

MASTER

**Detection of Hydrogen Buildup in Initially
Pure Nonhydrogenous Liquids**

S. R. McNeany
J. D. Jenkins

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION · FOR THE DEPARTMENT OF ENERGY

ORNL/TM-6466
Distribution
Category UC-77

Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION
ENGINEERING TECHNOLOGY DIVISION

HTGR BASE TECHNOLOGY PROGRAM

HTGR Fuel Recycle Development Program
(189a OH045)

DETECTION OF HYDROGEN BUILDUP IN INITIALLY
PURE NONHYDROGENOUS LIQUIDS

S. R. McNeany and J. D. Jenkins

Date Published: December 1978

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

NOTICE This document contains information of a preliminary nature. It is subject to revision or correction and therefore does not represent a final report.

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37830
operated by
UNION CARBIDE CORPORATION
for the
DEPARTMENT OF ENERGY

CONTENTS

ABSTRACT	1
INTRODUCTION	1
HYDROGEN DETECTION PRINCIPLE	4
Analytical Model of a Hydrogen Monitor	5
Numerical Model of a Hydrogen Monitor	7
DESCRIPTION OF EXPERIMENTAL DEVICE	10
EXPERIMENTAL RESULTS	12
DISCUSSION	17
CONCLUSIONS AND RECOMMENDATIONS	18
ACKNOWLEDGMENTS	19
REFERENCES	19

DETECTION OF HYDROGEN BUILDUP IN INITIALLY
PURE NONHYDROGENOUS LIQUIDS

S. R. McNeany* and J. D. Jenkins*

ABSTRACT

A technique for monitoring hydrogen buildup in initially pure nonhydrogenous liquids is described in this report. The detection method is based upon the neutron-moderating properties of hydrogen. The analysis leading to the selection and design of a hydrogen-monitoring device is described. An experimental mockup of the device was then constructed and tested for hydrogen sensitivity. A hot cell was used for these tests.

A device proved capable of measuring hydrogen concentrations in the range of 0 to 13.0×10^{27} atoms/m³, with an accuracy of about 1.0×10^{27} atoms/m³. A typical measurement can be made in 3 to 5 min. The experimental results confirmed the sensitivities predicted by the analysis and demonstrated that such a device would be practical for hydrogen concentration measurements for criticality control in an HTGR fuel refabrication plant.

INTRODUCTION

Fuel-handling equipment in nuclear-fuel processing facilities is often designed to be critically safe, under the assumption that no hydrogenous material will be mixed with the fuel. Hydrogen is a highly effective neutron moderator that can severely reduce the critical mass of nonhydrogenous systems. If it can be shown that mixing of fuel and other hydrogenous sources is highly unlikely, then equipment can be designed for higher throughputs and improved economic efficiency.

In the High-Temperature Gas-Cooled Reactor (HTGR) Fuel Recycle Program, in several cases designs are based on the assumption that hydrogenous materials will not be mixed with the fuel. Two of these involve fluidized-bed furnaces¹ (see Fig. 1) for carbonizing and coating fuel-bearing microspheres.

*Engineering Technology Division.

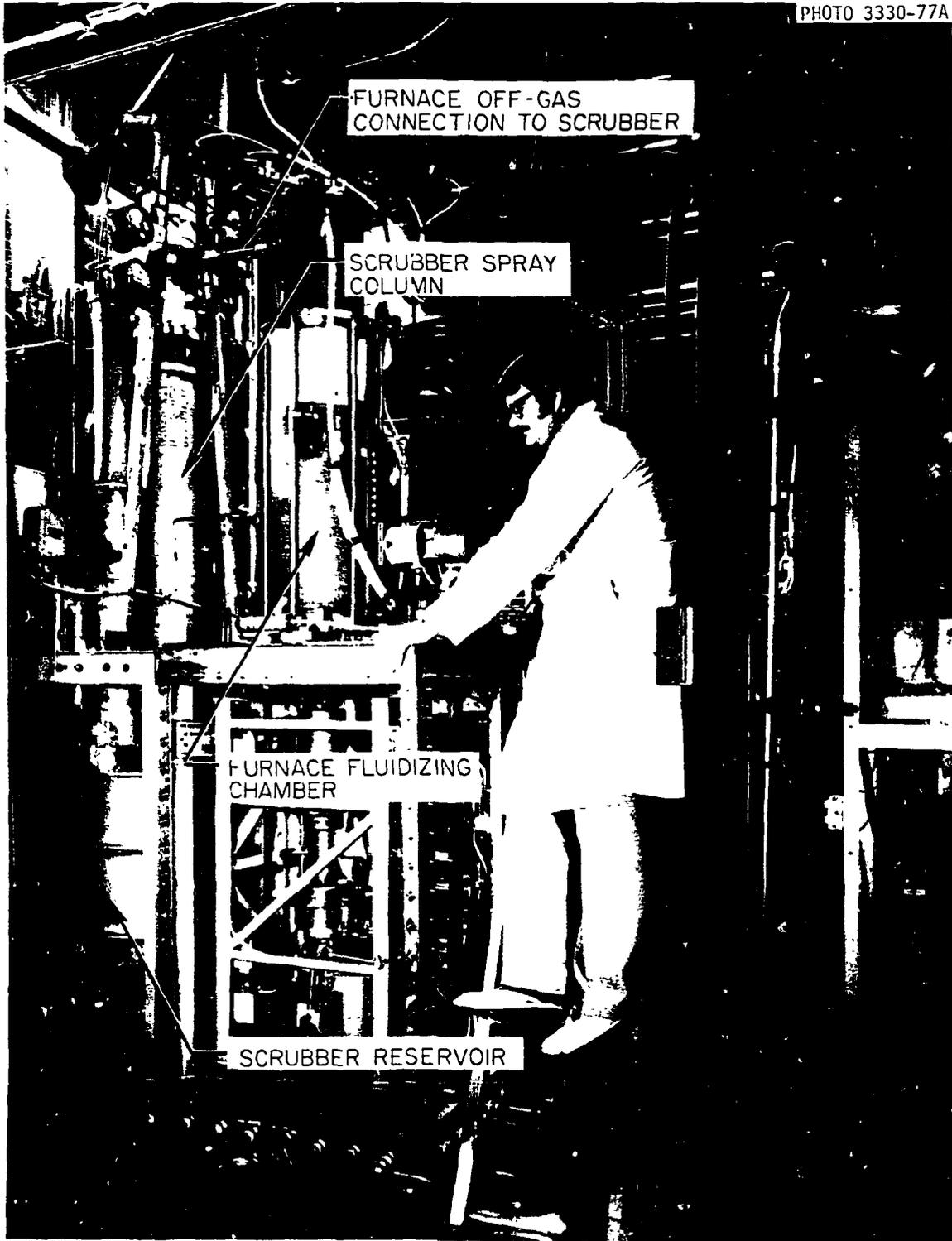


Fig. 1. HTGR Microsphere Processing Furnace.

Immediately adjacent to each furnace is an off-gas scrubber column that removes soot and condensable hydrocarbons from the furnace effluent. Most systems of this type employ water or a petroleum-based solvent such as kerosene as the scrubber liquid. Because a possible blockage can cause the liquid to overflow back into the furnace and cause a criticality accident, a nonhydrogenous liquid is desirable. For the HTGR process equipment, perchloroethylene, (C_2Cl_4), commonly called "perc," was selected as the scrubber solvent.¹

Substitution of "perc" for kerosene as the scrubbing medium would be a simple solution to criticality concerns were it not for the fact that some hydrogenous material from the off-gas dissolves in the "perc" and accumulates in the scrubber reservoir. Consequently, a monitor is required to measure the hydrogen buildup in the "perc" reservoir and to sound a warning when hydrogen levels are too high. A criticality study² was performed to determine how much hydrogen could be allowed in the scrubbers used in the HTGR program before becoming a hazard. The results of the study indicate that the amount varies considerably depending on factors such as the amount of fissile material in the furnace, the chemical composition and density of the fissile material, the quantity of residual water accompanying the fissile material, and whether the hydrogen in the scrubber is soluble or insoluble in "perc." In some highly unlikely, but crucial furnace conditions, as little as 3.78 liters (1 gallon) of water in the scrubber could cause a criticality accident. In this case water, which is insoluble in "perc," would be floating on top of the "perc" reservoir with the potential of being introduced into the furnace by itself. A hydrogen monitor, sampling from the "perc" surface, could detect the presence of water. When hydrogenous material is dissolved in the "perc," much more of it can be tolerated. Again, a monitor sampling from anywhere in the reservoir can easily detect unsafe levels of hydrogen — that is, more than

$$\left(\frac{20 \text{ Kg (H)}}{\text{m}^3}\right) \text{ or } \left(\frac{1.2 \times 10^{28} \text{ atoms H}}{\text{m}^3}\right) .$$

This report describes one possible technique for hydrogen monitoring.

HYDROGEN DETECTION PRINCIPLE

Hydrogen atoms, due to their low mass, very effectively slow down fast neutrons. A hydrogen atom can accept all of a neutron's kinetic energy in one head-on collision. No other element has this property. Thus, if fast neutrons impinge on an unknown sample, the speed of the transmitted neutrons can be used to indicate the hydrogen content in that sample.

For example, consider a spherical void around a point source of fast neutrons. Enclose this void in a spherical neutron detector shell in which slow neutrons are detected more readily than fast neutrons. If the detector shell is made thin enough so that only a small fraction of the neutrons incident upon it are absorbed, while the rest are transmitted, and if the shell is surrounded by an infinite void, the result is a monitor that can sense the presence of hydrogen atoms introduced to the central void.

To understand how the monitor operates, consider Eq. (1), which gives a neutron balance for the absorber shell:

$$\left[\begin{array}{c} \text{Rate of Neutrons} \\ \text{Entering Shell} \end{array} \right] = \left[\begin{array}{c} \text{Rate of Neutron} \\ \text{Absorption in Shell} \end{array} \right] + \left[\begin{array}{c} \text{Rate of Neutron} \\ \text{Transmission Through Shell} \end{array} \right] . \quad (1)$$

If no neutrons are absorbed in the central void region, then the rate of neutrons entering the shell is a constant equal to the neutron-emission rate of the point source. This is true also if nonabsorbing neutron moderators are present in the central region. However, the neutron absorption rate in the shell depends on the velocity of the neutrons entering it. Therefore, as the density of moderators increases in the central region, the neutron absorption rate in the shell increases as well. Thus, the absorption rate counter can be calibrated to indicate neutron moderator density in the central region.

This hypothetical situation is closely approximated when perchloroethylene (C_2Cl_4) replaces the central void and an adjacent BF_3 neutron

detector replaces the absorbing shell. This was the experimental arrangement used to validate the technique. A detailed discussion of the experiment is given later.

Analytical Model of a Hydrogen Monitor

The sensitivity of such a technique to variations in the hydrogen density can be estimated by using an approximation to the Boltzmann equation, which yields the neutron spectrum in a finite medium with hydrogen scattering and no absorption.

It is well-known³ that the collision density of neutrons in an infinite medium containing only hydrogen can be represented by the equation

$$F(E) = \frac{Q}{E_0} + \int_E^{E_0} \frac{F(E')}{E'} dE', \quad (2)$$

where $F(E) = \Sigma_S(E)\phi(E)$; the number of neutrons colliding per unit volume per unit time at energy E , and $Q =$ the neutron source strength at energy E_0 . For a finite system, the collision density must be reduced by the number of neutrons that escape the medium and hence, do not collide. These can be accounted for by including a "nonescape probability" term, P_S , in Eq. (2), where P_S is given by the Wigner rational approximation:⁴

$$P_S = \frac{\Sigma_S \bar{\lambda}}{1 + \Sigma_S \bar{\lambda}}, \quad (3)$$

and $\bar{\lambda}$ is taken as the mean distance from point of birth of the neutron at energy E to the point where it escapes from the medium. In this case $\bar{\lambda}$ can be approximated by one half the mean cord length of the medium,⁵ since the neutron-scattering events occur randomly within the medium and the neutrons must traverse, on the average, one half of a cord to escape. Thus, for an infinite cylinder

$$\bar{\lambda} = \frac{1}{2} \frac{4V}{S} = R, \quad (4)$$

where

V = the volume of the medium ($\pi R^2 L$) and

S = the surface area of the medium ($2\pi RL$).

With these approximations, the equation for the collision density becomes

$$F(E) = \frac{QP_s}{E_0} + \int_E^{E_0} \frac{F(E')P_s}{E'} dE', \quad (5)$$

with solution

$$F(E) = F(E_0) \left(\frac{E_0}{E} \right)^{P_s}; \quad F(E_0) = \frac{QP_s}{E_0}. \quad (6)$$

Since the neutron flux is given by

$$\phi(E) = \frac{F(E)}{\Sigma_s}, \quad (7)$$

and the flux per unit lethargy is related to flux per unit energy by

$$\phi(u) = E\phi(E), \quad \text{and that } u = \ln \frac{E_0}{E}, \quad (8)$$

one obtains as a final solution

$$\phi(u) = \frac{QR}{1 + \Sigma_s R} \exp[-u/(1 + \Sigma_s R)]. \quad (9)$$

To apply this solution to an actual situation, the container is assumed to be a 0.152-m-diam (6-in.) cylinder filled with hydrogen corresponding to 0-10% kerosene in "perc." Also, a hydrogen-scattering cross section of $2.0 \times 10^{-27} \text{m}^2$ is assumed. The detector

cross section is assumed to be $1/v$. With these assumptions, the reaction rate in a cadmium-wrapped detector between 10 MeV and 0.55 eV (cadmium cutoff) can be expressed as

$$\text{Reaction Rate} = K \frac{R}{1 + \Sigma_s R} \int_0^{16.72} \exp\left[\left(\frac{1}{2} - \frac{1}{1 + \Sigma_s R}\right)u\right] du . \quad (10)$$

Substituting the numerical values given above yields the relative reaction rates shown in Table 1.

This simple analysis indicates that the proposed method for the detection of hydrogen in nonhydrogenous liquids should have sufficient sensitivity to discriminate between concentrations in the range of interest, provided background counts from room-return neutrons can be held at a sufficiently low level.

Table 1. Calculated Response of a $1/v$ Detector to Hydrogen Additions in Perchloroethylene

Kerosene Content of Sample (wt %)	Hydrogen Atom Density (10^{27} atom/m ³)	Detector Reaction Rate (Relative)
0	0	7.6
2	2.51	12.0
4	4.94	21.2
6	7.28	37.5
8	9.54	61.5
10	11.72	92.1

Numerical Model of a Hydrogen Monitor

As explained in the previous section, the transport of neutrons from a central source through a sample region and into the surrounding environment is governed by the Boltzmann equation. In its differential form

this equation is exact, but in general, it cannot be solved analytically. However, many computer codes have been written to solve this equation by numerical techniques. One such code is YSDRNPM, which is part of the AMPX code system.⁶ We used it to study a one-dimensional model of a hydrogen monitor and to assist in the design of an experiment to measure hydrogen concentrations.

Figure 2 illustrates the geometry of the calculational model. It resembles the example proposed earlier in this section, except that here a thick concrete shell simulates the neutron reflection properties of shield walls. In addition a thin cadmium filter is wrapped around the "perc"-hydrogen mixture to remove all well-thermalized neutrons that are reflected back toward the "perc" from the concrete walls. This model only roughly approximates a real application. For instance, the model shows the monitor to be in the center of a 5.2-m-diam (17-ft) spherical concrete shell, when in reality it would probably be off-center in a rectangular room. However, the model is a useful tool for estimating the relative sensitivity to hydrogen and the effects of detector position.

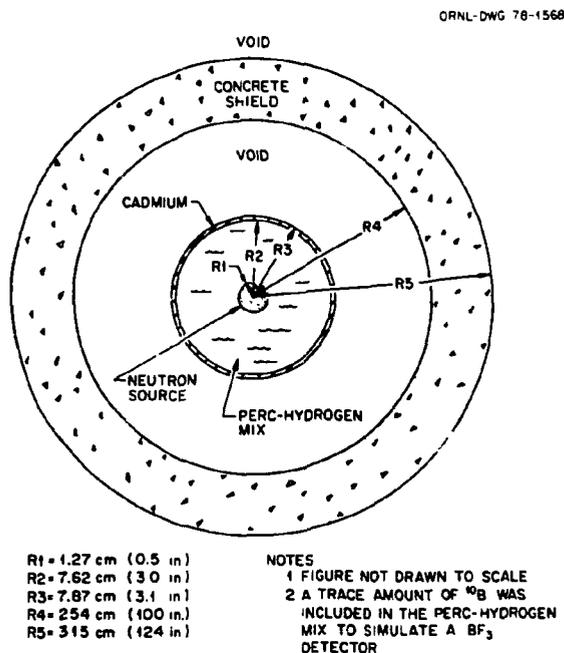


Fig. 2. XSDRNPM Model of Hydrogen Monitor.

A trace amount of ^{10}B is dispersed throughout the "perc"-hydrogen mixture so that detector activity can be calculated as a function of position. The hydrogen concentration is varied by varying the amount of kerosene in a "perc"-kerosene mixture. The results of the calculations are shown in Table 2 as ^{10}B absorption activity at various positions for various "perc"-kerosene mixtures.

Four observations can be made about the data in the table. The first is that activity varies inversely with the square of the distance from the source. So, if the distance between the detector and the source is doubled, the detector count rate will drop by a factor of four. Secondly, the monitor appears rather highly sensitive to the hydrogen content of the "perc"-kerosene mixture. That is, a detector's count rate increases rapidly as the density of hydrogen in a sample increases. However, because neutron reflection from the walls of a real hot cell may differ significantly from that in this model, the best estimate of hydrogen sensitivity is given by the experiment described later. Thirdly,

Table 2. Calculated ^{10}B Absorption Activity for One Source Neutron per s and One ^{10}B Atom per m^3

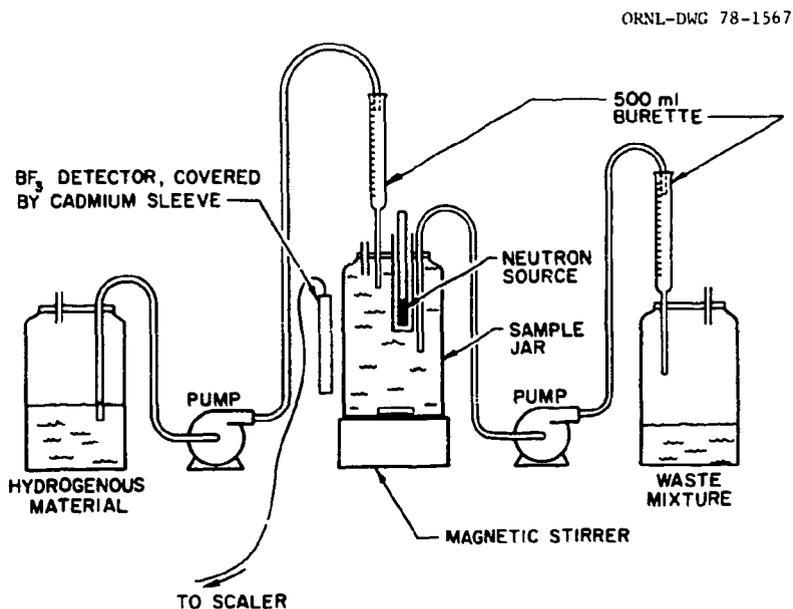
Kerosene Content of Sample (wt %)	Hydrogen Atom Density (atom/ m^3)	^{10}B Absorption Activity, absorptions/s m^3 , at Each Distance from Source Center to Detection Point (mm)		
		17	48	72
0	0×10^{27}	31×10^{-28}	4.2×10^{-28}	2.1×10^{-28}
2	2.51	34	5.9	2.9
4	4.94	39	9.3	4.6
6	7.28	49	16	7.5
8	9.54	64	27	12
10	11.72	87	42	19

the monitor is more sensitive to high than to low levels of hydrogen. Finally, based on these data, the optimum location for a detector is at a point 48.0 mm from the center of the neutron source.

From these calculations, a bench-scale mockup of a hydrogen monitor was designed and constructed to experimentally determine its hydrogen sensitivity in a hot-cell environment. The following sections describe the equipment and give the results of the measurements.

DESCRIPTION OF EXPERIMENTAL DEVICE

Based on the calculated result that a 152 mm-diam (6.0-in.) sample container gives good hydrogen sensitivity, an experimental device was designed from a standard 3.78-liter (1-gallon) sample jar, which was approximately 150 mm in diameter. Figure 3 shows how the apparatus was arranged. In the center is the sample jar, which was initially filled with pure "perc" and mounted on a magnetic stirrer. Through the center



NOTES

1. ALL JARS ARE 1 GALLON CAPACITY
2. ALL TUBING IS COPPER
3. SAMPLE JAR SEPARATED FROM OTHERS BY AT LEAST 24 in

Fig. 3. Arrangement of Hydrogen Monitor Mock-Up Experiment.

of the jar cover is a blind tube for placement of a fast-neutron source, and immediately adjacent to the jar is a BF_3 neutron detector covered by a cadmium sleeve. The detector was placed outside the jar for ease of setup, rather than at the optimum location 48 mm from the center. According to the calculations the hydrogen sensitivity just outside the jar was nearly as good as at the optimum radius, and though the count rate was only one half the optimum, it was sufficiently high that statistical uncertainties were small.

Two other jars are shown in the figure. One of these contains a hydrogenous liquid that is miscible in "perc," and the other contains waste mixture that has been removed from the sample jar to make room for more hydrogenous liquid. Both the hydrogenous liquid and the waste mixture are measured in 500-ml burettes as they are added to or removed from the sample jar. A minimum distance of 0.533 m (21 in.) separates the outside of the sample jar from the other jars so they do not contribute significantly to the background count rate, that is, the detector count rate when no hydrogen is in the sample jar.

The neutron source used in this experiment was ^{252}Cf , which has a total half-life of 2.646 y and a half-life for spontaneous fission of 85.5 ± 0.5 y. The average number of neutrons emitted per fission is 3.733 ± 0.083 , which, when used in conjunction with the half-life for spontaneous fission, leads to a calculated specific neutron activity of 2.29×10^{12} neutrons/s g for ^{252}Cf . The particular source used in our experiment contained 1085 μg of ^{252}Cf on its calibration date of April 20, 1976. At that time the ^{252}Cf in the source was responsible for 99.81% of all the neutrons emitted and the source had an activity of 2.49×10^9 neutrons/s. At the start of this experiment there remained 762 μg of ^{252}Cf with an activity of $\sim 1.75 \times 10^9$ neutrons/s.

The experimental procedure used to collect data was as follows. First, the neutron source was inserted in the sample jar filled with 3.63 liters of pure "perc" and three or more 1-min counts were accumulated on the scaler. The average of these was taken as the monitor's background activity. Next, a measured volume of the sample liquid was removed and an equal volume of the hydrogenous liquid added. Three or

more 1-min counts were taken of the new sample and averaged. The difference between this average and the background activity was taken as the net signal due to the presence of hydrogen. Again, a portion of the sample was removed, more hydrogen was added, and three 1-min counts were taken. This continued until the sample solution was fully saturated.

To determine whether the chemical form of the hydrogenous liquid had any effect on the response of the device, this procedure was performed twice under identical conditions, once with kerosene, and once with benzene.

EXPERIMENTAL RESULTS

The experimental monitor's response to kerosene and benzene additions to initially pure "perc" is tabulated in Tables 3 and 4. In the tables the volumes of kerosene and benzene added to the sample equal the

Table 3. Experimental Monitor's Response to Kerosene Additions to "Perc"

Addition	Volume of Kerosene Added (ml)	Monitor's Response to Hydrogen Additions, cpm			
		Total		Net Hydrogen	
		Mean	95% C.I. ^a	Mean	95% C.I. ^a
0	0	92,678	352	0	0
1	80	99,560	364	6,882	506
2	80	110,705	384	18,027	521
3	80	126,722	412	34,044	542
4	80	147,984	444	55,306	567
5	148	199,638	516	106,960	625
6	148	266,204	596	173,526	629
7	80	304,989	638	212,311	729
8	80	347,807	680	255,129	766

^aC.I. = Confidence Interval.

Table 4. Experimental Monitor's Response to Benzene Additions to "Perc"

Addition	Volume of Benzene Added (ml)	Monitor's Response to Hydrogen Additions			
		Total		Net Hydrogen	
		Mean (cpm)	95% C.I. ^a (cpm)	Mean (cpm)	95% C.I. ^a (cpm)
0	0	92,420	248	0	0
1	80	96,830	254	4,401	355
2	80	102,469	370	10,040	445
3	80	109,054	382	16,625	455
4	80	117,178	396	24,749	467
5	80	127,622	412	35,193	481
6	80	139,250	430	46,821	496
7	80	152,060	348	59,631	427
8	90	169,488	476	77,059	537
9	80	187,162	500	94,733	558

^aC.I. = Confidence Interval

volumes of waste mixture removed from the sample just before each addition. The total sample volume did not change noticeably over the course of the measurements using this procedure. The columns listing the monitor's response to hydrogen additions give the total detector count rate and the net count rate due to the hydrogen additions as the mean of several 1-min observations. Also listed here is the 95% confidence interval calculated for the means.

The confidence interval for the total response was obtained by doubling the calculated standard deviation of the mean. From the fact that a statistical model, the Poisson distribution, governs the total count rate, the standard deviation of the mean is given by Eq. (11):

$$\text{S.D. } \bar{x}(\text{TOTAL}) = \sqrt{\frac{(\text{MEAN})^2}{N^2}}, \quad (11)$$

where N^i is the number of observations contributing to the i th mean. The mean net response to hydrogen was obtained by subtracting the total count rate when hydrogen was absent from all the total count rates when it was present. The confidence interval for these values was again twice the standard deviation of the net count rate given by Eq. (12):

$$\text{S.D.}^i(\text{NET}) = \sqrt{[\text{S.D.}^i(\text{TOTAL})]^2 + [\text{S.D.}^0(\text{TOTAL})]^2} . \quad (12)$$

Since kerosene and benzene differ in density and hydrogen content, the volume additions of these need to be expressed in terms of the hydrogen atom densities in the sample jar to determine whether the device's response is indeed independent of the chemical form of the hydrogenous component. Table 5 lists the physical and chemical properties of the "perc," kerosene, and benzene needed to perform the translation. All the values in the table except the hydrogen content of benzene were obtained — by direct analysis of experimental samples — from the ORNL Analytical Chemistry Division. Since high-purity benzene was used in this experiment, its hydrogen content was calculated from its chemical formula, C_6H_6 . The reported uncertainties are only rough estimates

Table 5. Some Physical and Chemical Properties of Liquids Used in Hydrogen Sensitivity Measurements

Material	Density at 25°C Mg/m ³	Hydrogen Content (wt %)
Perchloroethylene (C_2Cl_4)	1.6130 ± 0.0005	0.000 ± 0.000
Kerosene (C_xH_y)	0.7978 ± 0.0013	14.5 ± 0.5
Benzene (C_6H_6)	0.8735 ± 0.0013	7.742 ^a ± 0.000

^aCalculated value.

of the standard deviation of the measurements obtained by submitting identical samples for analysis. Of particular interest is a rather large uncertainty in the hydrogen content of kerosene. This has a direct impact on the calculated hydrogen density in the "perc"-kerosene mixtures.

Tables 6 and 7 give the results of the hydrogen atom density calculations along with the measured net hydrogen signals of the monitor. Figure 4 presents the same information graphically. The tabulated uncertainties in the hydrogen atom densities and the horizontal error bars on the figure are estimates of the 95% confidence interval resulting from the uncertainties in Table 5. The hydrogen atom density uncertainties in "perc"-benzene and "perc"-kerosene mixtures were obtained from several atom density calculations, which used varying densities and hydrogen contents for benzene and kerosene selected from an uncertainty range around the measured values.

Table 6. Hydrogen Atom Density and Monitor Response for Kerosene Additions to "Perc"

Addition	Hydrogen Atom Density in Sample (atoms/m ³)	Net Monitor Signal (counts per minute)
0	0.0×10^{27}	0.0
1	1.523 ± 0.107	$6,882 \pm 506^a$
2	3.013 ± 0.166	$18,027 \pm 521$
3	4.470 ± 0.201	$34,044 \pm 542$
4	5.895 ± 0.265	$55,306 \pm 567$
5	8.472 ± 0.381	$106,960 \pm 625$
6	10.95 ± 0.49	$173,526 \pm 692$
7	12.23 ± 0.55	$212,311 \pm 729$
8	13.48 ± 0.61	$255,129 \pm 766$

^aUncertainties represent region beyond 95% confidence interval.

Table 7. Hydrogen Atom Density and Monitor Response for Benzene Additions to "Perc"

Addition	Hydrogen Atom Density in Sample (atoms/m ³)	Net Monitor Signal (counts per minute)
0	0.0×10^{27}	0.0
1	0.891 ± 0.009	$4,401 \pm 355^a$
2	1.763 ± 0.018	$10,040 \pm 445$
3	2.615 ± 0.026	$16,625 \pm 455$
4	3.449 ± 0.034	$24,749 \pm 467$
5	4.264 ± 0.043	$35,193 \pm 481$
6	5.062 ± 0.051	$46,821 \pm 496$
7	5.842 ± 0.038	$59,631 \pm 427$
8	6.699 ± 0.100	$77,059 \pm 537$
9	7.443 ± 0.112	$94,733 \pm 558$

^aUncertainties represent region beyond 95% confidence interval.

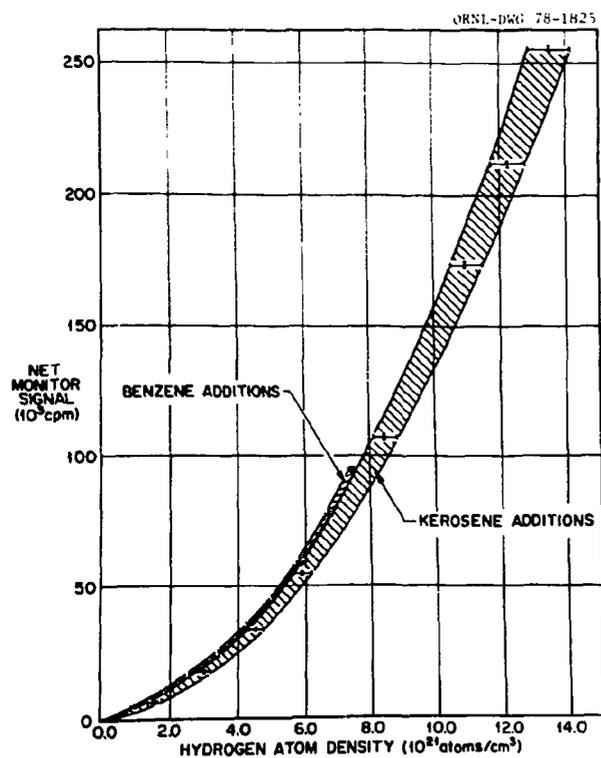


Fig. 4. Hydrogen Monitor Response to Kerosene and Benzene Additions.

DISCUSSION

The goal of this work is to develop and demonstrate a technique for monitoring the hydrogen buildup in an initially pure nonhydrogenous liquid. The degree to which the described technique meets this goal is best seen in Fig. 4. A fairly large change in the net response of the neutron detector to the additions of benzene and kerosene to "perc" can be seen. Note that the final benzene addition resulted in a "perc"-benzene solution with 8% benzene by weight and that the final "perc"-kerosene solution was 11% kerosene. As another point of reference, the hydrogen atom density in ordinary water is 66.7×10^{27} atoms/m³, about five times the maximum investigated in this experiment.

The statistical uncertainties, that is, the vertical error bars, are quite small, even with the relatively short (3-5 min) counting times used for these measurements. This remark is qualified by the fact that a rather large neutron source was used. The source consisted of 762 μ g of ²⁵²Cf with a corresponding unshielded dose rate of 3 rem/h at 1 m. Thus, source size and shielding are important concerns in the application of this technique. A hot cell was used for the present work, since the device is intended for in-cell application in the HTGR Fuel Recycle Program.

Due to the large uncertainty in the hydrogen content of the kerosene, the kerosene calibration curve is broad compared with the benzene curve. Note that the two curves do overlap; therefore, there is no reason to believe that the response of the monitor depends significantly on the chemical form of the hydrogenous liquid.

A final interesting feature of Fig. 4 is the increasing slope of the calibration curves with increasing hydrogen content over the complete range studied. This indicates higher sensitivity to changes in hydrogen content with higher hydrogen levels. Thus, one might want to add a known amount of hydrogen to a sample so as to accurately detect small changes in the hydrogen content.

CONCLUSIONS AND RECOMMENDATIONS

Over a range of hydrogen concentrations, 0.0–20.0 kg/m³ or more, the hydrogen content of liquids can be measured rapidly (~5 min) and reasonably accurately (± 2.0 kg/m³) by neutron-moderating techniques. In particular, a fast-neutron ²⁵²Cf source placed near the center of a 0.152-m-diam (6-in.) sample with a BF₃ neutron detector placed outside it has been found to perform well. A signal-to-background ratio of 2.7 was found for 10% kerosene in perchloroethylene.

The accuracy of the method depends on its application, but it could doubtless be used in many instances to detect gross changes in hydrogen concentrations. This application is most useful in certain criticality control areas.

If the technique is desired for precise analytical measurements, it should be carefully examined with respect to calibration procedures. Also, additional experiments should be conducted to investigate effects of the chemical form of the hydrogenous materials on measurement accuracy.

Finally, we present Fig. 5 as a suggested application of a hydrogen monitor in an HTGR fuel refabrication facility to detect high hydrogen concentrations in perchloroethylene-based scrubber systems. Shown is a 0.152-m-diam (6-in.) monitoring chamber installed on a perchloroethylene recirculation line. At the center of the chamber is a fast-neutron source surrounded by the sample liquid. A 0.5-mg ²⁵²Cf source would perform well in a hot-cell application. A BF₃ detector tube wrapped in a 0.762-mm-thick (0.03-in.) cadmium sleeve is located near the edge of the monitoring chamber. The detector signal is directed to a count rate meter. The meter is biased such that its output reading is zero when no hydrogen is in the chamber and a full-scale reading is obtained at a high hydrogen level. An intermediate level could be set to indicate an alarm condition.

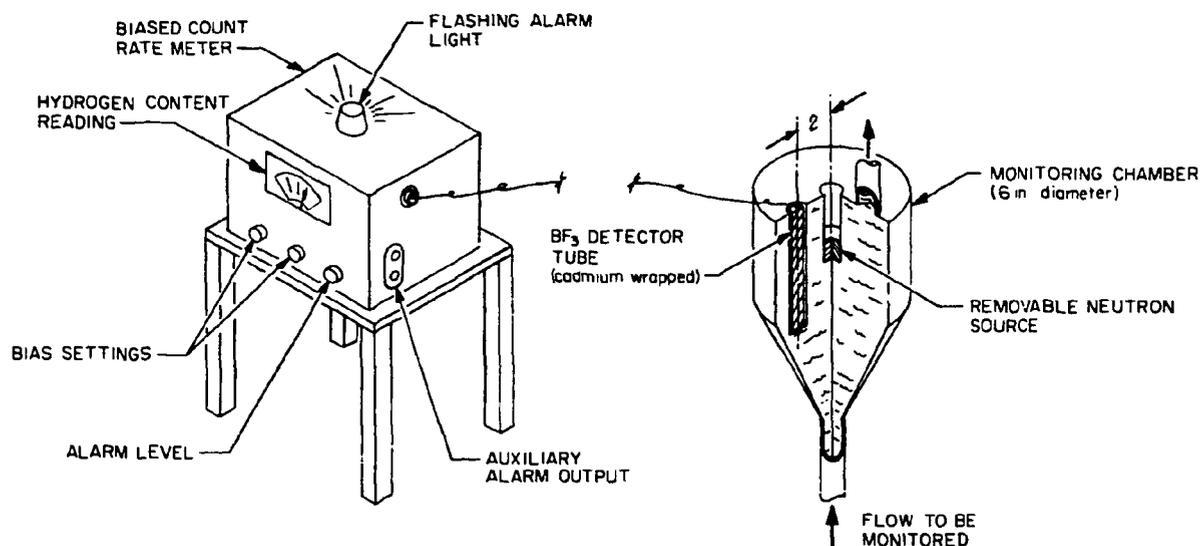


Fig. 5. Hot-Cell Application of Hydrogen Monitor for Criticality Control.

ACKNOWLEDGMENTS

The authors would like to thank the many people who assisted in the completion of this work. Thanks go to A. G. Mason and J. C. McLaughlin for performing the experiments; to L. J. Brady and his staff in the Analytical Chemistry Division for the chemical analyses; to E. J. Allen and S. M. Tieggs for their excellent technical reviews; to H. Livesey and M. Patrizio for preparing the graphs and figures; to the Metals and Ceramics Reports Office staff: Nan Richards for editing and Denise Jackson for preparing the report for reproduction.

REFERENCES

1. W. J. Lackey, J. D. Jenkins, F. J. Homan, and R. S. Lowrie, *Assessment of Coater Size for the Fuel Refabrication Prototype Plant*, ORNL-TM-4567 (July 1974).

2. S. R. McNeany and J. D. Jenkins, *Criticality Considerations for ^{233}U Fuels in an HTGR Fuel Refabrication Facility*, ORNL/TM-6136 (January 1978).
3. S. Glasstone and M. C. Edlund, *The Elements of Nuclear Reactor Theory*, p. 149, D. Van Nostrand, Princeton, New Jersey, 1957.
4. J. R. Lamarsh, *Introduction to Nuclear Reactor Theory*, p. 395, Addison-Wesley, Reading, Massachusetts, 1972.
5. K. M. Case, F. de Hoffman, and G. Placzek, *Introduction to the Theory of Neutron Diffusion*, p. 32, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, 1953.
6. N. M. Greene et al., *AMPX: A Modular Code System for Generating Coupled Multigroup Neutron-Gamma Libraries from ENDF/B*, ORNL/TM-3706 (March 1976).