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SODIUM SAMPLING AND IMPURITY  
MONITORING SYSTEMS IN U.S. BREEDER REACTORS

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US/JAPANESE SODIUM TECHNOLOGY AND LMFBR INSTRUMENT  
AND CONTROL EXCHANGE SEMINAR, JUNE 1978

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# SODIUM SAMPLING AND IMPURITY MONITORING SYSTEMS IN U.S. BREEDER REACTORS

J. J. McCown

W. H. Olson

## INTRODUCTION

In the U.S. Breeder Reactor Program (BRP) the capabilities for sodium sampling and impurity monitoring have undergone extensive changes and improvement over the past 10 years. Devices for obtaining bulk samples of sodium and for on-line distillation have been developed and are in use. Samplers which can perform multiple functions reducing plant hardware needs have been designed and tested. Improved plugging temperature indicators and meters for in-situ measurement of carbon, hydrogen, oxygen and tritium have been developed and tested on sodium loops, large test facilities, and excepting carbon meters, in reactor service.

The Experimental Breeder Reactor-II (EBR-II) at the Idaho National Engineering Laboratory (INEL) has been in operation since 1964 with a comprehensive program for sodium characterization in place since 1967. Details of this monitoring program, early sampling methods, samplers in use through 1974 and data on the behavior of measurable impurities have

been reviewed extensively in several Argonne National Laboratory (ANL) documents.<sup>(1,2)</sup> The status of the continuous on-line monitoring programs used at EBR-II on reactor sodium and cover gas systems (for purity control and for leak detection purposes) was presented in several papers at the 1976 International Conference on Liquid Metal Technology in Energy Production.<sup>(3,4,5)</sup>

The Fast Flux Test Facility (FFTF), outside Richland, Washington, will provide expanded BRP test capabilities in the U.S. The FFTF will start operations in 1978 and complete sodium characterization is planned on all FFTF sodium systems: primary and secondary, main reactor and closed loops. The Hanford Engineering Development Laboratory (HEDL) has been involved with ANL in sampler and meter development work since the start of the FFTF project. Much of this work was done on the Prototype Applications Loop (PAL) and the High Temperature Sodium Facility (HTSF) and has been reported.<sup>(6,7,8,9)</sup> A Multipurpose Sampler (MPS), designed for use on FFTF, combines the functions of several sodium sampling devices. It will be used on 12 separate sampling stations on the FFTF to provide a total sampling capability for all reactor sodium systems.

A new insert, using small oxygen and hydrogen meter probes, has been designed and is presently being tested on PAL.<sup>(10-16)</sup> The insert will be used in the FFTF MPS units utilizing the sampler controls to condition the sodium, thus simplifying on-line meter hardware significantly.

This paper presents the current status of the samplers and meters on EBR-II, a description of the FFTF systems and their performance capabilities.

## SODIUM SAMPLING SYSTEMS

### EBR-II

The primary and secondary sodium systems on EBR-II have used two types of sodium sampling devices, "flow-through" and "overflow", for bulk sampling. The "overflow" technique used presently, provides to the laboratory cups containing 3 to 60 g of sodium with the device shown in Figure 1. Dissolution of the total sample avoids impurity segregation errors. The samples are used in determining all impurities except oxygen, hydrogen and carbon.

Primary system overflow sampling is done on the purification loop, as indicated in Figure 2. Secondary system sampling utilizes a similar arrangement with the samplers located on a side loop of the sodium purification system.

One other system is used to provide additional primary sodium monitoring and sampling capabilities. A Radioactive Sodium Chemistry Loop (RSCS)<sup>(17)</sup>, consisting of a pumped loop in parallel with the purification system, supplies sodium to several shielded cell areas which contain an on-line meter module<sup>(18)</sup> and a metal equilibration device<sup>(19)</sup> used to expose wires and foils for analyzing carbon, hydrogen and oxygen. This sampler has been in use since 1972 performing 24 hour equilibration runs in 750°C sodium. Details of the analytical methods are presented in a separate seminar paper.

Two additions to the EBR-II sampling devices are the segregated-iodine sodium sampler (SISS), shown in Figure 3; and the distillation sampler, shown in Figure 4. SISS is essentially a vertical mounted flow-through tube sampler which uses the temperature-dependent segregation behavior of iodine isotopes to collect active iodine on the tubing wall

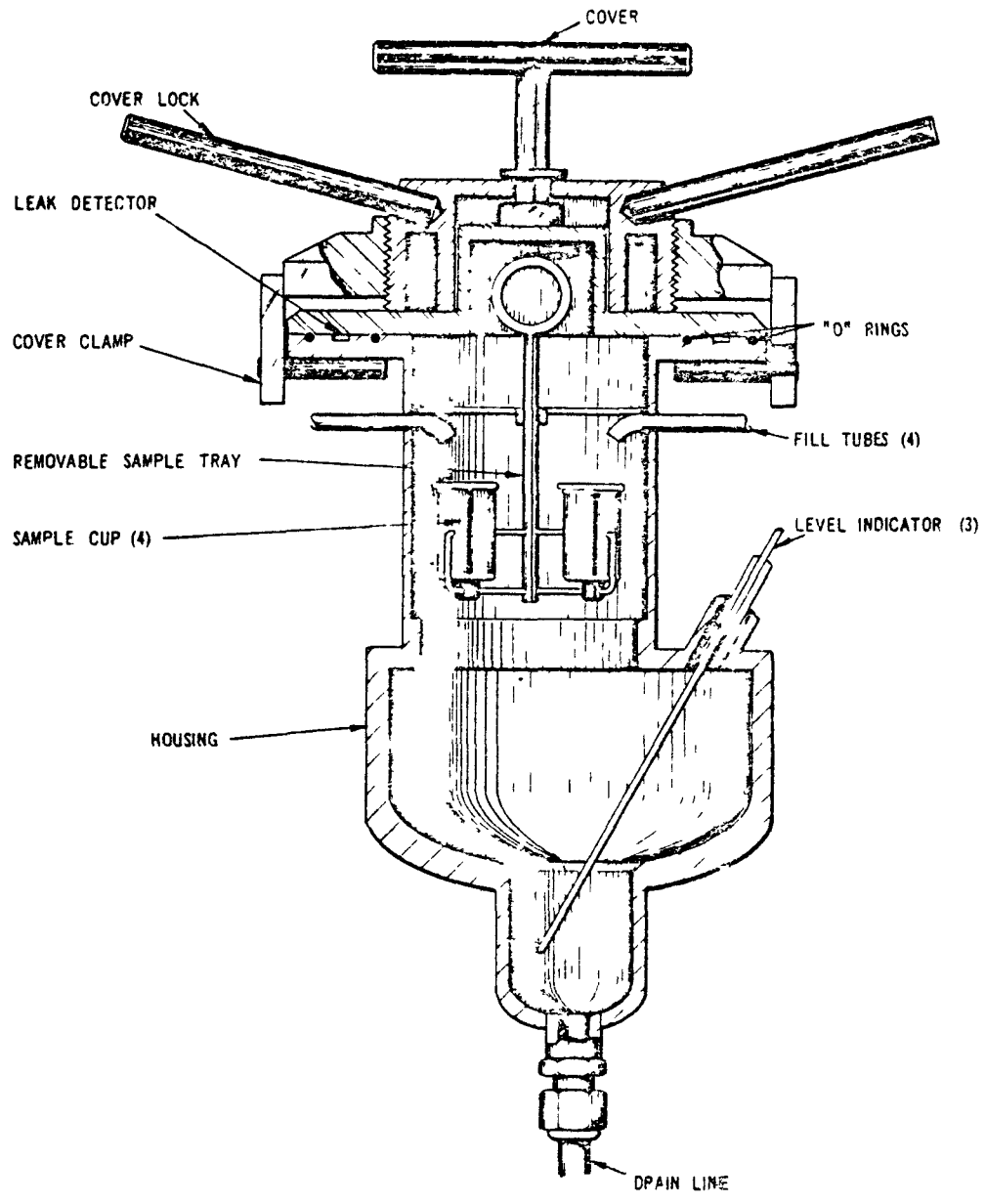


Fig. 1 EBR-II overflow sampler

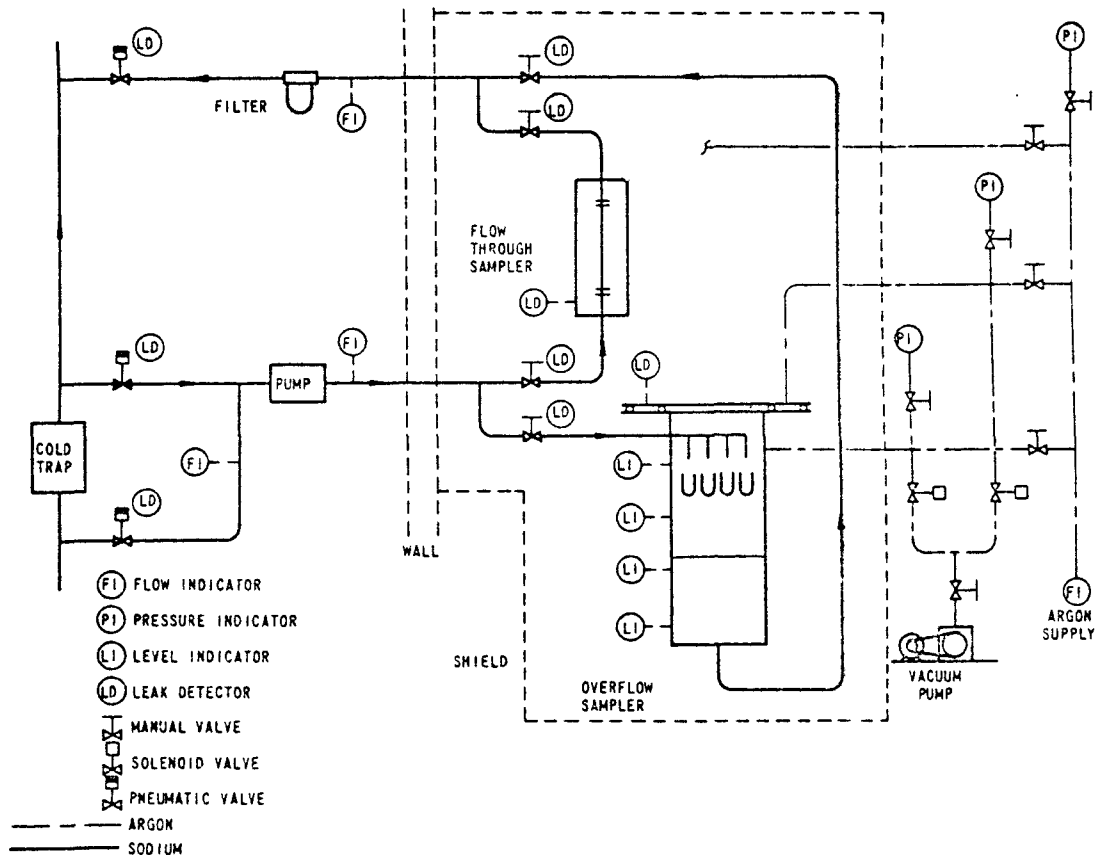


Fig. 2 EBR-II Primary-sodium sampling system

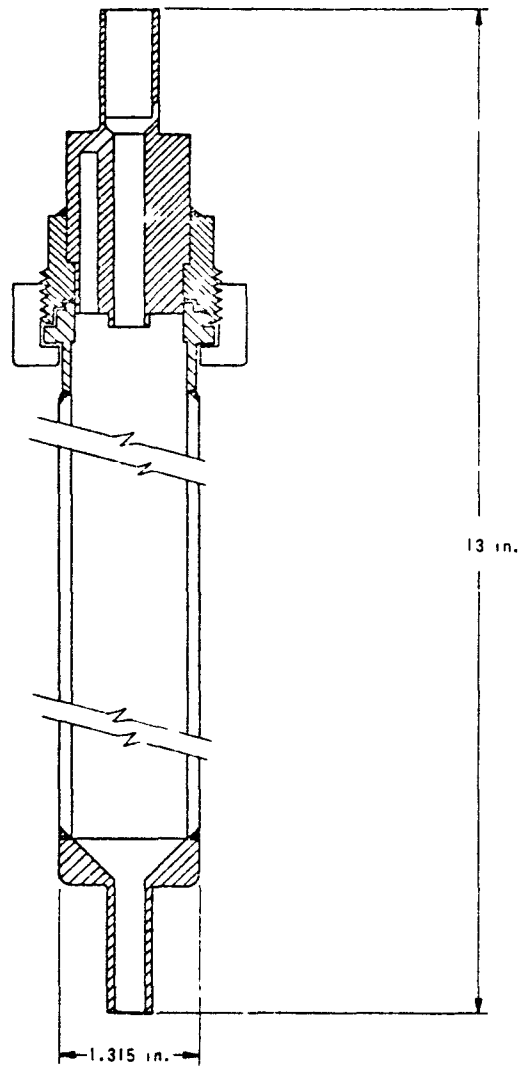


Fig. 3 Segregated-iodine sodium sampler (SISS)



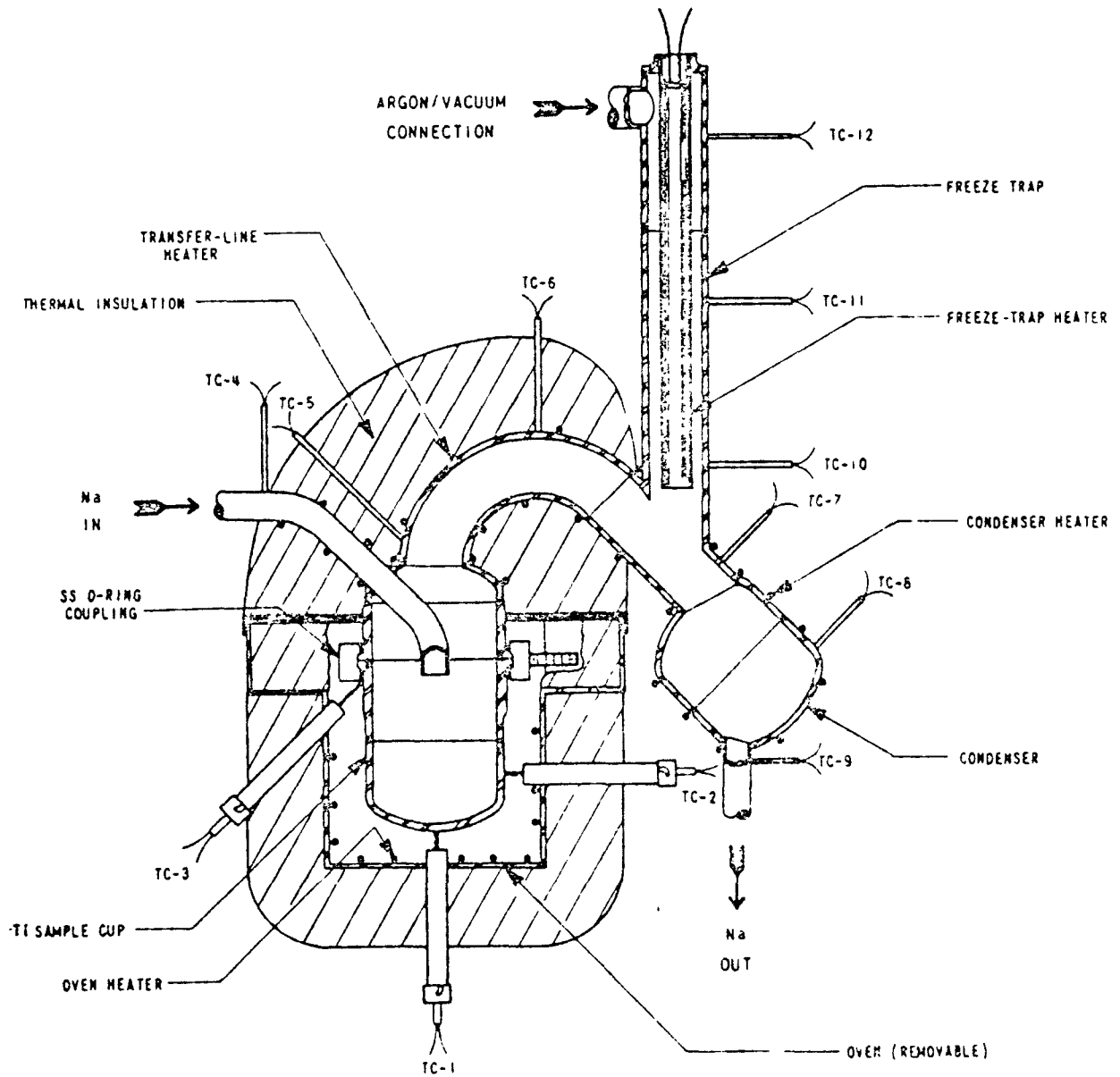


Fig. 4 EBR-II on-line vacuum-distillation sodium sampler

as the sodium cools to about 230°F prior to draining the chamber. The distillation sampler provides capability for concentrating impurities from bulk sodium by on-line distillation which greatly reduces the sample activity and simplifies subsequent handling problems in the laboratory. Both units are to be located in shielded cells on RSCL.

#### FFTF

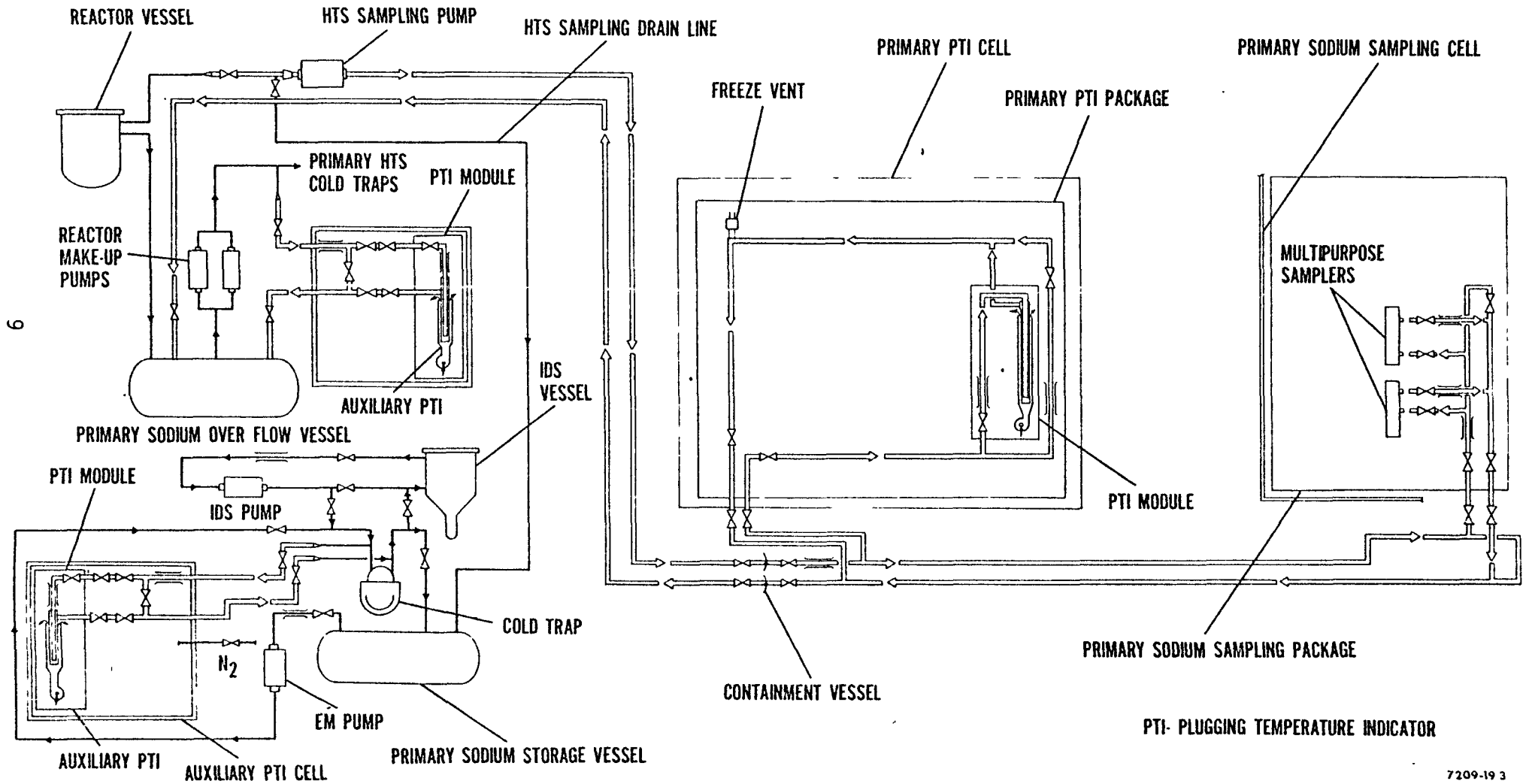
The Impurity Monitoring and Sampling Systems on FFTF provide Multipurpose Samplers (MPS) and Plugging Temperature Indicators (PTI)<sup>(20)</sup> on all reactor and closed loop primary and secondary sodium systems, as shown in Figures 5-8. In all cases, a side loop of sodium equipped with em pump, flowmeter and flow control valve provides up to 10 gpm which is passed through several parallel flow paths to the MPS and PTI equipment. A typical sampling package arrangement is shown in Figure 9 for the secondary systems. All primary sodium sampling systems are arranged with the PTI's in shielded cells and with two MPS units per system (6 at startup), located in a common manipulator equipped hot cell.

Development work done in support of the FFTF design included testing sampler prototypes of the flow-through tube, metal equilibration, overflow cup and multipurpose units. This work was done on several sodium facilities at HEDL with the primary testing performed on a controlled chemistry system, the PAL. The chemistry changes on PAL and on the HTSF were followed using the three MPS inserts shown in Figures 10-12 to provide three types of samples; metal equilibration, overflow cup and particulate filtration.

Installation of the sampling systems on FFTF is almost complete. The sodium sampling system piping has been installed and twelve MPS units have recently been put into place. Plant chemistry conditions at startup and during initial operations will be followed via sampling

# PRIMARY SODIUM CHARACTERIZATION SYSTEM

## FFTF HEAT TRANSPORT SYSTEM



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Fig. 5 Primary sodium characterization system for FFTF heat transport system

# PRIMARY SODIUM CHARACTERIZATION SYSTEM

## FFTF CLOSED LOOP SYSTEMS

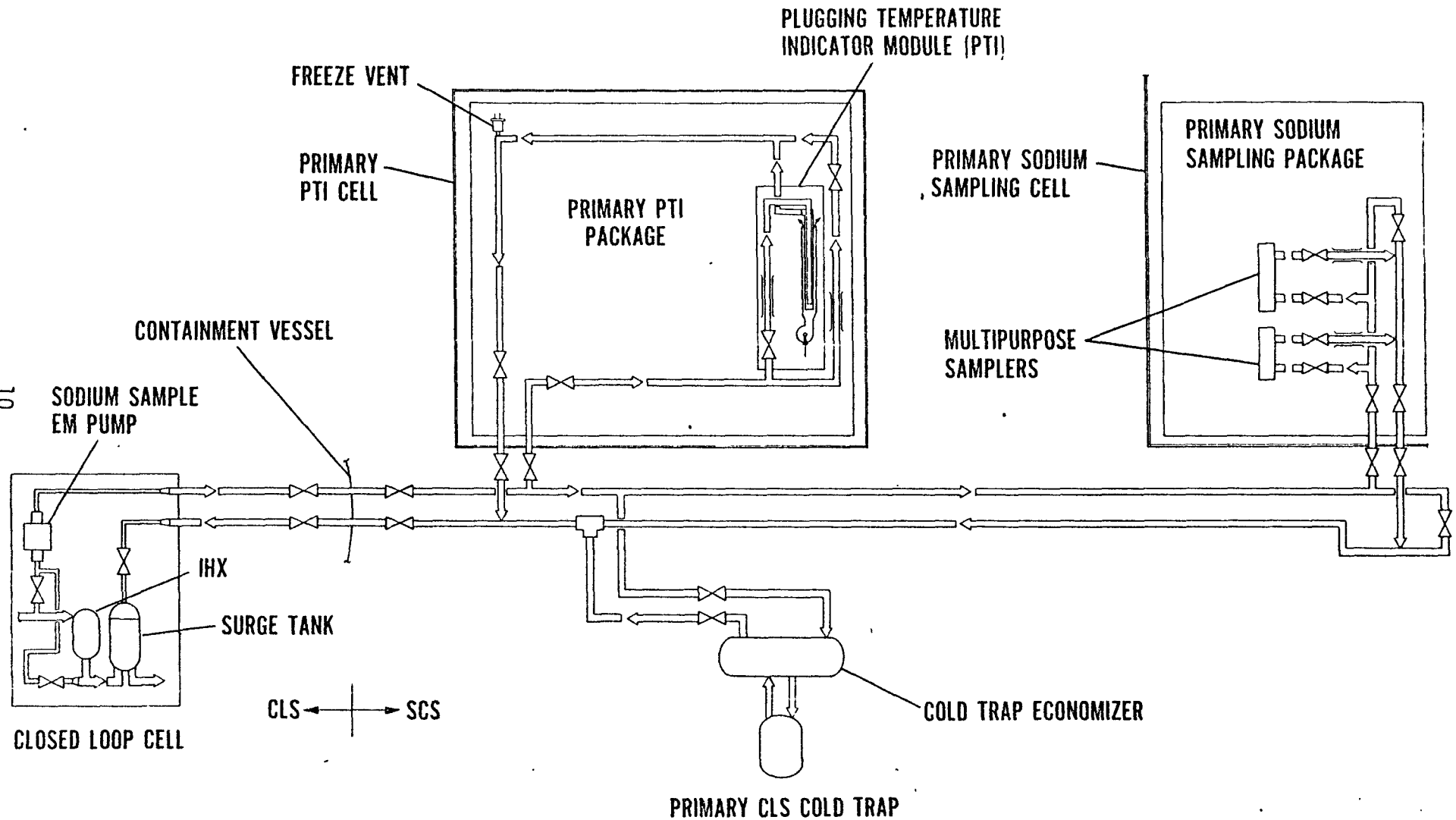


Fig. 6 Primary sodium characterization system for FFTF closed loop systems

# SECONDARY SODIUM CHARACTERIZATION SYSTEM

FFTF HEAT TRANSPORT SYSTEM SECONDARY SODIUM LOOPS NO. 1,2&3

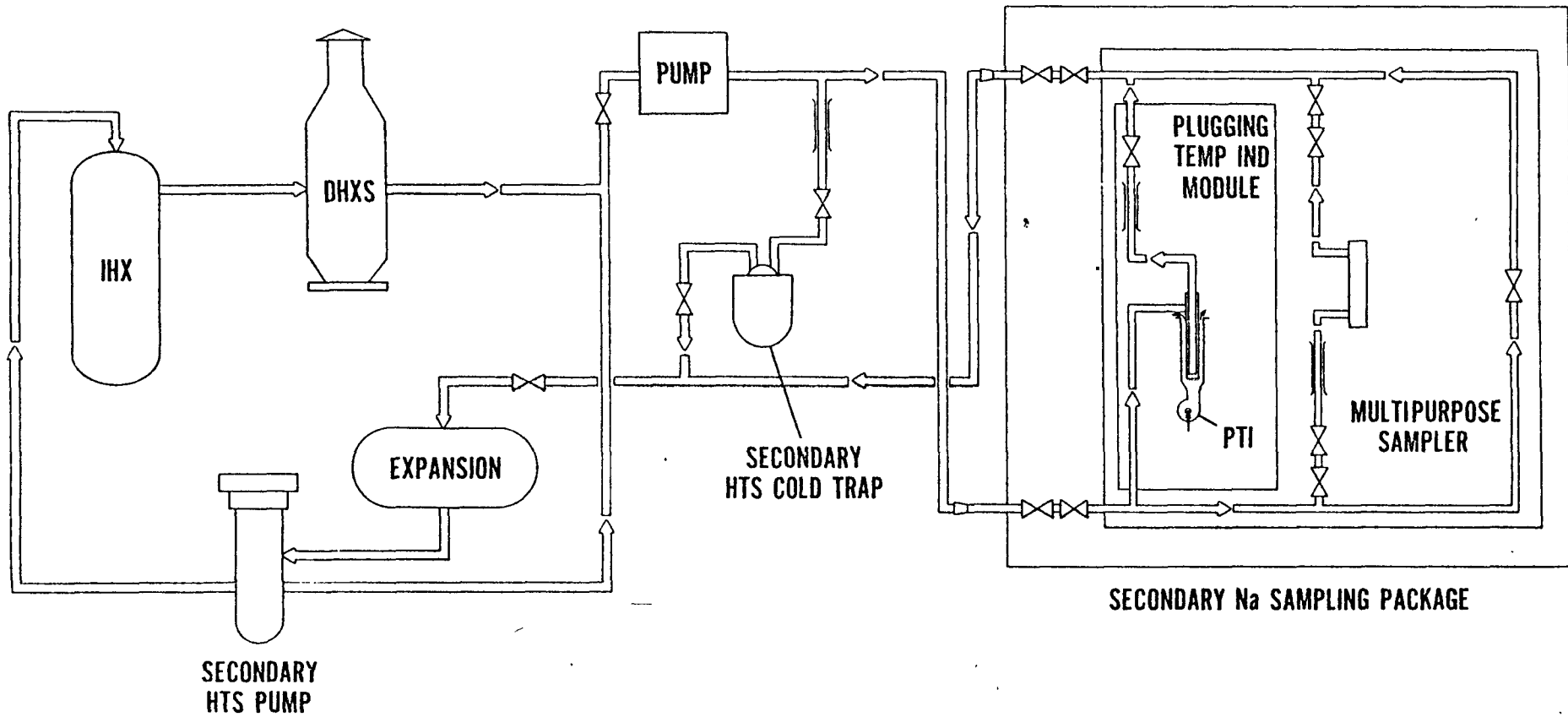


Fig. 7 Secondary sodium characterization system for FFTF heat transport secondary loops

# SECONDARY SODIUM CHARACTERIZATION SYSTEM

## FFTF CLOSED LOOP SYSTEM SECONDARY SODIUM LOOPS

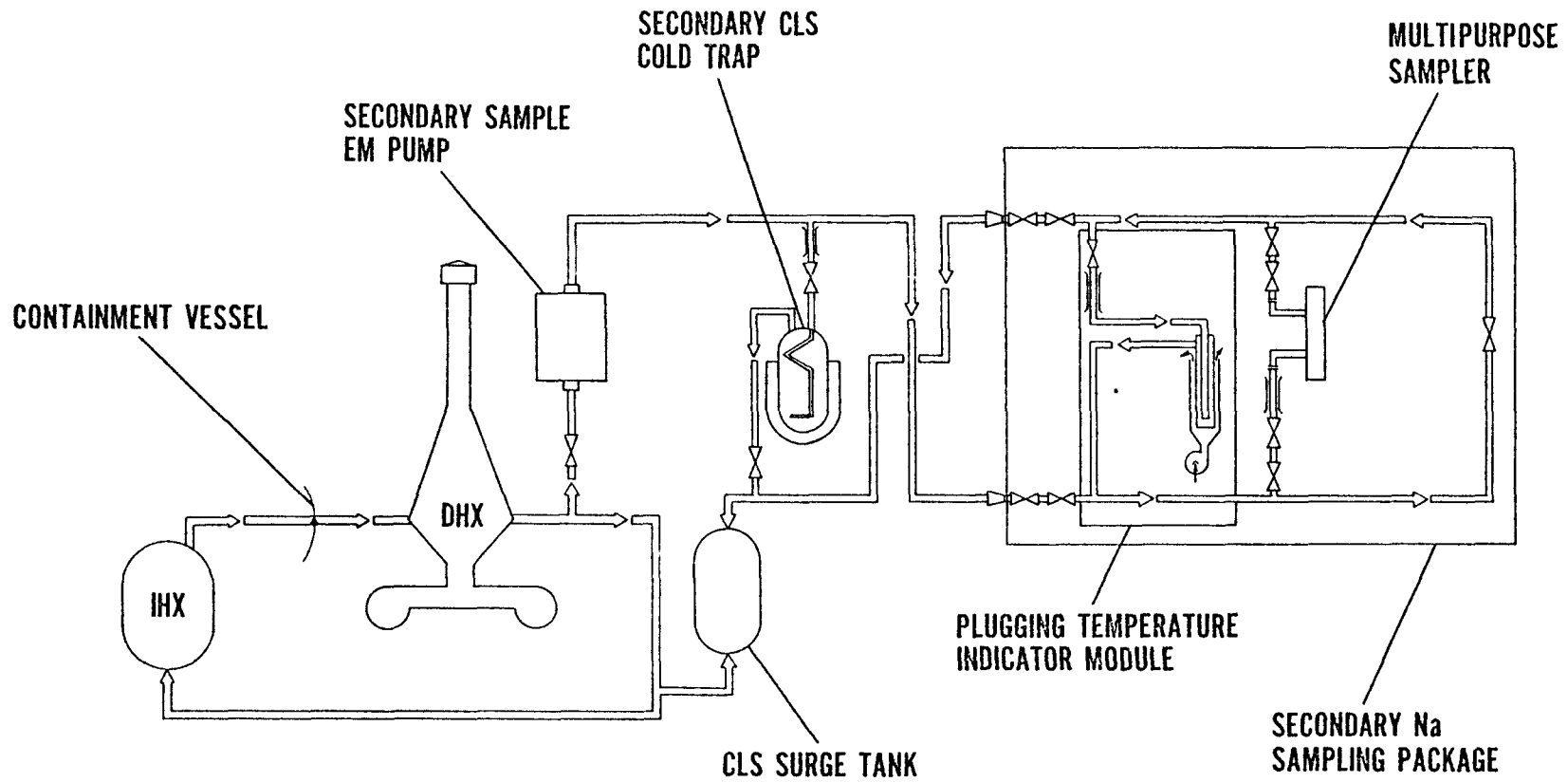
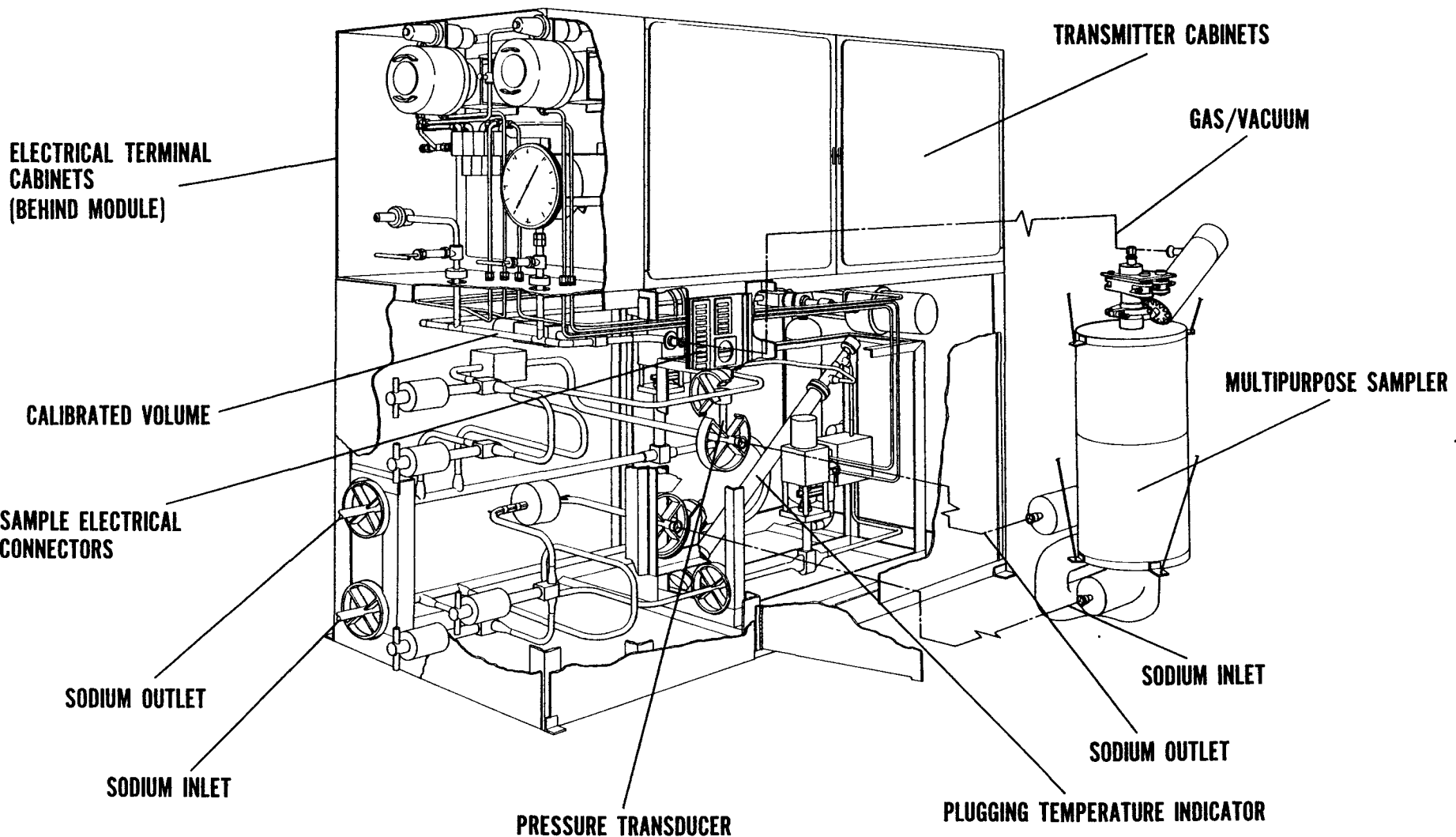


Fig. 8 Secondary sodium characterization system for FFTF closed loop secondary systems

# SECONDARY SODIUM SAMPLING PACKAGE



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Fig. 9 FFTF secondary sodium sampling package

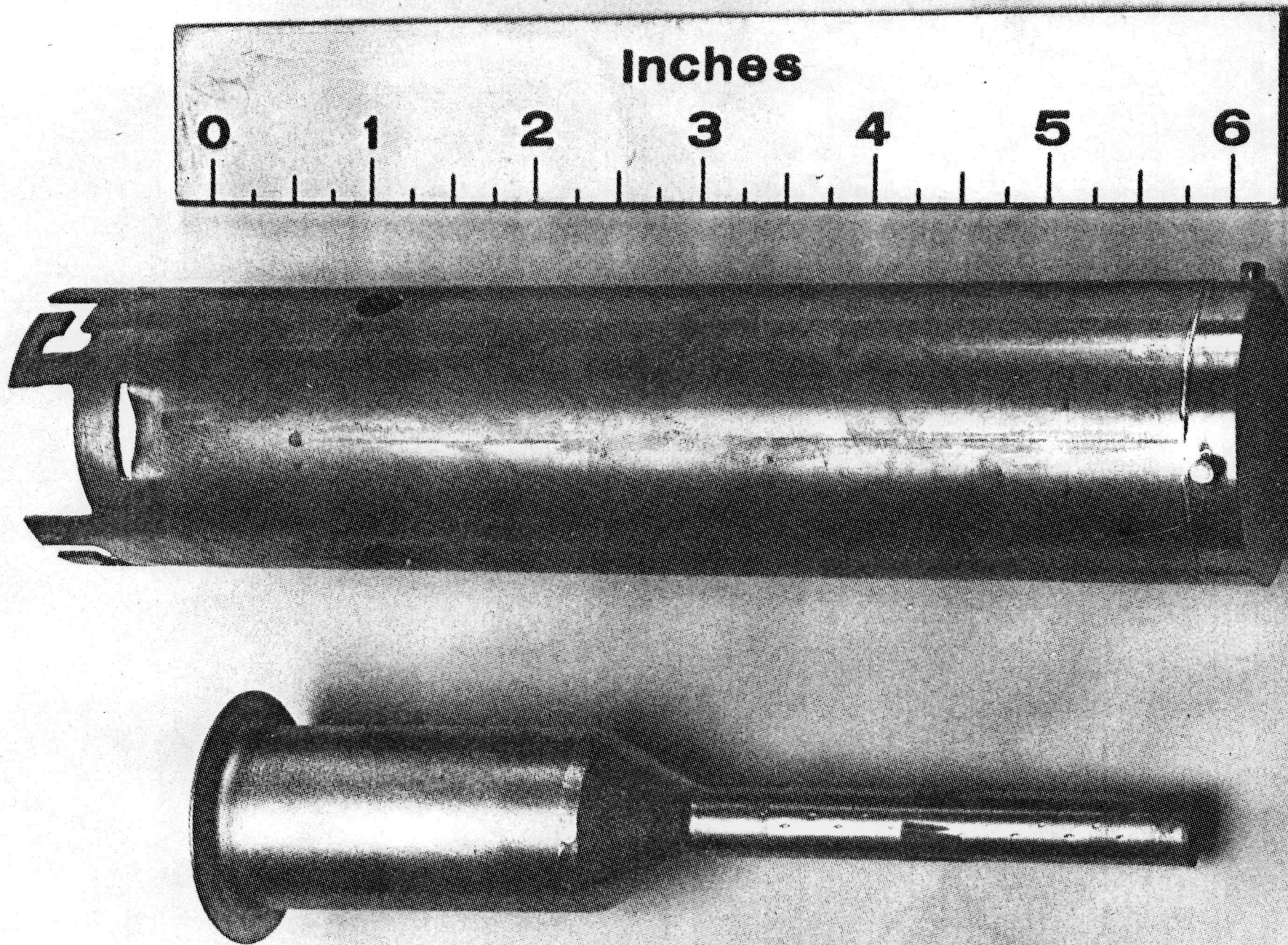


Fig. 10 Multipurpose sampler insert cup for metal wire equilibrations



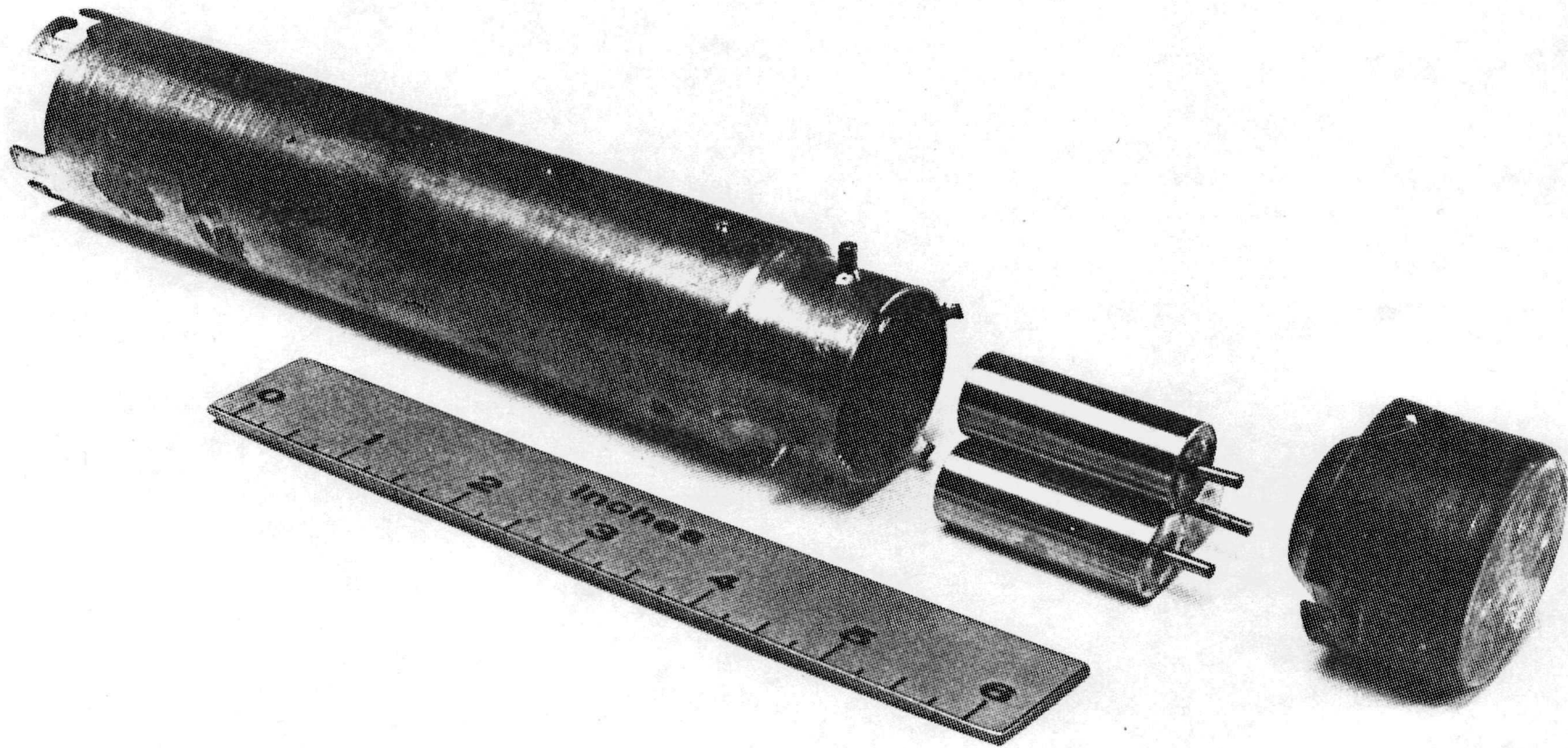


Fig. 11 Multipurpose sampler insert cups for overflow sampling

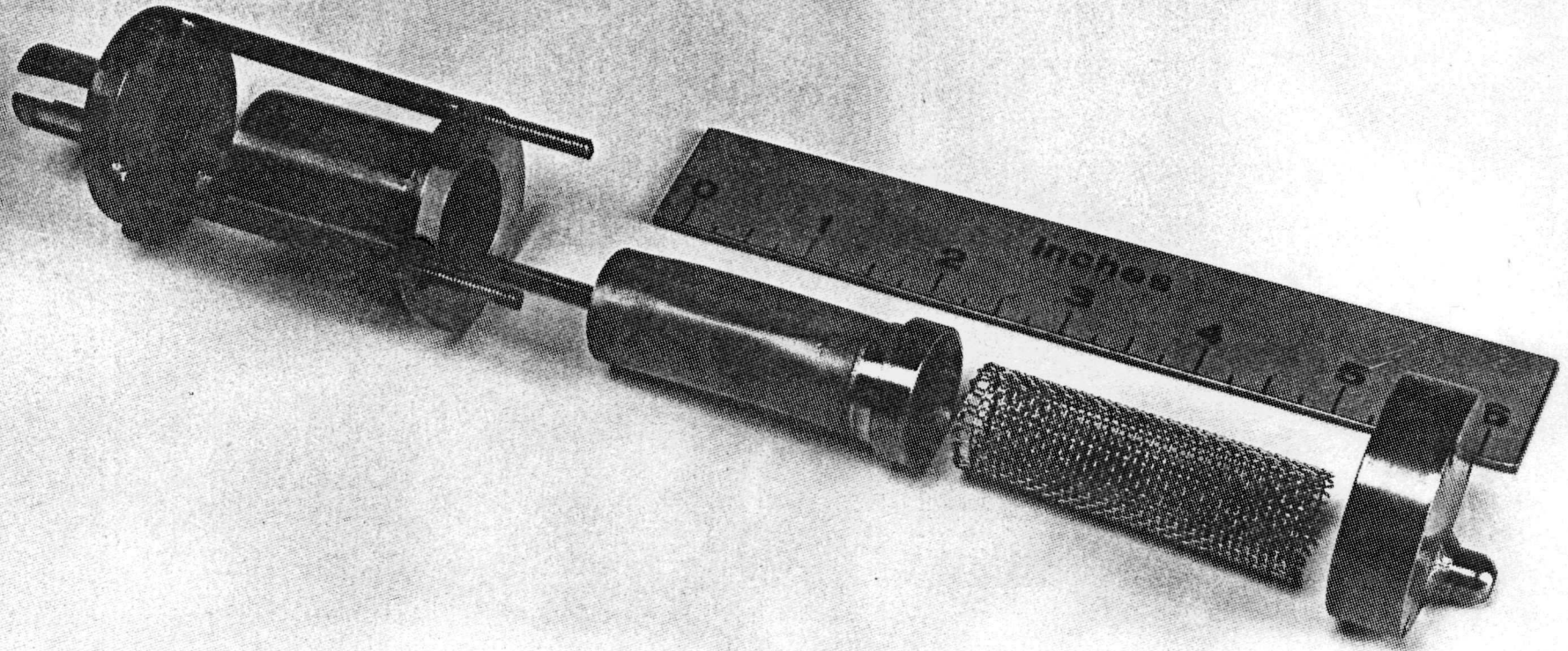


Fig. 12 Multipurpose sampler insert particulate filtration device

and with PTI measurements. During all subsequent reactor operations a comprehensive sodium characterization program similar to the EBR-II work will be conducted.

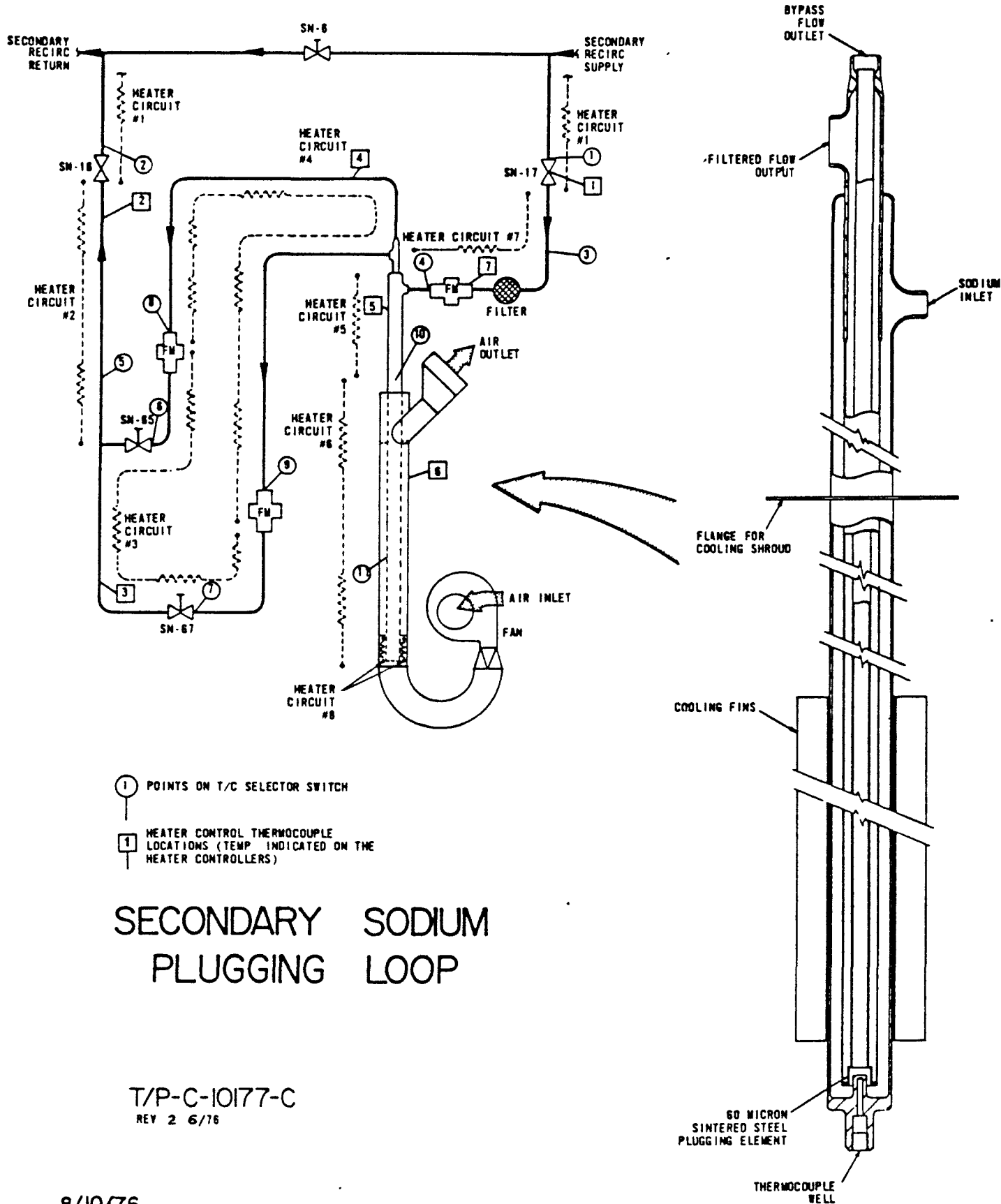
## CONTINUOUS IMPURITY MONITORING IN SODIUM SYSTEMS

### EBR-II

Two types of continuous methods of analysis are done in-situ on EBR-II sodium systems. One uses the non-specific device commonly known as a plugging meter; the other uses meters specific for given impurities. Three such meters have been developed for sodium systems for measuring oxygen,<sup>(21)</sup> hydrogen,<sup>(22)</sup> and carbon:<sup>(23,24)</sup> and have been tested on several loops. EBR-II has been equipped with plugging meters since startup and with oxygen and hydrogen meters in the RSCL since 1972. Starting in 1975, oxygen and hydrogen meters have been used on the EBR-II secondary system for water-sodium leak detection purposes.

Continuous signals from the oxygen and hydrogen meters are monitored and used to detect impurity trends in the primary system or to detect leaks in the steam generator heat exchanger. Monitoring small changes in oxygen and hydrogen during various phases of plant operation has been useful in detecting impurity sources, i.e. water on cladding surfaces and in establishing time limits on cold trap outages for maintenance or replacement.

The plugging meters on EBR-II measure whether impurities are accumulating which could plug flow passages and also provide a guide as to cold trap purification system performance. Recently, a more sensitive and accurate type of plugging meter has been developed and added to the EBR-II systems, shown in Figure 13. This device can measure plugging temperatures as low as 250°F. It operates automatically and



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Fig. 13 EBR-II secondary sodium plugging loop with filter type plugging meter

with little operator attention required. A typical temperature-flow curve is shown in Figure 14.

A continuous monitor for tritium in sodium has recently been developed by ANL<sup>(25,26)</sup> and the unit shown in Figure 15 is now in service on EBR-II. The tritium meter uses a hydrogen diffusion membrane at 900°F with argon sweep gas carrying hydrogen and tritium through a gas flow proportional

#### FFTF

At the present time, the FFTF is equipped only with on-line plugging temperature indicators for continuous sodium purity monitoring. The MPS units, which were designed as samplers, can be used to provide continuous oxygen and hydrogen meter coverage by the addition of an MPS insert containing small O-H sensitive probes. The insert shown in Figure 16 has been designed for reactor use and a hydrogen meter probe has been tested successfully in this insert. Several miniature ceramic-metal sealed oxygen probes have been tested for use in the same insert. Ceramic-metal braze sealed electrodes have been tested in the PAL MPS with varying degrees of success. The unit shown in Figure 17 was produced by GE and has performed well in PAL tests. By the time FFTF starts fueled operations, oxygen and hydrogen meters mounted in MPS units will be in use on the primary sodium systems.

On-line carbon meters are in use on sodium meters in several other countries. The carbon meters tested on sodium systems in the U.S. were found to either lack sensitivity for the concentration expected (UNC diffusion type - methane detector) or to be subject to interference from hydrogen in the sodium (combination diffusion membrane - oxygen emf cell). HEDL has recently developed a meter<sup>(12-16)</sup> using a diffusion membrane of iron-iron oxide, as shown in Figure 18. Helium is used as sweep gas and a helium ionization detector is used to measure the CO<sub>2</sub> produced by

PLUGGING ELEMENT TEMPERATURE (°F)

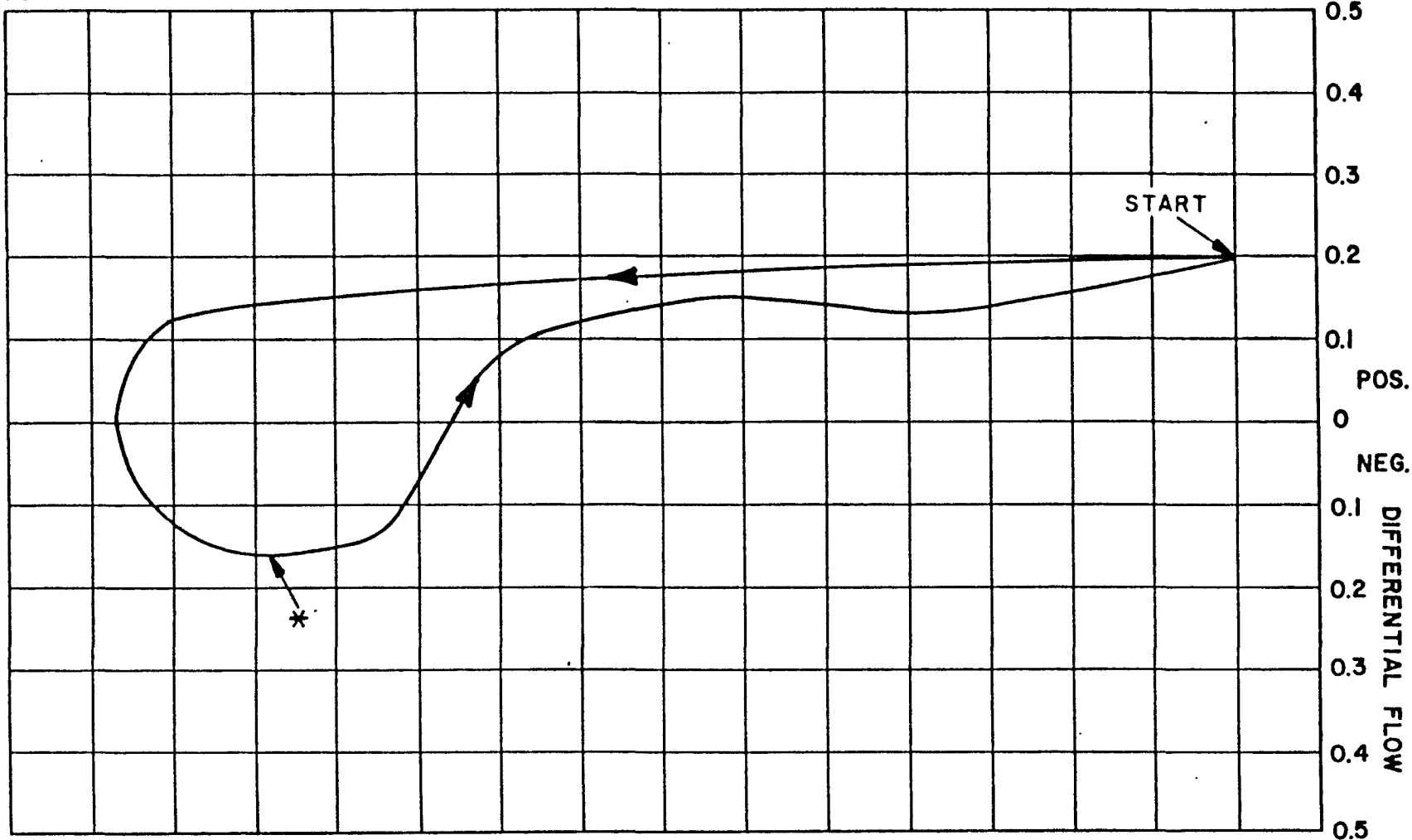
200

300

400

500

600



\* PLUGGING TEMP. = 280°F (The temperature at which the plugging element unplugs has been found to be the most accurate indication of the saturation temperature for a given impurity)

Fig. 14 Typical temperature ramp plugging run with filter type meter T/P-C-10277-A

TEMPERATURE RAMP PLUGGING RUN (X-Y PLOT)

GAS FLOW DIAGRAM FOR TRITIUM METER

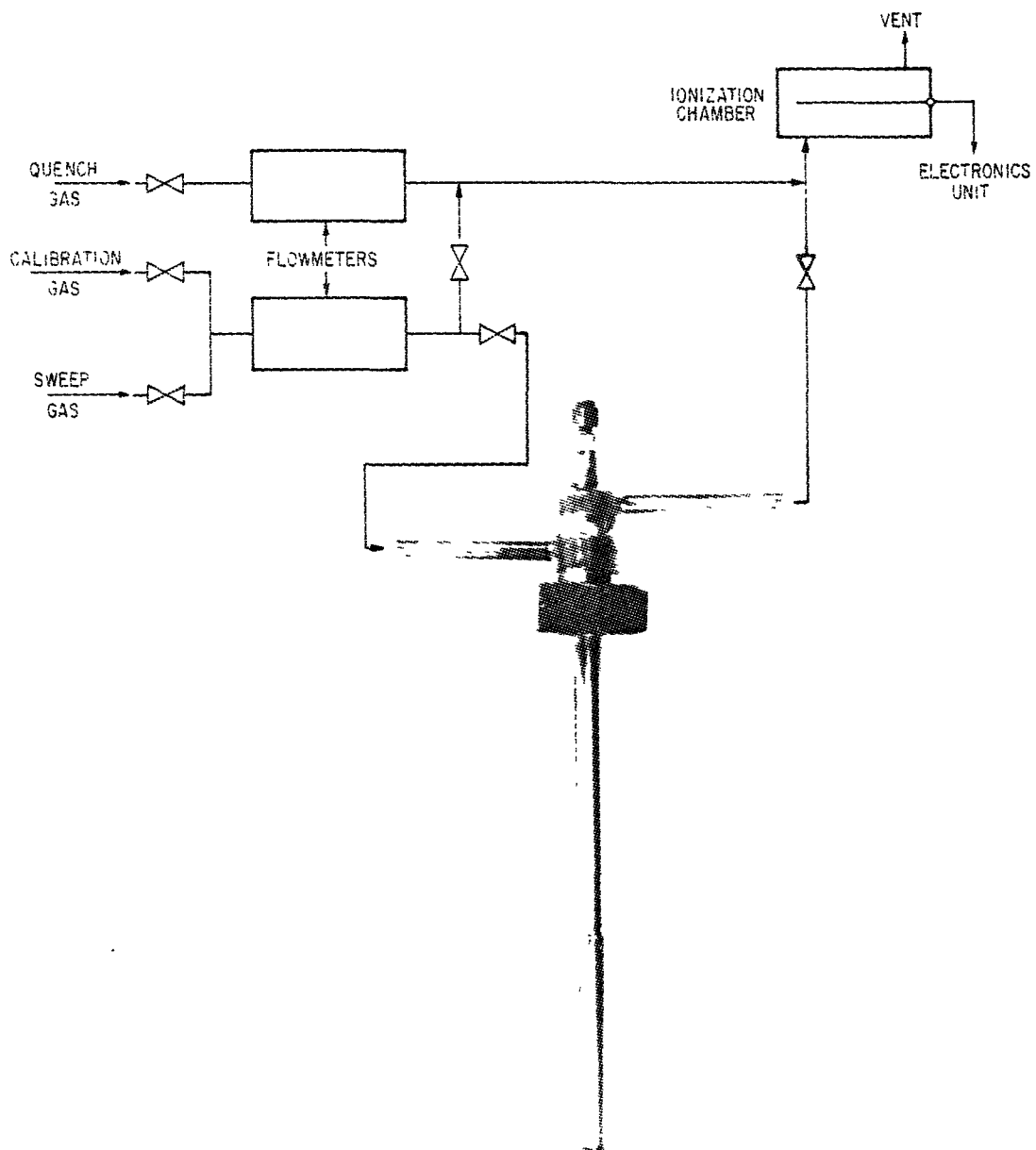


Fig. 15 EBR-II tritium meter and gas flow schematic

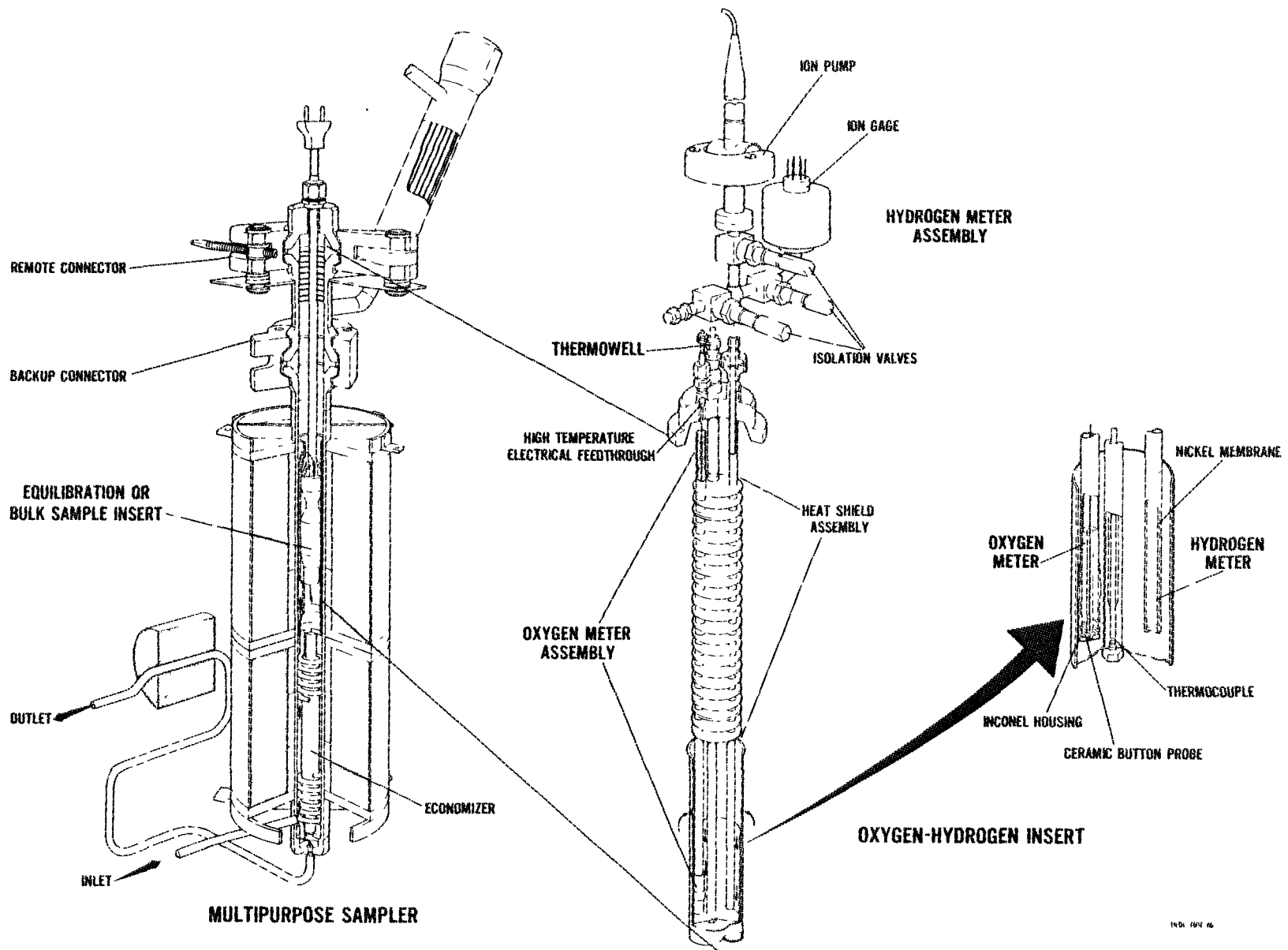
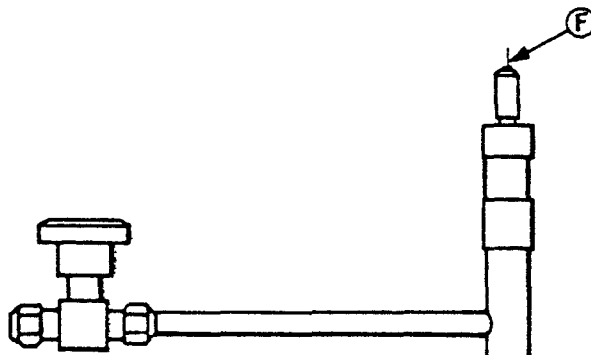


Fig. 16 O<sub>2</sub>-H<sub>2</sub> insert for multipurpose sampler applications

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## DESCRIPTION OF THE EQUIPMENT

Major features of the meter (and the housing supplied as an option) are shown on Figure 1.

- The sensing device (A) is installed in a housing (B) through which passes liquid sodium. The sensing device supports the electrochemical cell structure.
- A Marman flange (C) is provided at the upper end of the sensing device. This flange mates with a corresponding flange which is part of the outer housing (B).
- The outer housing (B) is welded into the outer wall of the pipe or vessel which contains the liquid sodium. The housing positions the sensing device in a fixed location and serves as a flow director for the sodium.
- The key component of the electrochemical cell is the solid electrolyte cup (D). The cup is machined from hi-purity yttria-doped thoria. The lower end (outside) of the cup is in contact with the sodium. The cup is brazed to a holder made from a low-expansion alloy. The holder is welded to the sensing device. (A)
- The inside of the cup is filled with a mixture of a metal (gallium, indium or tin) and the oxide of the metal. This mixture (E) forms the reference electrode of the electrochemical cell. The sodium acts as the other electrode.
- The difference in oxygen activity between the sodium electrode and the reference electrode causes ionic transport of oxygen through the electrolyte cup. This results in an electrical potential.
- The value of this potential is a direct measurement of the oxygen concentration in the sodium. The potential is measured by a voltmeter connected between the pipe wall and an electric lead (F) in contact with the metal-oxide mixture.

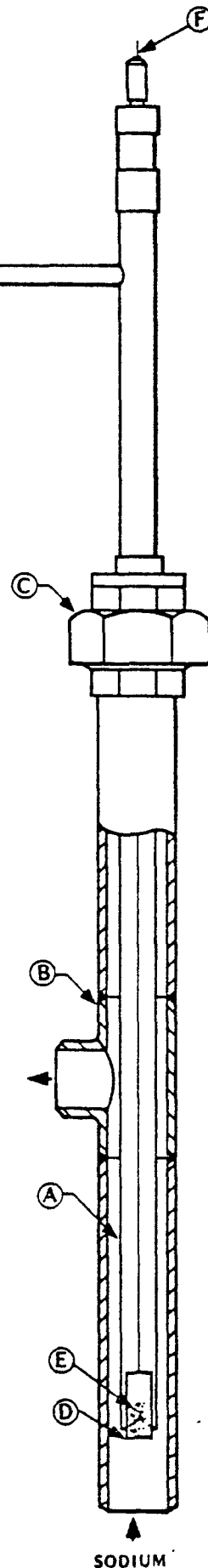
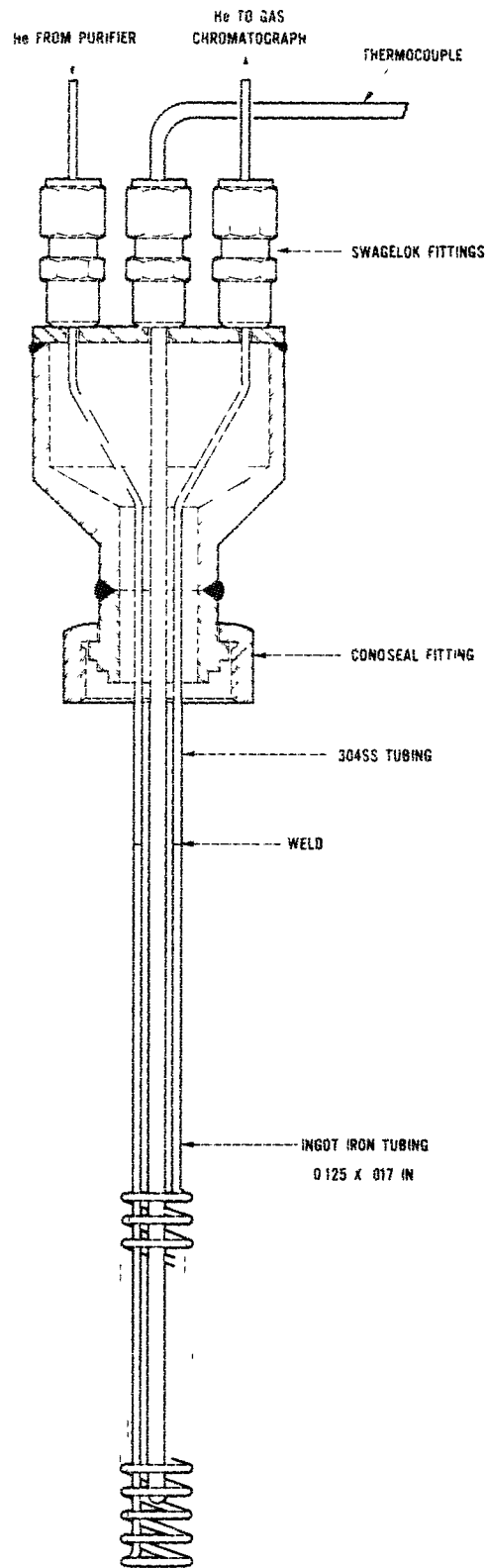


Fig. 17 General Electric type electrochemical oxygen meter

# DIFFUSION CARBON METER PROBE



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Fig. 18 HEDL diffusion carbon meter probe

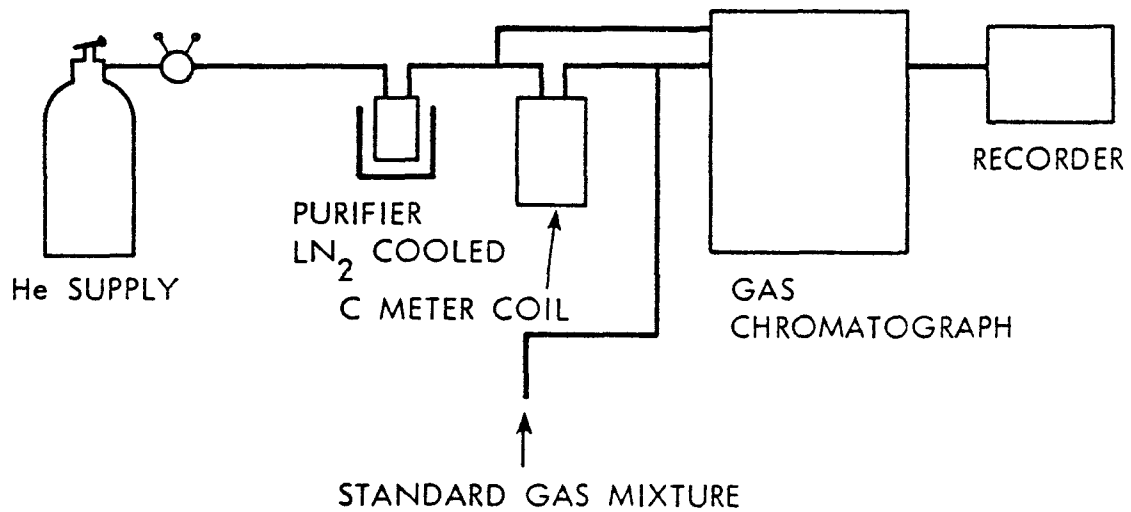
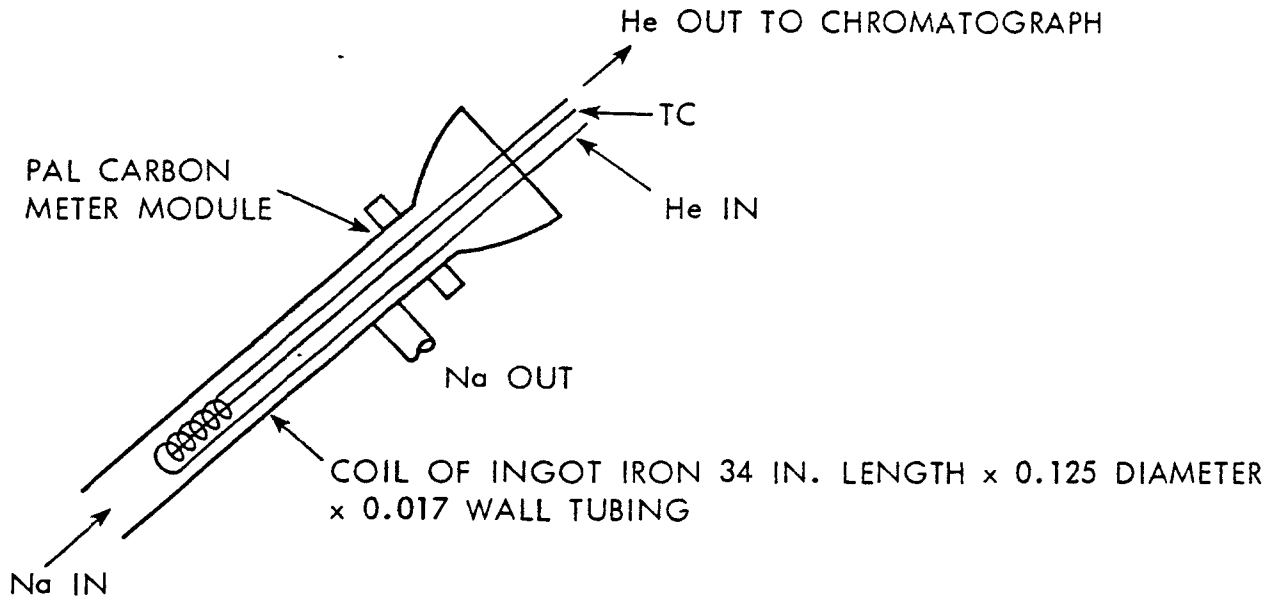
carbon diffusion. The system shown in Figure 19 has been tested on the PAL loop and provides a signal well above background at the  $10^{-3}$  carbon activity level present in stainless systems containing high purity sodium. This meter is being automated by the addition of an integrator and valve control circuitry and will be available for reactor use if needed.

#### PERFORMANCE CAPABILITIES

The samplers on EBR-II provide sodium samples on a routine basis for the laboratory analysis of twelve radionuclides, twenty metallic elements and six non-metallic elements. The overflow cup sampler is used for the majority of sampling since it provides total consumption samples, can provide four samples simultaneously for different analyses and will provide up to 50 g samples if needed. Several of the non-metals are measured by metal equilibration methods which use an equilibration sampler module.

The oxygen and hydrogen meters on the EBR-II primary measure oxygen at 0.5 to 1 ppm level and hydrogen at the 0.05 to 0.1 ppm level during normal reactor operations. During one cold trap outage when the reactor was being refueled, changes as small as 0.2 ppm oxygen and 0.01 ppm of hydrogen were monitored with measured source rates of about 9 grams/day and 0.9 grams/day respectively. Changes in the cover gas system made prior to Run-Beyond-Clad-Breach work have decreased this source rate.

The tritium meter in use on the primary system RSCL and in the O-H module on the secondary purification loop, has been shown to be sensitive to as low as  $0.04 \mu\text{C/gNa}$  with the normal level in primary running about  $50 \mu\text{C/gNa}$ . The meter has a delay time of  $\sim 2$  hours in responding to changes of the tritium level in the plant due to both the volume of tubing from the probe to the counter and to the counting tube volume itself.



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Fig. 19 Carbon meter on PAL - schematic of installation and gas flow equipment

The PTI units originally used on EBR-II were limited to measuring plugging (saturation) temperatures in the range of 270 and above. The new filter type plugging meters will measure plugging temperatures as low as 250°F.

On FFTF the MPS units provide the capabilities of overflow sampling, metal equilibration sampling and particulate filtration sampling in one piece of hardware by utilizing several types of inserts. With the development of the fourth insert containing oxygen and hydrogen meter probes, the MPS can also be used to provide continuous on-line monitoring capability.

The PTI's on FFTF use an orifice plate design and are not capable of measuring plugging temperatures below ~265°F. They are designed to operate automatically in an oscillating mode providing both plugging and unplugging temperature measurements when sufficient material is in solution in the sodium to cause adequate plug formation. This requires at least 1.5-2 ppm oxygen and/or 0.15-0.2 ppm hydrogen. The concentration of other materials which will form plugs at these temperatures, i.e. carbon species, has not been accurately determined.

#### SUMMARY

In the U.S. Breeder Reactor Program, sodium samplers and on-line impurity monitoring devices are used to provide complete sodium characterization data. The EBR-II has had such a program in place for over 10 years following the behavior of all detectable metals, non-metals and radionuclides. FFTF will be equipped with samplers and monitors which will be used to characterize all its sodium systems. This program will start in the summer of 1978 during sodium fill operations and continue for the life of the plant.

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