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TRITIUM MONITORING IN THE GCFR SWEEP GAS FUEL IRRADIATION CAPSULE GB-10*

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Tritium Monitoring in the GCFR Sweep Gas Fuel Irradiation Capsule GB-10, Uri GAT, M. E. PRUITT, A. W. LONGEST, Oak Ridge National Laboratory,* Oak Ridge, Tennessee, and B. D. EPSTEIN, General Atomic Co., San Diego, California -- The release of tritium and its transport pathways were studied in a vented, pressure-equalized fuel rod which simulated a fuel rod in a Gas-Cooled Fast Reactor (GCFR). The purpose was to determine the fraction of total tritium production transported via the various pathways and to determine its chemical form (tritiated hydrogen or water). It was concluded that the fuel rod and its effluent venting lines retained low concentrations of HT (or T_2) and any HTO (or T_2O) present. However, the addition of 1% hydrogen to the helium carrier gas quantitatively eluted the tritium from the charcoal trap integral to the fuel rod and from the effluent lines. The chemical composition of the tritium arriving at the monitoring system could be determined by means of converters which convert HT to HTO and vice versa. HT was the dominant species in the samples measured. Interference from ^{24}Ne (source probably impurities in the fuel) impeded some ionization chamber measurements and necessitated cumulative sampling of tritium collected on a molecular sieve. Only about 10% of the calculated tritium produced by ternary fission was measured. The whereabouts of the other tritium was not determined. Measured venting of tritium was proportional to capsule power. Transmission rates through the integral fuel rod charcoal trap were shown to be temperature dependent.

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INTRODUCTION

The Gas Cooled Fast Reactor (GCFR) has a unique pressure equalized and vented fuel [1,2]. The outer surface of the fuel cladding is roughened to enchance heat transfer. Tritium generated in the GCFR fuel primarily by ternary fission can remain in the fuel region, be vented with the gaseous and volatile fission products, pass through or be trapped in the fuel rod and traps of the system, or diffuse through the 316 SS, 20% cold worked, cladding.

To determine tritium release and venting, its chemical forms (either tritiated water or hydrogen) and transport pathways, the irradiation capsule, GB-10 [3], which simulated a pressure equalized and vented GCFR fuel rod, was modified to enable tritium measurements. The prime purpose of the GB-10 capsule irradiation was to determine fission gas and volatile fission product release and venting fractions and fuel irradiation characteristics under simulated GCFR, controlled conditions.

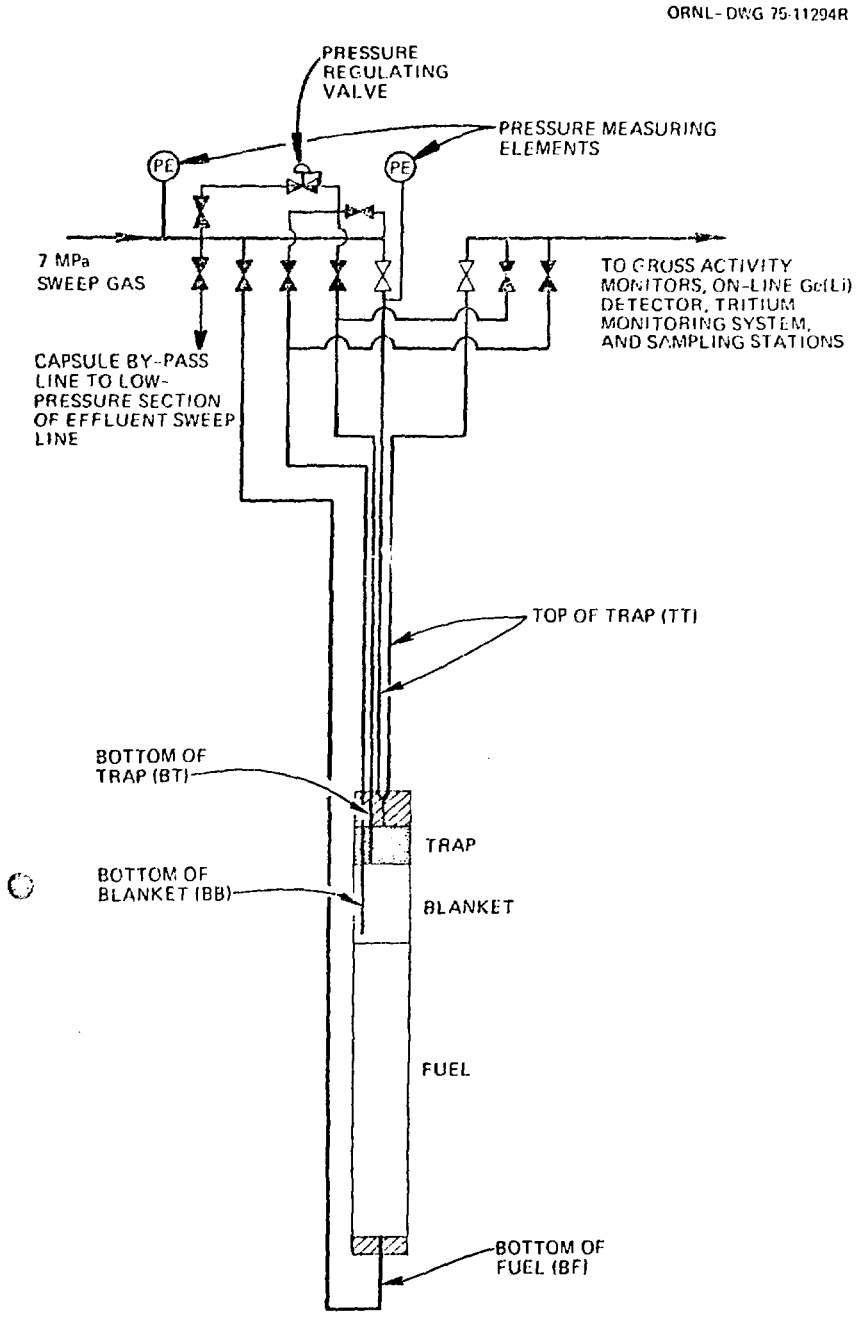
THE GB-10 IRRADIATION CAPSULE

The GB-10 irradiation capsule was designed and constructed to simulate the GCFR pressure equalized and vented fuel and was irradiated in the Oak Ridge Research Reactor. The capsule contained a fuel rod which was comprised of a mixed-oxide fuel, short

upper and lower blankets, and a cladding with an artificially roughened surface of SS-316 20% cold-work 9 mm. A 25-mm-long charcoal trap was incorporated in the cladding above the upper blanket. An arrangement of gas lines enabled the sweeping with gas through various parts of the capsule at will. The line to the bottom of the fuel rod was designated BF; the bottom of the (upper) blanket was named BB; bottom of (charcoal) trap BT; and the two lines across the top of the trap, allowing sweep of the vented gases and volatiles, were each dubbed TT. A sweep mode is characterized by the entrance and exit ports in that order, making a sweep through the entire fuel rod from bottom to top a BF-TT sweep [3]. A schematic of the capsule indicating the various sweep lines is depicted in Fig. 1. The normal sweep gas was pure helium at controlled pressure (up to 7 MPa) and flow-rate (from 2 to 40 ml[STP]/s). A modification allowed the addition of various amounts of hydrogen and tritiated hydrogen to the carrier gas for calibration and to determine tritium transport characteristics of the system.

THE TRITIUM MONITORING SYSTEM

The tritium monitoring system, operated at 70 kPa (gage), Fig. 2, was comprised of in flow sequence, a HTO to HT converter consisting of a Mg



SCHEMATIC OF GCFR-ORR CAPSULE GB-10 SHOWING SWEEP GAS LINES

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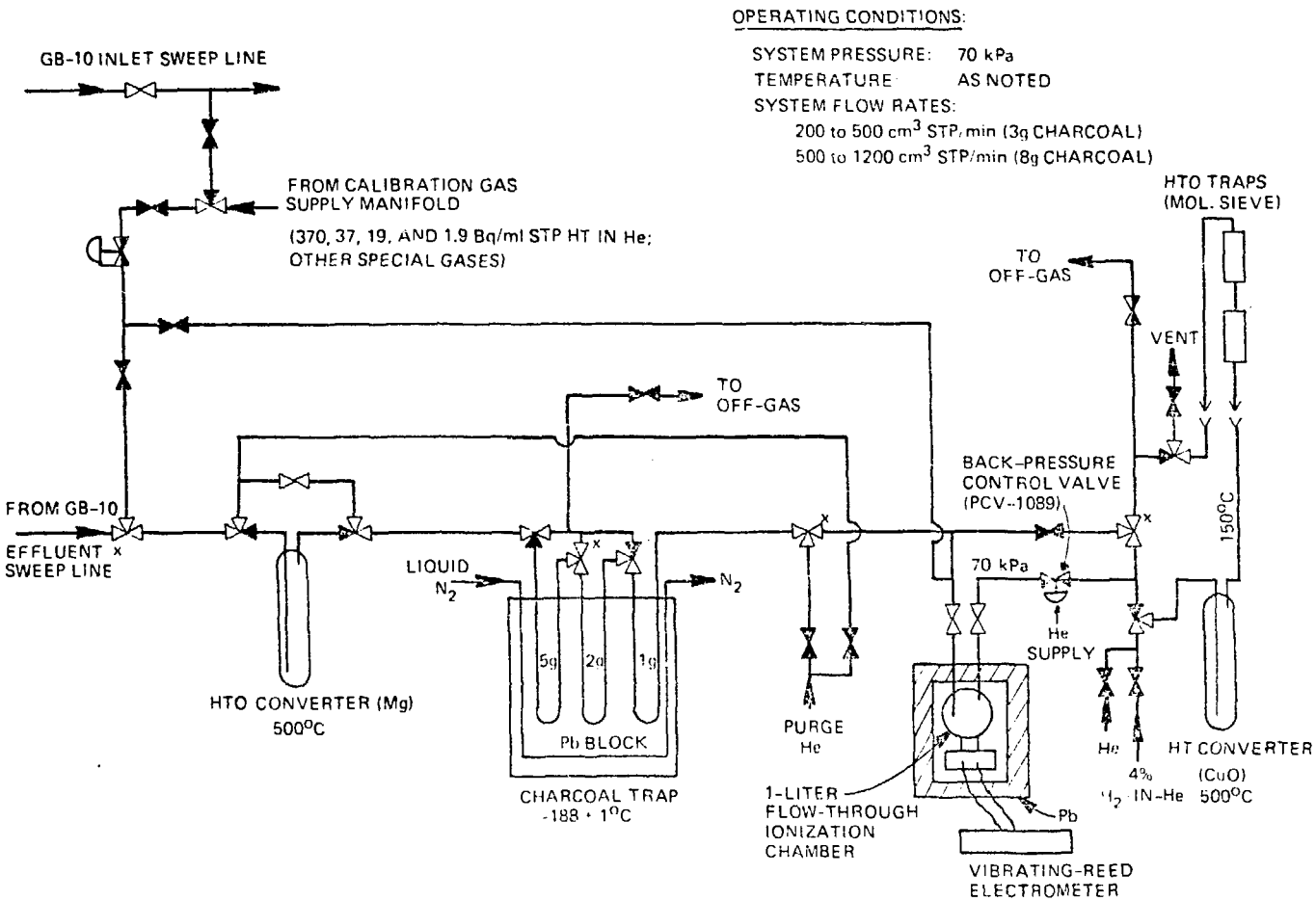

SCHEMATIC OF CAPSULE GB-10 TRITIUM MONITORING SYSTEM

bed operated at 500°C which could be bypassed to determine HT, or inserted in line to determine total tritium (the sum of HT and HTO and, also, if present, any T_2 or T_2O respectively). A liquid-nitrogen-cooled charcoal trap operated at -188 ± 1°C removed gaseous and volatile fission products and tritiated moisture and could be operated at various combinations of 1, 2, and 5 g sections to provide suitable tritium delay times, of typically 12 to 32 minutes, over a range of flow rates. The sweep gas required very clean and dry helium to avoid clogging of the trap. The delay times were temperature sensitive; the 1°C fluctuations could be readily detected on the tritium monitor response. Purge or additional carrier gas could be added at different locations in the lines and was used for cleanup between experiments or for flow enhancement. The tritium monitoring was done in a 1 liter flow-through ionization chamber equipped with a vibrating-reed electrometer. A direct line, bypassing the charcoal trap and HTO converter, permitted the unperturbed flow of calibration gases to the ionization chamber. Following, or bypassing the ionization chamber, the gas could be fed to an HT to HTO converter consisting of a CuO bed operated at 500°C, followed by two 60 ml molecular sieve (4A) HTO traps

arranged in tandem. The moisture trapped on the molecular sieves was eluted and analyzed for tritium, off line, by a liquid scintillation beta counting technique. Finally, effluent gases were released to the GB-10 capsule gas cleanup system. The system components were laboratory tested and calibrated prior to installation.

TRITIUM SYSTEM CALIBRATIONS AND OPERATION

The tritium monitoring system components were assembled, tested, and calibrated in the laboratory [4]. A linear calibration curve was established for the electrometer using calibration gases. The curve was further confirmed by liquid scintillation countings of the molecular sieve samples. Additional in situ tritium calibration runs (utilizing the high purity HT in helium, i.e., a few ppm of hydrogen, bypassing the capsule) yielded essentially complete transmission of the tritium. Tritium concentration in the calibration gases was in the range 1.8 to 350 kBq/l (STP). The same calibration gas in the TT-TT flow mode measured about 10% of the tritium injected, and in the BT-TT, that is, flow through the fuel rod charcoal trap, measured less than 1% of injected tritium [5]. Helium carrier gas containing tritium and 1% hydrogen flowing through the system except through the fuel or blanket regions of the fuel rod yielded

tritium quantitatively. To preserve the fuel rod flow constriction, observed to develop with increasing fuel burnup, for the postirradiation examination no attempt was made to flow any hydrogen through the fuel or blanket regions. When flowing through the HTO converter, tritium was retained when no hydrogen was added to the carrier gas. The hydrogen laden helium carrier gas would also release tritium trapped in the system in preceding experiments.

A rather severe interference with the ionization chamber measurements of tritium was encountered, when sweeping the fuel, by the presence of 3.38-minute-half-life ^{24}Ne in the gas and the buildup of its daughter product ^{24}Na in the ionization chamber. The neon passes through the cryogenic charcoal traps faster than the tritium. The neon source is attributed to activation of magnesium impurities in the fuel. To supplement ionization chamber readings, molecular sieve sample measurements were made.

Response of the electrometer connected to the ion chamber showed an initial peak which then decreased toward a steady state reading of approximately 90% of the maximum. This phenomena occurred in the absence of hydrogen addition to the sweep

gas. The steady state value was used in establishing the calibration curve in high purity helium. The addition of hydrogen to the carrier gas eliminated the peaking phenomenon. A small rate of flow response dependence, observed in laboratory calibrations, was also eliminated with the addition of hydrogen. A second calibration curve was determined for the tritium in helium containing 1% hydrogen at actual in situ conditio and it was confirmed by molecular sieve sample measurements.

EXPERIMENTS AND RESULTS

Selected experiments, from a larger series of experiments [5], are summarized in Table I. A flow restriction in the fuel rod [3,6] necessitated the use of a technique employing simultaneous flow through the capsule and capsule bypass flow to maintain practical breakthrough times in the tritium monitoring system. Experiment 9, devised to clean up the HTO converter of previous tritium depositions, swept out tritium, deposited in previous experiments which had no hydrogen added to the sweep gas, and thus demonstrated the sensitivity of the tritium monitoring system to hydrogen concentrations below 1%. It was concluded, somewhat arbitrarily, that a hydrogen concentration of 1% or higher in helium sweeps the tritium quantitatively

Table I. Selected GB-10 tritium monitoring experiments

Experiment No.		Major purpose	Fuel-rod power kW/m	Sweep gas	Flow rate[a] ml[STP]/min	Flow mode	HTO converter	Tritium concentration E - electrometer M - molecular sieve	Comments
9	a.	clean up accumulated tritium in the system	n.a.	He + 1% H_2	200	By-pass	in	varying	Tritium calculated to approximate earlier deposition in monitoring system
10	a.	determine tritium (HT) in capsule sweep gas	39.4	He[b]	1200[b]	BF-BT	out	382 Bq/min[M]	Approximately 10% of calculated tritium detected
	b.	determine total tritium (HT + HTO)					in	not determined	
11	a.	recalibrate	n.a.	He + HT[c]	1200[c]	By pass	in	~20 kBq/l [E]	Recalibration by-pass determined HT decrease in bottle with time
	b.	examine trans-	39.4			TT-TT	in	increasing drift	Determined line dependency
	d.	mission line	39.4	He + HT[c]		BT-TT	in	decreasing drift ~0.2 kBq/l [E]	of system absorptions
	e.	dependency	39.4			TT-BT	in	decreasing drift ~2 kBq/l [E]	
13	a.	remove tritium				TT-TT	in	varying	Hydrogen proved effective in
	b.	accumulations	48.6	He + 1% H_2	1248	TT-BT	in	varying	sweeping accumulated tritium
14	a.	measure tritium release	48.6	He + 1% H_2	1200	TT-BT	in and out	3.6 kBq/min (E+M)	Tritium release was predominantly HT.
	b.	measure power dependency	0				in	~0	Tritium release correlated well with power
15	c.	tritium transmission and chemistry	48.6	He + 1% H_2 + HT	1200	TT-BT	in and out	~ as calibrated	Tritium transmitted completely, chemically unaltered
	e.	power dependency	48.6 24.3	He + 1% H_2	1200	TT-BT	in	~4 kBq/l [E] ~2 kBq/l [E]	Proportionality of tritium and power
	f.	charcoal trap	0.	He + 1% H_2	1200	TT-BT	in	various [E]	charcoal trap temperature determining tritium delay

a. Total flow rate in the tritium monitoring system.

b. Combination of 745 ml/min through the capsule 155 ml/min capsule by-pass and 300 ml/min He + 4% H_2 added in tritium monitoring system.

c. Calibration gas at 900 ml/min and 4% H_2 in He at 300 ml/min.

through the monitoring system. Experiment 10, sweeping helium without hydrogen through the fuel and blanket region of the fuel rod but adding hydrogen upstream of the monitoring system, resulted in the measurement of a small fraction of about 10% of the calculated ternary fission tritium as HT in molecular sieve samples. The ion chamber could not be used for these measurements because of overwhelming ^{24}Ne signals.

The gas bottle tritium recalibration, performed as the first part of experiment 11, showed that the tritium concentration in this particular calibration gas bottle decreased with time and reached about 50% of initial value of 38 kBq/l a few months earlier. The reason for this change is not known. Experiment 11 was planned to determine tritium losses in the lines by flowing the same route through a fuel rod area in two directions. The BT-TT and TT-BT flow modes were chosen thus flowing through the fuel rod charcoal trap in two directions. The measurements showed that readings of tritium varied with time, slowly decreasing over a period of hours, in the BT-TT and TT-BT flow modes. A similar experiment in the TT-TT flow mode resulted in readings which slowly increased with time. The reason for these slow drifts was not determined. Final results showed a transmission

rate of 1 to 10% of injected tritium with the TT-BT flow mode tritium transmission rate being almost an order of magnitude higher than the rate in the BT-TT flow mode. The BT line was considered to be the cleaner line since it was rarely used. It was concluded that tritium could not be quantitatively transported in the GB-10 capsule to the monitoring system unless significant amounts of hydrogen are added to the helium sweep gas. Neither tritium nor hydrogen were ever injected to the fuel or blanket regions of the fuel rod because the preservation of the chemical state of the fuel rod relative to flow blockage for the post irradiation examination was considered more important. A plan to inject a balanced H_2/H_2O ratio that would not upset the fuel oxygen potential was cut short by the completion of the capsule irradiation. Subsequent experiments did not involve sweeping through the fuel or the blanket areas and utilized helium sweep gas containing 1% hydrogen.

Experiment 13 was designed to elute accumulated tritium by flowing 1% hydrogen in helium in the TT-TT and TT-BT flow modes. The tritium indication in either flow mode varied, decreasing with time as the previously accumulated tritium was eluted. It was also observed that the ^{24}Ne was comparatively low and that essentially all of the

tritium appeared as HT. The impact of hydrogen addition on the chemical form of the tritium was not determined. Experiment 14, a continuation of experiment 13 and using 1% H_2 in He, determined the tritium release in the TT-BT flow mode and capsule power of 48.6 kW/m to be 3.6 kBq/min and to be predominantly HT. The experiment continued with the rod power reduced to zero. As a result of the power reduction the fuel rod charcoal trap temperature decreased from 300 to 250°C. In the final experiment, number 15, tritium calibration gas with 1% hydrogen in helium, was passed through the TT-BT pathway. It was shown that tritium was transferred quantitatively through the fuel rod trap and that no detectable change in chemical composition occurred during the passage. In the last parts of this experiment, for which the gas was changed to 1% H_2 in helium a qualitatively proportional correlation between capsule power and tritium release was determined. A positive correlation between the charcoal trap temperature and the tritium released was also established.

Related experiments on tritium transport in GCFR fuel are being carried out in the Helium Loop Mol [7].

CONCLUSIONS

The major accomplishments of the experiment are the useful contributions to tritium technology and future tritium transport experiments. The following conclusions were made.

1. Accumulations of tritium over selected time periods can be measured by elution of tritium by the addition of H_2 to the carrier gas. The addition of 1% H_2 to He was found to be adequate in these experiments to recover quantitatively the tritium previously accumulated in the components of both the capsule and the tritium monitor.
2. The addition of H_2 to the sweep gas is necessary for quantitative conversion of HTO to HT in the Mg converter and for its transmission through the converter.
3. The use of a Mg converter to convert HTO to HT enables the determination of the tritiated chemical composition. However, the impact, if any, of the excess hydrogen on the chemical form of the tritium was not determined.
4. Cryogenic charcoal beds have been shown to be useful in separation of tritium from other radioisotopes except for Ne.
5. The ^{24}Ne, presumed to originate in the fuel, was identified from its decay curve and that

of its daughter product ^{24}Na. The ^{24}Ne decay signal in the ion chamber interferes with that of the tritium, and completely masking the tritium signals when flowing through the blanket and fuel regions of GB-10.

6. The initial goal of measurement of transport of tritium in the GB-10 fuel rod, which simulated GCFR pressure equalized and vented fuel, was not fully accomplished. The desire not to alter in any way the chemical or physical properties of the fuel rod, which had developed a flow restriction, was overriding. Only about 10% ofcalculated ternary fission-tritium (based in 0.001 fission yield) was measured. The whereabouts of the other tritium was not determined. Nevertheless, the accounted tritium was proportional to capsule power. The tritium accounted for was also positively correlated with the fuel rod integral-charcoal-trap temperature.

7. Tritium concentrations can be measured to below 1 Bq/l (STP) (1 fmol/l) in nuclear fuel helium sweep gas utilizing samples accumulated by the molecular sieve. The on-line ionization chamber can be used to below 100 Bq/l in the absence of interference from ^{24}Ne.

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