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# Experience on the Removal of Impurities From Liquid Metal Systems By Cold-Trapping

by

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## SYNOPSIS

This paper reviews the experience of impurity removal by cold-trapping which has been obtained on DFR and its associated liquid metal rigs. It outlines the development of the present DFR cold-trapping system and describes the operation of the additional pumped loops which were required in order to control the reactor impurity levels. The operation of the liquid metal rigs ancillary to the reactor project is discussed with particular reference to the control of impurity levels.



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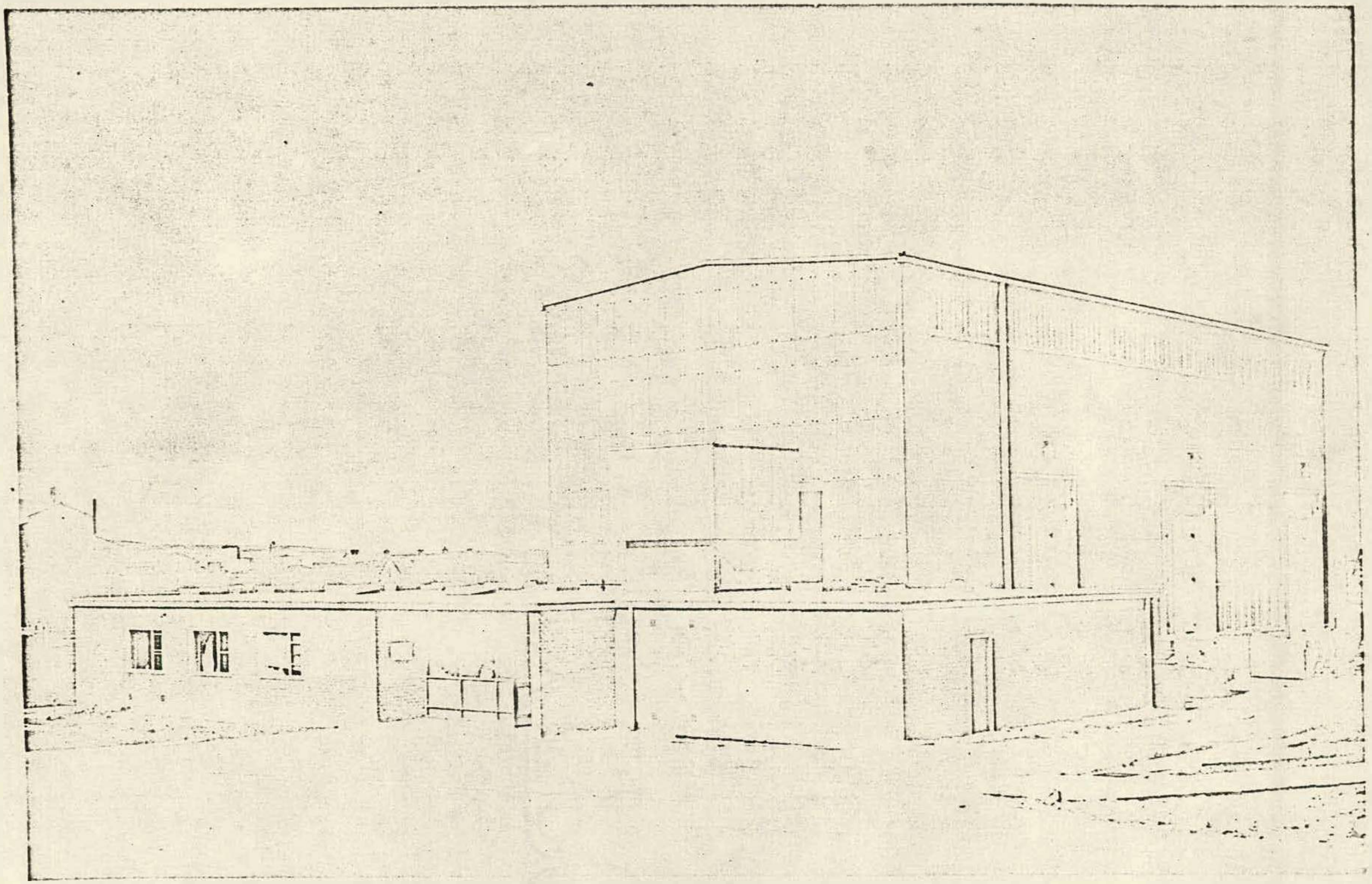

Frontispiece

Sodium Rig Building D.E.R.E.

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# INTRODUCTION

1. The levels of oxygen and hydrogen impurities in sodium and sodium/potassium alloy systems may be controlled by making use of their positive temperature coefficients of solubility (Fig. 1). If part of the circuit is maintained at a temperature lower than the saturation temperature of the liquid metal for the impurities present, the excess impurities will precipitate out and may be retained in some way. Impurity control by this means was originally demonstrated by Voorhees and Bruggeman in the Knolls Atomic Power Laboratories 1951 (1). This method is capable of reducing oxygen contamination to less than 5 ppm. If clean-up beyond this figure is required it becomes necessary to use chemical methods, e.g. reaction of oxygen with an insoluble getter such as zirconium.

2. Three methods of cold trapping may be used to effect clean-up of a liquid metal circuit. In the first one the cold trap has a portion of the system flow passing through it. The liquid metal is cooled as it flows through the trap, the excess impurities precipitate out and are retained by means of packing material, filters or other suitable surfaces in the trap. This method is most commonly used for the control of liquid metal systems in reactors and experimental rigs.

3. The second method relies upon the diffusion of the impurities to a cooled limb connected to the main liquid metal system. There is no forced flow in the trap container and precipitated impurity settles out as diffusion from the bulk liquid metal occurs. Diffusion cold-trapping is a slow process and is considered to be of only limited value in cleaning up a contaminated circuit. It could, however, have application in the continued control of circuits where relatively clean conditions have already been attained.

4. The third method is that of batch cold-trapping, where the liquid metal is heated to above the saturation temperature of the system and then dumped into a storage tank. The metal is then allowed to cool to a temperature which is just above its freezing point and maintained at this for several hours so as to precipitate the impurities. It is then recharged to the system through a filter. The process may be repeated until the impurity level has reached the required level and clean up to less than 5 ppm of oxygen has been obtained in this way. The method is suitable for initial clean up of circuits, for removing accidental gross oxygen contamination and for use on circuits where no installed cold trap is provided.

5. This paper is confined principally to discussion of experience on the through-flow cold trapping methods employed in liquid metal systems at D.E.R.E.

## SOME GENERAL PRINCIPLES AFFECTING THROUGH-FLOW COLD TRAPPING

6. Cold-trapping is essentially a crystallisation process, and although most of the data available on crystallisation was obtained in aqueous systems it appears reasonable to apply these as general principles to liquid metal work. The main problem in a cold trap is to ensure that the rate of deposition is high in regions designed to contain the deposit

and does not occur in regions where complete blockage could take place. The amount, distribution and rate of cooling applied to the cold trap and the residence time of the liquid metal within it, will largely determine the mechanism of crystallisation and the region where deposition will take place. The liquid metal flowrate is dependent on these factors but also influences the degree of agitation and turbidity which in turn also affect the crystallisation mechanism.

7. As the liquid metal is cooled below its saturation temperature, oxide nuclei are produced and growth continues in the stream. The crystals are retained in the filter medium and may in fact become part of it (2). Nucleus formation is induced by the presence of foreign bodies and by agitation. The rate of nucleation is affected by the extent to which super-saturation occurs i.e. the difference between the saturation temperature and the temperature at which precipitation, in fact, takes place. Where the degree of supersaturation is small, precipitation should result mainly in the growth of large crystals. Where the degree of supersaturation is large, numbers of fresh nuclei will be formed on precipitation resulting in the formation of numerous small crystals. The extent to which crystal growth occurs initially may thus affect the extent to which the cold trap packing material will act as a filter.

8. The situation in practice is complicated by the simultaneous presence of oxide, hydride and possibly other impurities with the result that individual precipitation mechanisms may be modified. In the presence of sodium monoxide and other impurities it is improbable that any degree of supersaturation exists. If this is so it would appear that when once some precipitate has been collected in the trap the process will be one of continued deposition and growth. The surface condition of the trap and packing material may also affect the process of crystallisation.

9. To summarise therefore the factors in cold trap design and operation which must be influenced by the mechanism and kinetics of the crystallisation process are:-

i) The range and rate of cooling,

ii) The residence time,

iii) The flow rate,

iv) The packing material density, surface area and condition,

v) The influence of the presence of one impurity on another.

10. A number of different designs of cold trap have been in use on the liquid metal circuits at D.E.R.E. which include the Dounreay Fast Reactor and the rigs associated with the project. It has been found possible to operate all of these cold traps successfully but a lack of fundamental data so far prevents any true optimization of cold trap design or operating parameters.

## RIG COLD TRAP EXPERIENCE

### General

11. Five dynamic liquid metal rigs fitted with cold traps have so far been commissioned at D.E.R.E. All the traps have been of the through flow type, but some experiments with diffusion type traps and with batch cold-trapping have been carried out. A number of types and designs have been used and their main features are summarised in Table I and illustrated in Figs. 2 to 7.

12. The cold-traps have been used for impurity level control i.e. they have not only been used to clean up the liquid metal to the lowest possible impurity level, but have been used to provide specific impurity levels for experimental purposes by acting both as traps and dispensers. The solubility data of Fig. 1 may be used to indicate the impurity levels in any liquid metal system by measuring the temperature of its cold trap and this method is used for adjusting the oxygen content of the rigs. The methods of verifying the actual impurity level as against the predicted level of impurity obtained from solubility data have included:

i) Direct sampling followed by analytical determinations for a particular impurity e.g. oxygen, carbon, hydrogen.

ii) Plugging meter observations, where dissolved impurities are precipitated out by cooling an orifice and noting the temperature at which flow reduction commences.

iii) Rhometer readings, where the electrical resistivity of the liquid metal is continuously measured and the impurity level of the system deduced from this.

13. It has been noted that although response to large changes in cold trap temperature is rapid, a period of fluctuation in impurity levels lasting as long as several days may be required before steady conditions prevail once more. For this reason changes in cold trap temperature are made very gradually and in normal practice are usually less than, and do not exceed 10°C per hour. Sudden increases in cold trap temperatures have led to wide fluctuations in the impurity level probably due to large particles being detached and flowing round the circuit. Sudden large decreases in temperature almost invariably lead to blockages in the cold trap lines and to fluctuations in impurity level.

14. Because of the scatter in analytical results and the delays associated with the sampling and analytical methods in use it is not possible to see these effects easily where this means of determination is the only one available. This is less true of the plugging meter technique which can be performed on a semi-continuous basis and from which results can be obtained in a very short time. Truly continuous monitoring of impurity is only possible at the moment using the rhometer.

15. The liquid metal rigs fitted with cold traps for clean-up purposes at D.E.R.E. include those listed below.

### ¼" Loop 'F'

16. This rig was designed for instrument development and testing at temperatures of up to 600°C. It is built of ¼" nominal bore stainless steel pipework and contains approximately 0.3 cubic feet (10 litres) of sodium. Impurity control is by a cold trap but several hot traps have been fitted to the rig in the course of experimental work. The cold trap is a simple air-jacketed cylindrical vessel (Fig. 2) packed with stainless steel rings and fitted into a by-pass across the electro magnetic pump. The performance of the trap has proved quite satisfactory but it tends to block easily and better control could be achieved if it were possible to obtain a more accurate packing temperature measurement.

### ½" Loop 'J'

17. This rig is a development of the small loops which carried out the early British liquid metal studies at Harwell and later at Culcheth. The pipework is of ½" nominal bore stainless steel and the liquid metal capacity is approximately 0.5 cu. ft. (15 litres) and this loop is filled with 70/30 NaK. It is used mainly for corrosion studies on reactor materials at temperatures up to 650°C and impurity control is by a cold trap and a zirconium filled hot trap. The cold trap is a unpacked oil cooled vessel (Fig. 3) and although the capacity is limited it has proved to be suitable for accurately controlling oxygen levels to within ± 2 ppm. Trapping probably occurs in and below the sintered plate and in the gap between inner and outer walls.

### Components Test Rig 'B'

18. This rig is larger in size and is constructed of ¾" nominal bore stainless steel tubing. Its capacity is approximately 4 cubic feet (120 litres) of 70/30 NaK and temperatures up to 600°C can be obtained in its three test zones. Two of these are simple vessels which have accommodation for small specimens and also provide for the removal of liquid metal samples. The remaining facility can house up to three complete DFR fuel elements and subject them to flow and temperature conditions of the same order as those in the DFR core. A plugging meter and rhometer are installed on this rig and impurity control is provided by an oil cooled cold trap. The trap (Fig. 4) is of large capacity and internally cooled with a silicone oil cooling coil passing through the stainless steel packing rings. In service it has performed well and has shown no tendency to block. Control of plugging temperatures by changing cold trap temperatures has been possible to within ±5°C but temperature measurement could be improved by repositioning the thermocouples to read more accurately the packing material temperature.

### 1" Loop 'A'

19. This rig is of 1" nominal bore stainless steel pipework designed to test and commission the liquid metal sampling gear for DFR. The loop contains approximately 7 cubic feet (200 litres) of 70/30 NaK and impurity control is achieved by using an air-cooled cold trap and a zirconium filled hot trap. A rhometer is fitted and the maximum operating temperature is 220°C. The trap (Fig. 5) is of large capacity

and fitted with a simple regenerative heat exchanger to economise in total heat requirements.

20. During experiments on the measurement of impurity levels using the rhometer the rig was operated at 220°C and the cold trap temperature varied from 110°C to 220°C. No increase in resistivity took place above a cold trap temperature of 180°C (Fig. 6) and investigation showed that parts of the rig were forming 'dead legs' through which there was no flow and whose temperature was lower than the main rig temperature by approximately 50°C. These 'dead legs' appeared to act as efficient diffusion cold traps and cleaned up the rig to a plugging temperature of 180°C. After several months running the 'dead legs' became reservoirs of impurities and dispensed them into the system whenever the cold trap temperature was reduced below 180°C rendering impurity level control a difficult and slow process despite the ample capacity of the cold trap.

Twin Zone Loop 'K'

21. This rig is similar in concept to the 'J' rig already described but is fitted with two specimen chambers which enable corrosion tests to be carried out in the same liquid metal at different temperatures. The test zone temperatures can reach 650°C and 750°C respectively and the loop has a capacity of approximately 1.2 cu. ft. (40 litres) of sodium. A zirconium filled hot trap and a wire mesh packed cold trap with oil cooling (Fig. 7) provide impurity control. The trap is an improved version of the 'J' design and has proved to be satisfactory in operation.

4" Loop 'G'

22. This rig is similar in size and layout to one of the 24 DFR primary circuits. It is intended for fuel element and reactor component testing. The liquid metal capacity is approximately 62 cubic feet (1800 litres) and the impurity control is by a cold trap similar in design to the DFR type but fitted with internal oil cooling in the basket to supplement the external air cooling (Fig. 8). The rig is at present under construction.

## D.F.R. COLD TRAP EXPERIENCE

Original Cold Traps

23. The original design of the DFR primary circuit had cold traps fitted in fifteen of the twenty four primary circuits (3). The pressure drop due to friction along a section of the main circuit was used to create a flow in a by-pass (Fig. 9) and at full primary pump flow the by-pass flow was about 5 gpm.

24. The arrangement of the cold trap is shown at Fig. 10. Cooling was provided by blowing air over a finned exterior and an internal regenerative heat exchanger was incorporated to minimise heat loss. The basket was packed with stainless steel rings and the flow through it was moderated by an internal by-pass which left a flow through the basket of approximately 2 gpm. The height of the trap in the system

was arranged to allow a free NaK surface to the blanket gas. This allowed the basket to be raised above the liquid metal so that hot trapping operations would not merely cause migration of oxide from cold traps to hot traps. It also permitted complete removal of the baskets from traps, which are of course situated within the biological shield, by a flasking technique.

25. After liquid metal was charged into the primary circuit in 1959 it became clear that these traps were inadequate for the conditions under which they were required to operate at that time (4). Many of the by-pass circuits were found to be blocked and those which were open tended to block when the trap cooling system was brought into operation due to the high impurity levels which caused precipitation of oxide and consequent blockage in the regenerative heat exchanger and the annulus adjacent to the cooled wall.

Temporary Cold-Trap Loop

26. The by-pass cold traps can only have flow through them when the main flow is greater than 50%, as below this the liquid metal level does reach the outlet pipe. In the early stages of commissioning it was not possible to operate the reactor at anywhere approaching the full design flow rate because of gas entrainment difficulties (5,6). The electromagnetic pumps cannot be run at above 10% flow at any time when the reactor is de-pressurised for fuel changing or maintenance, and thus operation of the by-pass traps is impossible during these periods, when the possibility of oxygen contamination is most likely to occur. A simple external pumped cold trap loop was therefore designed and installed temporarily on the reactor top. Dip legs fitted into one of the original by-pass cold-trap vessels and an expansion tank in adjacent circuits provided flow and return points. The loop consisted of two cold traps of similar layout to the existing ones, an electromagnetic pump and plugging meters which could be used to monitor either the inlet or outlet side of either trap. Shielding was not provided and neither was double containment of the pipework and vessels and this meant that the loop was only suitable for operation at shutdown conditions.

27. The trap baskets were similar to the previous ones but there was no internal by-pass i.e. the whole of the trap flow passed through the basket. With a inlet temperature of 160°C and the design flow of 2 gpm, full cooling reduced the basket temperatures to 120°C. Under these conditions the traps blocked after two or three days operation. It was found possible to clear the blockages by isolating the loop from the reactor and re-circulating the NaK after heating. Re-trapping was then carried out until the plugging temperature fell as the impurities were redistributed through the system. This procedure could be repeated until the maximum loop temperature (approx. 300°C) was approached by the plugging temperature of the recirculating coolant.

28. The blockages were found to be occurring in the annulus between the trap body and the basket and in the regenerative heat exchanger. In order to minimise deposition in the annulus the trap cooling was controlled to give a basket temperature of only 10°C below the inlet plugging temperature. At that time the reactor plugging

temperature was about 130°C when the coolant was at 150°C but rose to 180°C when the reactor temperature rose to 220°C. This behaviour was similar to that already discussed for the 1" loop 'A' (para 20) and it appeared that there were regions in the circuit where oxide was trapped and being dispensed out to control the plugging temperature. Continuous cold-trapping operations over several months reduced the plugging temperature to 100-110°C but none of the blocked by-pass circuits cleared. Mechanical means together with a series of batch cold-trapping operations were required before all the by-pass flows were established. The loop was dismantled in 1962 when it was replaced with a permanent facility.

Permanent Cold Trap Loop

29. The experience gained on the temporary loop confirmed the advisability to fit a permanent separate pumped cold-trap circuit on the reactor which could operate independently of main circuit flows. This facility was designed and manufactured to the same rigorous specification laid down for the reactor primary circuits. It consists of two cold-traps, a separate regenerative heat exchanger, an electromagnetic pump, a plugging meter and a rhometer. All the vessels and pipework are fully leak-jacketed (the valves are double bellows sealed and leak-jacketed). The whole unit is housed in a lead shielded cubicle with a steel lined base which can act as a liquid metal containment of 1000 gallon capacity in the unlikely event of a failure of both a vessel or pipe and its leak jacket. The flow and return lines are fitted to adjoining installed cold trap vessels in the primary circuit. A simplified flow sheet of the loop is given in Fig. 11 and a general view of the loop is in Fig. 12.

30. The cold trap bodies used in this loop are again virtually identical to the original D.F.R. design and have external air cooling. The basket interiors (see Fig. 13) differ from previous designs in the following ways:

i) the annulus between basket and body has been increased from 0.25" to 2.70" in order to reduce blocking in this region.

ii) stainless steel knitmesh is used instead of Raschig rings to improve filter efficiency.

iii) the internal regenerator is omitted as the loop is fitted with a separate regenerative heat exchanger.

In addition, the internal by-pass is omitted as the circuit, like the temporary loop, has its own positive pumping system.

31. During the early stages of its operation in mid 1962 blockages occurred in the trap outlet pipes where basket cooling air was leaking on to them. Remedial work to correct this was successful but other leaks in the cooling ducts have prevented the full design figures being reached. Basket temperatures of 135°C at 1 gpm are however obtainable from an inlet temperature of 204°C. During high power operation of the reactor the radiation levels are too high to permit continuous manning of the loop and this has not permitted the

fullest use to be made of the facility. Improvements to the shielding are in hand and these should enhance the value of this facility in the future.

Modified Main Circuit Traps

32. The cold trap capacity in the reactor has been supplemented by the recommissioning of four of the main circuit cold traps. The air cooling to these four trap bodies has been improved by additional fans. The basket design is similar to that used on the cold trap loop but the following additional features are incorporated (Fig.14).

i) Provision has been made for the basket to be lifted above NaK level during shut down and a 1½ Kw immersion heater has been provided at the trap inlet. These measures ensure that any tendency of the inlet pipe to block during shut down is minimised.

ii) The internal by-pass has been reintroduced as the air cooling could not be increased sufficiently to deal with full by-pass flow (full flow must exist in the by-pass circuit to avoid lowering the level in the expansion tank to a point at which vortex formation and consequent gas entrainment might occur).

33. The performance of these traps cannot be checked directly because the trap outlet plugging temperature and the flow through the basket cannot be measured. However temperature measurements and heat balances indicate the following:

| | |
|---|---|
| Total by-pass circuit flow | 4.36 gpm |
| Flow through basket | 0.7 - 1 gpm |
| Inlet temperature | 204°C |
| Basket temperature | 141°C |
| Outlet temperature | 199°C |

Modifications to improve the air cooling system still further are in hand and it is hoped to reach a minimum basket temperature of 115°C in this way.

34. During the most recent power run these traps together with the Permanent Cold Trap Loop operated and the plugging temperature dropped from 135°C to 110°C over one week and then remained steady. When the traps were shut down the plugging temperature rose again to 135°C and this tends to confirm that there is still a source of undissolved impurity present in the circuit.

35. The first of the redesigned main circuit baskets to be installed indicated that due to the balance of pressure drops between the basket and the internal by-pass it was possible to have a condition of reverse flow in the basket. This fault has been overcome by fitting a restriction in the basket but it was thought desirable to have a full flow basket which could not possibly recirculate. This design is shown at Fig. 15. In order to make maximum use of the air cooling an economiser has been fitted and a 5 Kw heater is situated at the outlet to eliminate

any possibility of blockage. The $1\frac{1}{2}$ Kw inlet heater has been omitted as it has been found to be unnecessary. Further development of the main circuit traps may include internal cooling as in the experimental trap in the 4" loop (para. 21) possibly using NaK instead of silicone oil as the cooling medium.

## ACKNOWLEDGEMENTS

36. The author is indebted to many members of the Fast Reactor Project for their assistance, but would like particularly to acknowledge the help of Dr. R. A. Davies, Mr. J. Gray, Mr. R. E. Godfrey and Mr. L. C. James.

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## TABLE I

### D.E.R.E. RIG COLD TRAP DATA

| RIG | | | COLD TRAP | | | | | |
|---|---|---|---|---|---|---|---|---|
| Title | Metal | Volume ($ft^3$) | Flowrate g.p.m. | Residence Time (Mins) | Packing | Method of Cooling | Method of Checking | Fig. No. |
| ¼" Loop 'F' | Na | 0.3 | 0.1 | 2.5 | Lessing Rings | External Air | Sampling | 2 |
| ½" Loop 'J' | 70/30 NaK | 0.5 | 0.1 | 1.3 | Nil (Sintered S.S. Filter plate) | External Oil | Sampling | 3 |
| Components Test Rig 'B' | 70/30 NaK | 4.0 | 1.0 | 9.0 | Raschig Rings | Internal Oil | Sampling Plugging Meter Rhometer | 4 |
| 1" Loop 'A' | 70/30 NaK | 7.0 | 1.0 | 32.0 | Lessing Rings | External Air | Rhometer Sampling | 5 |
| Twin Zone Loop 'K' | Na | 1.2 | 0.1 | 1.0 | Woven Wire Mesh | External Oil | Sampling Plugging Meter | 7 |
| 4" Loop 'G' | 70/30 NaK | 62.0 | 1.5 | 5.0 | Woven Wire Mesh | Internal Oil and External Air | Sampling Plugging Meter | 8 |

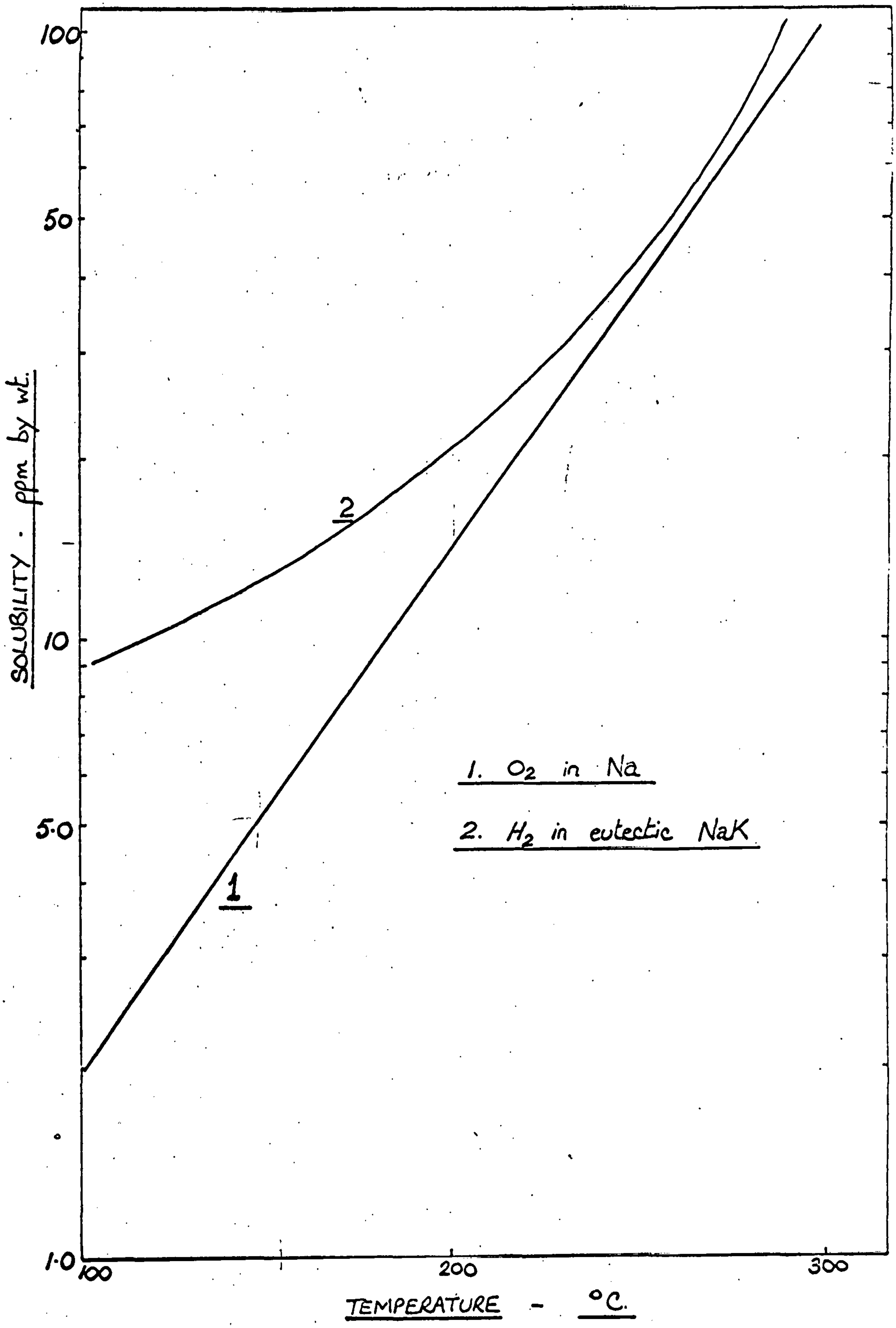



Fig. 1 Solubility Data for $O_2$ and $H_2$ in Na(K)

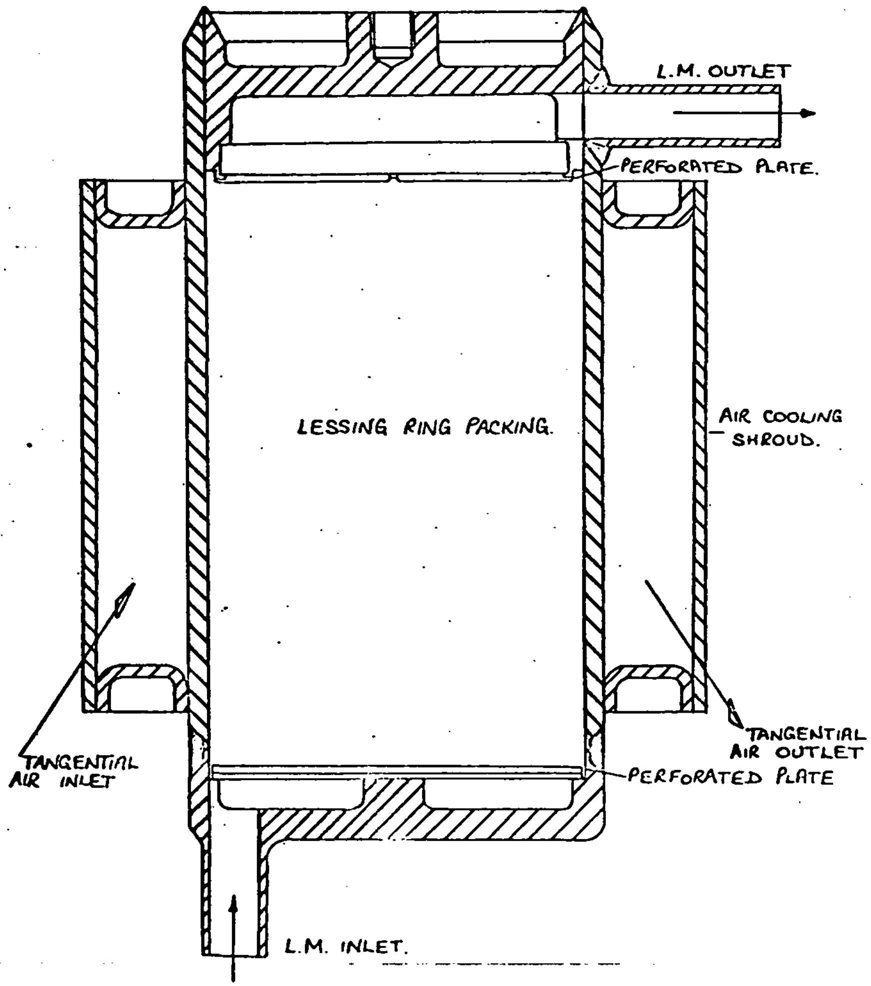



Fig. 2 Cold Trap Layout 1/4" Loop 'F'

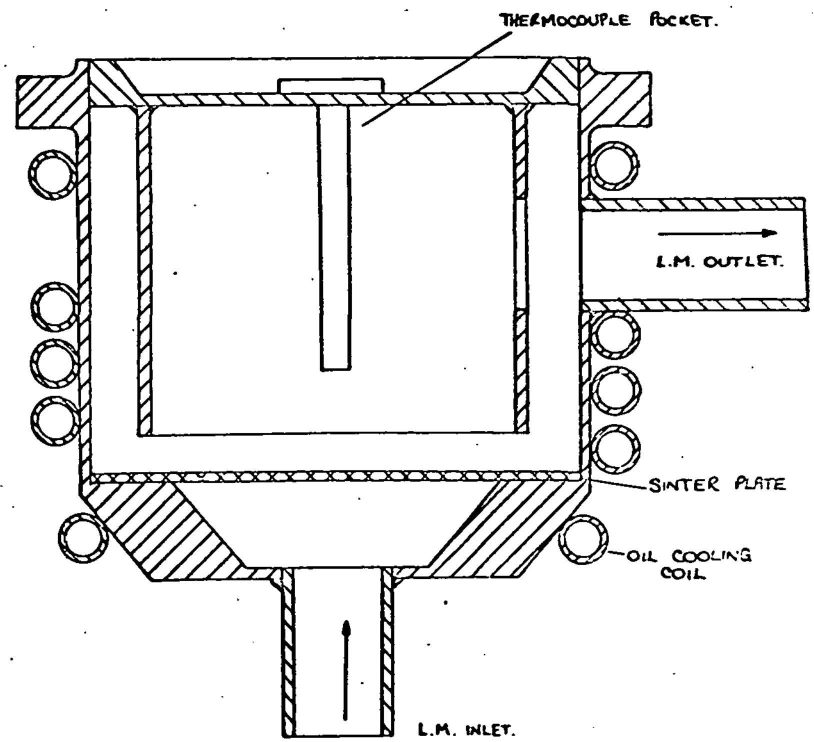



Fig. 3 Cold Trap Layout 1/2" Loop 'J'

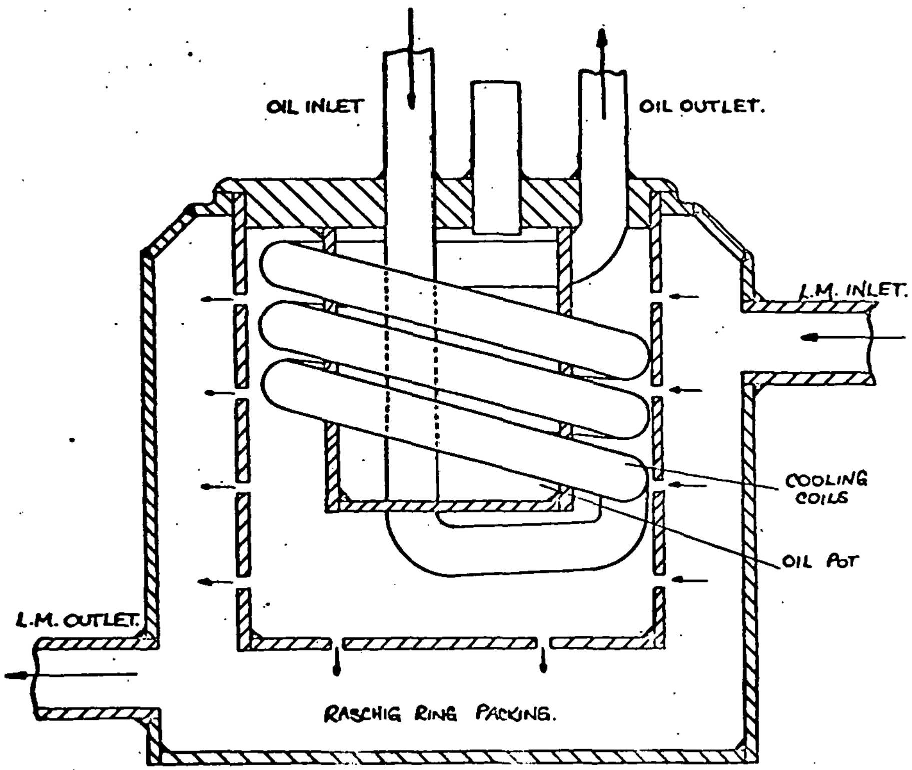



**Fig. 4** Cold Trap Layout Components Test Rig 'B'

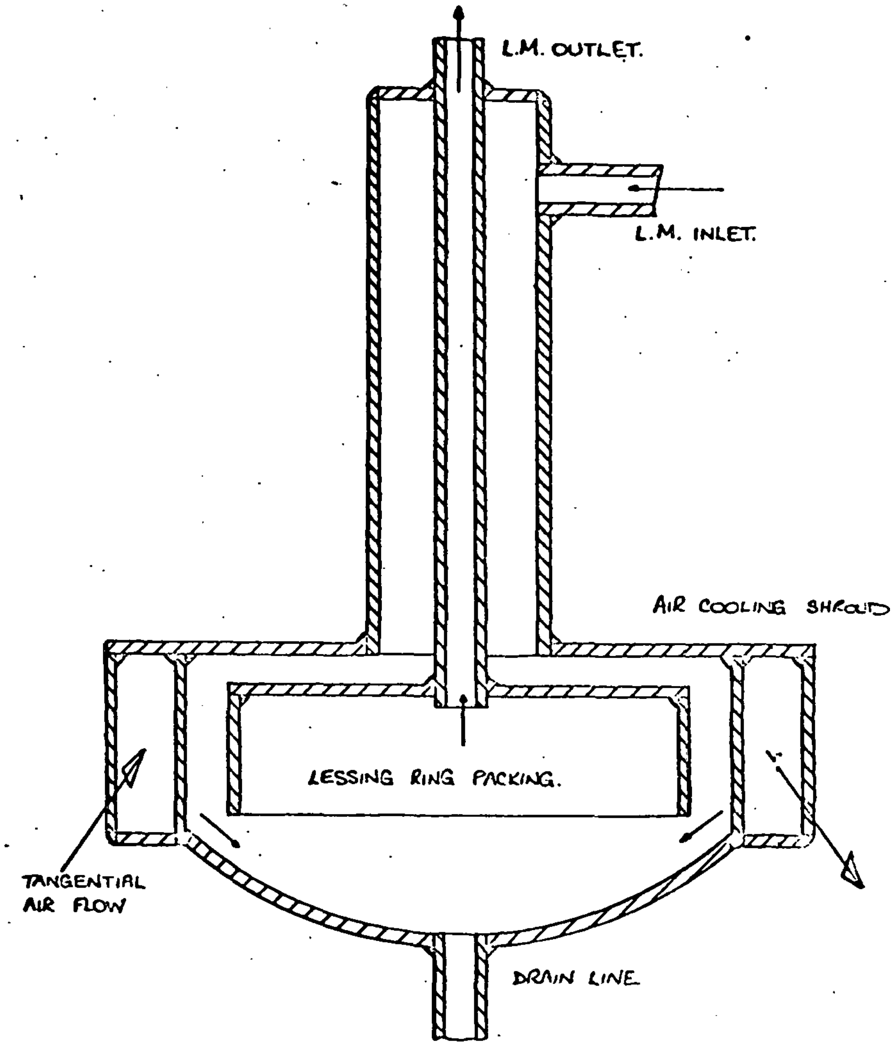



**Fig. 5** Cold Trap Layout 1" Loop 'A'

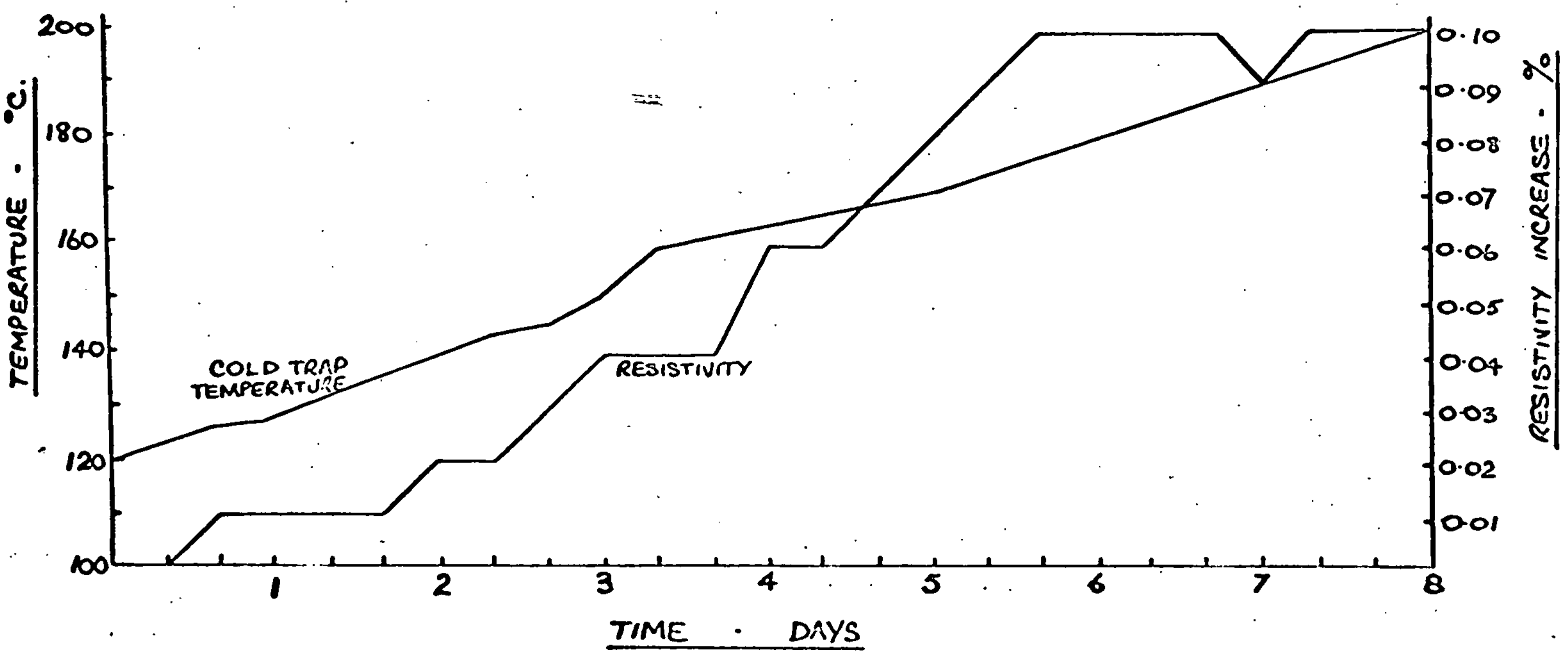
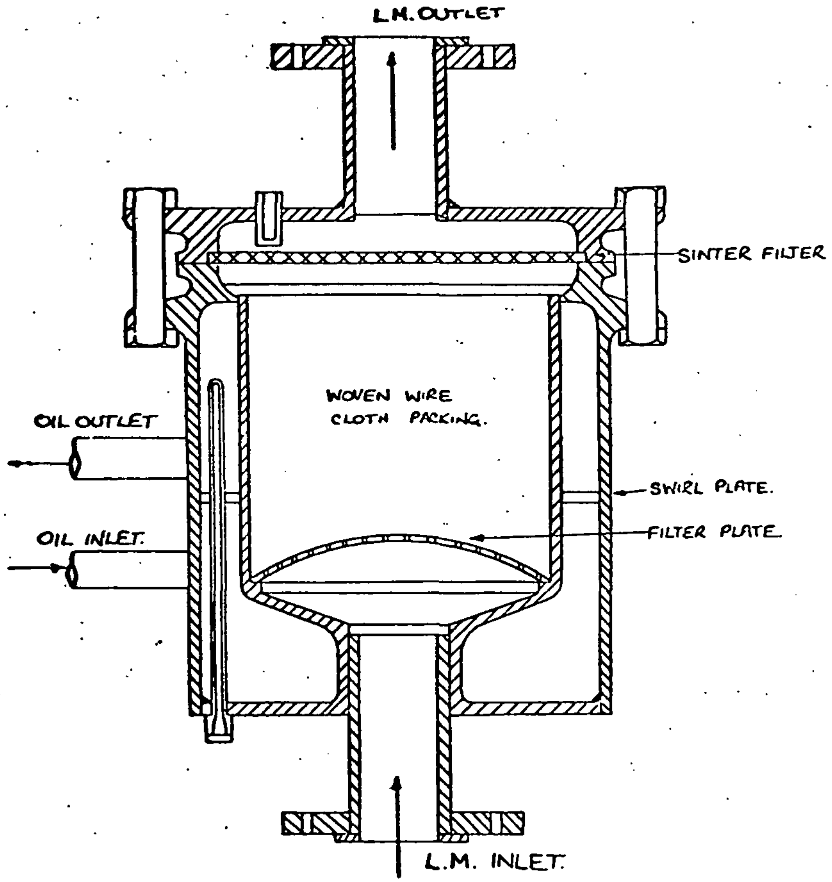



Fig 6. Comparison of Cold Trap Temperatures with Rhometer Reading (1" Loop "A")



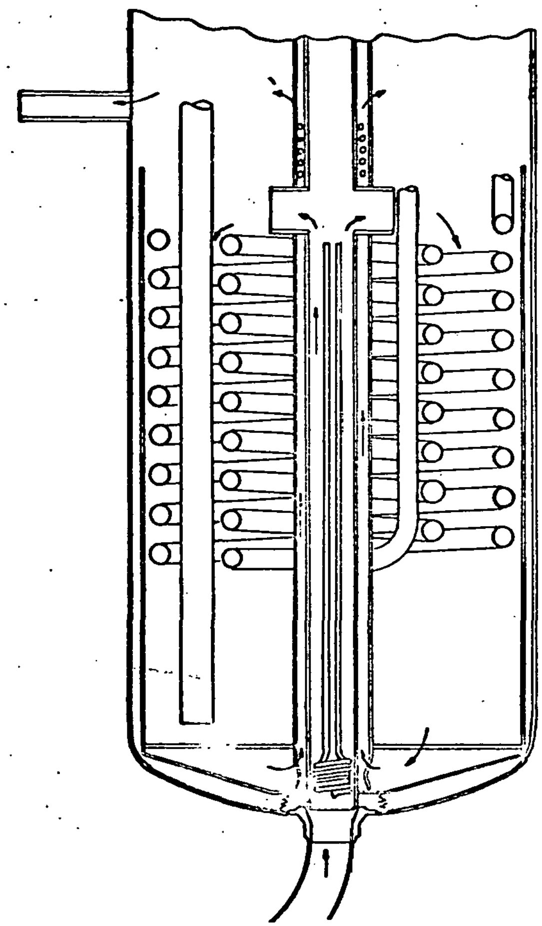



Fig. 7 Cold Trap Layout, Twin Zone Loop 'K'

Fig. 8. 4" Loop Cold Trap

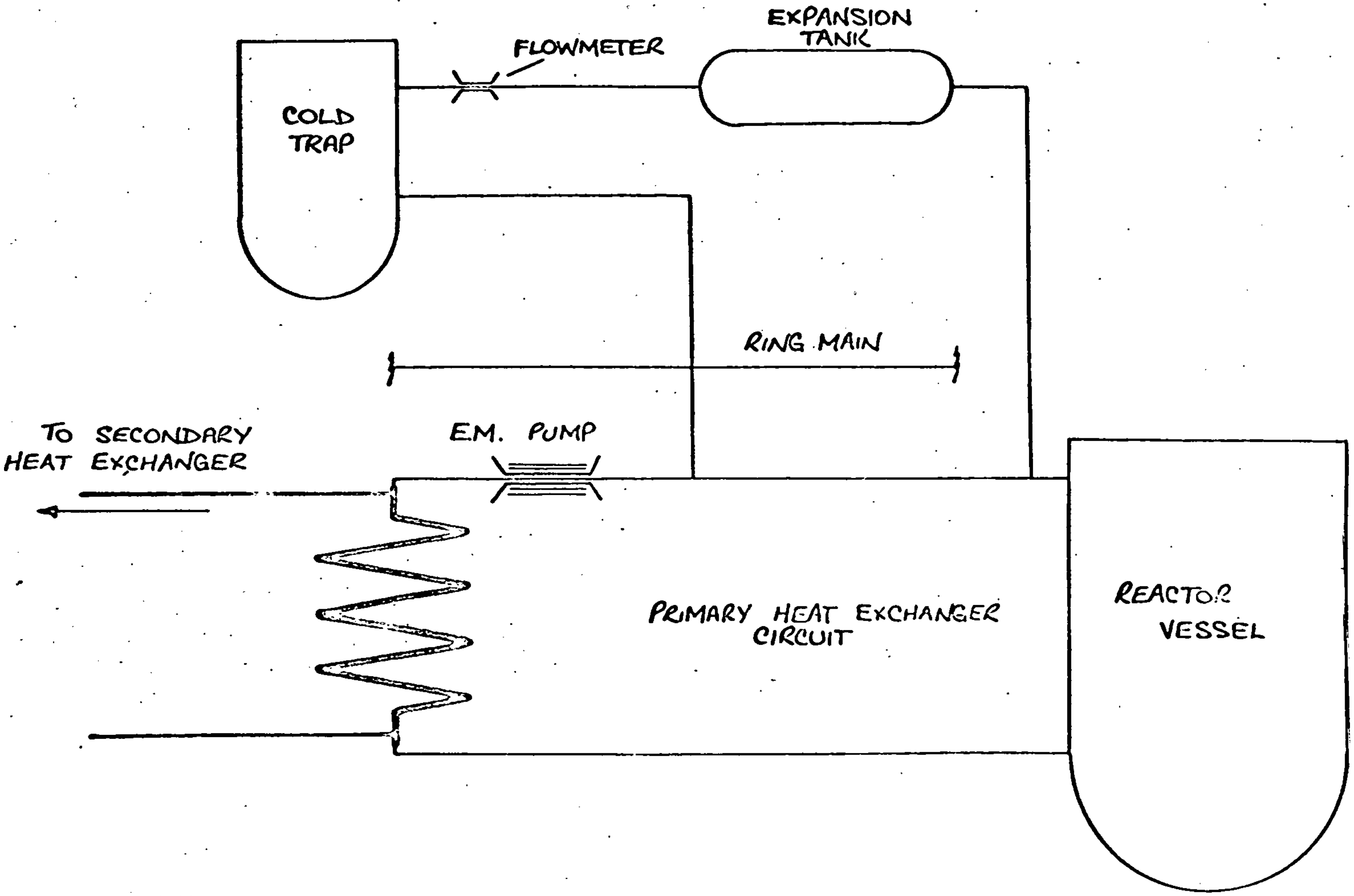



Fig. 9 Schematic Diagram of D.F.R. Coolant By-pass Circuit

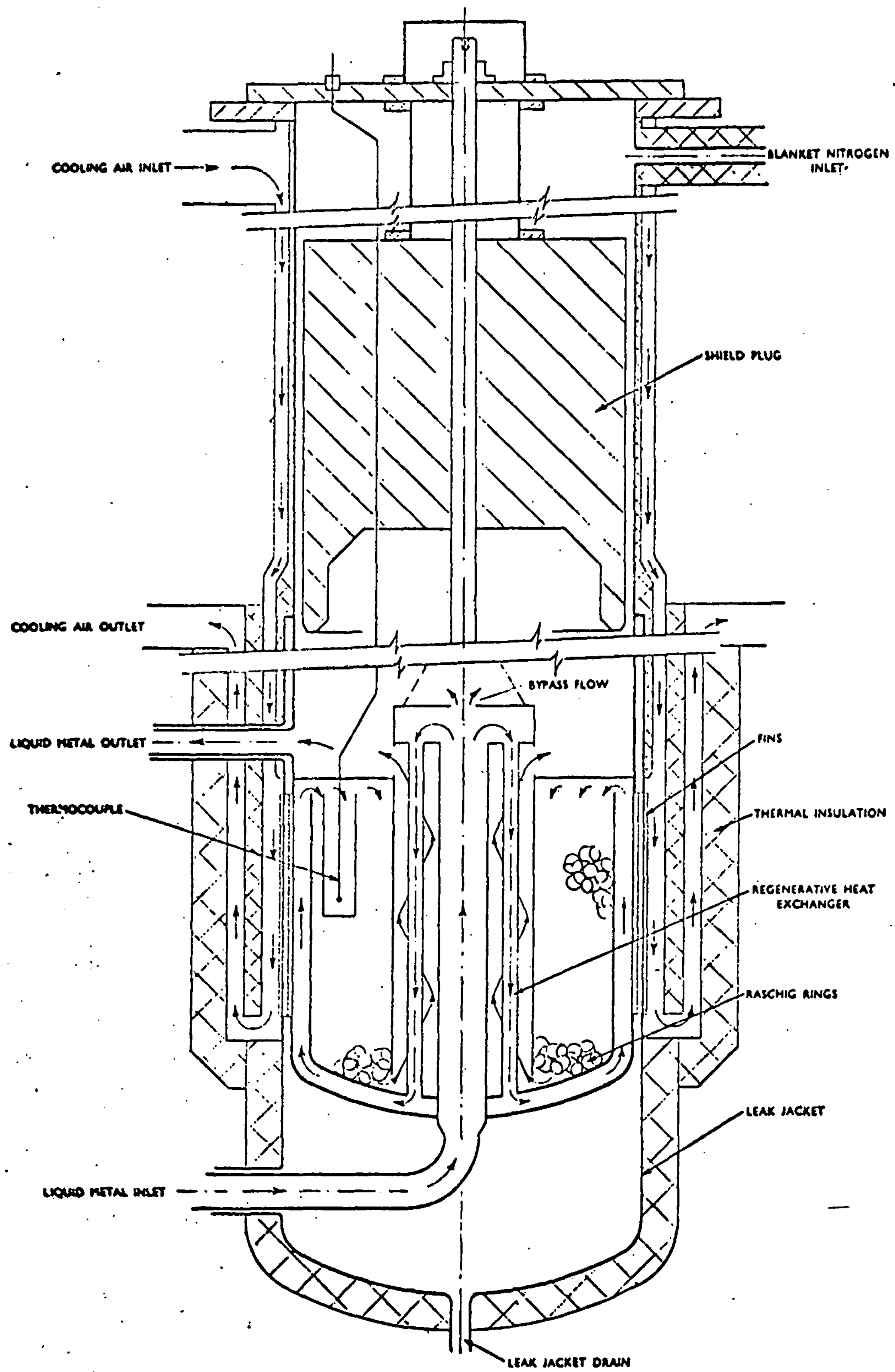



Fig. 10 D.F.R. Primary Cold Trap Arrangement

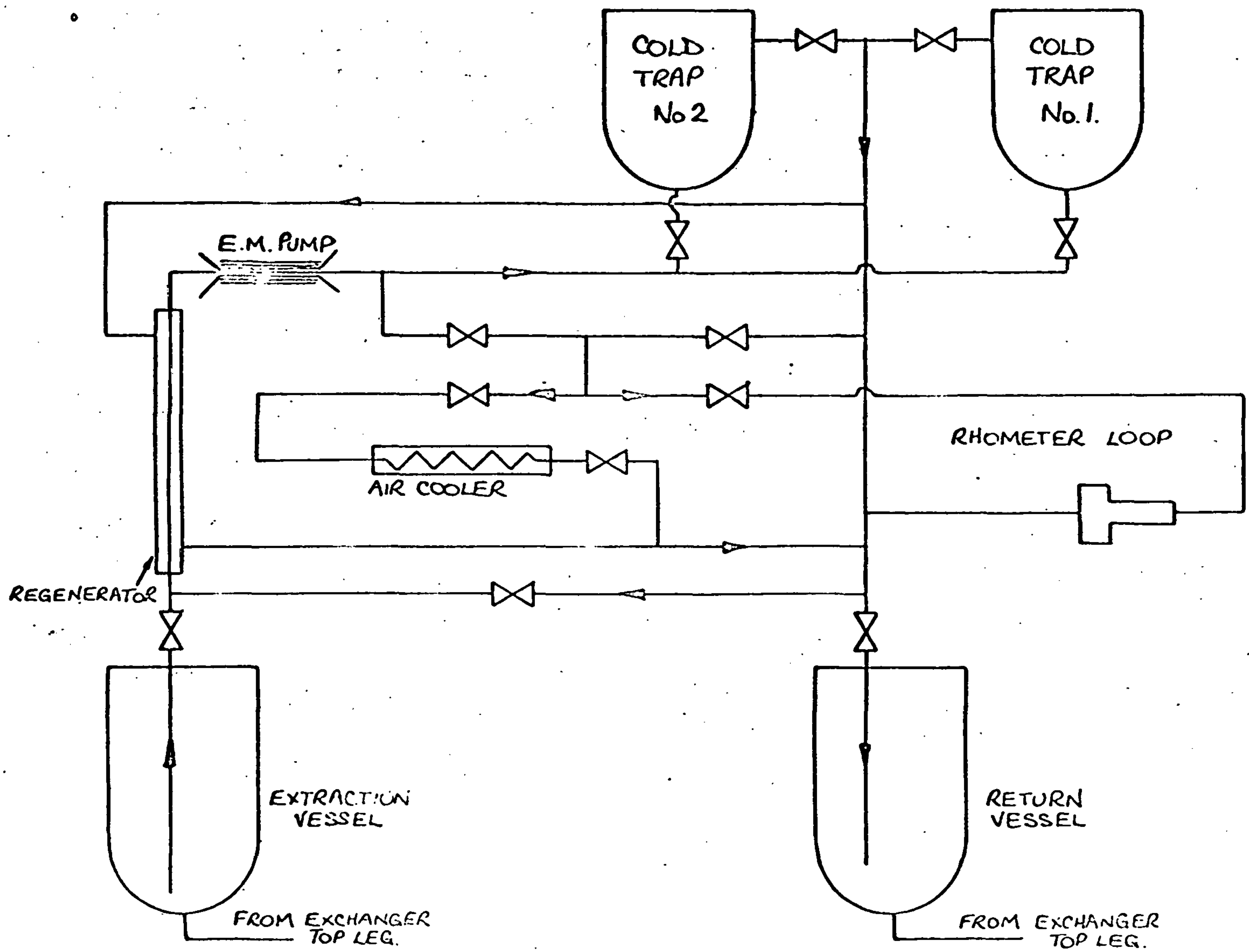



Fig. 11 D.F.R. Permanent Cold Trap Loop NaK Circuit

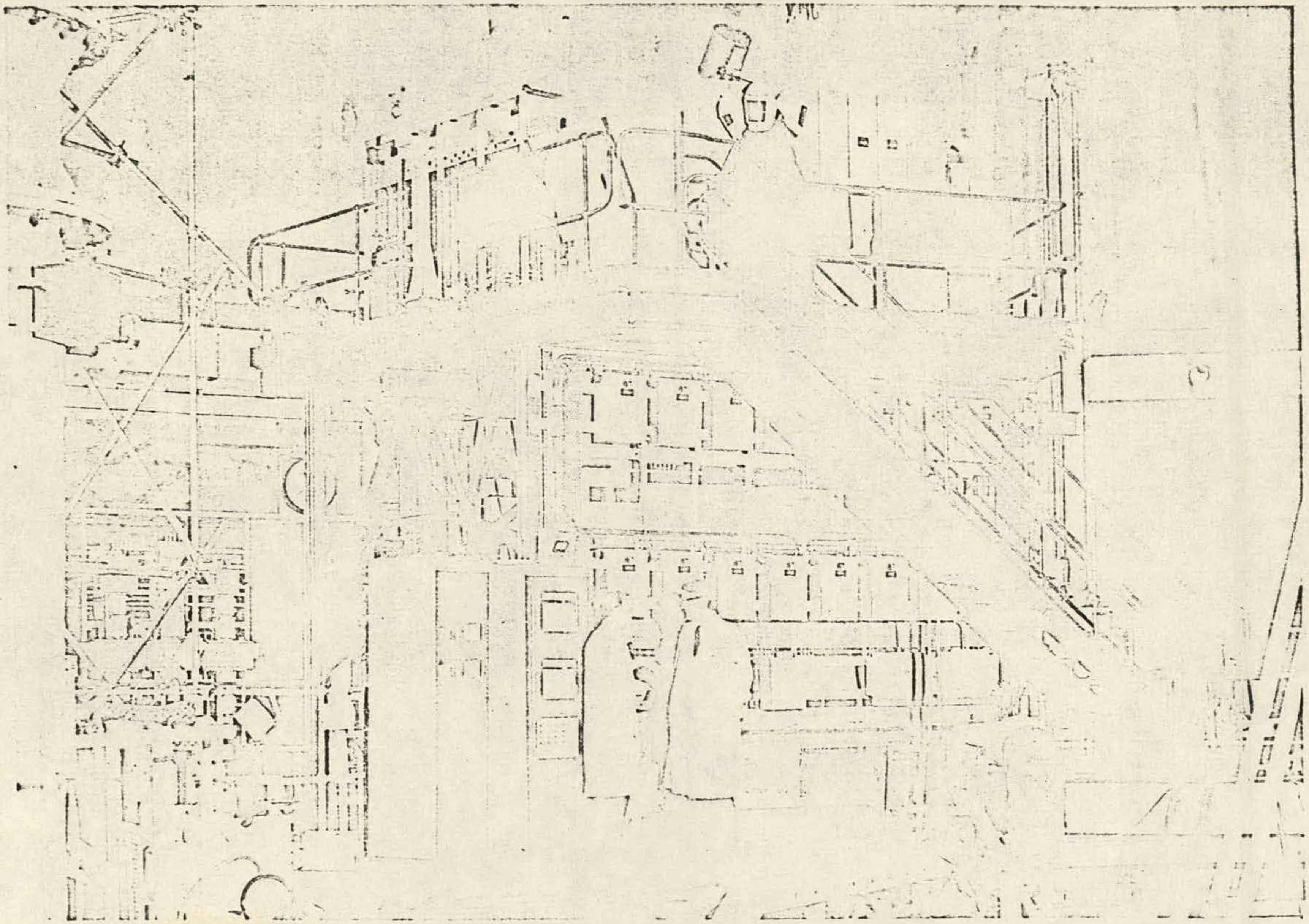


Fig. 12 D.F.R. Perma[illegible] Cold Trap Loop - General View

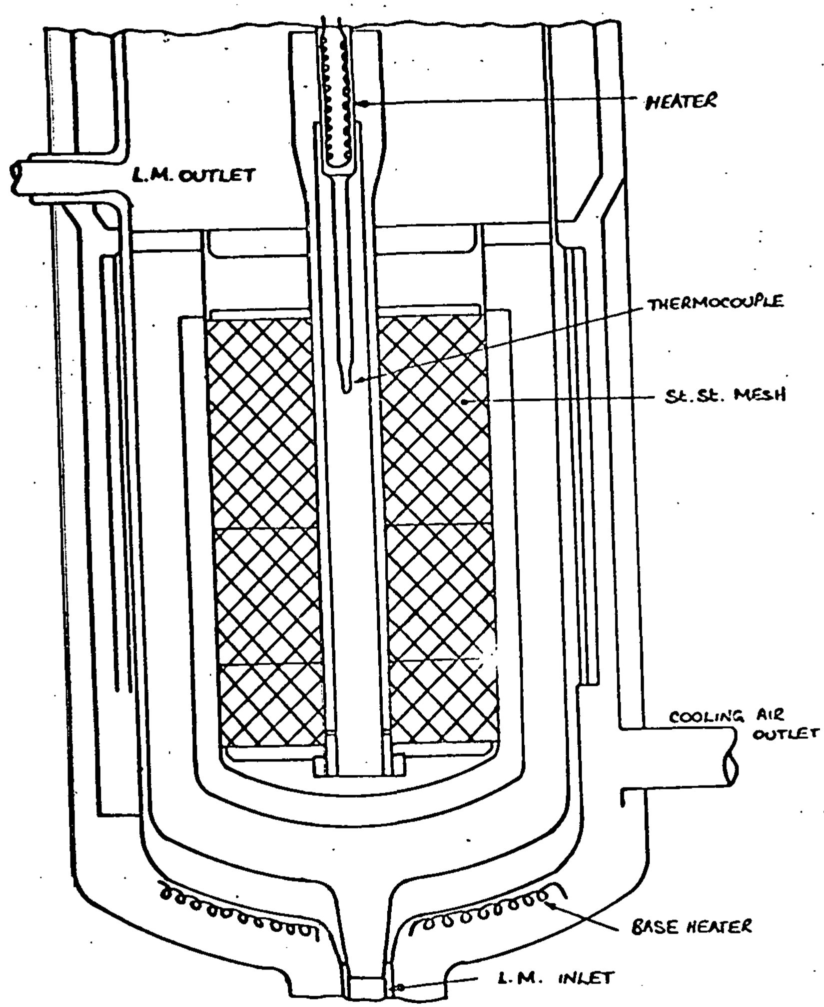



Fig. 13 D.F.R. Permanent Cold Trap Loop - Cold Trap

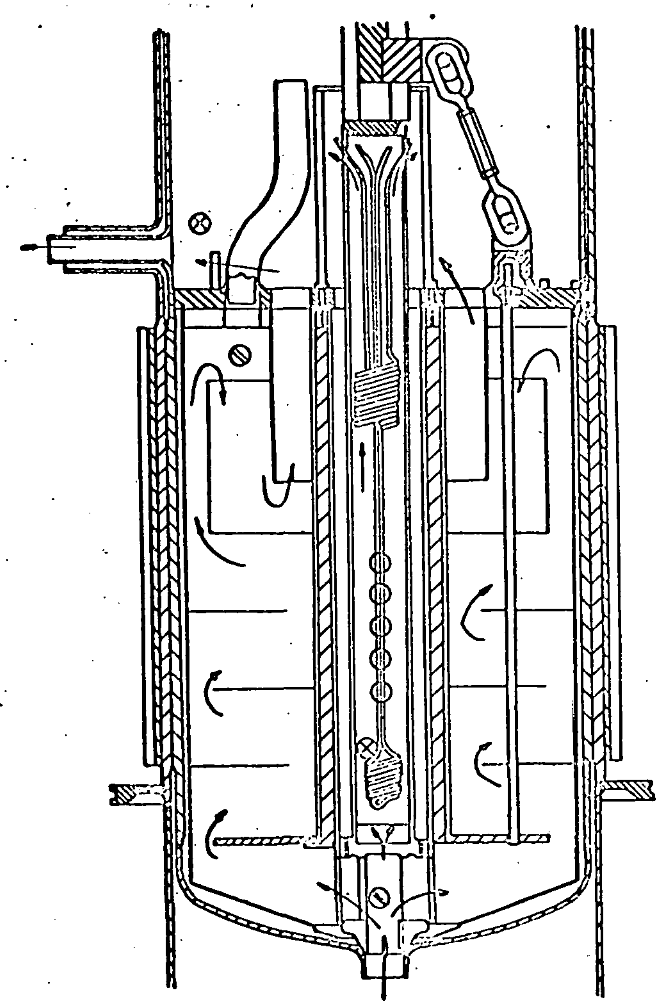


Fig. 14 D.F.R. Main Circuit Cold Trap (Modified Basket)

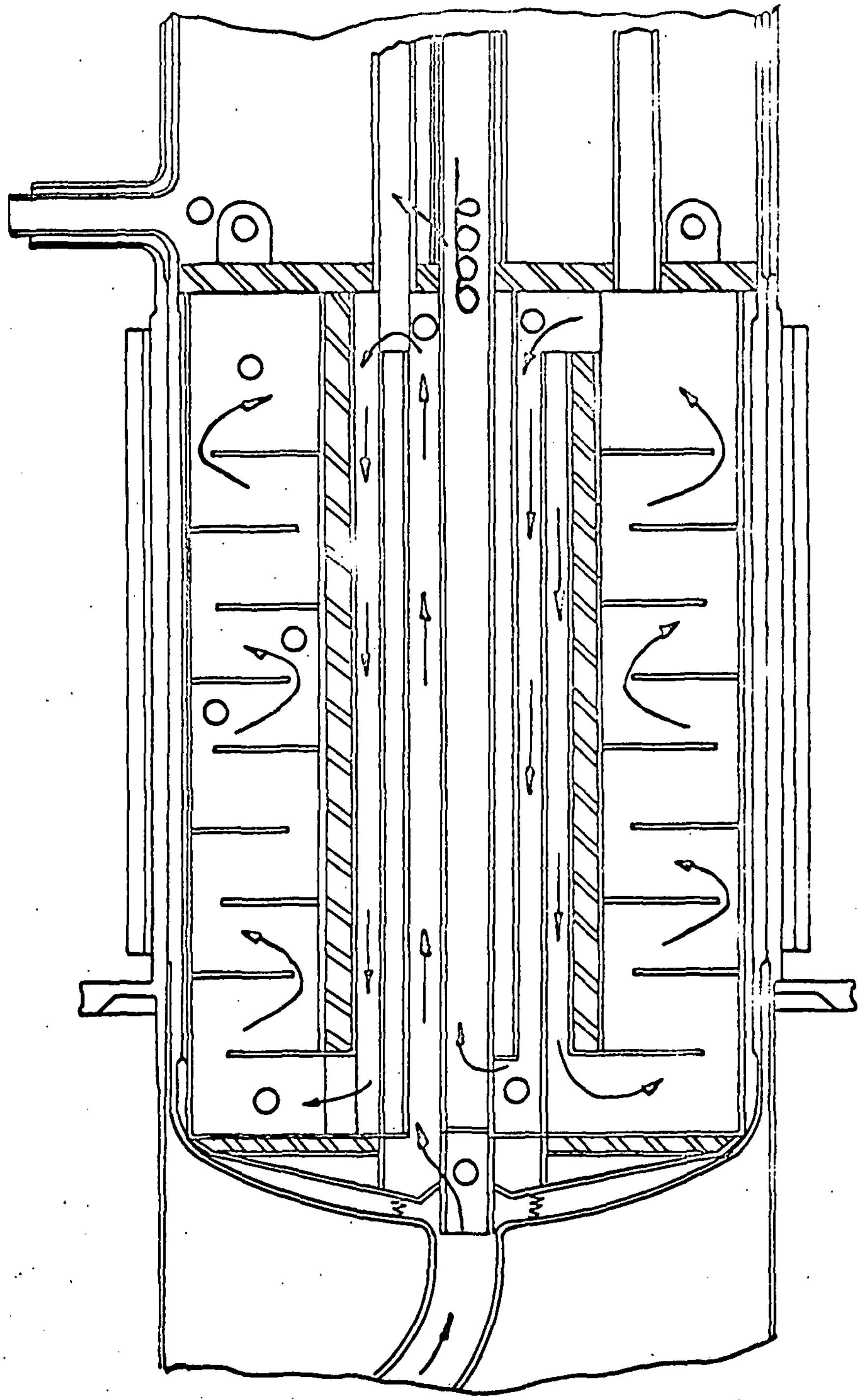


Fig. 15 D.F.R. Main Circuit Through-Flow Cold Trap