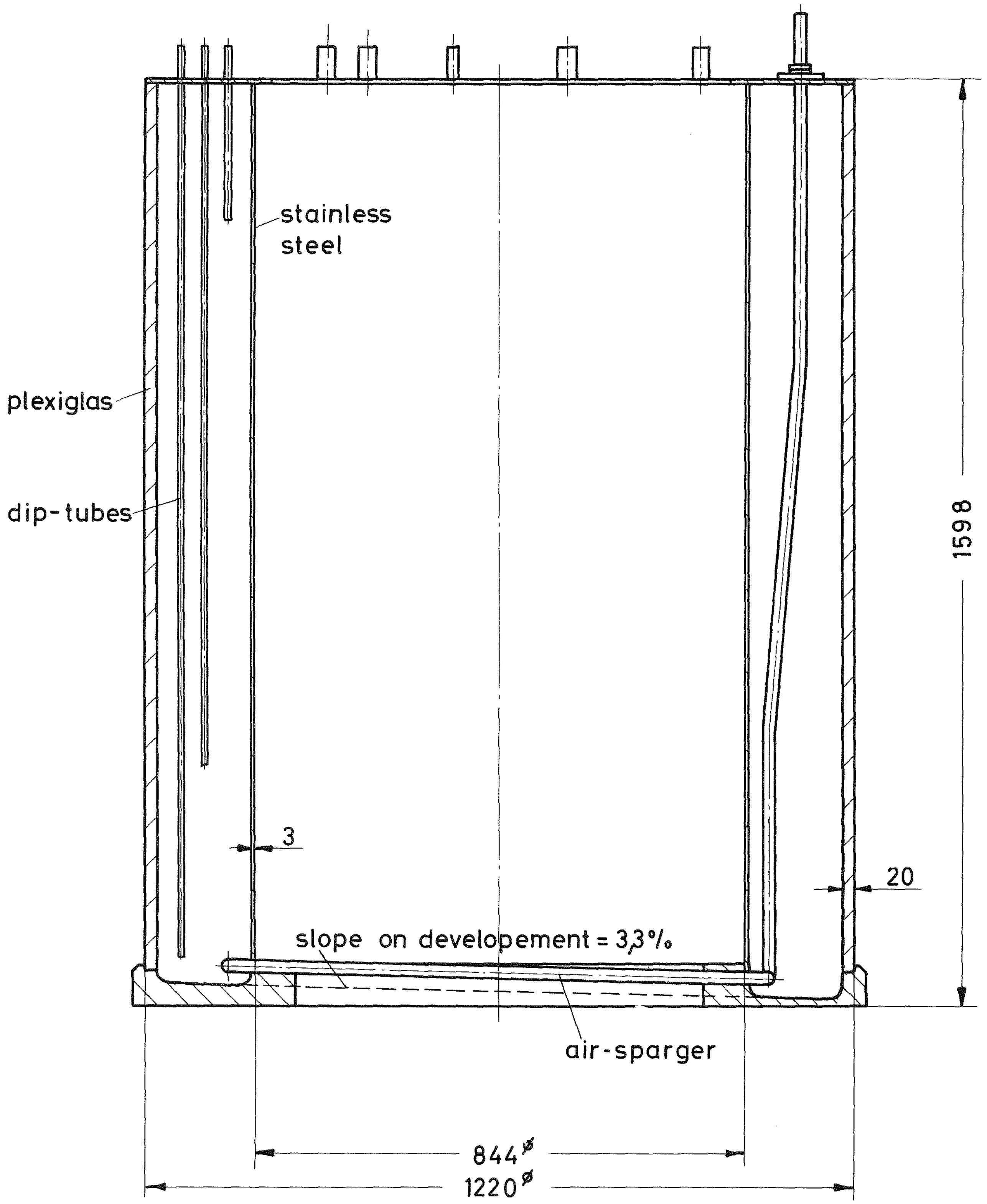
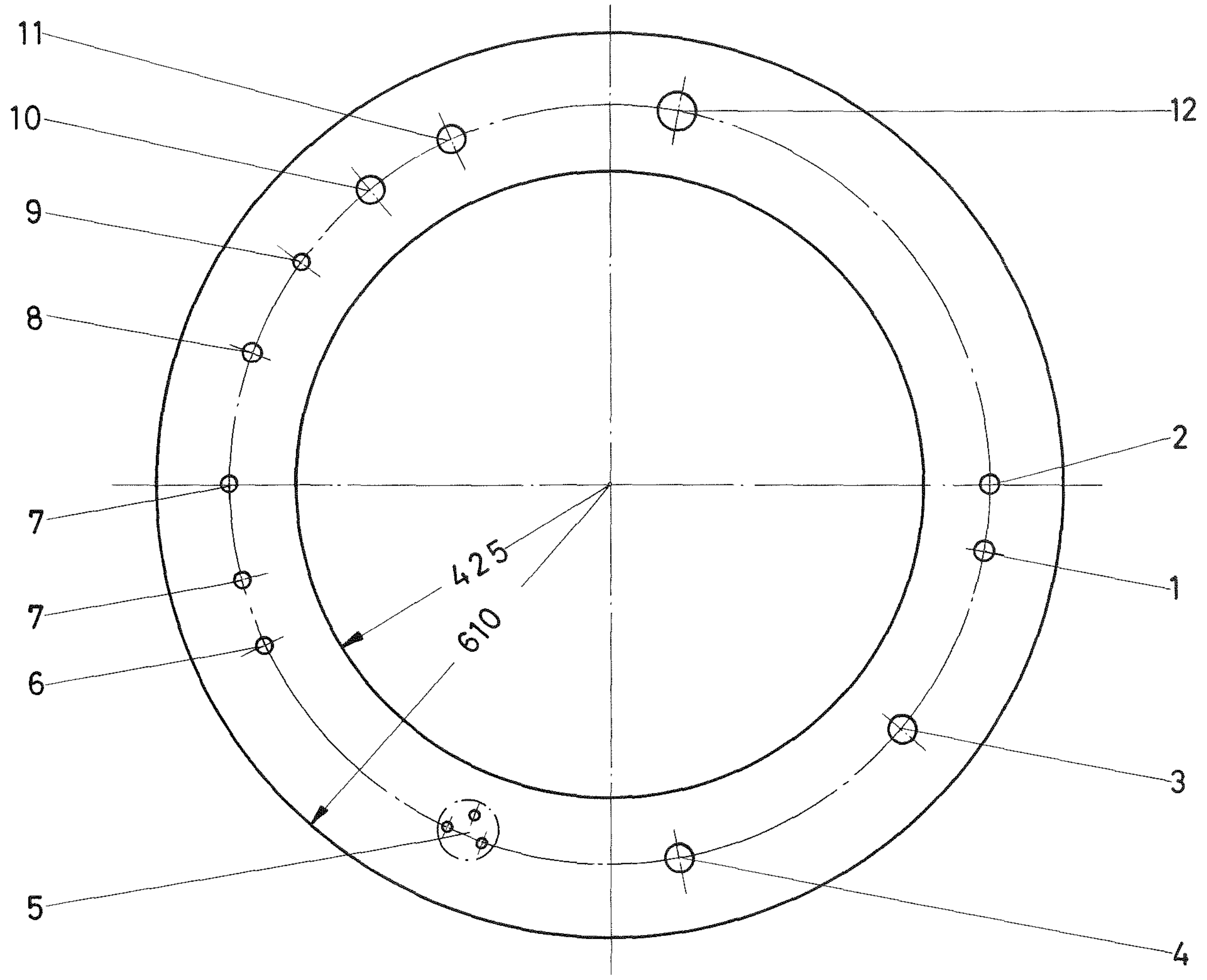


Fig. 2 Cross section of Tank I



1. overflow - line
2. air - sparge
3. suction line
4. sampling line
5. dip - tubes
6. return sampling line
7. spare
8. temperature measurement
9. spare
10. spare
11. ventilation duct
12. feed line

Fig. 3 Top view of Tank I and denomination of connections

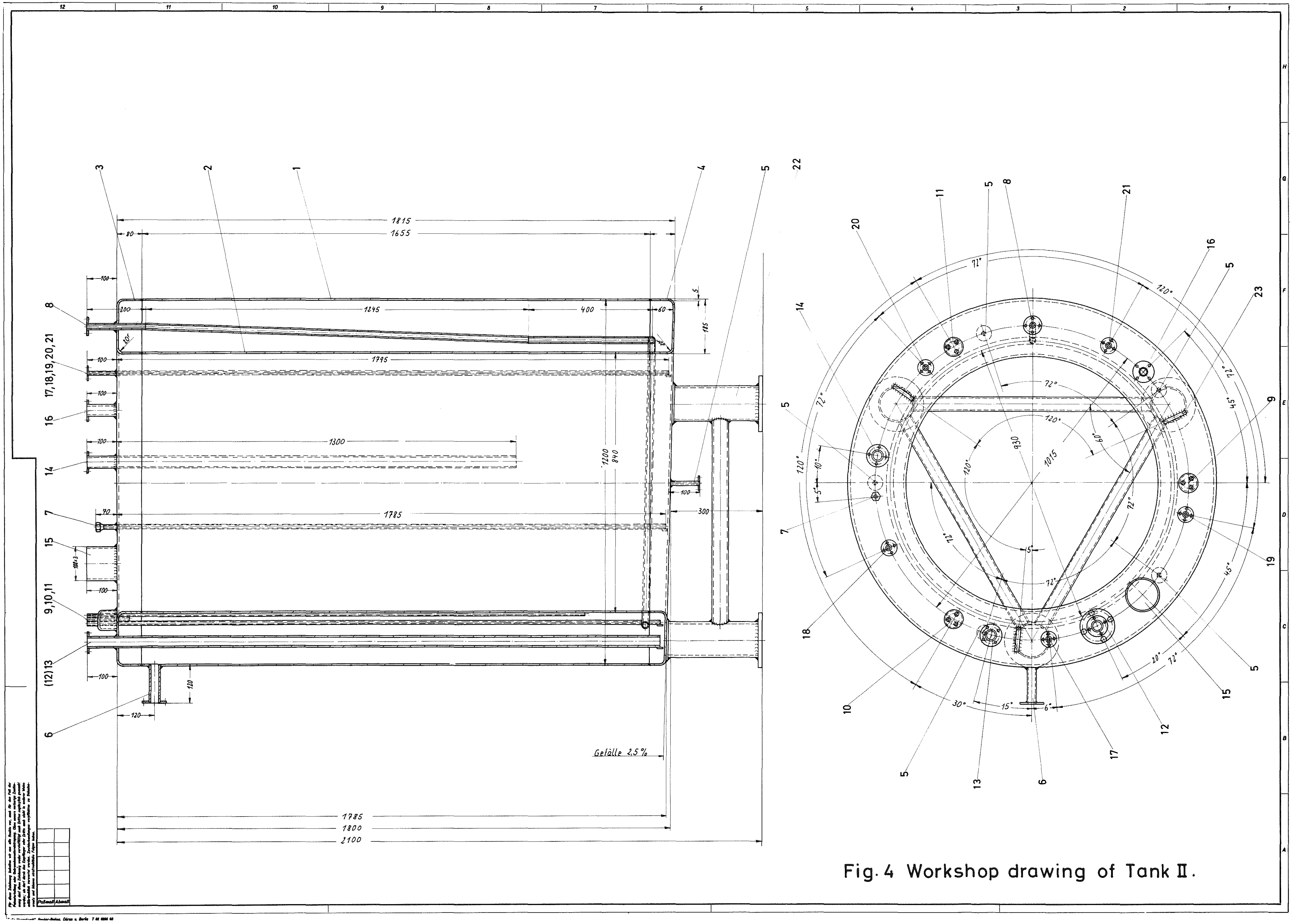


Fig. 4 Workshop drawing of Tank II.

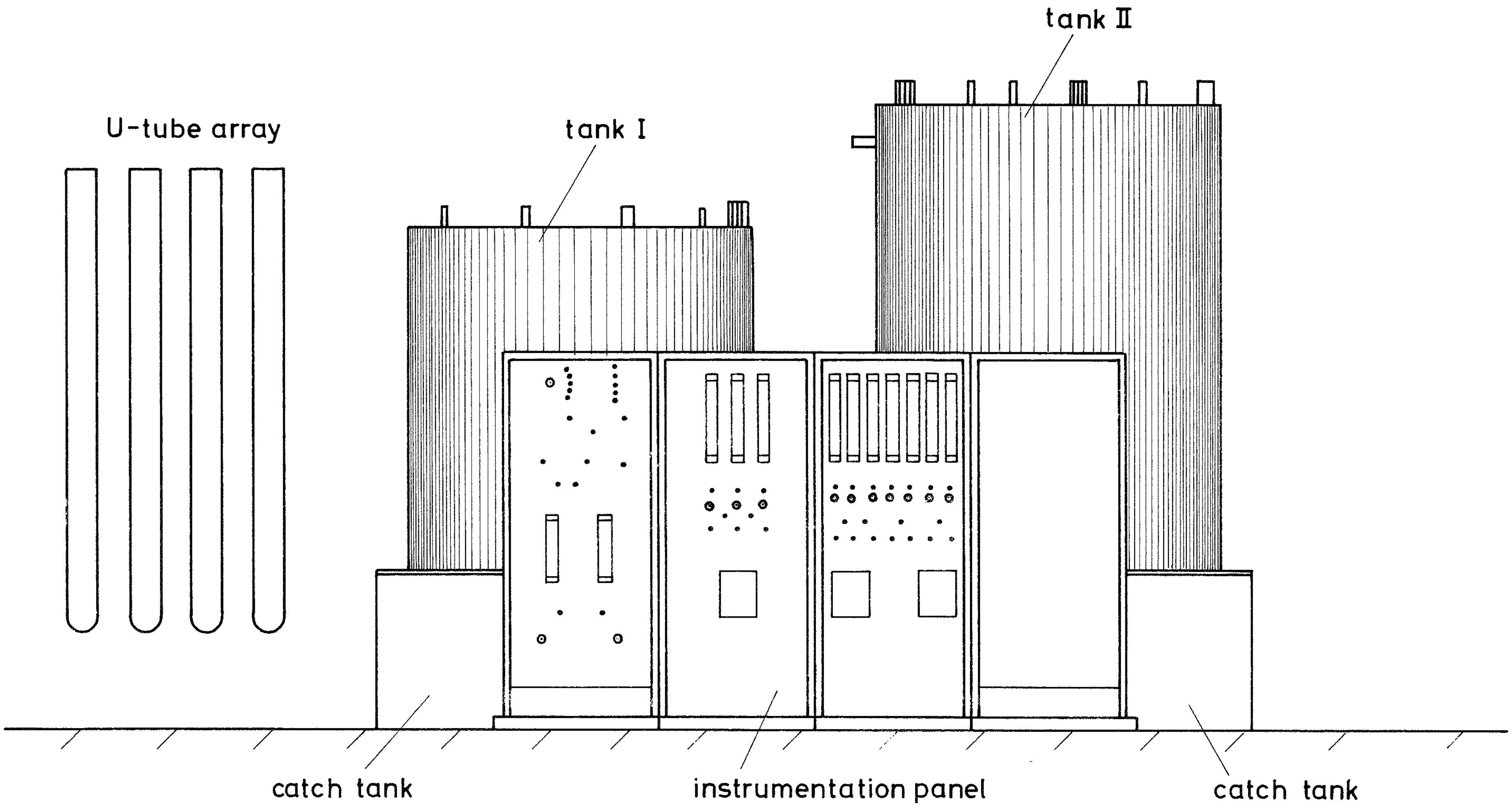


Fig. 5 Experimental set - up (schematic)

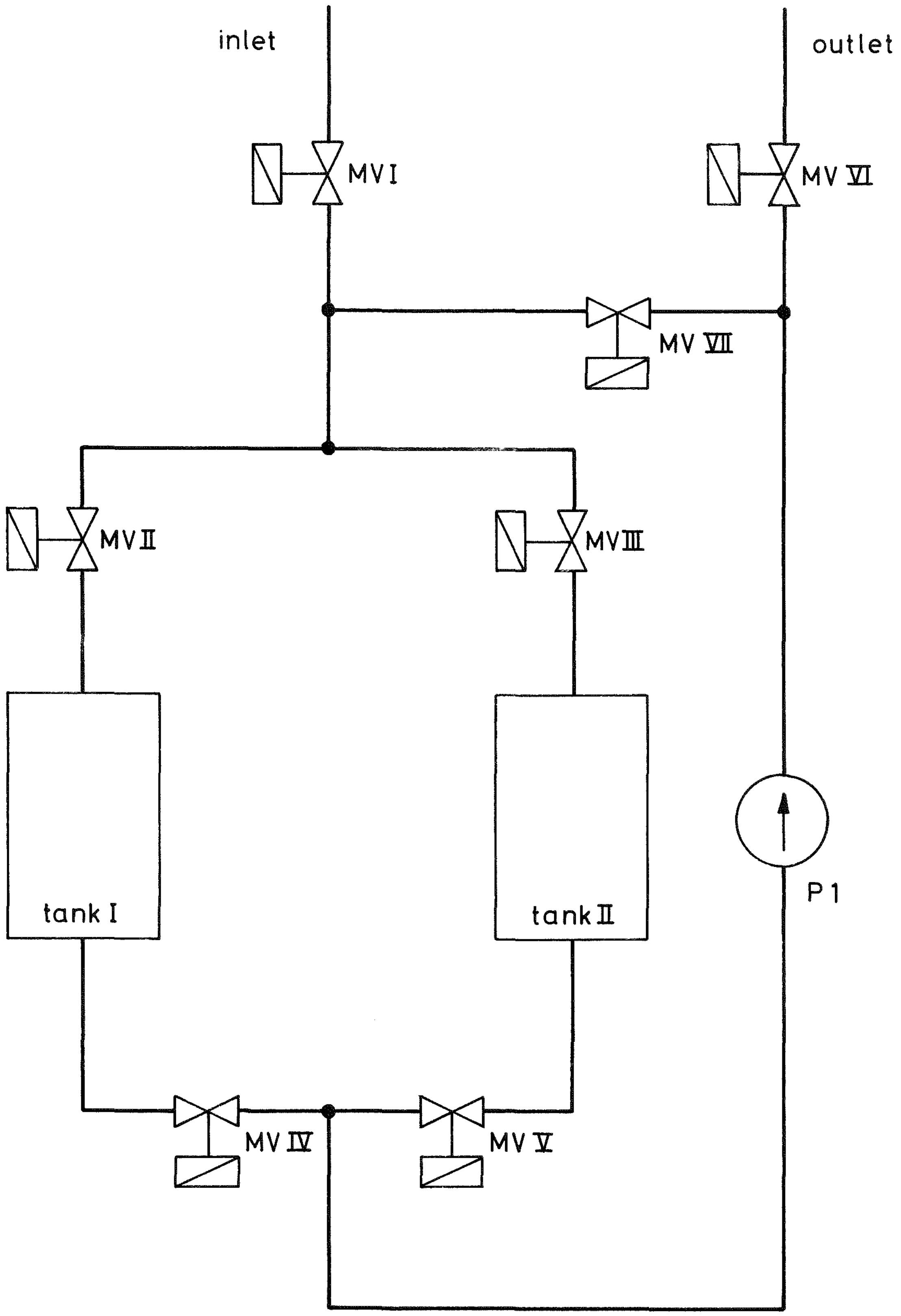
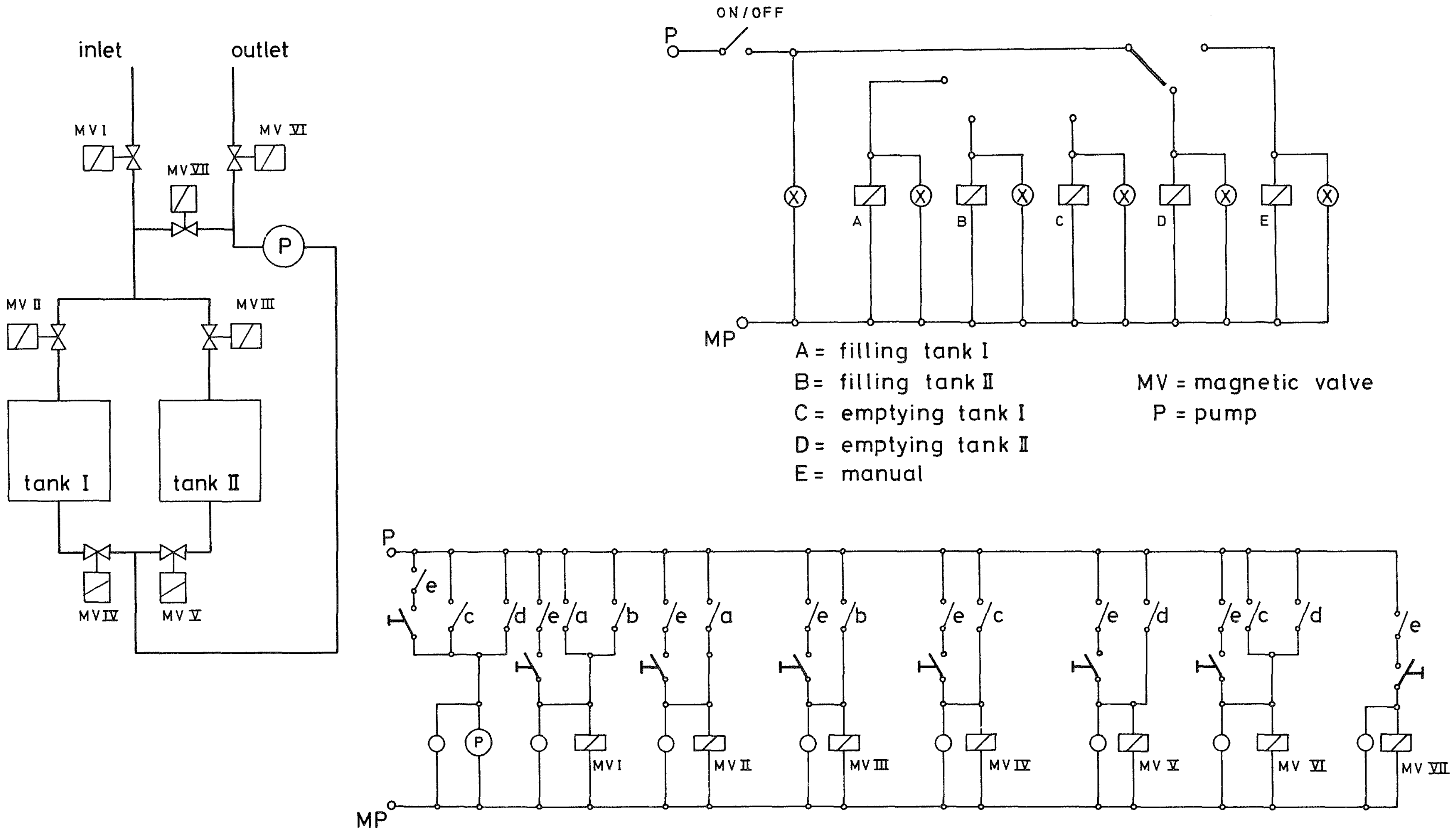


Fig. 6 Pipe connection diagramme



A = filling tank I
B = filling tank II
C = emptying tank I
D = emptying tank II
E = manual

MV = magnetic valve
P = pump

Fig. 7 Electrical connection diagramme

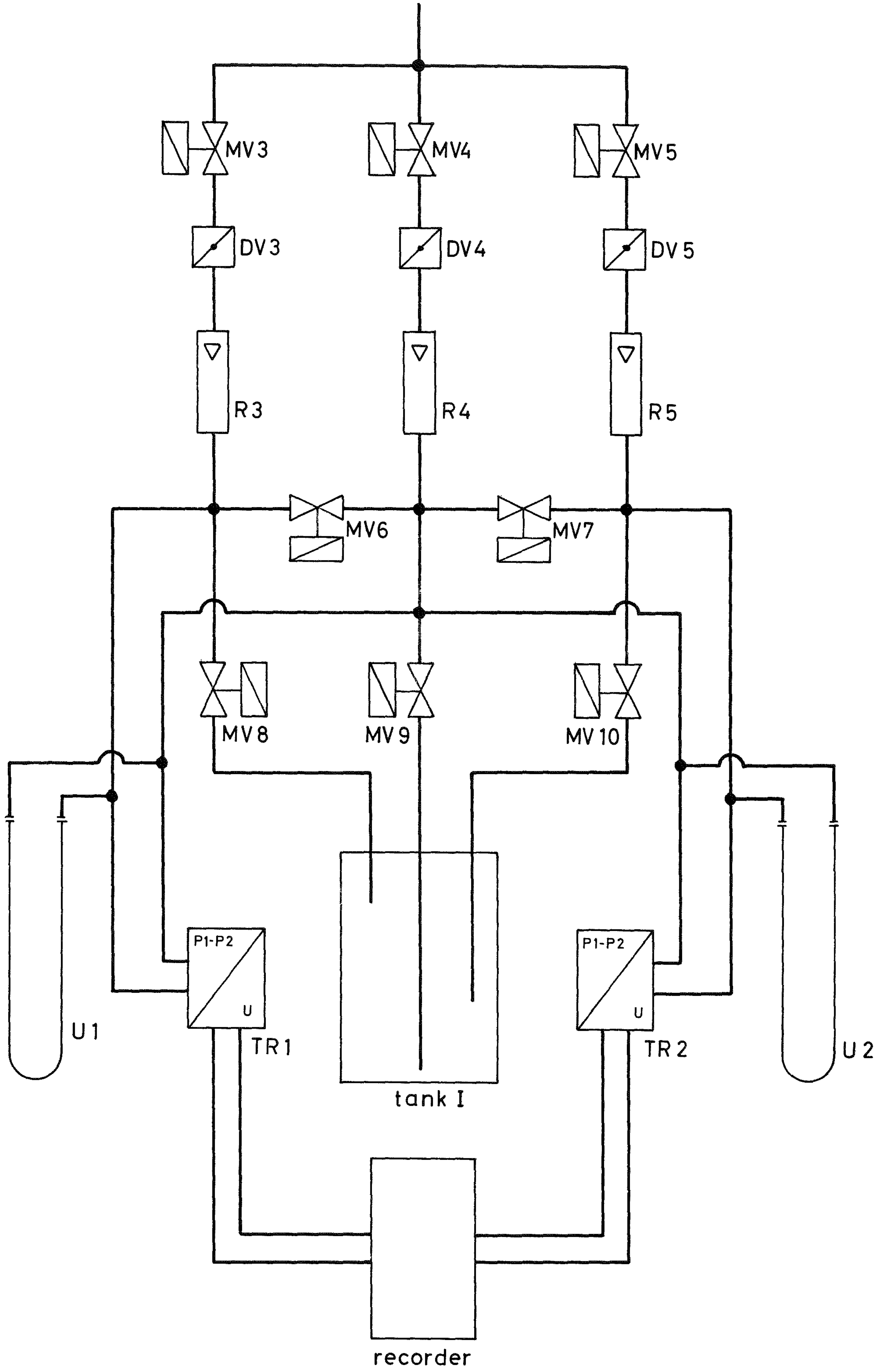


Fig. 8 Level and density measurement array for Tank I (schematic)

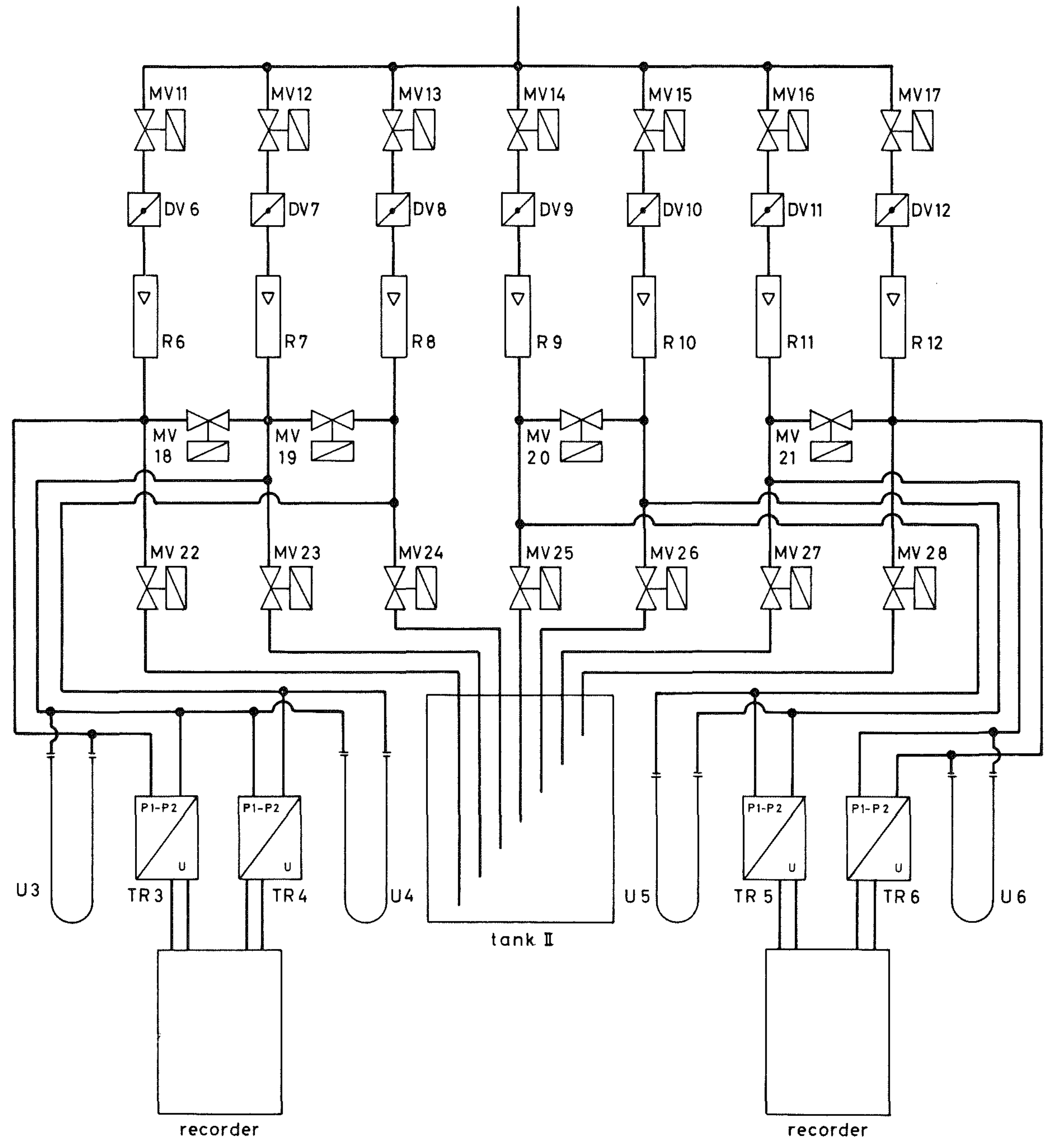


Fig. 9 Level and density measurement array for Tank II (schematic)

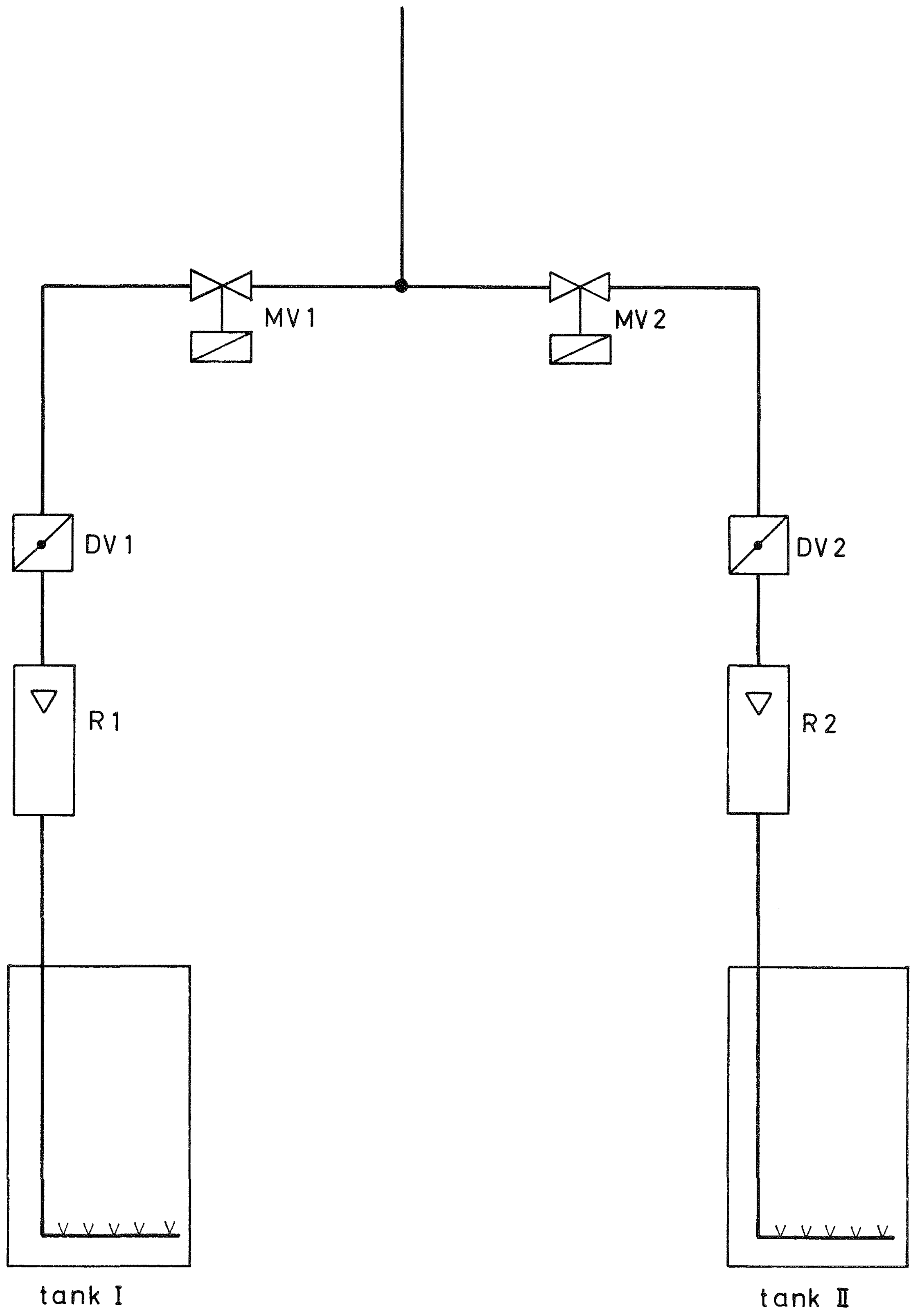


Fig. 10 Air-sparge supply for Tank I + Tank II

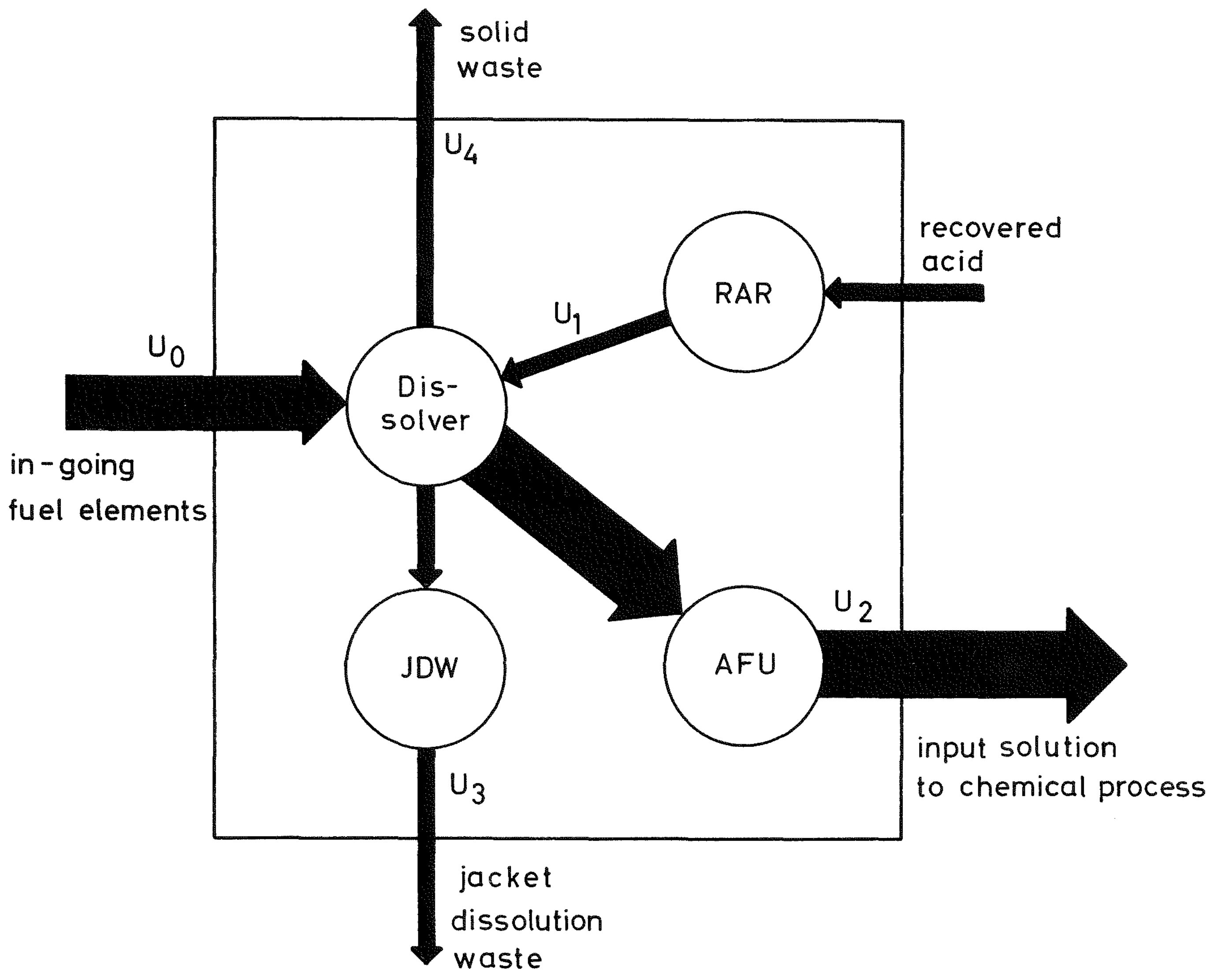


Fig. 11 Input accountability area of a EUROCHEMIC-type reprocessing plant (schematic).

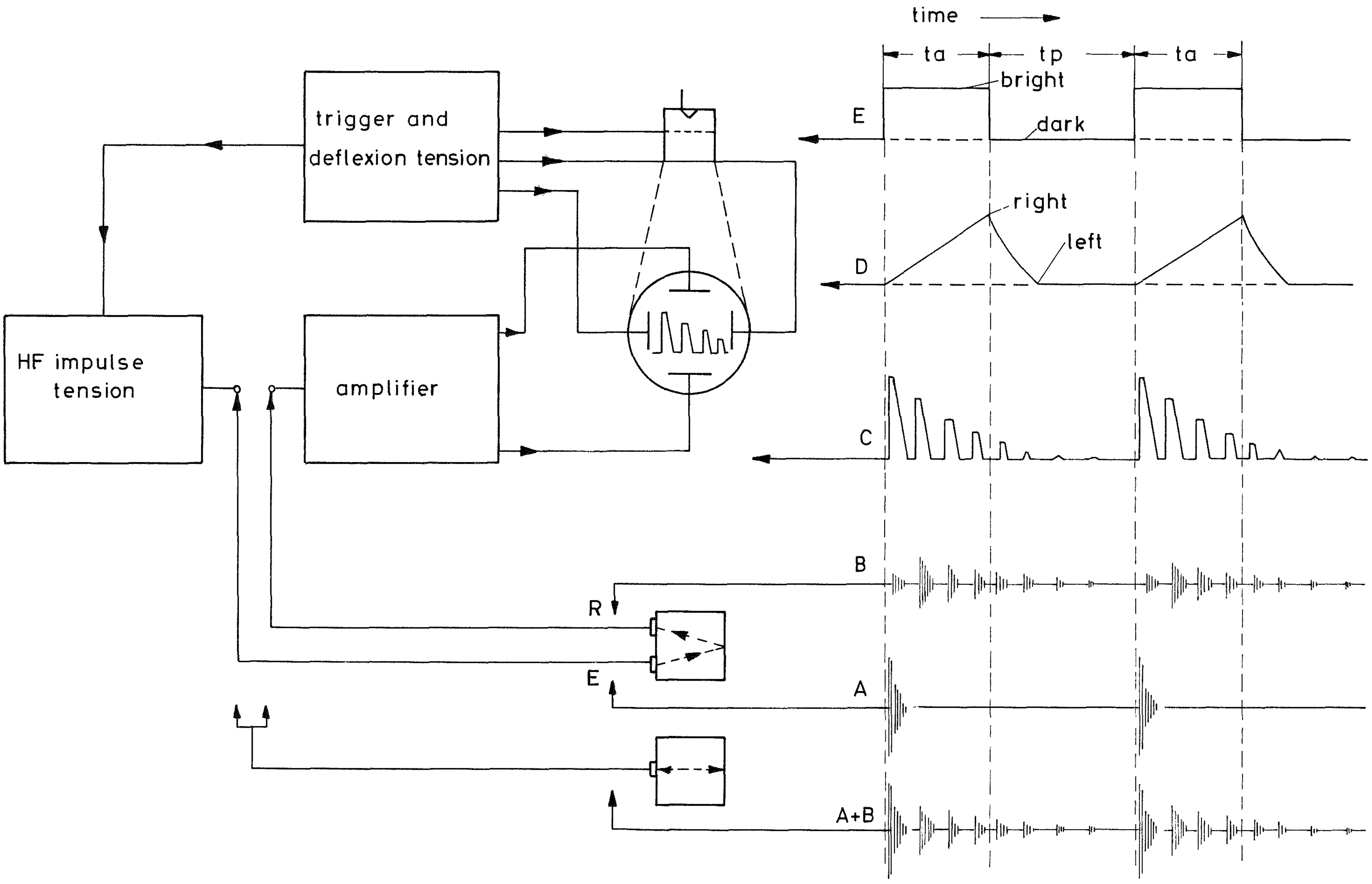


Fig. 12 Block diagramme of a impulse-echo-unit, with resp. tension-time diagramme

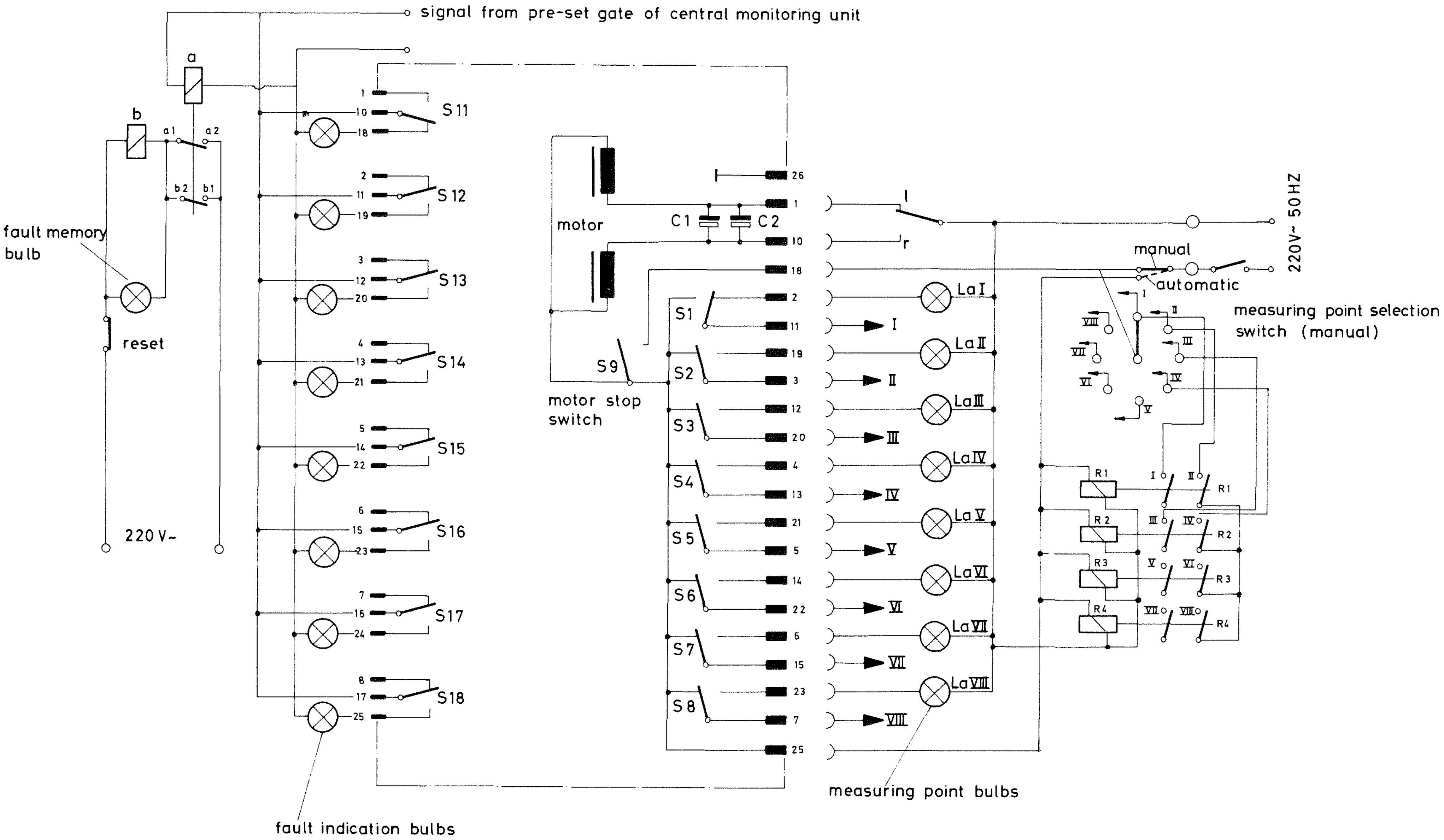
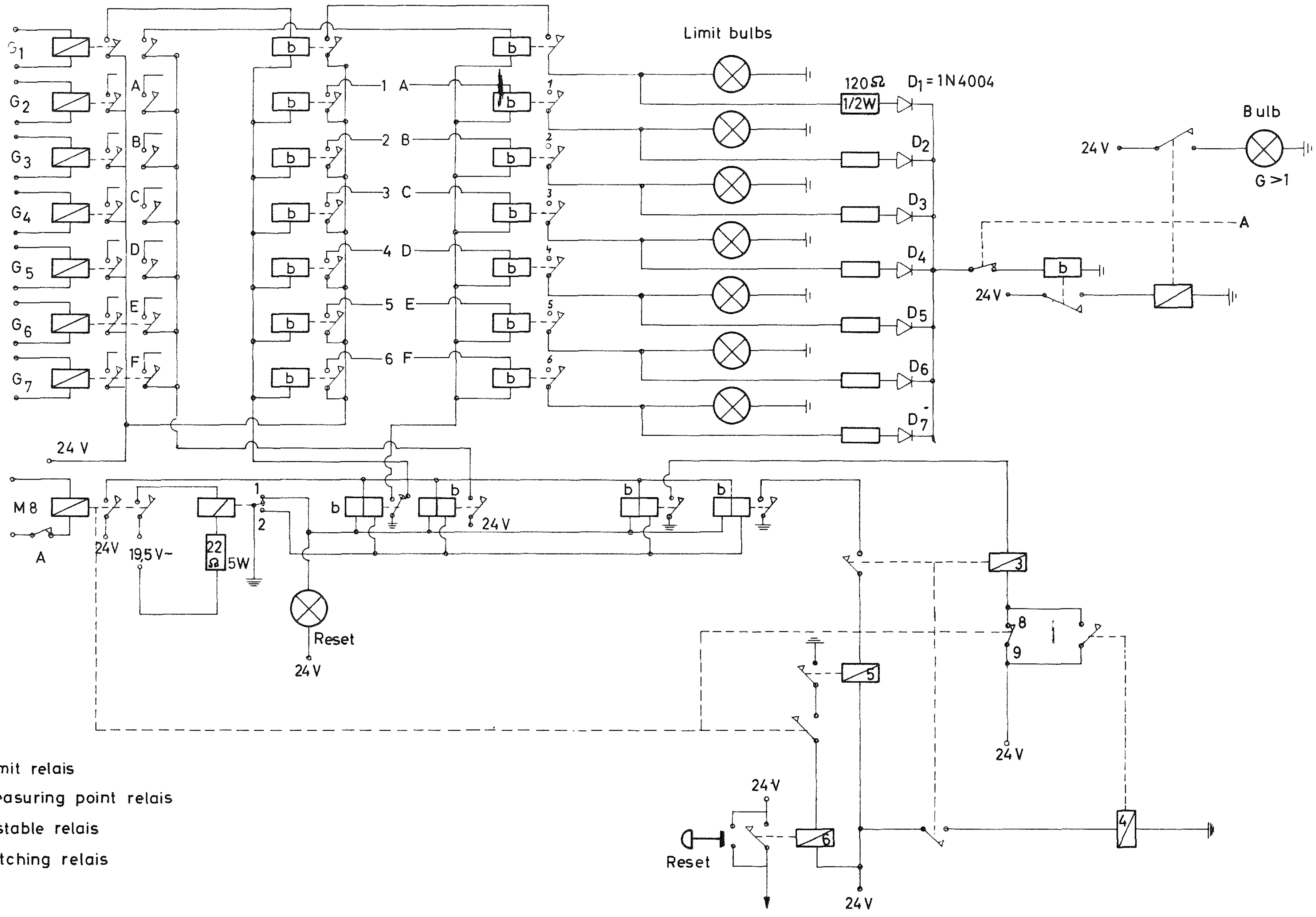


Fig. 13 Measuring point switch



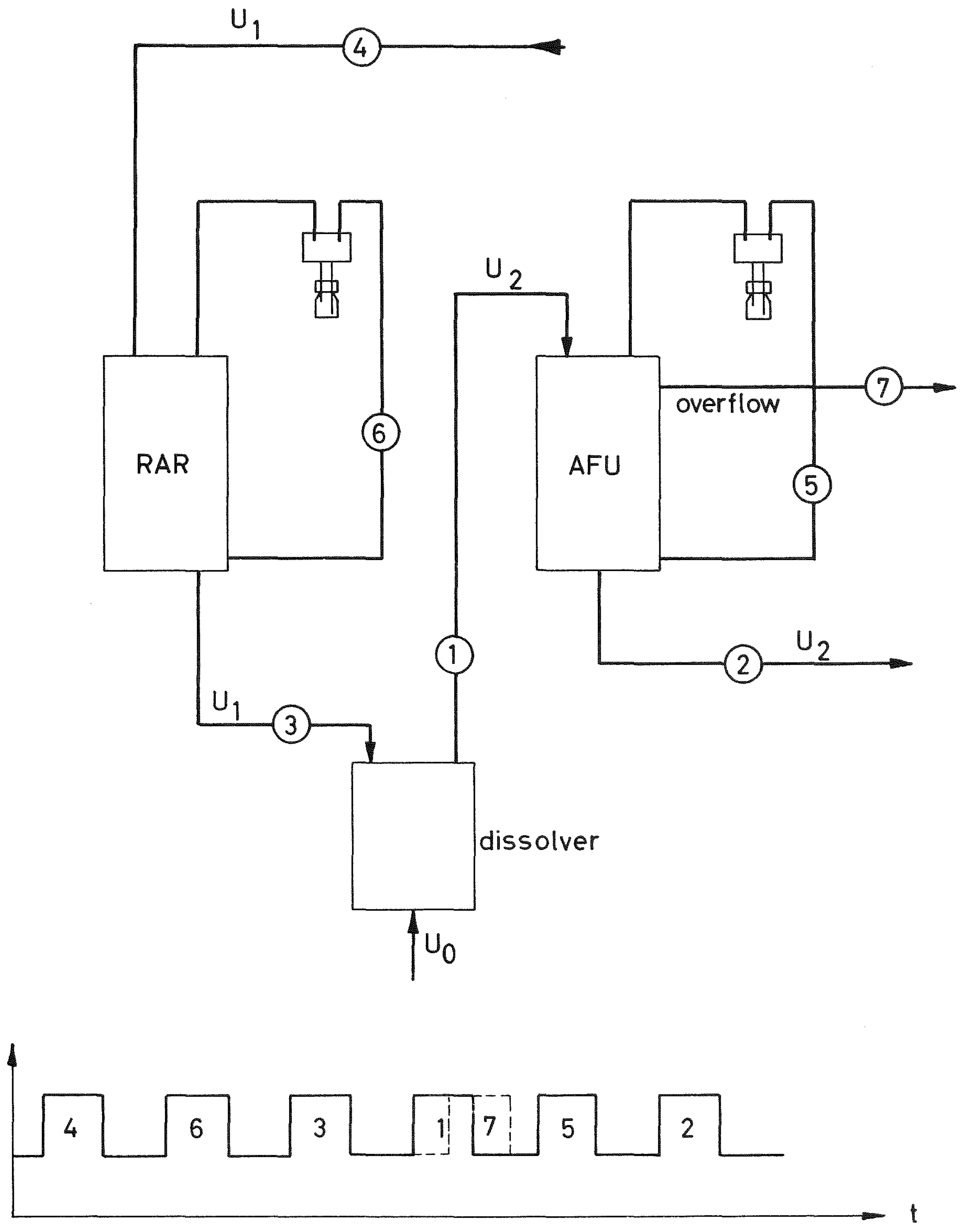
G_1-G_7 = Limit relais

M 8 = Measuring point relais

b = Bistable relais

Rel. 7 = Latching relais

Fig. 14 Signal storage and alarm triggering unit



time sequence of sensor signals with a properly performed input measurement

Fig. 15 Array of liquid sensors and signal sequence.

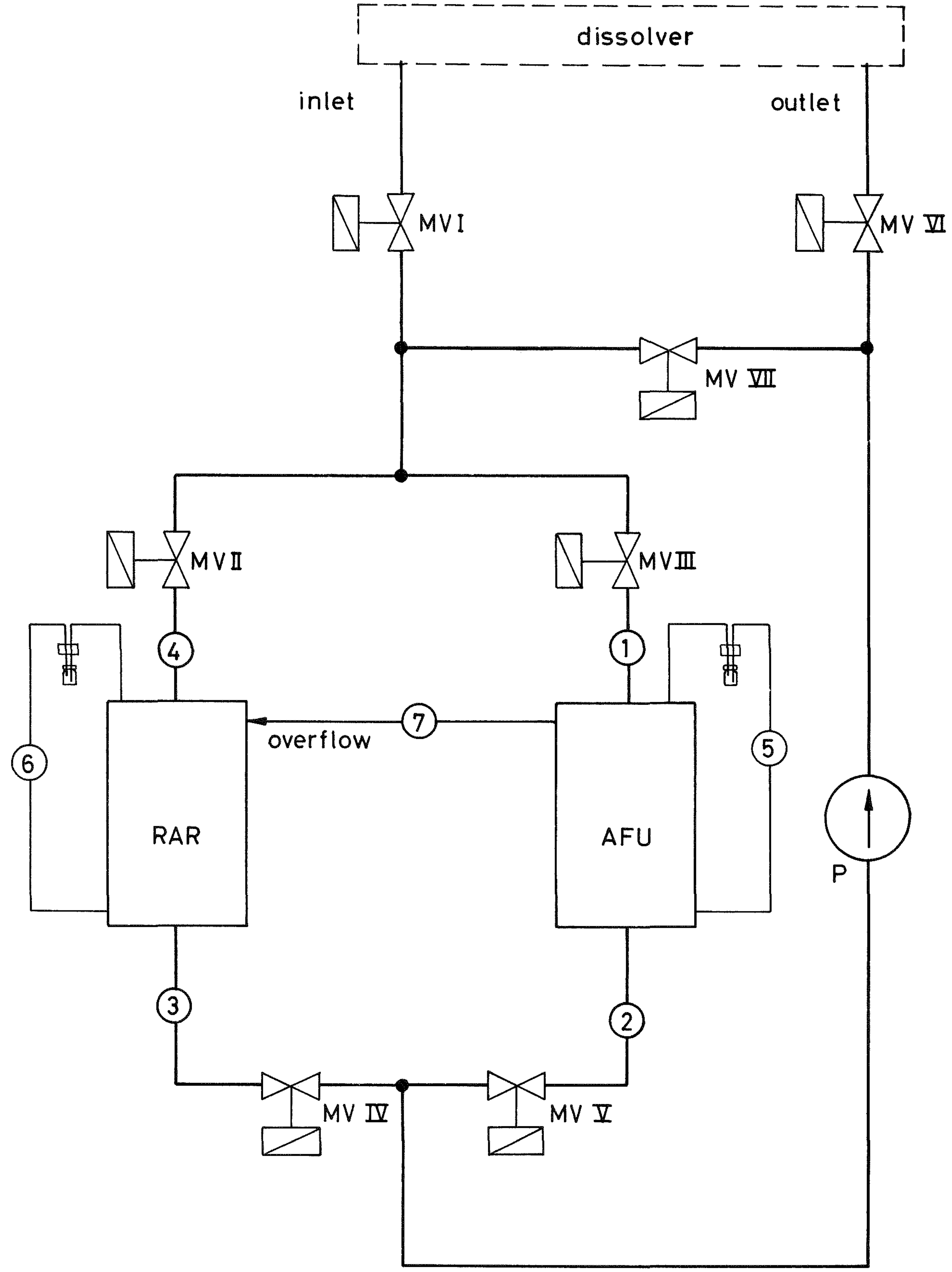


Fig. 16 Tube connection for liquid sensor tests.

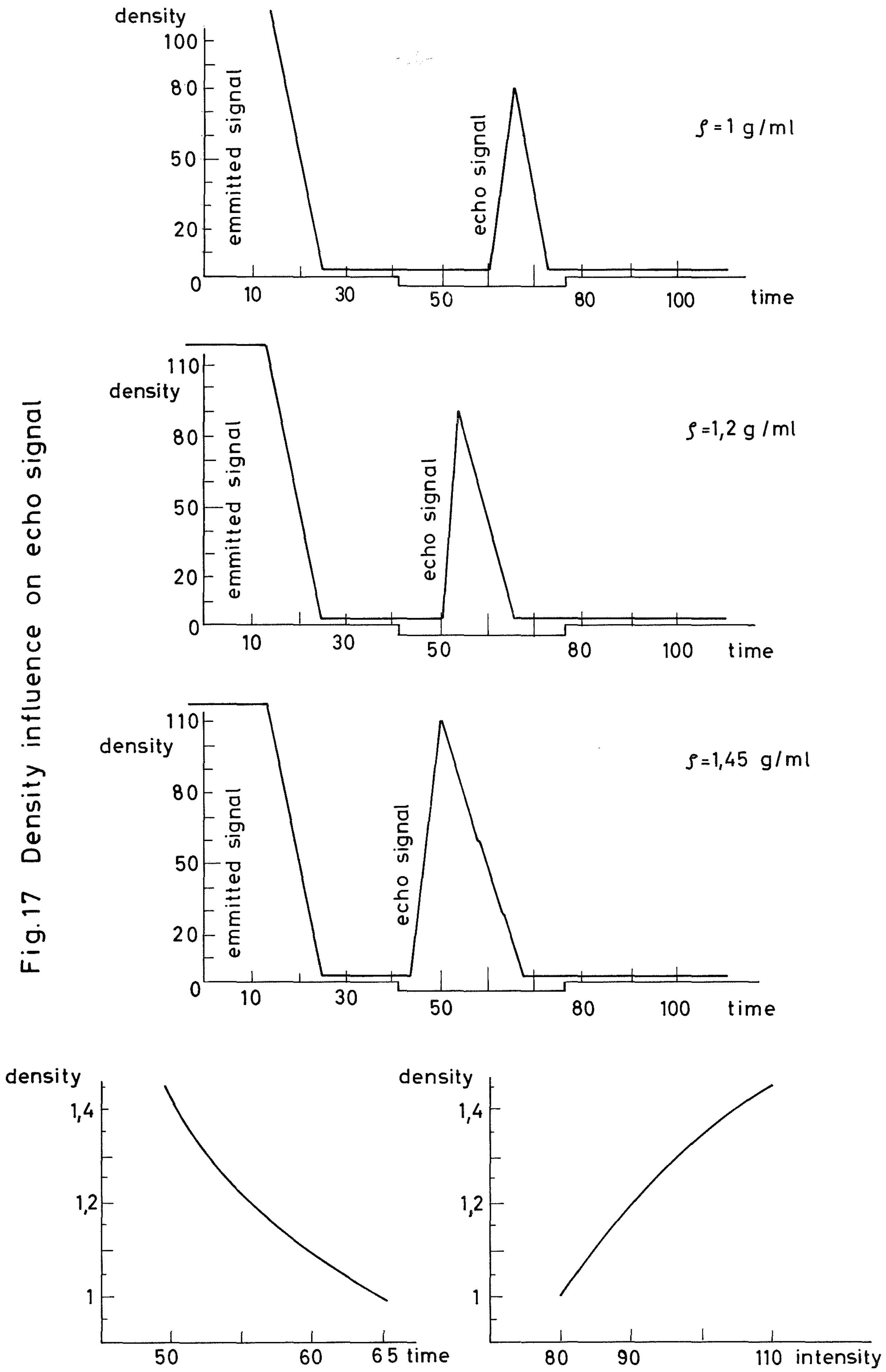
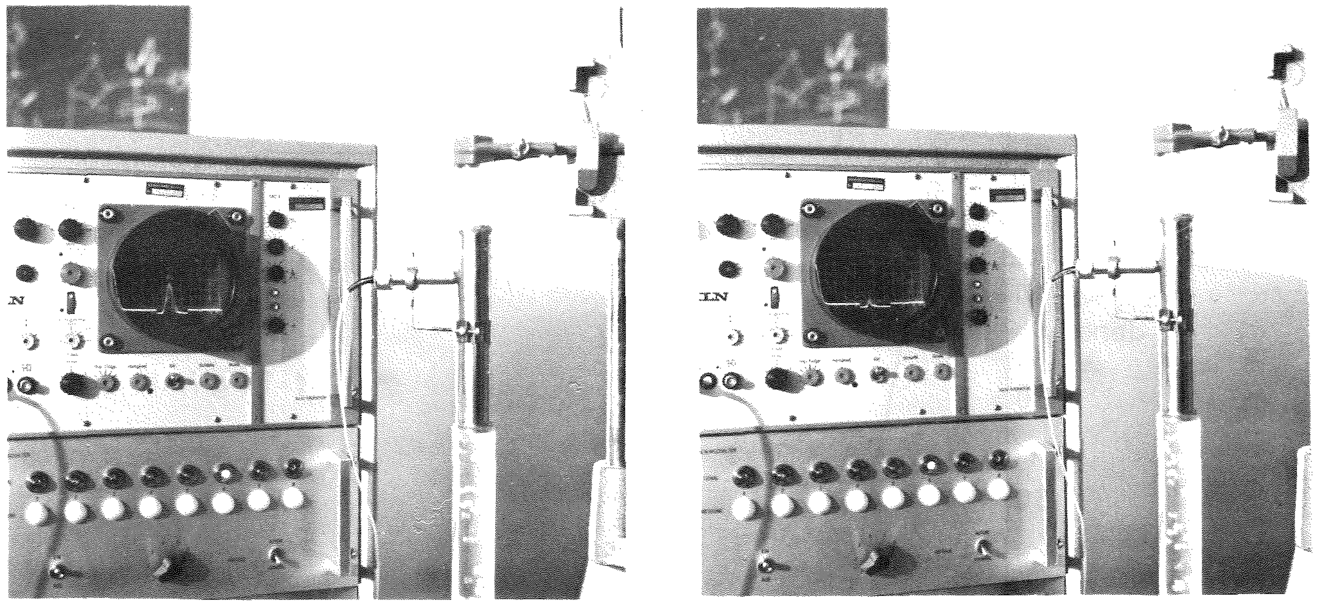


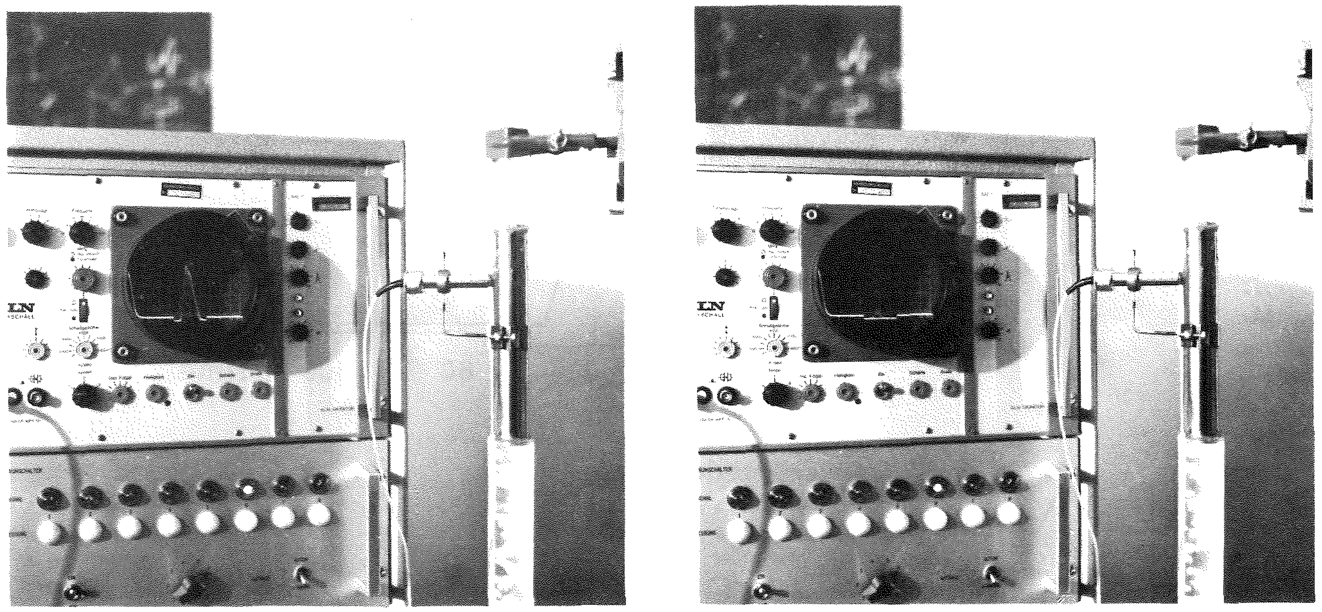
Fig.17 Density influence on echo signal

Fig. 18 Effects of mixed media (air/liquid) on echo signal



Air-throughput at 12 l/h. The sound signal is partly scattered after hitting a small air bubble. Echo signal therefore weaker.

Air throughput at 12 l/h. Sound beam hitting mainly a big bubble. Only a minor part is reflected on rear wall.



Air throughput at 30 l/h. Sound beam crossing entirely the filled tube without hitting a bubble. Echo signal at its maximum intensity.

Air-throughput at 30 l/h. Same situation as above.

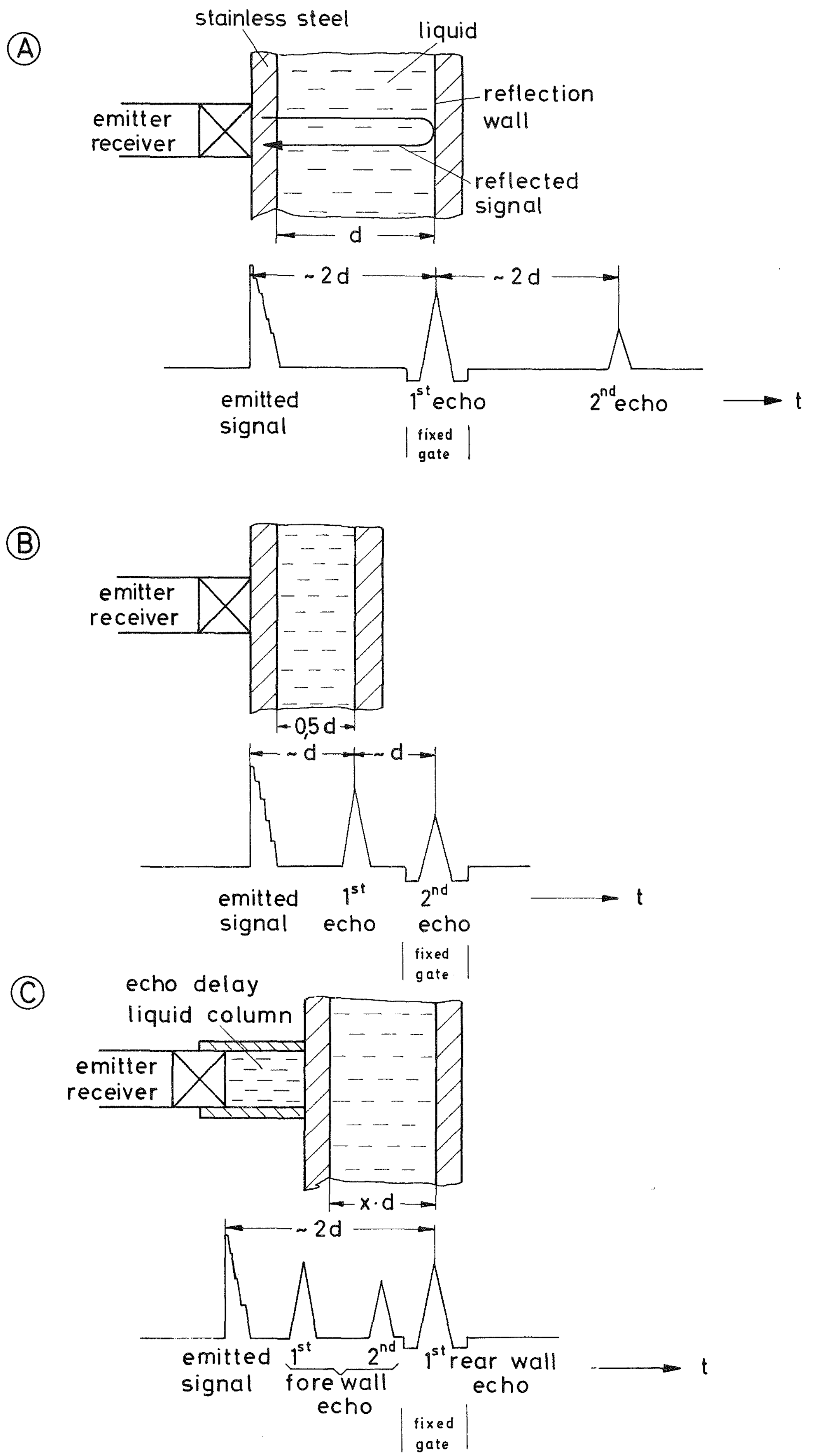


Fig. 19 Different time lapses for different tube diameters.

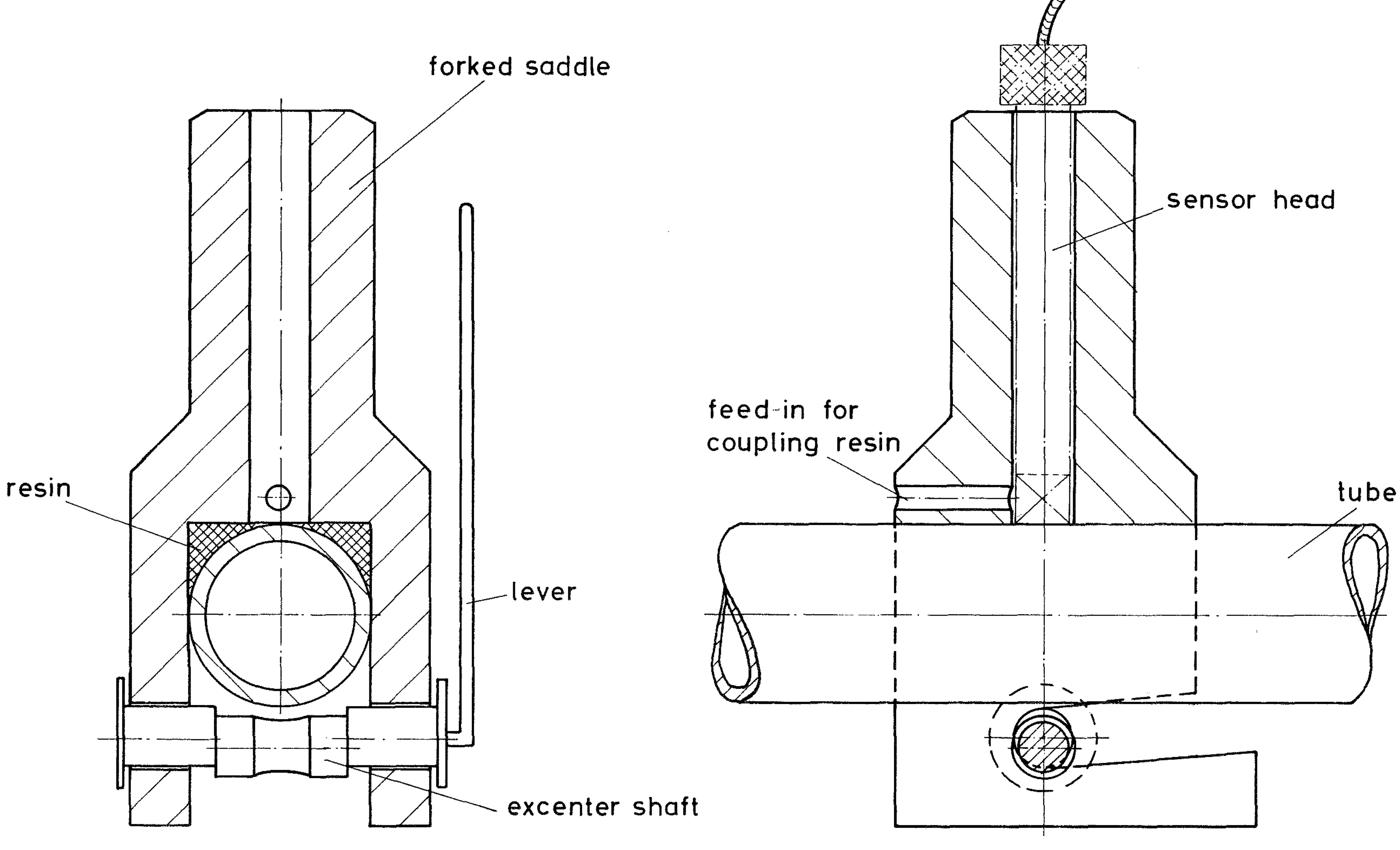
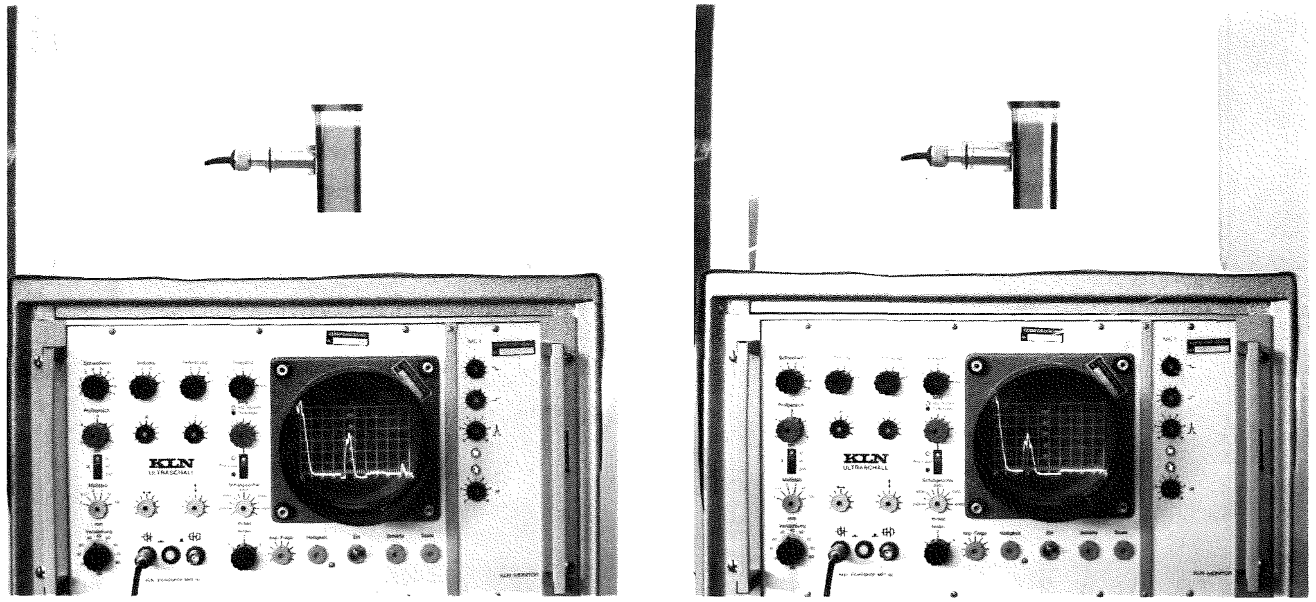


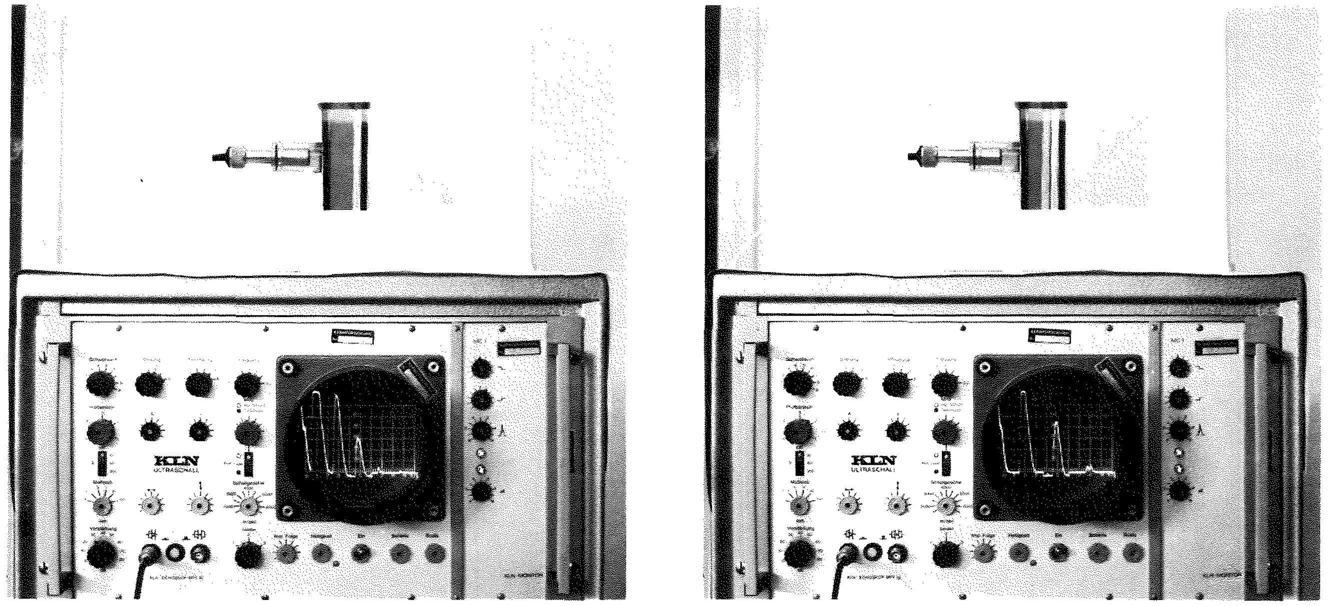
Fig. 20 Sensor head coupling device

Fig. 21 Monitoring different tube diameters



Monitoring a tube diameter x without liquid foreleg and using the 1st echo to trigger the alarm.

Monitoring a tube diameter 0.75 x without using a liquid foreleg. 1st echo falls outside the gate.



Monitoring a tube diameter 0.75 x, using a liquid foreleg. The first two peaks are 1st and 2nd forewall echo, the 3rd is 1st rear wall echo.

Monitoring a tube diameter 0.5 x, using liquid foreleg. 1st peak is 1st forewall echo, 2nd is 1st rear wall echo.

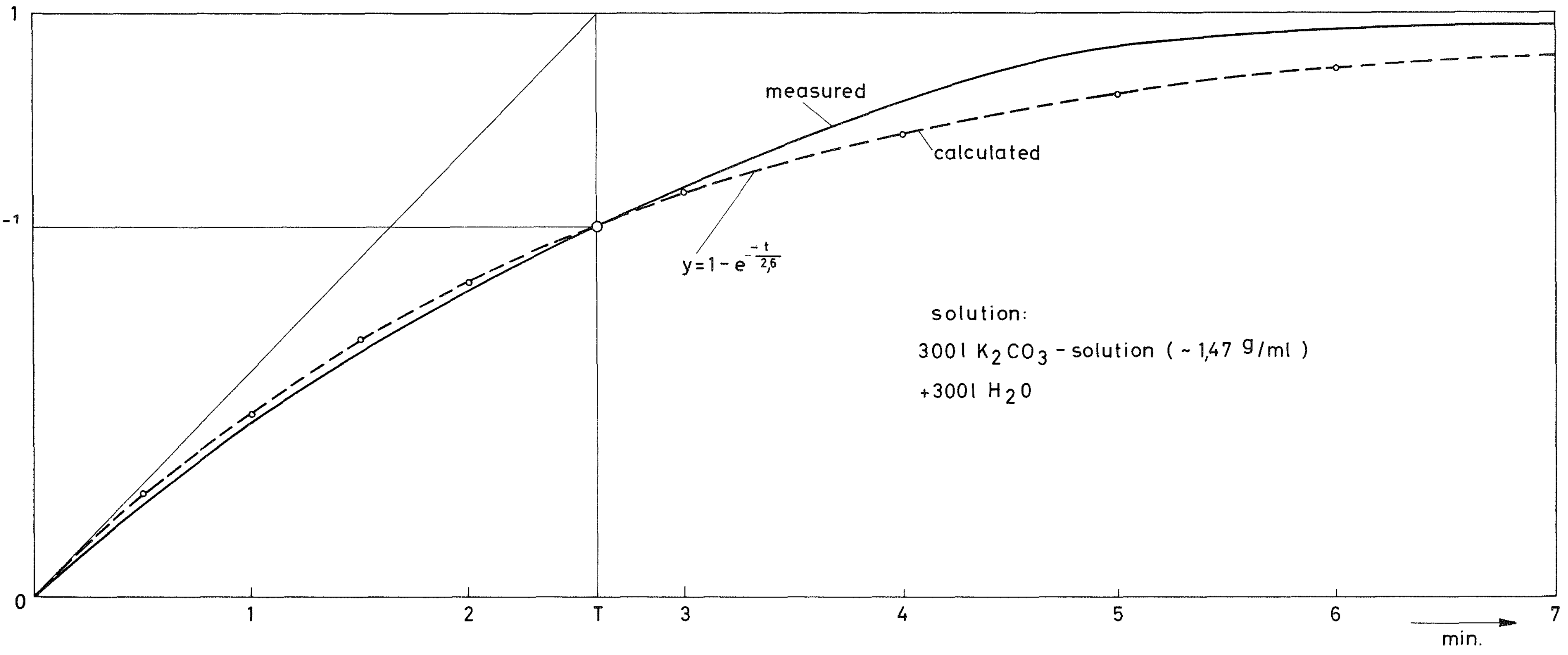


Fig. 22 Mixing behaviour of 2 liquids with different densities

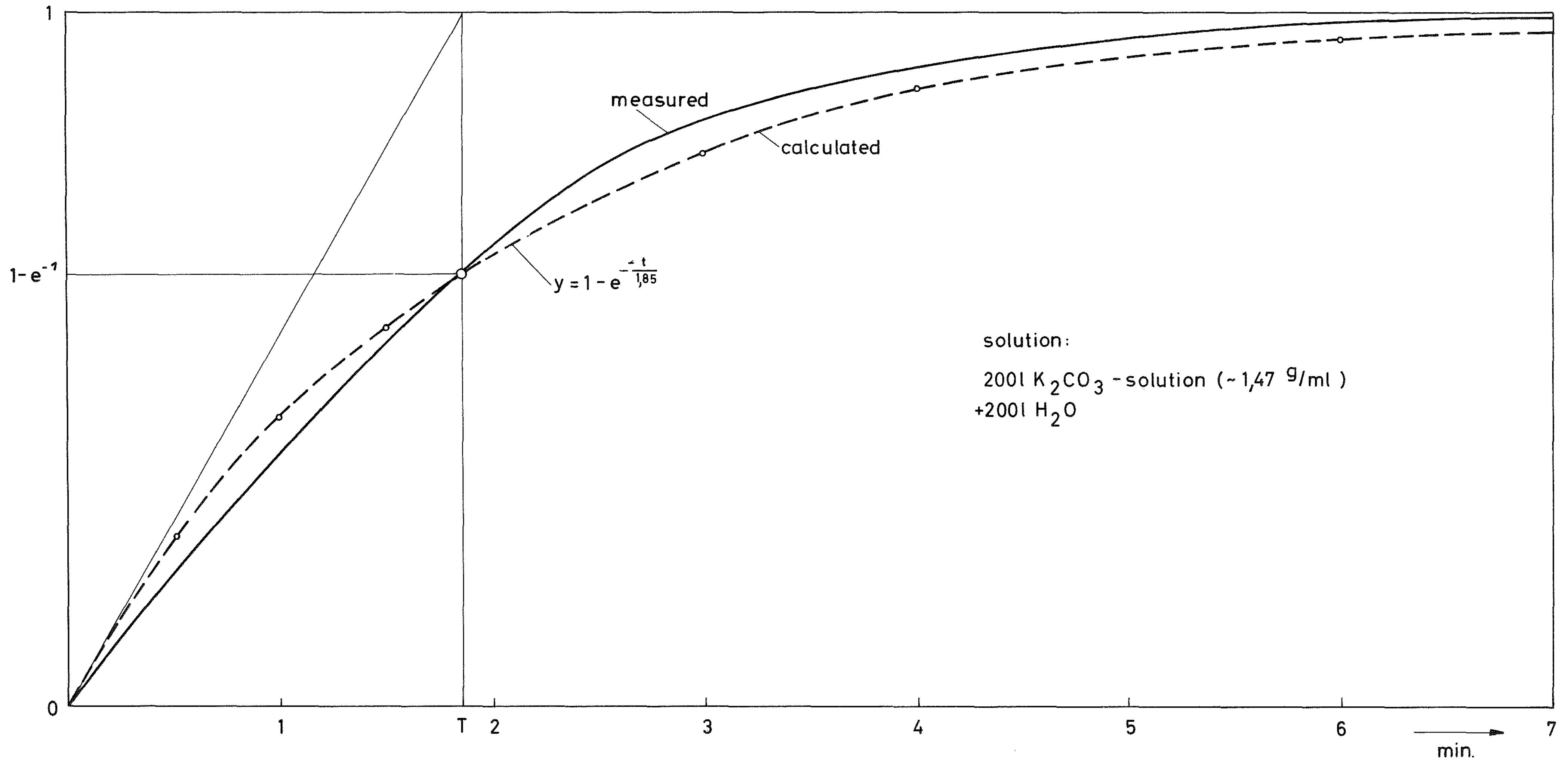


Fig. 23 Mixing behaviour of 2 liquids with different densities

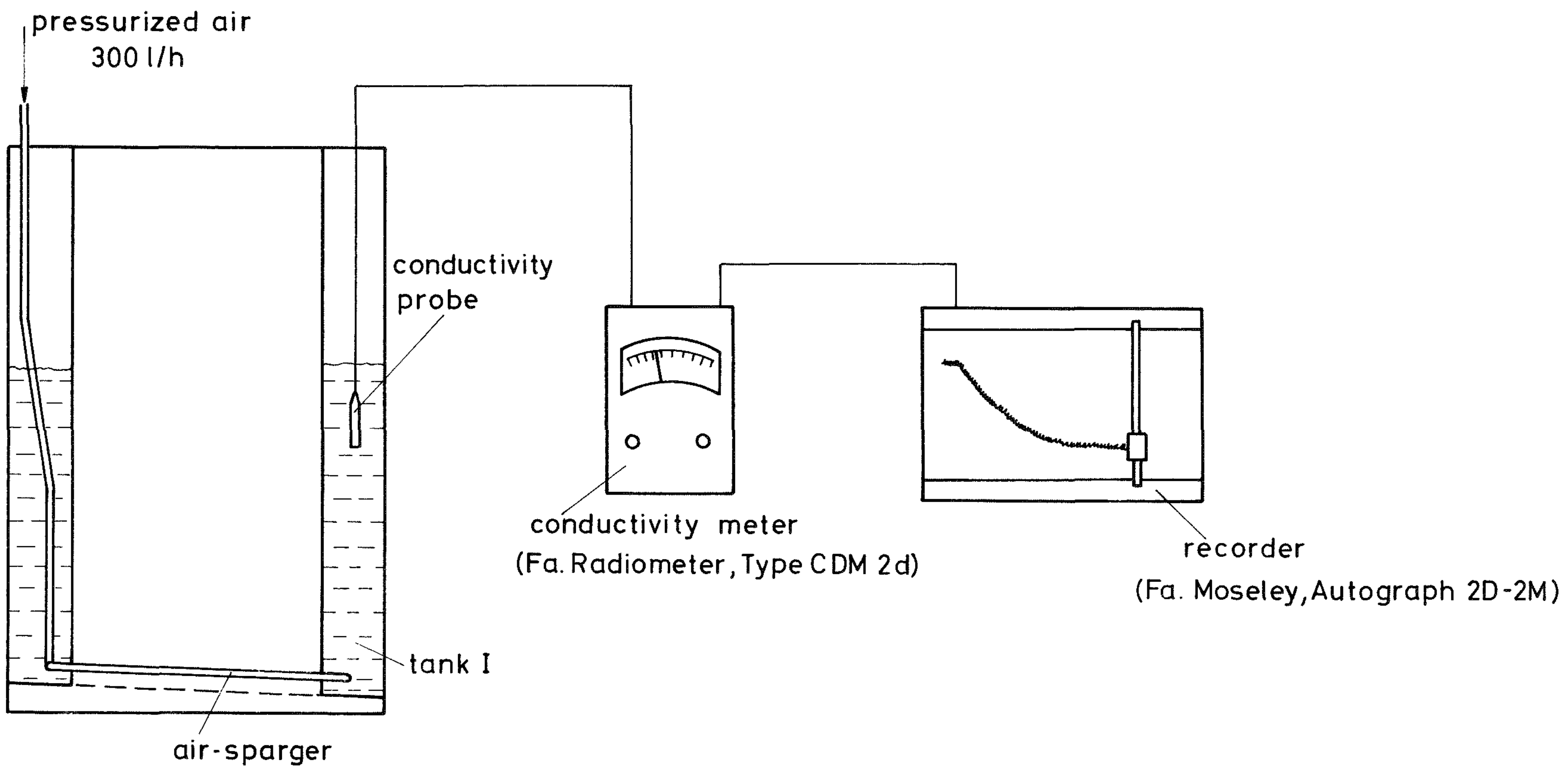


Fig. 24 Experimental set-up for conductivity measurement

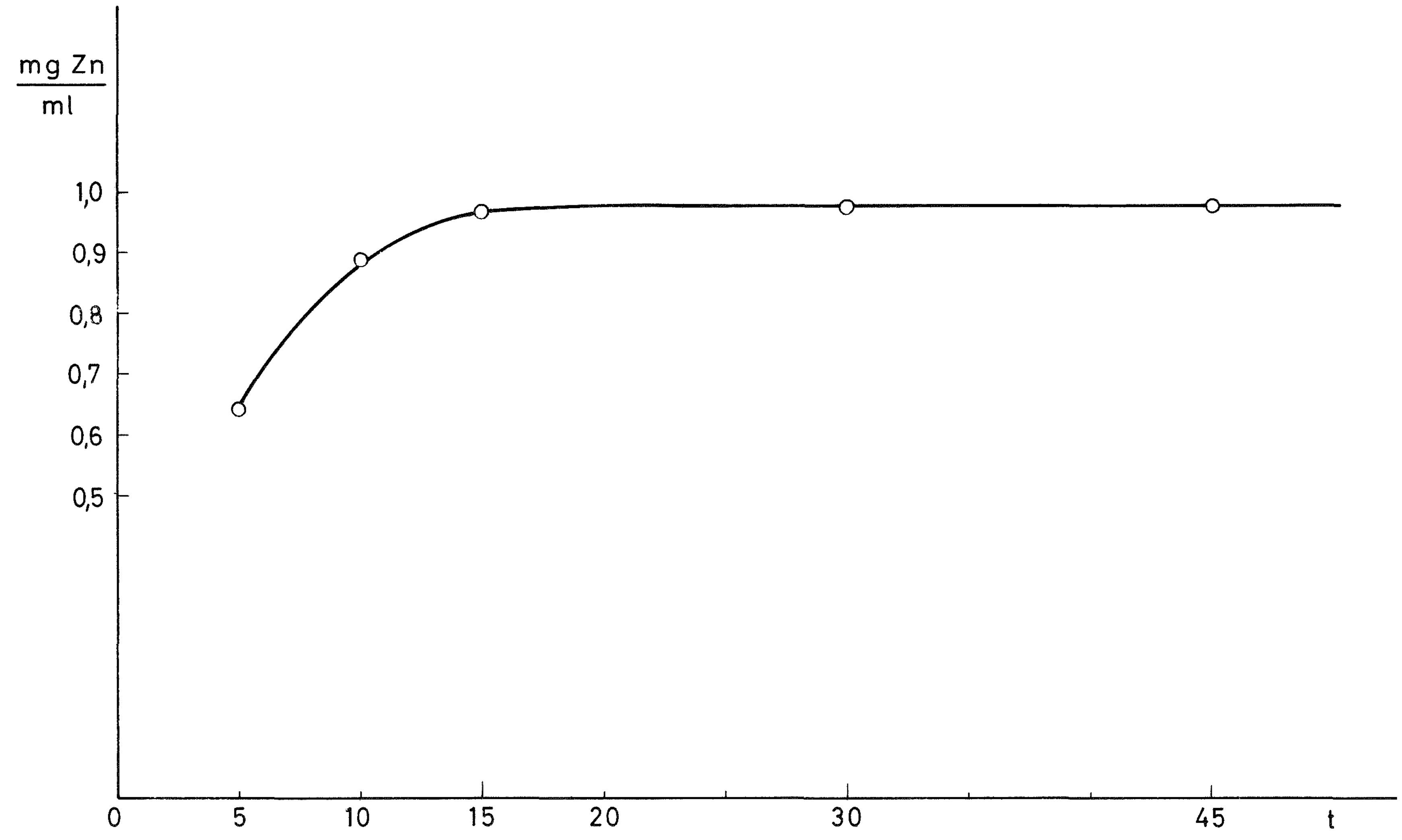


Fig. 25 Change of sample concentration dependent on circulation time

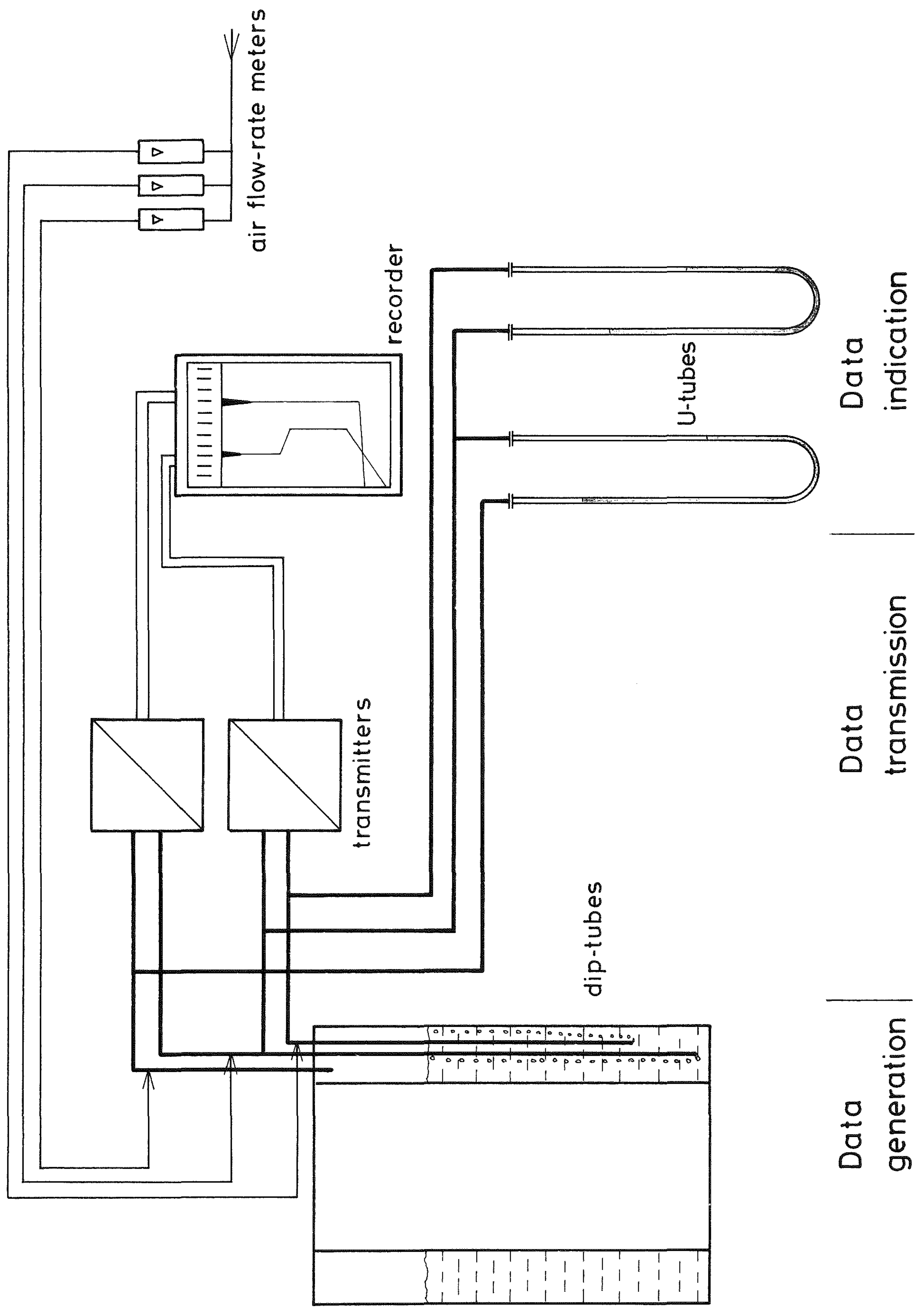


Fig. 26 Level and density measuring system with devision in data generation, transmission and indication areas.

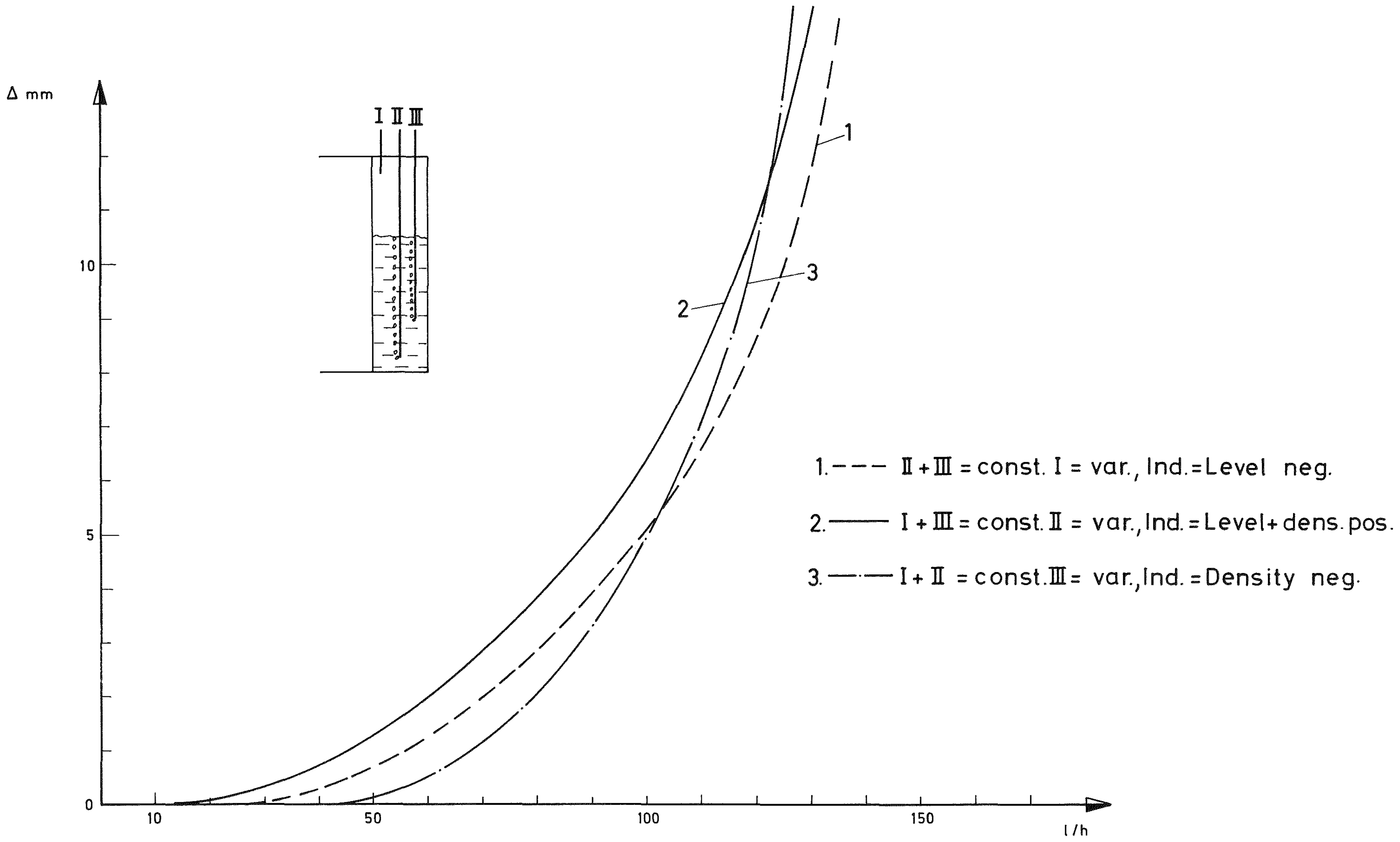


Fig. 27 Dependence of U-tube readings from dip-tube air flow rate.

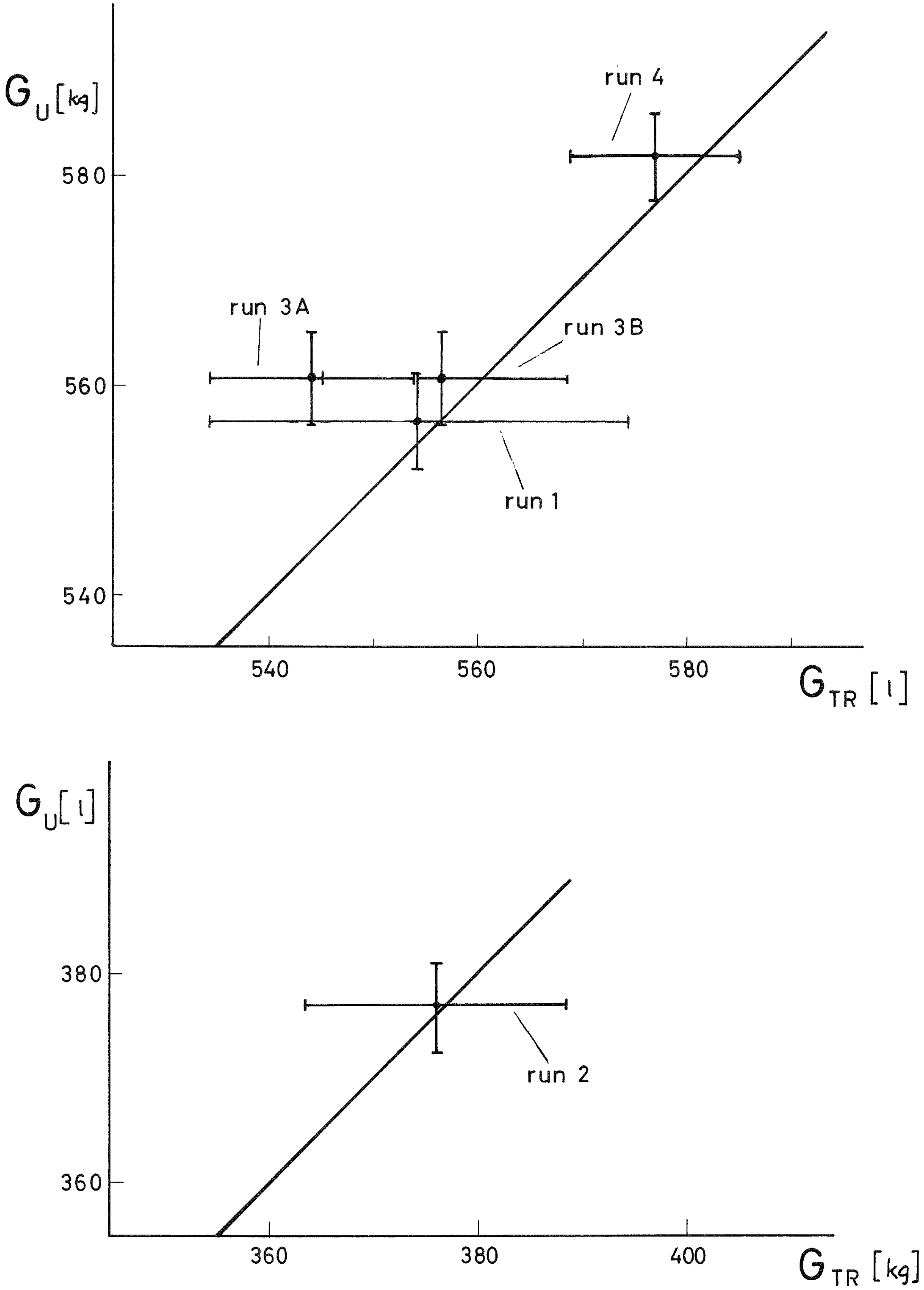


Fig. 28 Confidence region for points measured by U-tube and Zn-tracer method.

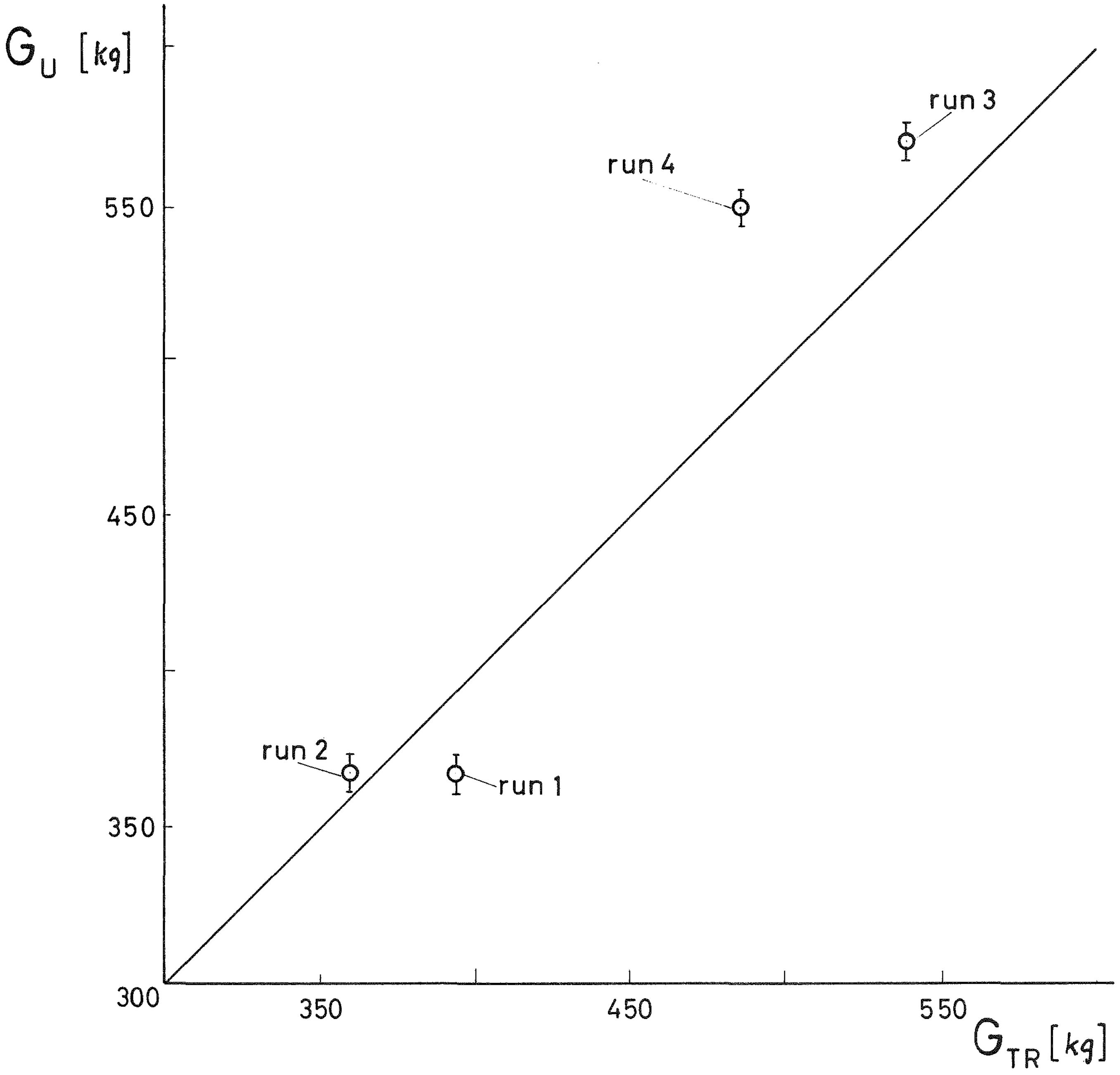


Fig. 29 Points measured by U-tube and Re-tracer method with confidence interval for U-tube measurement.

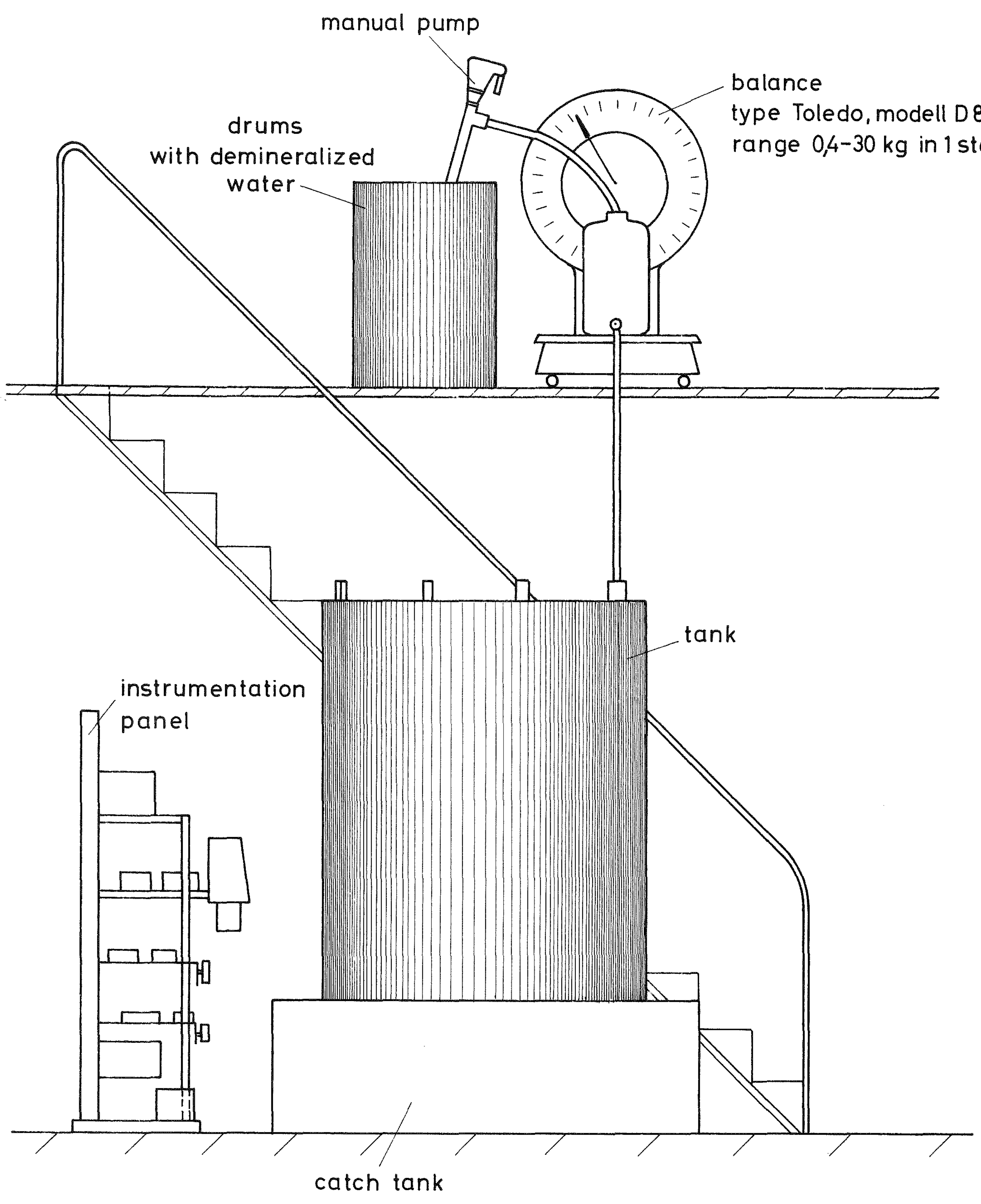


Fig. 30 Set-up for tank calibration

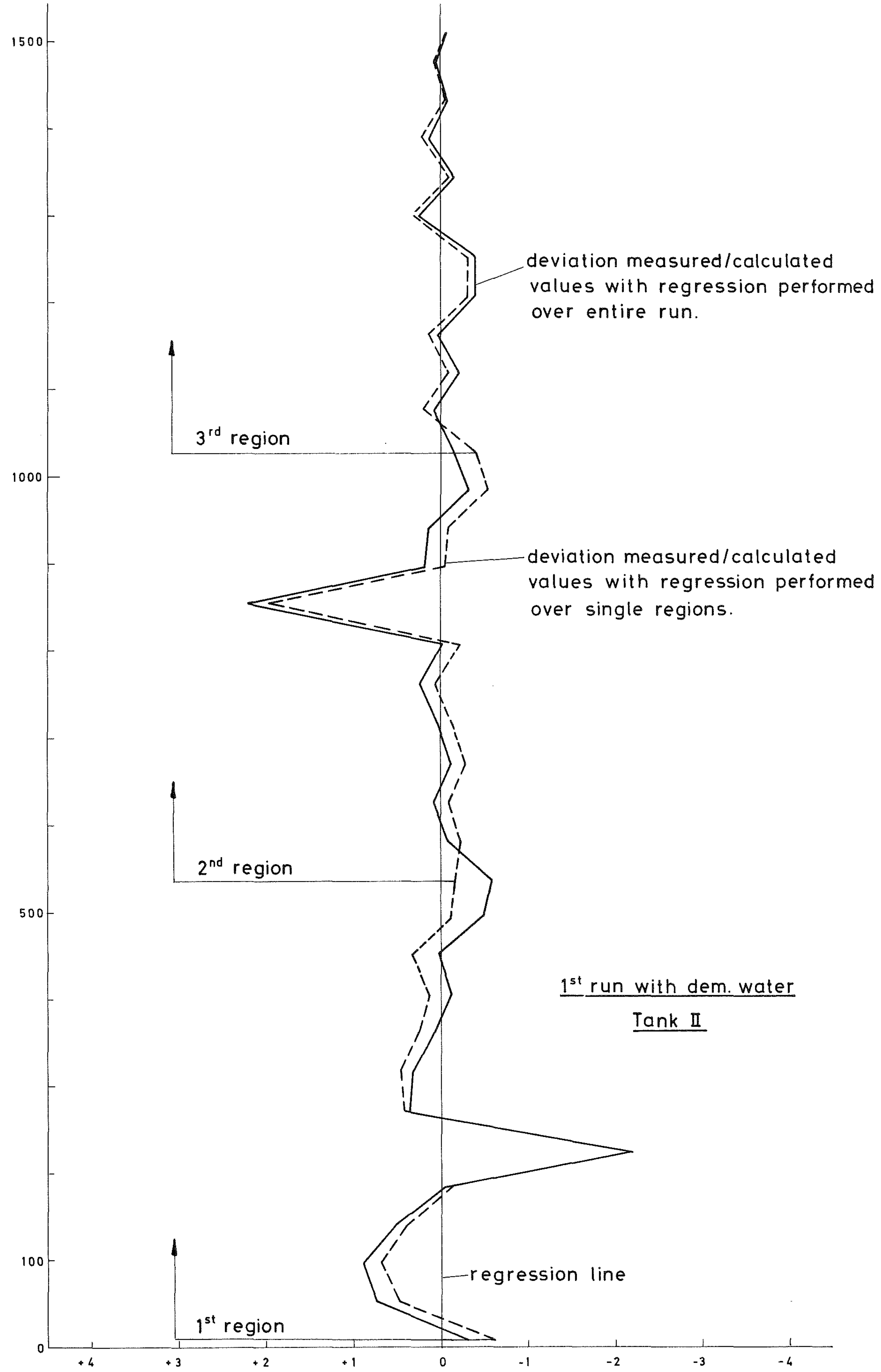


Fig. 31 Deviation of measured from regression values.

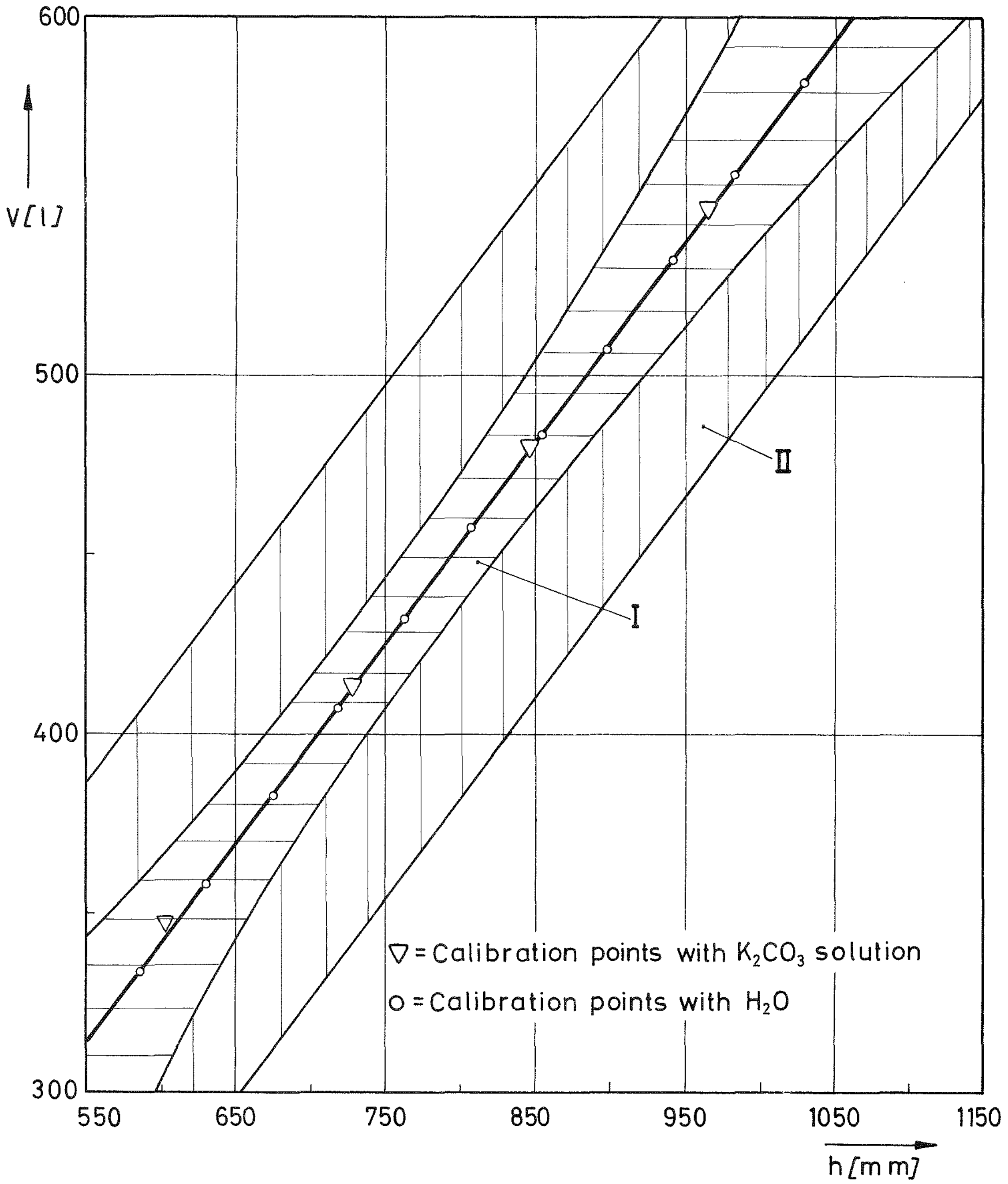
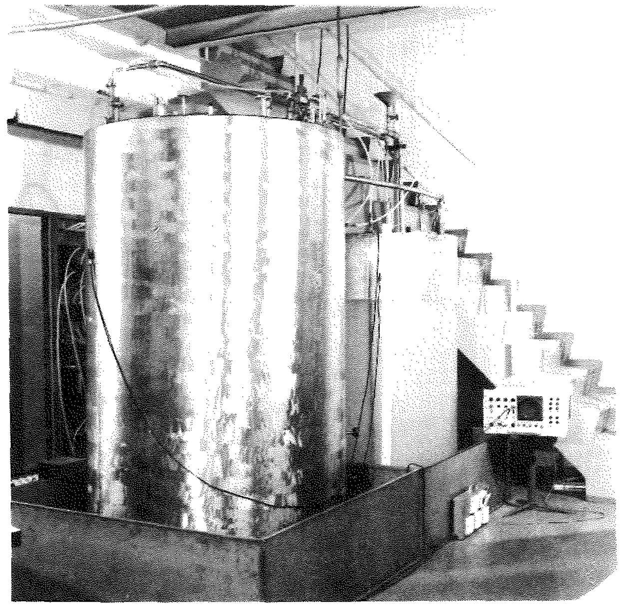
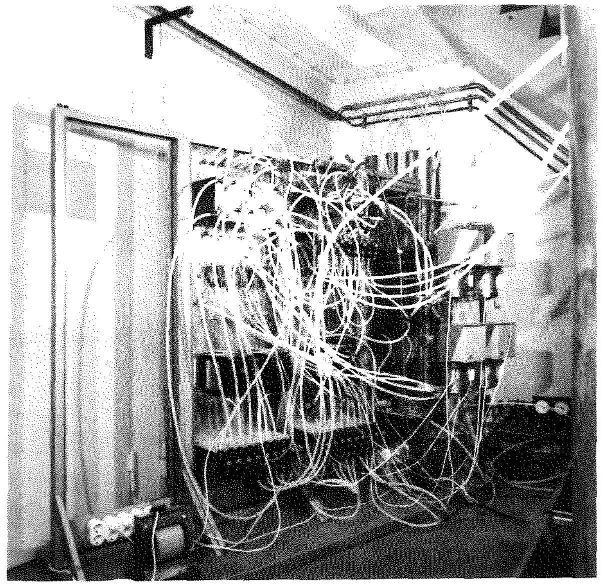


Fig. 32 Part of the calibration line with
100 times magnified confidence region for I. calibration,
20 times magnified confidence region for II. measurement after calibration.

AII.1 Mock-up

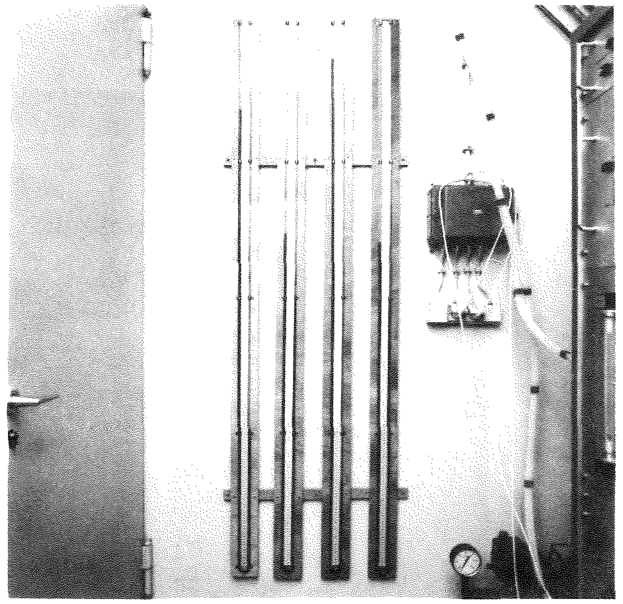


F1. General view of the mock-up. In front, Tank II, behind, Tank I, each in its corresponding catch-tank. On the right, the liquid sensing monitor. On the left, part of the instrumentation panel.

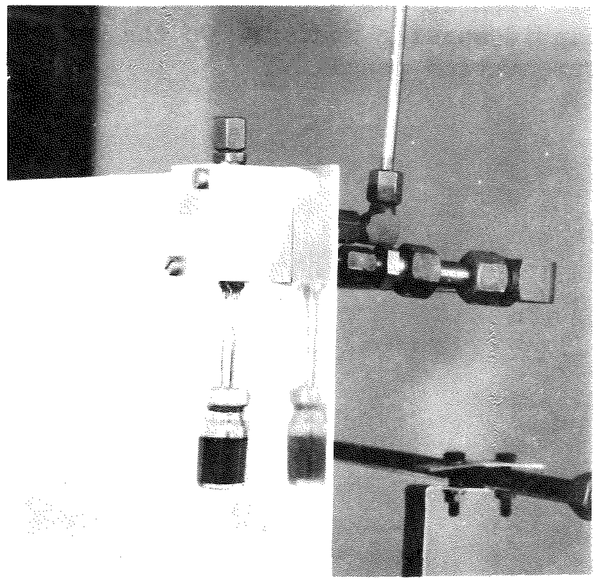


F2. Instrumentation panel, rear view during construction.

AII.2 Instrumentation

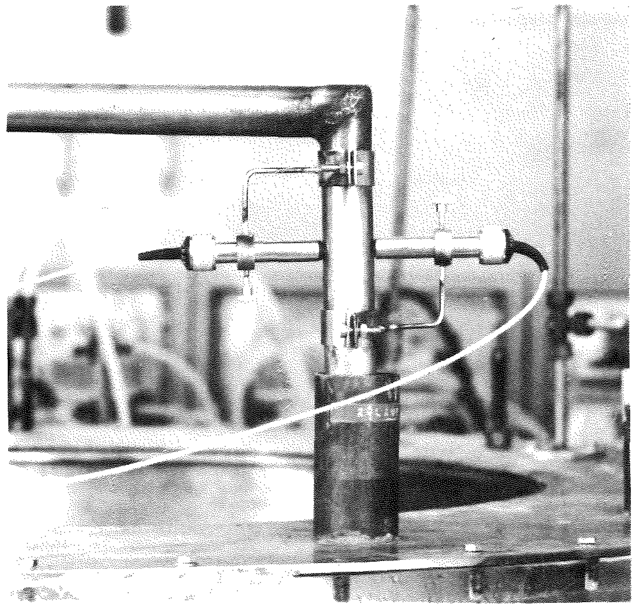


F3. U-tubes for level and density for both Tank I and II. The manometer liquid has been dyed for contrast reasons.

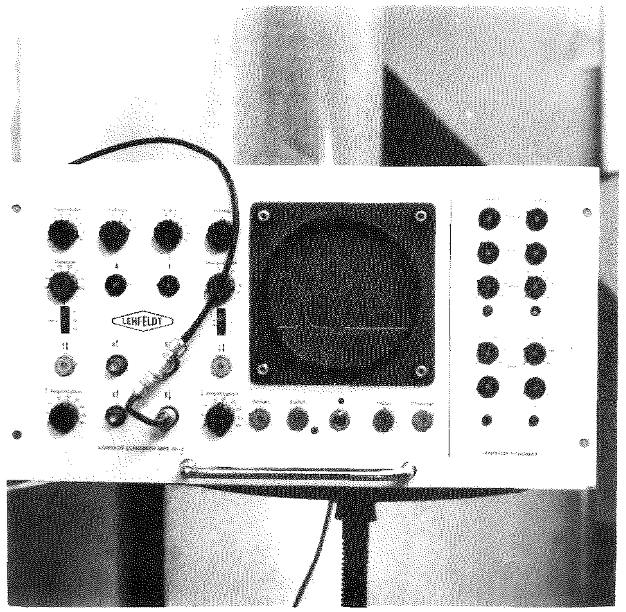


F4. Needle-block with attached sample vial for sample taking in Tank I.

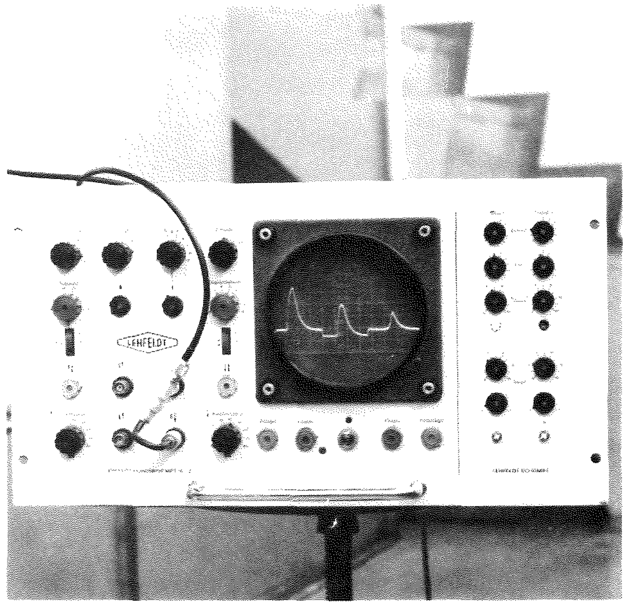
AII.3 Liquid indicating sensor



F5. Experimental set-up of 2 ultra-sonic sensors, one functioning as emittor, the other as receiver.



F6. Ultra-sonic monitor. The signal on the screen is the emitted signal.



F7. The same monitor. The first peak is the emitted signal, the second is the first echo signal (which will be used to trigger the alarm), the third is a second echo (circulation echo).