

TRACE ANALYSIS IN THE SODIUM COOLANT OF FAST BREEDER REACTORS

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Abstract - Liquid sodium is the preferred coolant in fast breeder reactors. While sodium itself is compatible with the structural materials in the coolant circuits, presence of even trace levels of impurities such as oxygen, carbon and hydrogen can lead to corrosion and mass transfer. In addition, metallic impurities, activation products and fission products have to be monitored in the coolant circuit to keep track of corrosion, mass transfer, activity transport, incursion of impurities and fuel failure. This paper summarises the development of trace analysis techniques as applied to sodium in support of India's fast reactor programme. The techniques for handling sodium are discussed and laboratory techniques for analysing trace level metallic and non-metallic impurities in sodium are described. On-line meters being developed for monitoring oxygen, carbon and hydrogen are mentioned. The main features of the analytical sodium loop that is being set up for meter calibration and intercomparison of results from different techniques are brought out. The methods proposed to be used for the determination of radioactive impurities in the primary sodium in the reactor are indicated.

INTRODUCTION

Fast Breeder Reactors are designed with high specific power and hence require a very efficient heat transfer fluid as coolant. Sodium meets this requirement satisfactorily. Sodium is a liquid having low vapour pressure at the operating temperature of fast reactors and its high thermal conductivity, high specific heat and good flow characteristics make it an ideal heat transfer fluid. Another desirable property is its good compatibility with structural materials in the coolant loops. A fast reactor has two kinds of coolant loops: the highly radioactive primary sodium loop which carries heat from the reactor core to the intermediate heat exchanger and the secondary sodium loop which transfers this heat further on to the steam generator. The structural materials in the primary loops are austenitic stainless steel of the type 316 and 304, while the secondary loops contain in addition ferritic steels such as 2½ Cr - 1 Mo used in the steam generator. While sodium itself is quite compatible with these steels, presence of impurities such as oxygen, hydrogen, carbon, nitrogen etc., even in parts per million (ppm) levels, can lead to corrosion and mass transfer. Oxygen in sodium causes general corrosion and contributes to mass transfer from high temperature regions in sodium loops. Carbon in sodium is associated with the carburization and decarburization in the coolant circuits. Presence of hydrogen can lead to hydriding and that of hydroxide accelerates the corrosion process. Hence there is need to control and monitor these trace level impurities. Further the periodic measurement of the corrosion products is also necessary to monitor the corrosion process. Some of the corrosion products get activated leading to the transport of long-lived radioactive isotopes which can cause problems in maintenance. Radioactive isotopes are also injected into the primary coolant as a result of fuel failure. Measurement of these fission product isotopes would help not only in understanding the mechanism of activity transport but can also aid in the detection of fuel failure. Incursion of impurities during fuel handling operation etc., can also be kept track of by periodic monitoring.

Sodium arrives at the reactor site in the form of the commercial product. Commercial sodium contains impurities at tens of ppm levels or even more in some cases. The major impurities are oxygen and hydrogen arising from exposure to atmosphere, carbon, magnesium, calcium and chloride originating from the manufacturing process where graphite electrodes are used in the electrolysis of fused sodium chloride containing calcium chloride, and trace metals. This sodium is purified to reactor grade before it is charged into the reactor system. Quality control analysis is required at every stage of these operations. Once the sodium is in the reactor loop its purity level is monitored. Figure 1 shows the points at which sodium has to be analysed and the levels at which they are measured. In addition to the reactor, a fast reactor research centre has several experimental sodium loops for studying material behaviour, corrosion and mass transfer and for testing components. Purity of the sodium in all these loops have to be monitored by periodic sampling and analysis.

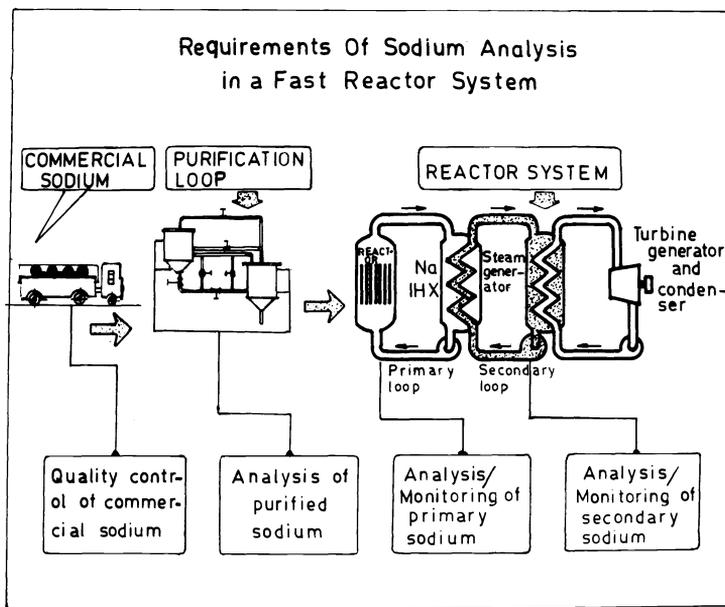


Fig. 1. Requirements of sodium analysis in a fast reactor system

This paper summarises the development of trace analysis techniques as applied to sodium in support of India's fast reactor programme. A Fast Breeder Test Reactor (FBTR) is under construction at Kalpakkam. FBTR is a sodium-cooled fast reactor with a nominal power output of 40 MWth. It has two primary coolant loops and two secondary coolant loops which together have a total inventory of about 90 tonnes of sodium. Some of the early work in connection with sodium analysis was carried out by a group working as part of the Analytical Chemistry Division, BARC, but this was later merged with the main programme on sodium chemistry in RRC.

HANDLING TECHNIQUES

Sodium being a very reactive element requires special techniques for handling. It is well known that sodium catches fire in contact with air and moisture. In handling sodium we have to design against this fire hazard. Brief exposure to air or even commercial argon would lead to the pick up of oxygen, moisture and carbon dioxide thus vitiating the measurement of oxygen, hydrogen and carbon impurities at trace levels. Samples for analysis are, therefore, handled in high purity inert atmosphere glove boxes in which oxygen and moisture levels are maintained at sub-ppm levels by recirculatory purification. Sampling techniques are also carefully designed to eliminate pick-up or loss of impurities during such operations.

A number of inert atmosphere glove boxes have been specially designed and fabricated by us for handling sodium. Figure 2 gives a schematic of the box with its purification system. The cylindrical glove boxes are designed for evacuation, if necessary. The purity of the argon atmosphere is maintained by recirculating it continuously through a purification column using a sealed blower. The purification column consists of a bed of molecular sieves followed by BASF R3-11 catalyst. The box is maintained at a pressure slightly above the atmospheric pressure to minimise the in-leakage of air and moisture. The boxes are leak tight to better than 0.001% of box volume per hour. Hence the purity achieved is very high, with sodium surface remaining untarnished for several hours.

Samples of commercial sodium are received as cut pieces of bricks. These samples are taken inside the glove box and the surface contamination is removed by cutting off all the outer surfaces with stainless steel knives. Only the inside portion is used for analysis. In the case of samples from sodium loops, they are received in nickel tubes crimped at the ends. The ends of these tubes are cut off and only the central portion is used for analysis.

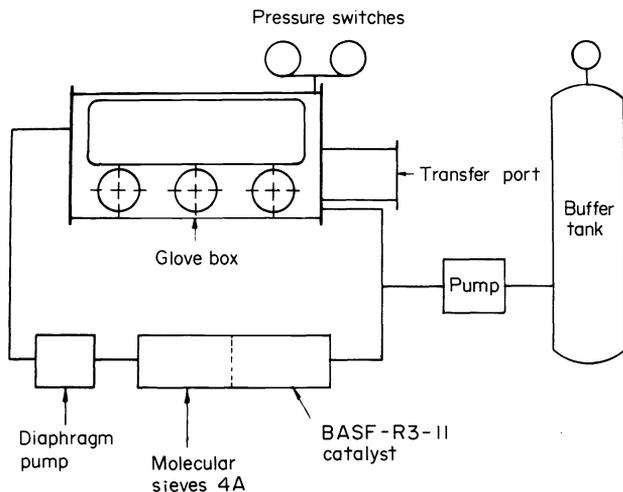


Fig. 2. Schematic of the inert atmosphere glove box with its recirculatory purification system

ANALYSIS OF SODIUM SAMPLES

The demand made by FBTR for trace analysis in sodium has already been shown in Fig. 1. Methods for the analysis of metallic and non-metallic impurities have been standardised to cater to the requirements of the quality control of both commercial sodium and the reactor grade sodium produced by the purification loops. The specifications for these two types of sodium are given in Tables 1 and 2. In addition, sodium from several experimental sodium loops with impurity levels sometimes even lower than purified sodium are being routinely analysed. The sensitivities and accuracies of the methods are adequate to meet the requirements of practically all these samples. When the levels of oxygen, carbon and hydrogen are very low, on-line meters are more appropriate for their measurement.

TABLE 1. Commercial sodium: Specifications and typical results

Impurity	Specification (ppm)	Measured values* (ppm)
Oxygen	< 300	5 - 20
Carbon	< 50	20 - 24
Hydrogen	< 25	7 - 20
Calcium + Magnesium	< 500	1 - 100
Chloride	< 50	5

* Batch to batch variation

TABLE 2. Specifications of reactor grade sodium

Impurity	specified maximum (ppm)	Impurity	Specified maximum (ppm)
Al	5	In	30
Ag	20	K	200
B	5	Li	10
Ba	10	Mg	10
C	30	Mn	10
Ca	10	O	10
Cd	10	P	20
Co	10	Pb	10
Cr	5	Rb	50
Cs	6	S	10
Fe	25	Si	10
H	10	Sn	10
Halogens	15	U	0.2

The sodium metal content shall be 99.95% or better

Oxygen Analysis

Oxygen was initially analysed using the mercury amalgamation method (1). This method depends on the physical separation of sodium from sodium oxide by repeated extraction with mercury. The insoluble sodium oxide residue is dissolved in water and estimated by titration. It was possible to bring down blank values and oxygen could be estimated at ppm levels. However, the amalgamation method proved cumbersome and hence the distillation method (2) was adopted. In this method sodium is distilled off under a vacuum of about 10^{-5} torr. and the residual sodium oxide is estimated by atomic absorption spectrophotometry (AAS). Figure 3 shows the experimental arrangement. The distillation vessel is made of glass and the sodium sample is taken in a clean nickel crucible and placed inside the vessel inside a glove box. The sample is heated by induction heating. The heating stops when the distillation is complete and the temperature is not allowed to go beyond 400°C. To minimise blank values, distillation is carried out in a vessel in which a pre-analysis distillation has already been carried out.

Table 3 gives the oxygen content of sodium samples saturated in oxygen at 125°C, 150°C and 200°C. Precision of these values and their agreement with oxygen solubility data taken from literature (3) are also indicated in the table.

TABLE 3. Oxygen solubility data

Temperature	Oxygen concentration (ppm)	
	Our value	Eichelberger (ref.3)
125°C	1.02 ± 0.1	1.24
150°C	3.16 ± 0.2	2.86
200°C	11.56 ± 0.8	11.67

Carbon Analysis

In the early stages of our programme carbon in sodium was estimated by total combustion with Al_2O_3 and SiO_2 as flux (4). This method was found to be too tedious and time-consuming. Further, the blank values were high. Hence the total combustion method was given up. At present carbon is routinely measured using the distillation-combustion method. The distillation arrangement is the same as that for oxygen except that the sample is taken in a quartz crucible. After the distillation the crucible is covered with quartz wool and the residue is combusted in a stream of oxygen at 1100°C and CO_2 thus produced is estimated by manometry or by gas chromatography. The recovery in this method was established by carrying out the estimations after standard additions of graphite and calcium carbonate.

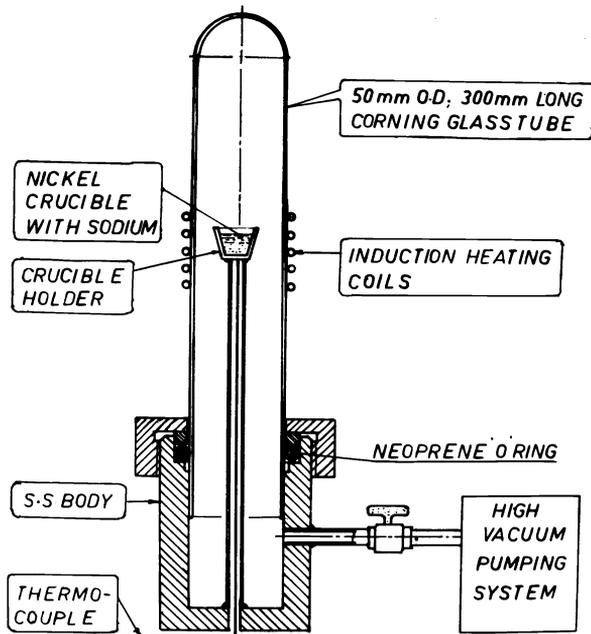


Fig. 3. Set up for sodium distillation

Some results are shown in Table 4 along with standard deviation for a set of measurements which reflects not only the precision of the method but also the inhomogeneity of the samples.

TABLE 4. Carbon analysis

Nature of sodium sample	Carbon content (ppm)	Standard deviation (ppm)
Molten sodium from the pot in the sodium glove box	9.8	3.5
Commercial sodium (1st lot)	27.6	5.4
Commercial sodium (recent lots)	19.8	1.4
Loop sodium - initial stages	13.5	2.8
Loop sodium - final stages of purification	1	-

Hydrogen Analysis

Hydrogen in sodium is estimated by the amalgam reflux method (4). Hydride and hydroxide hydrogen are estimated separately by refluxing at 200°C and 370°C respectively. The evolved hydrogen is estimated using a gas chromatograph with a thermal conductivity detector. Sample addition is done inside the glove box after successive refluxing which was found necessary for reducing blank. The sodium sample taken from the pot at 200°C gives the solubility of hydrogen at that temperature. This has been measured and found to be 0.64 ± 0.01 ppm. This value agrees with that quoted in literature (5), viz. 0.63 ppm. The values quoted are for hydride hydrogen. No hydroxide hydrogen was observed by following the above procedure.

Trace Metal Analysis

Trace metals are being analysed by AAS in the residue obtained after distilling off sodium. The distillation is carried out in tantalum or alumina crucibles. The residue is dissolved in dilute acid and analysed by AAS with a flame atomiser. For impurities present at low levels a flameless (graphite furnace) atomiser is employed. Table 5 gives some typical results.

TABLE 5. Analysis of trace metals in 'Nuclear grade' sodium

Element	Flame AAS Concentration (ppm)	Flameless AAS			
		Direct method		After distillation	
		Concentration (ppm)	Detection limit (ppm)	Concentration (ppm)	Detection limit (ppm)
Calcium	< 0.5	-	-	-	-
Magnesium	< 0.05	-	-	-	-
Chromium	< 0.3	0.17	0.06	0.16	0.01
Manganese	< 0.1	0.04	0.02	0.03	0.005
Cobalt	< 0.3	< 0.08	0.08	< 0.01	0.01
Copper	< 0.2	0.15	0.08	0.12	0.01
Lead	-	0.16	0.07	-	-

More recently a technique of direct determination of trace metals without a separation and pre-concentration step has been developed in our laboratory. As this is discussed in a paper (6) being presented at this Symposium it will not be further described here. This technique is particularly useful for elements such as Cd, Zn etc., which volatilise off during distillation.

Other Trace Elements

Trace elements which are not covered by the above methods require individual attention (7). B and Si are determined by colorimetry making use of the colours developed in the curcumin oxalic acid complex and molybdenum blue respectively. Chloride is estimated using an ion selective electrode and uranium by fluorimetry. K and Li are measured by flame emission spectrometry. Some typical results are given in Table 6.

TABLE 6. Analysis of trace elements in Commercial sodium - Typical results

Element	Technique	Range of values (ppm)
Potassium	Flame emission	100 - 200
Lithium	Flame emission	0.5 - 1.0
Boron	Colorimetry	0.20
Cadmium	Colorimetry	0.1
Uranium	Fluorimetry	0.1
Aluminium	Colorimetry	2.0
Silicon	Colorimetry	5 - 20

ON-LINE MONITORS

The laboratory techniques described above apply to sodium samples received in the laboratory. Sampling procedure itself could change the characteristics of the sodium to be analysed. Impurities are likely to be picked up during sampling and in the subsequent handling of the sample. With sufficient care this error can be minimised. Secondly there is the possibility of impurity segregation during sampling. The solubilities of most impurities drop sharply as the sample cools to its melting point and, as the cooling is non-uniform the impurities tend to segregate to the surface and get absorbed on the walls of the container. On-line methods would minimise such errors and would also provide a continuous record of the changes in impurity levels. Sampling followed by analysis takes a good deal of time. In order to monitor the sudden variations in concentration levels, an on-line monitor is particularly

advantageous. For example, oxygen and hydrogen meters are proposed to be used to detect the leakage of water into sodium in the steam generators of fast breeder reactors (8). Only on-line monitors can provide the fast response that is desired in such applications. As has already been shown, the laboratory techniques do not give accurate values when the levels of oxygen, carbon and hydrogen are very low. The on-line meters discussed below have proved very useful in such cases. The laboratory techniques discussed above measure only the total concentration of an impurity, whereas the on-line meters measure the thermodynamic activity. This is particularly important for carbon where the total carbon measured include the dispersion of undissolved carbon whereas the meter measures the activity due to dissolved carbon which is more important in the carburization-decarburization process.

Oxygen Meter

Oxygen meters have been developed in several laboratories (9,10). They are electrochemical cells in which the electromotive force (e.m.f) acting between two oxygen electrodes is measured. One is a reference electrode and the other the sodium containing dissolved oxygen. They are separated by an oxygen-ion-conducting solid electrolyte made of yttria doped thoria.

The cell may be represented as follows:

Na, Na₂O (dissolved) // ThO₂, Y₂O₃ // O₂ (reference). The e.m.f. output of this cell is given by

$$E = -\frac{RT}{nF} \ln \frac{a_o(\text{Na})}{a_o(\text{ref})}$$

where $a_o(\text{Na})$ and $a_o(\text{ref})$ are the activities of oxygen in sodium and the reference electrode respectively. The Harwell oxygen meter (10) uses a 15 cm long electrolyte tube one end of which dips in sodium. The GE meter (9) uses a thimble of solid electrolyte brazed to a metal tube so that the entire thimble is immersed in sodium. The electrochemical oxygen meter that we have set up is shown in Fig. 4 (a) and (b).



Fig. 4 (a). A view of the oxygen meter

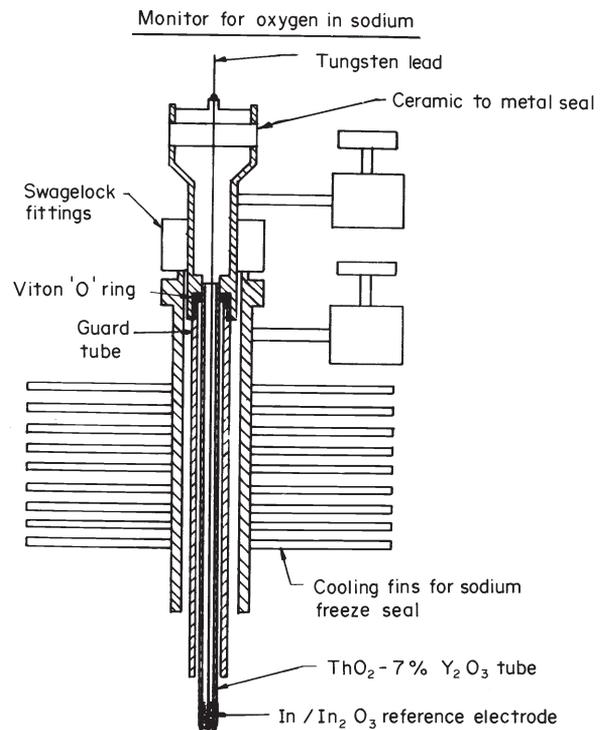


Fig. 4 (b). Schematic diagram of the oxygen meter

It consists of a long solid electrolyte tube dipping in sodium. An In/In₂O₃ reference electrode is located inside the electrolyte tube with a tungsten wire dipping in it for electrical contact. Care is taken to ensure that the atmosphere above the reference electrode as well as of sodium is of high purity argon. The e.m.f. generated due to the difference in oxygen activity between the sodium and the reference electrode is measured with a high impedance digital voltmeter connected between the tungsten wire and the sodium container.

Table 7 gives the data from our oxygen meter obtained in static pot tests. Here the oxygen was first purified by gettering with magnesium. This highly purified sodium was contained in a stainless steel container and the oxygen activity was controlled by the Na-Cr-O system. The measured e.m.f. values and the corresponding oxygen concentrations are given in columns 2 and 3. The oxygen concentrations have been computed from the known thermodynamic data on the Na-Cr-O system (11). The oxygen activities were converted to oxygen concentration assuming Henry's Law to be valid and making use of Noden's (12) oxygen solubility data. The agreement between the measured and the theoretical e.m.f. is shown in Fig. 5. The observed values are slightly lower than the theoretical values and this may be due to (1) the O²⁻ ion transport co-efficient being less than unity and (2) the uncertainties in the thermodynamic data. As a practical tool for oxygen measurement in liquid sodium, the oxygen meter is undoubtedly very useful. For this purpose it has to be calibrated.

TABLE 7. Results from the Oxygen meter

T(K)	e.m.f. (Volts)	Concentration of Oxygen in sodium (ppm)
657.2	0.585	0.039
680.2	0.596	0.060
705.2	0.600	0.095
731.2	0.604	0.147
740.2	0.604	0.170
753.2	0.606	0.208
758.2	0.613	0.225
769.2	0.614	0.265
777.2	0.608	0.297
801.2	0.619	0.416
824.2	0.620	0.563

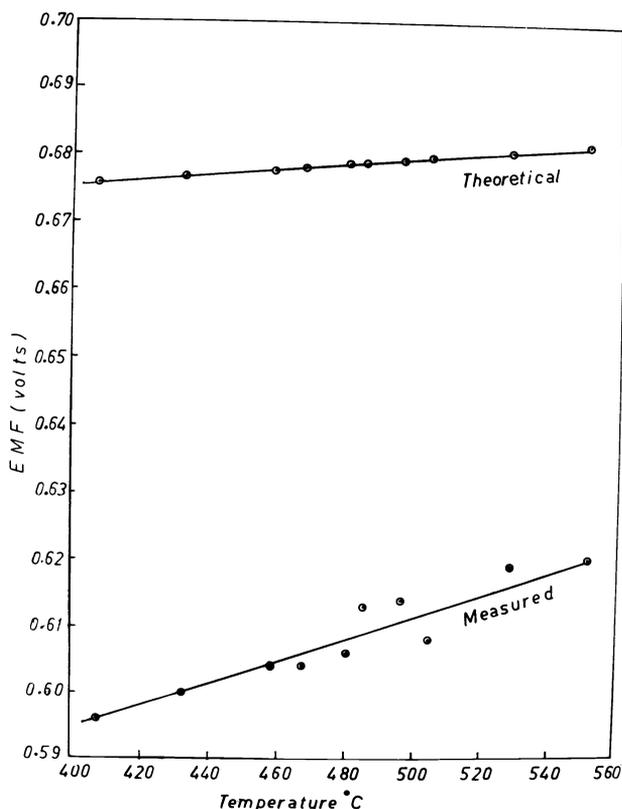


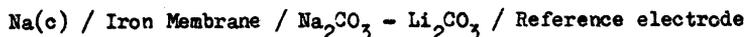
Fig. 5. Plot of the E.M.F. Vs. Temperature for the Oxygen meter

Carbon Meter

There are two types of carbon meters currently under development in the various laboratories: (1) the diffusion type and (2) the electrochemical carbon meter. The diffusion type of carbon meter represented by the Harwell Carbon Meter (13) makes use of an iron membrane as a sensor. This sensor is in the form of a helically wound iron tube, the inner surface of which is oxidised to form an iron oxide film. When exposed to sodium, preferably in the temperature range of 500 - 600°C, carbon diffuses through the membrane and reacts with the iron oxide on

the inner surface to form CO and CO₂ which are swept away by an inert carrier gas. The rate of diffusion of carbon through the membrane and the resulting flux of CO + CO₂ is directly proportional to the carbon activity of CO + CO₂ in sodium. The rate of production of CO + CO₂ is analysed with high sensitivity by first catalytically converting them to CH₄ and then measuring CH₄ in a Flame Ionisation Detector.

The electrochemical carbon meter (14) directly measures the activity of carbon in sodium (a_c) using an electrochemical cell in which an iron membrane in equilibrium with sodium forms one electrode and a carburized iron rod dipping in a molten electrolyte contained inside the membrane forms the other electrode. The electrolyte is a molten eutectic of sodium and lithium carbonates. The cell is represented by



The cell e.m.f. E_e is related to the carbon activity by the relation

$$E = -\frac{RT}{4F} \ln \frac{a_c}{a_{\text{ref}}}$$

An electrochemical carbon meter working on this principle has been set up in our laboratory and tested in a static sodium system (Fig. 6). These e.m.f. readings and the corresponding carbon concentrations calculated using the above equation in conjunction with Ainsley's solubility data (15) are given in Table 8.

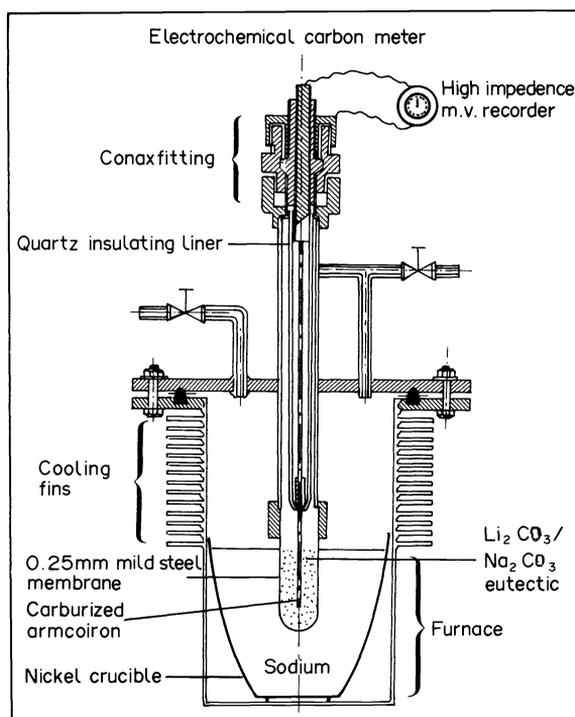


Fig. 6. Schematic diagram of the electrochemical carbon meter

Hydrogen Meter

The hydrogen meter is important especially in the secondary circuits as it aids the detection of leakage of water into sodium. All hydrogen meters in use or under development depend on the diffusion of hydrogen through a nickel membrane and the measurement of hydrogen pressure (8). The hydrogen meter being fabricated by the Technical Physics Division, BARC, for use in FBTR is schematically shown in Fig. 7. It is based on the French (EdF) design and incorporates a quadrupole mass spectrometer and a sputter ion pump in the detection system. The mass spectrometer affords high selectivity and sensitivity and it is expected that this system can measure hydrogen concentration down to 0.01 ppm (16). A dynamically pumped detector system like this has to be separately calibrated.

TABLE 8. Results from the carbon meter

T K	Measured e.m.f. (mV)	Solubility of carbon (ppm)	a_o	C (ppm)
793	57	1.30	0.012	0.0156
803	66	1.63	0.006	0.0098
813	63	2.02	0.0085	0.017
823	64	2.49	0.00835	0.021
833	104	3.06	0.00046	0.0014
843	72	3.73	0.0052	0.0194
853	72	4.54	0.0055	0.025
858	100	5.00	0.00077	0.0039

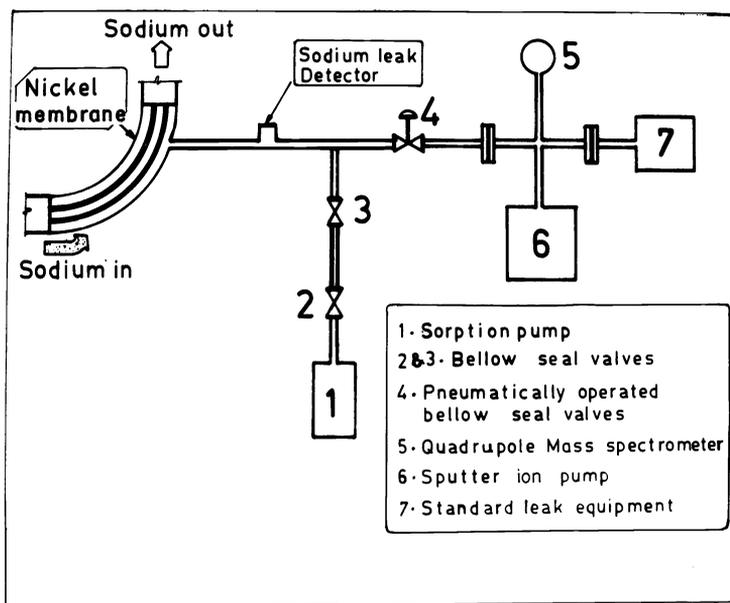


Fig. 7. Schematic diagram of the FBTR hydrogen meter

ANALYTICAL SODIUM CHEMISTRY LOOP

For trace analysis in sodium no standard is available. Even if a sodium sample with known concentrations of impurities were to be made, the composition is likely to change on handling. To increase our confidence in the analytical procedure we have to analyse the same sample by different techniques. We have, therefore, set up an Analytical Sodium Chemistry Loop (ASCL) in which analysis for trace levels of O, C and H can be carried out simultaneously using three different methods: (1) sampling and analysis; (2) on-line monitoring and (3) foil equilibration.

Figure 8 gives a schematic diagram of the loop. It is a small volume loop with a capacity of about 2 litres of sodium which can be operated isothermally using an electromagnetic pump. The material of construction is AISI 316 stainless steel. The d.c. electromagnetic pump was fabricated in our laboratory using a permanent magnet. A direct current of about 100 A is passed across the sodium in the specially shaped duct passing through the magnetic field.

Copper bus bars attached to the duct by silver soldering carry the current to the sodium. The loop incorporates a cold trap for impurity control, cooling coil, expansion tank and charge and dump tanks.

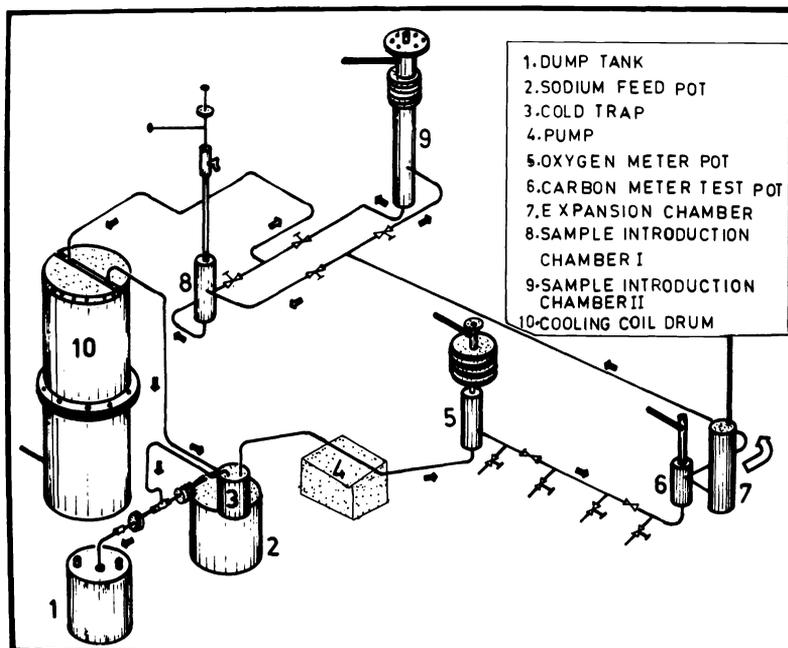


Fig. 8. Analytical Sodium Chemistry Loop

The loop is provided with ports for oxygen and carbon meters. Oxygen and carbon meters of the type described earlier are being fitted in these ports and can be operated in the temperature region of 500 - 600°C. There is also provision for testing other meters. Ports are provided for foil equilibration. Vanadium wire equilibration method is used for the analysis of oxygen (17) and iron alloys for carbon (18). Samples can be withdrawn for chemical analysis and impurities introduced. All these features make this a versatile facility for calibrating on-line meters and for standardising analytical techniques.

ANALYSIS OF PRIMARY SODIUM

In addition to inactive impurities, sodium in the primary loops of a fast reactor contains a number of radioactive impurities. These fall in three groups:

- (1) Activities such as ^{54}Mn , ^{58}Co and ^{60}Co which result from the activation of the corrosion products.
- (2) Fission products such as ^{137}Cs , ^{131}I and ^3H . Tritium is also produced in the control rods.
- (3) U and Pu which are fuel materials.

When the reactor is operating, the radioactivity of the primary sodium is mainly due to ^{24}Na . This isotope has a half-life of 15 hours and hence decays down to tolerable levels within a few days of reactor shut down. Only those activities which are significant after this period are considered here.

These impurities will be required to be analysed after FBTR is commissioned. Current effort is only in standardising the methods. ^{54}Mn , ^{58}Co and ^{60}Co remain in the residue left after the distillation of sodium and can be conveniently measured by gamma spectrometry. ^{137}Cs cannot be separated by distillation and, hence the sodium sample is dissolved and the activity is determined in the solution by gamma spectrometry. In general, this method of gamma spectrometry of the solution is used for all these isotopes. ^{131}I is determined by counting after separating the iodine by solvent-extraction technique.

Tritium determination involves its conversion to tritiated water and assay by liquid scintillation counting. Part of the tritium is evolved along with hydrogen during the dissolution of sodium and this is converted to water by passing over copper oxide at 600°C. The sodium hydroxide produced is acidified and distilled to obtain the other part of the tritiated water. The two portions are combined for counting.

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