PATENT SPECIFICATION (11) 1 548 868

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(21) Application No. 34776/75 (22) Filed 21 Aug. 1975
(23) Complete Specification Filed 23 Apr. 1976
(44) Complete Specification Published 18 Jul. 1979
(51) INT. CL.^{2} G01B 15/00
(52) Index at Acceptance
G1A A2 A4 A9 D10 D11 D2 DB G10 G17
G1 G2 G5 G6 G7 MR P16 P1 P5
RX S10 S11 S6 T14 T15 T22 T25
T26 T27 T2 T3 T7 T8

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(54) INTERFACE DETECTION BY NEUTRON SCATTERING

(71) We, SHELL INTERNATIONAL RESEARCH MAATSCHAPPIJ B.V., a company organised under the laws of the Netherlands, of 30 Carel van Bylandtlaan, The Hague, The Netherlands, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:-

The invention relates to a method and apparatus for detecting an interface of materials having different hydrogen content, present in a metal vessel or pipe, e.g. made of steel.

Interfaces as indicated above occur frequently in the process industry. Examples are levels of liquid hydrocarbons in settlers, in absorption columns, in distillation columns, interfaces in pipelines for gas transport if two-phase flow occurs, etc. Walls of columns, reactors, pipelines and the like are usually of steel and it has always been of importance to have available detection methods which do not require special constructions on the steel walls such as sight glasses, lead-in wires for measuring equipment, etc.

According to the present invention there is provided a method for detecting an interface of materials having different hydrogen contents and which are present in a metal-walled vessel or pipe, e.g., made of steel, which comprises effecting a series of measurements of the hydrogen density of the contents of said vessel or pipe by use of at least one californium-252 neutron source located near or at the outer side of the metal wall of the vessel or pipe emits neutrons to the inside of the vessel or pipe, the neutrons being scattered by the contents of the vessel or pipe and being detected by at least one neutron detector located near or at the outerside of the metal wall, the detector(s) having a larger sensitivity for scattered neutrons than for neutrons emitted by the source(s), and monitoring the so-determined hydrogen densities for a change therein indicative of an interface.

This method is based on the fact that neutrons are transmitted through layers of steel and other heavy metals about as easily as light through a glass window, but are strongly scattered by the light hyrdrogen nuclei abundantly present in hydrocarbons or in water. When a suitable source and a detector that picks up the scattered neutrons are placed in appropriate locations outside the vessel amounts of hydrocarbons or water in the vessel as small as a few grams can thus be traced from behind a five centimetre steel wall and this is sufficient to detect the presence of an interface. Such an interface may be liquid hydrocarbons against their vapours or air or other gases, the level of its vapour or air or other gases, the level of pulverulant solid organic material containing hydrogen. The method does not detect an interface between liquid hydrocarbons and water, because of the comparable hydrogen content of these liquids.

Californium-252 is a man-made transuranium isotope with a half value time of 2.7 year. This nuclide produces neutrons by spontaneous fission with a yield of $2.3\text{x}10^6$ neutrons per second per microgram. A source containing about 0.1-1 microgram produces sufficient neutrons for most technical applications. The dose rate from a $1\mu g$ source having an activity of about 0.5 mCi at 1 m distance in air is within the limit of 2.5 millirem per hour, permitted for daily exposure of the radiological worker. This makes safe handling rather simple in comparison with most gamma-radiation sources generally used in the process industry. Most other neutron sources have much lower neutron yields and therefore sources of much larger activities are needed than Cf-252 which make the applications in process industries of other type neutron sources like Am-241-Be less attractive.

In a counting tube filled with helium-3 at a pressure of about 10 bar an uncharged neutron is converted by a reaction with He-3 into a tritium nucleus and a proton with a total kinetic energy of 764 keV. The charged particles produce an electron avalanche between the wire-shaped anode (at a potential of 2000 V) and the cathode or wall of the tube. The avalanches produce current pulses which are counted by appropriate equipment. Such a detector is very sensitive for scattered neutrons which owing to the collisions with hydrogen they have made energies somewhere between the thermal energy distribution and that of fast neutrons from the source. The detector has a lower sensitivity for fast neutrons. It is therefore possible for the detector to be in the neighbourhood of the source. The detector receives scattered neutrons and a much larger number of fast neutrons from the source, which cannot be shielded from radiation directly to the detector. Owing to the properties of the detector as indicated above this creates no problem.

The intensity of the scattered neutrons decreases by increasing distance from the source and it is therefore of importance that the distance between source and detector is not larger than 50 cm and preferably not larger than 25 cm. This improves the sensitivity of the detection. For measurements on vessels with large diameters use is made mainly of returning neutrons by positioning the source and the detector close to each other one side of the vessel. This avoids special constructions on opposite sides of the vessel as is required for gamma-ray absorption with the accompanying problems of alignment.

An attractive possibility is to locate source and detector in such a way that the centre of gravity of both are in a plane which is perpendicular to the centre line of the vessel or pipe. This may be achieved by positioning source and detector along a circle line against the outside of the steel wall of a vessel. The two components may then be combined in one probe which can be moved along that wall. Another possibility is to position the source and the detector behind each other, the source being nearest to the wall. Both geometries have the advantage of sharp detection of levels in a vessel or column and the combination of source and detector in one probe promotes ease of handling.

Notwithstanding that the dose rate of a Californium-252 source is already very low, it may be further decreased by means of a shielding.

The apparatus suitable for use in the method according to the invention therefore comprises a neutron source of Californium-252, a detector having a larger sensitivity for scattered neutrons than for neutrons emitted by the source, a shielding enveloping said detector and a source at least partly, which shielding consists of a reflector, a moderator and a layer of material between said reflector and moderator, which pregents substantially the passage of slow neutrons.

The method and apparatus according to the invention is very suitable for the detection of liquid water and/or liquid hyrdrocarbons present in pipelines for gas transport. The presence of liquid in a gas transport pipeline should be avoided as much as possible because the resulting two-phase flow decreases the capacity of the pipeline for gas transport and it is of importance for the operator of the pipeline to be informed on the occurrence of two-phase flow in order to enable him to take measures. One source and one detector may be located around the wall of a pipeline opposite each other for a pipeline with a diameter of less than 10 cm. Any slug passing this location will be detected. Still bettter results are obtained with three sources and three detectors located alternatively around the wall of a pipeline at equal distances. Apart from the detection of the presence of liquid information is obtained on the volume fraction of liquid present at the measuring spot. In the light of the above it is clear that another number of sources and detectors, alternating and equally spaced, will be optimal under certain circumstances.

The invention is very suitable for the detection of the level of a liquid such a liquid hydrocarbon in a vessel or a column. It is furthermore possible to detect the foam height of such a liquid in a vessel or a column. An intersecting application is the detection of level of a fluid bed of particles of hydrogen containing solid material such as polymeric material.

The detection of levels as indicated above may be used for measurment of the height of a liquid column, a foam or a fluid bed, as well as for alarming and control purposes.

The invention will further be elucidated with reference to the drawings and number of examples.

Fig. 1a shows, schematically, a measuring set-up for a helium-3 detector 1 with a californium-252 source 2. The detector being provided with a pre-amplifier 3, a high-voltage bias supply 4, an amplifier analyser 5 and a digital rate meter 6.

Fig.1b and c show the detection system, like reference numerals denote like parts, which may be placed in a shielding 7, consisting of a graphite reflector 8, a paraffin wax moderator 9 and a layer 10 of cadmium between the graphite reflector 8 and the paraffin moderator 9. The graphite 8 reflects the neutrons from the source 2 towards the medium (not shown) to be measured. The neutrons which pass through the graphite 8 are absorbed by the paraffin wax 9. To prevent slow neutrons returning from the paraffin wax 9 to the detector 1 a layer 10 of cadmium is provided. This shielding increases the background signal at the detector 1, but this increase is more than compensated for by an increase in the signal from the measuring medium. The inclusion of the shielding 7 is thus equivalent to an increase in the source strength.

This shielding thus has two advantages:

1. Reduction of the dose rate by at least a factor of 3.

2. A reduction in background signal variation due to variations in the immediate surroundings. For example, if the shielding is omitted, a person approaching the detector causes a slight increase in the back ground signal.

This set-up is preferably used in the Examples I to V which will now be discussed.

EXAMPLE 1

A settler made of 2 cm thick steel, with an outlet at 2.55 m height and with a diameter of 2.00 m was used to separate an alkylate/propylene mixture from water. The neutron scattering method was used to detect the alkylate-propylene/gas interface for which purpose a probe was used containing 2.5 µg californium-252 as a neutron source and a helium-3 detector mounted on an aluminium pipe which could be moved along the outer surface of the steel wall. The neutron-scattering intensity is given in counts per second as a function of height above the bottom of the settler in figure 2. The plotted number are net counts being the difference between observed number of counts and the number of counts ($\sim$700 c.p. 10 sec) without any scattering medium present. A strong increase in signal is observed when the probe is moved outside the settler in downward direction. The level found at 2.55 ± 0.05 m corresponds exactly with the centre of the alkylate/polypropylene outlet.

The settler is indicated in figure 2 as 11, with an inlet 12, a bottom outlet 13 for water and an outlet 4 for alkylate/propylene. The height scale in the graph corresponds with the actual height at the settler.

EXAMPLE II

Similar measurements as described in example 1 have been carried out on a flasher of a propane deasphalting unit in order to detect foam and liquid levels.

In figure 3 the flasher is indicated by 14 and consists of a column of 10 m height and 3 m diameter made of steel. Two trays 15 and 16 are present. The feed inlet with a tray is indicated by 17. A wire mat 18 is present to prevent liquid droplets leaving the column. An outlet 19 for gas is present at the top and an outlet for liquid 20 near the bottom. The height scale in the graph corresponds with the actual height at the flasher.

It is shown in the graph that the liquid level in the column is present at 3.6 m. The smaller peaks in the counts indicate liquid present on the trays and the feed inlet tray. There is virtually no foam above the feed inlet as is clear from the low counting rate above 7m height.

EXAMPLE III

Similar measurements as described in examples I and II have been carried out on an asphalt flasher unit. In figure 4 the flasher is indicated by 21 and consists of a column of 7 m height and 2.3 m diameter made of steel. The feed inlet is indicated by 22. A wire mat 23 is present to prevent liquid droplets leaving the column. An outlet 24 for gas is present at the top and an outlet for liqud 25 near the bottom. There are no trays in this flasher. The height scale in the graph corresponds with the actual height at the flasher.

The liquid level is present at 1.1 m as is clear from the sharp increase in counting rate. No foam is present above the liquid.

EXAMPLE IV

Similar measurements as described above have been carried out on a flasher for deasphalted oil. In figure 5 the flasher is indicated by 31. It is provided with a feed inlet and tray 32, a gas outlet 33 and an outlet for liquid 34. A wire mat 35 is also present and the height scale in the graph again corresponds with the actual height at the flasher.

This flasher below the feed tray is filled with foam as is clear from the counting rates below a height of 8 m. In this flasher no liquid level could be detected because of the presence of a concrete support at the location where the level was expected. The measuring points show a distinct scatter which is probably caused by density fluctuations of the foam.

EXAMPLE V

Measurements have been carried out on a stripper which is used to dry polypropylene powder with nitrogen in order to detect the surface of the fluid bed. The stripper is a steel vessel with a height of 3.9 m and a diameter of 1.6 m. Fluidization of the contents was carried out with 750 kg n_2 per hour. Results of measurements along a vertical line outside the wall are shown in figure 6. It is clearly shown that a strong signal increase, due to the presence of polypropylene is found if the probe is moved down the reactor wall.

From the examples I to V it can be concluded that the gauge, consisting of a californium-252 source and a helium-3 detector, is an effective tool for external detection of hydrocarbon levels in vessels. Further experiments for testing the neutron gauge as an external level detector were carried out with an artificial hydrocarbon level, which was created behind a steel wall by piling paraffin wax bricks behind various steel plates with thicknesses from 2.5 to 14 mm. The gauge was located on the other side of the steel plate at a fixed position. By removing rows of paraffin wax bricks the level was effectively moved with respect to the gauge. It appeared to be possible to detect levels behind steel walls of up to 14 cm thickness.

Experiments also showed that depending upon the response time of the gauge the source strength may be recued to only 0.1μ gram for several applications.

It will further be shown that the present method, as well as apparatuses are extremely useful in acquiring a better understanding of two-phase flow phenomena.

In the production and processing of natural gas, the heavier components of the gas tend to form a liquid phase called condensate. This liquid may be either water or hydrocarbons with five or more carbon atoms in the molecule. The condensate is, in general, transported along with the natural gas until it is removed by gas condensate separators. Whilst in the pipeline, however, the condensate affects the transport of the natural gas. A better understanding of two-phase flow phenomena is thus especially important in the design of natural gas transport systems.

In a bech-scale set-up the detection of hydrogen containing materials with one Cf-252 source and a He-3 detector was studied. For this purpose use was made of a piece of 30 cm 3" I.D. gas transport pipe with a steel wall thickness of 15 mm. The ends were closed by two metal prices welded on the pipe. This container was connected to a supply of water so as to be able to fill it gradually, while it had an open connection with the atmosphere. The source and detector were located with respect to the pipe diametrically opposite each other. Gamma-ray attenuation measurements were made on the same pipe. The signals at zero hold-up, i.e. only air in the pipe were normalized to 100%; an increasing hold-up gave an increase in signal for the neutron scattering, while an exponential decrease was observed, as expected, for the gamma signals. The gamma-ray absorption only yields information about the liquid hold-up in the optical path of the gamma-ray beams while the neutron scattering gives a signal related to the total amount of liquid present in this pipe.

It appeared that the gauge has its lowest sensitivity for small hold-ups. For a hold-up increase from zero to 0.1 a signal increase of 50% is found while from zero to 0.2 liquid fraction, the signal increases by 300%. This initial low sensitivity is probably due to the fact that several neutron-proton collisions are needed to sufficiently slow down a neutron and to achieve a substantial increase in detection probability. If only a small amount of hydrocarbon is present the relative probability of more than one collision gets very small.

This low initial sensitivity can be a disadvantage if small hold-ups have to be measured accurately when no other scattering material such as high pressure gas-phase is present. To solve this problem polyethylene covers were constructed. The amount of atomic hydrogen (13 g) and the geometry needed to achieve optimal conditions have been determined experimentally. It shall be clear that any material, other than polyethylene, which contains hydrogen will be suitable for this purpose.

The location of the liquid phase in a gas transport pipe, with respect to the source and detector will affect the measured signal. The signal will also be affected by the position of the source with respect to the detector. These dependences are referred to as geometry effects.

In actual gas transport pipelines there is no a priori knowledge of the two-phase flow geometry. This will give a large inaccuracy in the hold-up determination if only one source and one detector are used. In the following a measuring set-up using several sources and detectors is discussed which almost completely eliminates geometry effects.

Fig. 7 shows, schematically, a measuring set-up employing three sources 42', 42" and 42"' and three detectors 43', 43" and 43"' placed alternatingly and symmetrically around a pipe 41, which is partly filled with water 44.

The signals of each separate detector will be different, although to a lesser extent for the detectors 43" and 43"', however the sum of the signals will be a measure for the quantity of water present. Experiments showed that for pipe diameters up to 5" an arrangement with three detectors and three sources is within 1% standard deviation independent of geometry effects. Said experiments were carried out by displacing the sources and detectors with respect to the water, as well as by means of three concentric rings of glass tubes, which were placed inside the pipe (not shown) and could separately be filled with water.

It was found that there is a well-defined correlation between the sum signal of the three detectors and the liquid hold-up in a gas transport tube. By calibration a signal can be converted into a hold-up value. It was further possible, by comparing the three separate detector signals, to determine the location of the liquid present in the pipe.

No scattering due to the gas phase occurred in the experiments which were hereinbefore discussed with respect to pipes. However, in actual gas transport lines, where pressures may be as high as 250 bar, the density of the gas phase is sufficiently high to contribute significantly to the neutron scattering.

To simulate an actual gas transport pipe on a laboratory scale a 15.1 gas cylinder with an I.D. of 14 cm, was pressurized with methane.

Fig. 8 shows the signal increase as a function of gas pressure, indicating that the neutron scattering gauge can also be used as completely external pressure gauge. It should be noted, however, that the neutron scattering signal is only indirectly related to the gas pressure, the gauge in fact determines the density of hydrogen atoms inside the cylinder.

To simulate the two-phases in natural gas transport lines as closely as possible, the condensate was replaced by water which has about equal hydrogen content, on a volume basis, is easier to handle and has a negligible solubility for methane. The gas phase was methane at 100 bar. In this experiment the detector-covers were not used since the gas phase has sufficient initial hydrogen at zero hold-up.

Fig. 9 shows the sum signal of the three detectors versus liquid hold-up in the cylinder. The signal offset of about 5% is due to neutron scattering in the gas phase. This result indicates that hold-up measurements for two-phase flow can be made on actual gas transport lines. If the gas pipeline is considerably larger than 14 cm. I.D. then more than three detectors and sources may be applied.

Time-dependent two-phase flow measurements were made with a pipeline for experimental purposes of 100 m length and 5 cm diameter. Air was pumped through this pipe together with water in various amounts. Three sources and three detectors were placed around the pipe in an aribtrary location. Scattering measurments were carried out as a function of time. It depends very much on the air velocity and the amount of water in which way the water passes through the pipe. The air blows over the surface of the water and causes waves, the magnitude of which varies from ripples to surges. The measurements of the three detectors have been added and used to calculate the hold-up of water with the aid of a calibration carried out previously. Results are shown in figure 10 and figure 11. In figure 10 the arrival of water at the measuring location occurred at 1.5 × 10^3 sec. The volume fraction of water was virtually constant after 2.5 × 10^3 sec. at a level of 0.33. Small, thin waves were visible at more or less regular intervals.

In figure 11 the same amount of water was used and a much higher air velocity. Large peaks were detected, almost reaching through the entire cross-section of the pipe.

WHAT WE CLAIM IS:–

1. A method for detecting an interface of materials having different hydrogen contents and which are present in a metal-walled vessel or pipe, e.g. made of steel, which comprises effecting a series of measurements of the hydrogen density of the contents of said vessel or pipe by use of at least one californium-252 neutron source located near or at the outerside of the metal wall of the vessel or pipe emits neutrons to the inside of the vessel or pipe, the neutrons being scattered by the contents of the vessel or pipe and being detected by at least one neutron detector located near or at the outerside of the metal wall, the detector(s) having a larger sensitivity for scattered neutrons than for neutrons emitted by the source)s), and monitoring the so-determined hydrogen densities for a change therein indicative of an interface.

2. A method as claimed in claim 1, in which the or each neutron detector is a proportional counting tube filled with helium-3.

3. A method as claimed in claim 1 or 2, in which the source(s) and the detector(s) are positioned along a circle line near the outside of the metal wall of the vessel or pipe in such a way that the centre of gravity of both are in a plane which is perpendicular to the centre line of the vessel or pipe.

4. A method as claimed in claim 1 or 2, in which the source(s) and the detector(s) are positioned behind each other respectively, the source(s) being nearest to the wall.

5. A method as claimed in any one of claims 1-4, which is applied for the detection of the level of a liquid in a vessel or a column.

6. A method as claimed in any one of the claims 1-4, which is applied for the detection of foam height of a liquid in a vessel or a column.

7. A method as claimed in any one of the claims 1-4, which is applied for the detection of the level of a fluid bed of particles of hydrogen containing solid material.

8. A method as claimed in any one of the claims 1-3, in which at least one source and one detector are located around the wall of a pipeline.

9. A method as claimed in claim 8, in which three sources and three detectors are located alternatively around the wall of a pipeline at equal distances.

10. A method as claimed in claim 8 or 9, which is applied for the detection of liquid water and/or liquid hydrocarbons present in pipelines for gas transport.

11. A method according to claims 8 or 9

and 10, wherein more than one detector is applied comprising comparing the signals of each of the detectors with each other so as to determine the location of the liquid in the pipeline for gas transport.

12. Apparatus suitable for carrying out the method according to any one of the claims 1-7, comprising a neutron source of californium-252, a detector having a larger sensitivity for scattered neutrons than for neutrons emitted by the source, a shielding enveloping said detector and source at least partly, which shielding consists of a reflector, a moderator and a layer of material between said reflector and moderator, which prevents substantially the passage of slow neutrons.

13. Apparatus according to claim 12 comprising as detector a proportional counting tube filled with helium-3.

14. Apparatus according to any one of claims 12 and 13, in which said layer of material is made of cadmium.

15. Apparatus suitable for carrying out the method according to any one of claims 1-11, comprising a detector being at least partly enveloped with a cover made of hydrogen containing material.

16. Apparatus according to claim 15 in which the cover is made of polyethylene.

17. A method as hereinbefore described with reference to the drawings and examples.

18. An apparatus as hereinbefore described with reference to the drawings.

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Printed for Her Majesty's Stationery Office, by Croydon Printing Company Limited, Croydon, Surrey, 1979.
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from
which copies may be obtained.

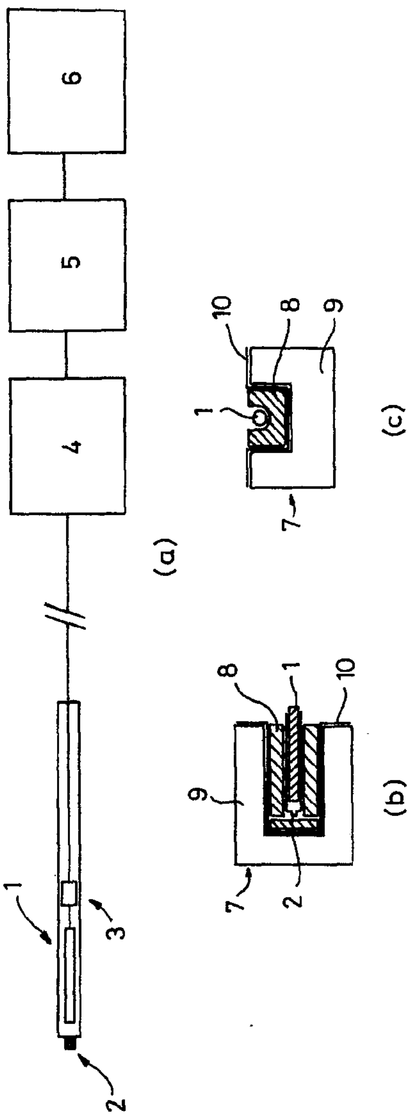

FIG.1

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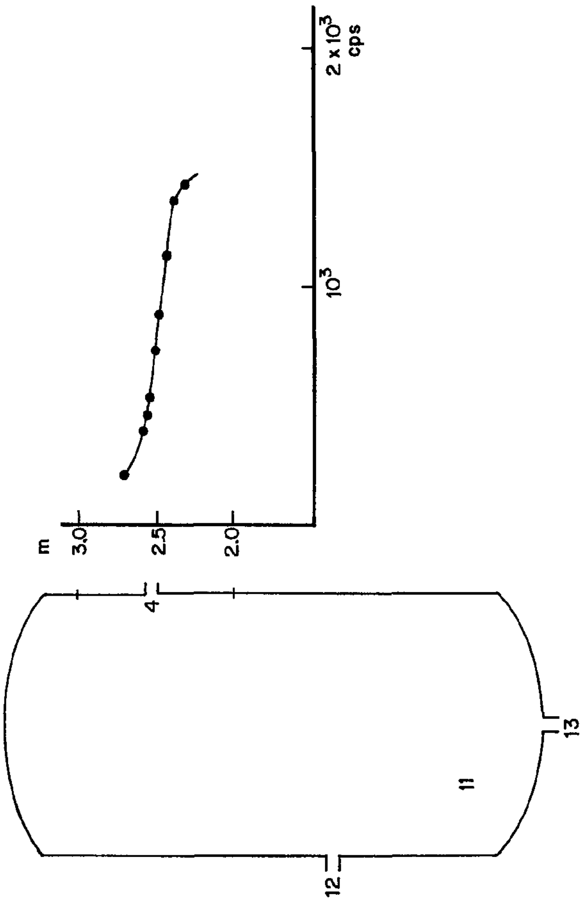

FIG. 2

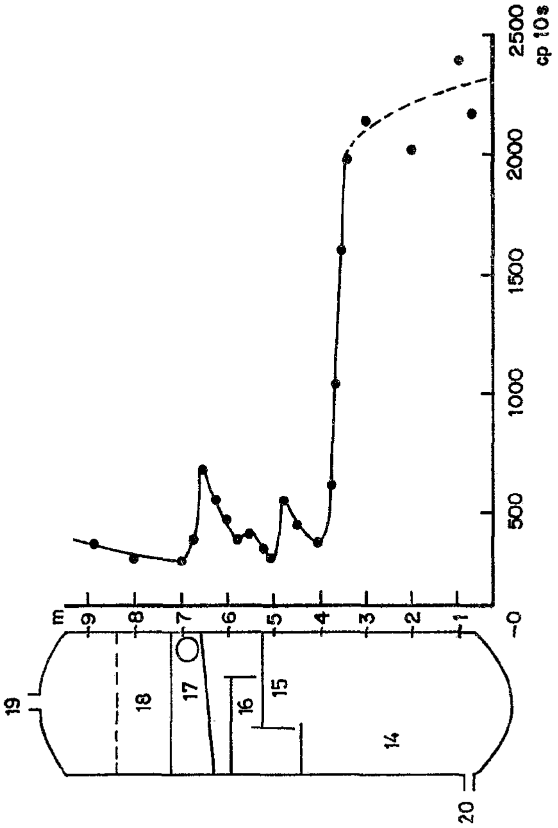

FIG.3

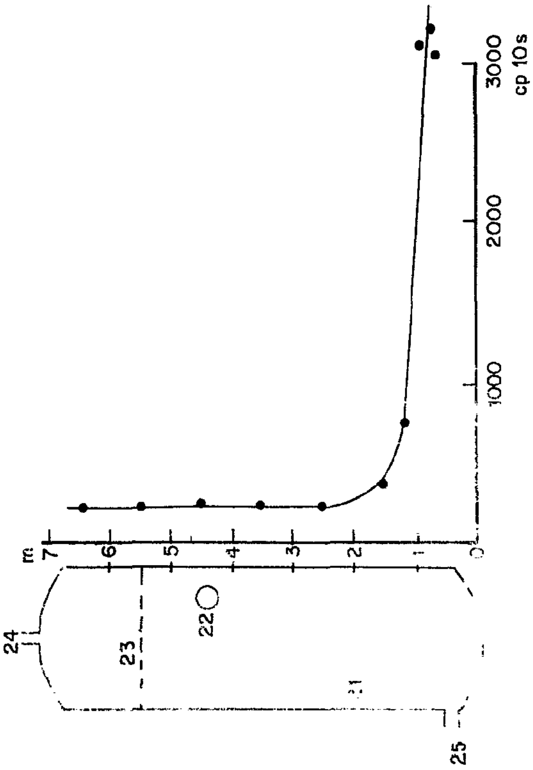

FIG 4

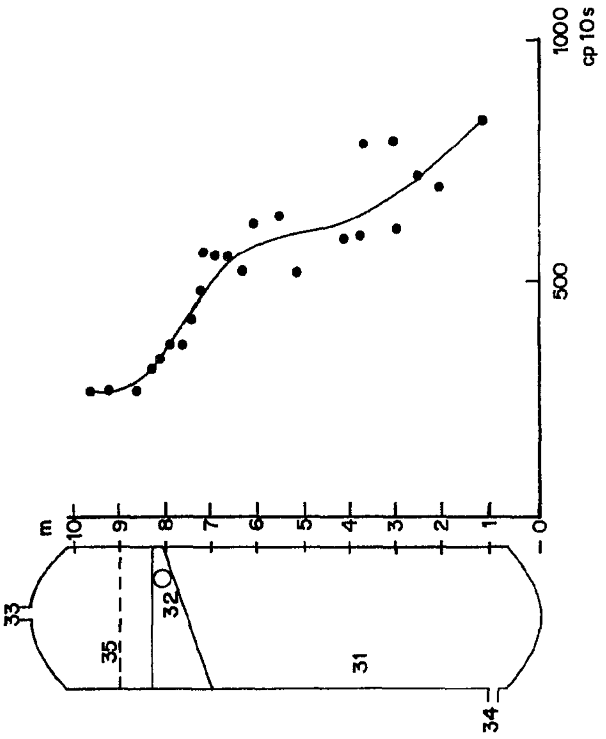

FIG. 5

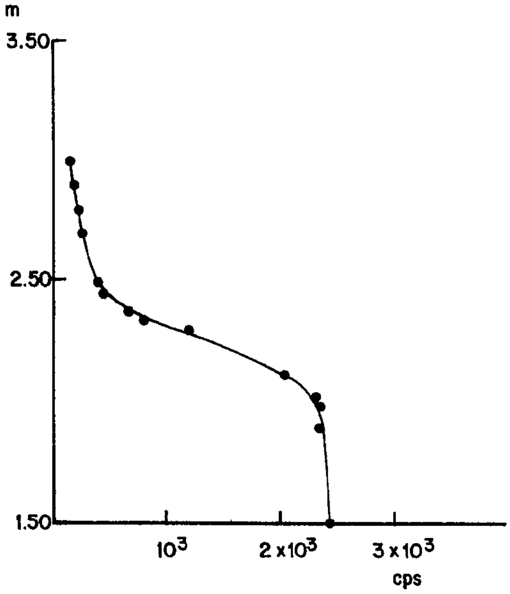

FIG. 6

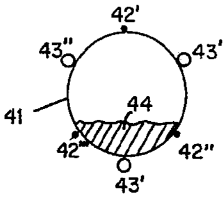

FIG.7

1548868 COMPLETE SPECIFICATION

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Sheet 7

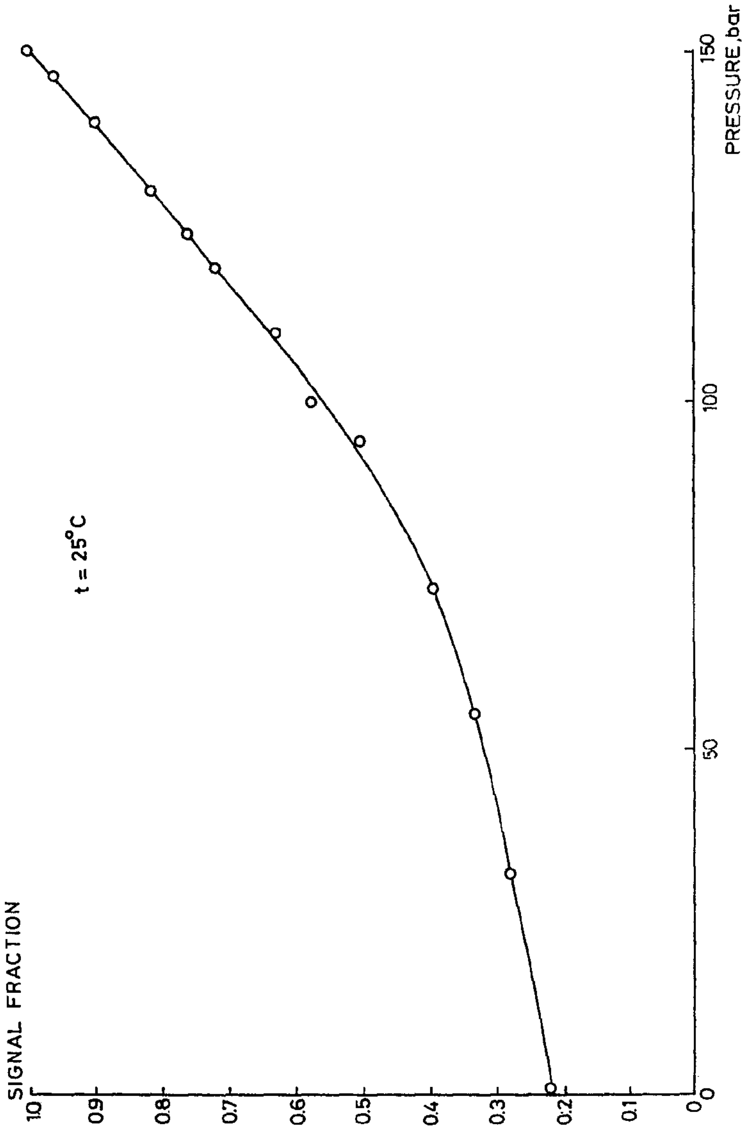

FIG. 8

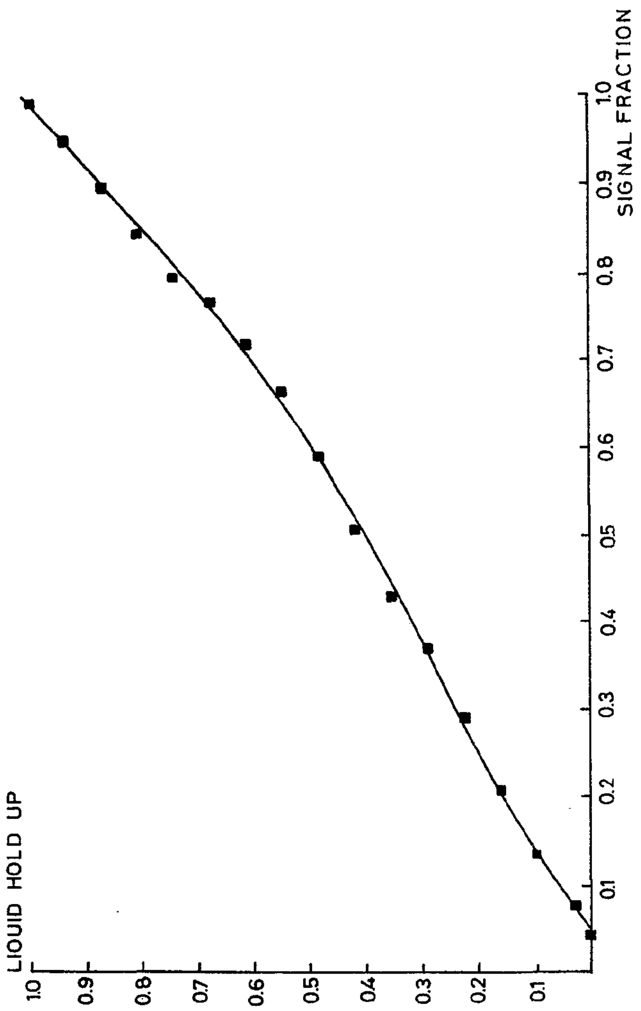
LIQUID HOLD UP
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
SIGNAL FRACTION
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0

FIG.9

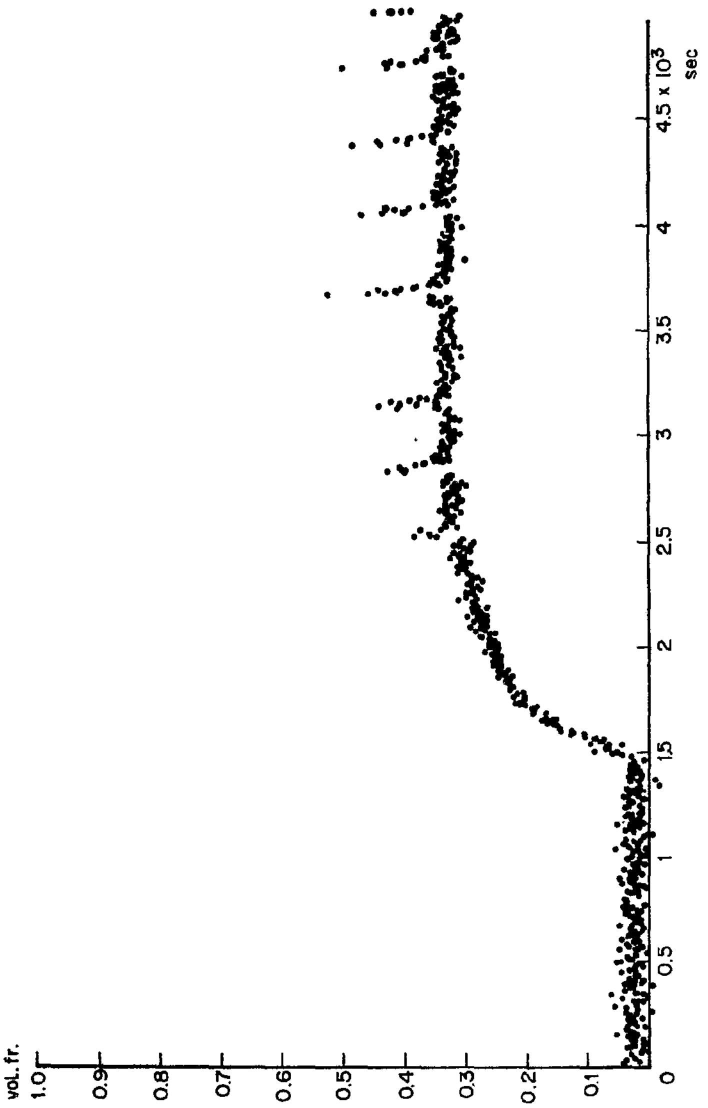

FIG.10

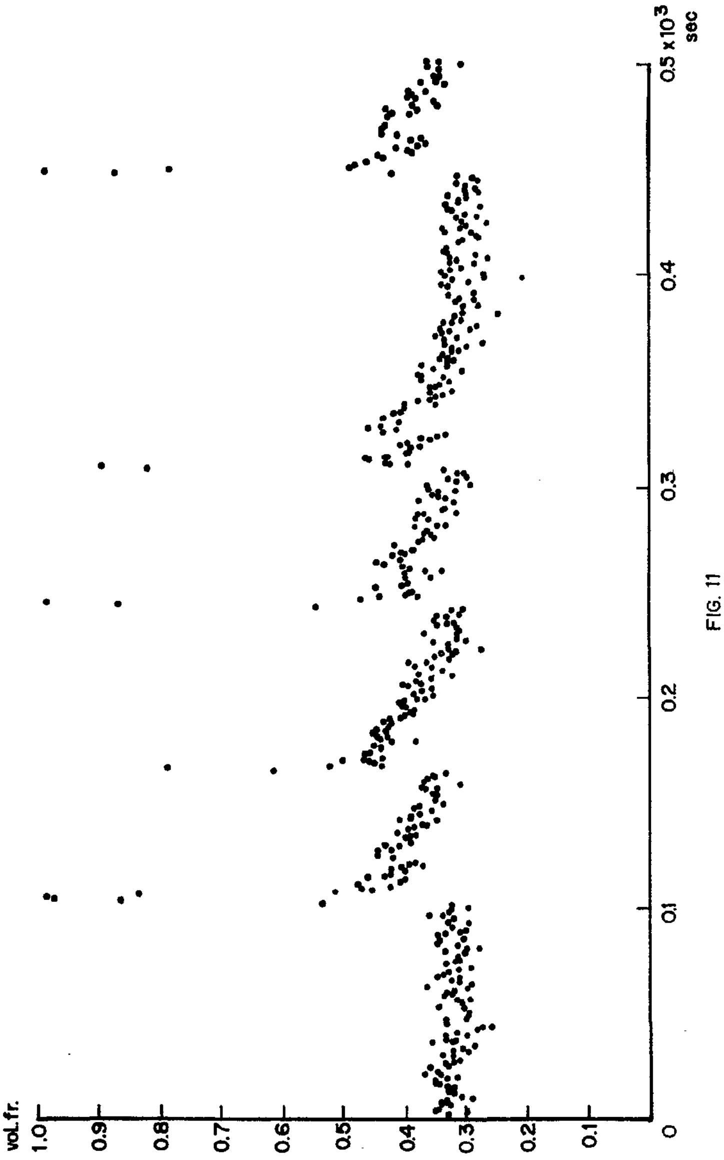

FIG. 11