



Feasibility Study of a Low-Energy Gamma Ray System for Measuring Quantity and Flow Rate of Slush Hydrogen

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Abstract

As part of a study to demonstrate the suitability of an X-ray or gamma ray probe for monitoring the quantity and flow rate of slush hydrogen, mass attenuation coefficients for Cd^{109} X- and gamma radiation in five chemical compounds have been measured. The Ag^{109} K X rays were used for water and acetic acid, whereas E3 transition from the first excited state at 87.7 keV in Ag^{109} provided the probe radiation for bromobenzene, α^2 -chloroisodurene, and cetyl bromide. Measurements were made for a single phase (gas, liquid, and solid) as well as mixed phases (liquid plus solid) in all cases. It has been shown that the mass attenuation coefficient for the selected radiations is independent of the phase of the test fluids or phase ratios in the case of mixed-phase fluids. This report describes the procedure and results for the five fluid systems investigated.

Introduction

Slush hydrogen is being considered (refs. 1 and 2) as a propellant for the National Aero-Space Plane (NASP). It has low mass, high energy content, and higher density than liquid hydrogen. However, no suitable technique is currently available for monitoring the quantity and flow rate of a fuel whose phase configuration is continuously changing. We have investigated the feasibility of a low-energy gamma ray system for gauging the quantity and flow rate for such fuels. It is based on the principle that the mass attenuation coefficient of a chemical compound for gamma radiation does not depend on the phase of the chemical. This report describes the results for five mixed-phase fluid systems.

Theoretical Background

When a narrow beam of gamma rays passes through matter, some of the rays are scattered or absorbed in the medium because of interactions with the atoms of the medium. For example,

$$I = I_0 e^{-\mu x} \quad (1)$$

where

I intensity of gamma radiation arriving at detector

I_0 intensity of gamma radiation incident on test medium

μ attenuation per unit path length in the medium (i.e., the linear attenuation coefficient)

x path length in the medium

The linear attenuation coefficient (μ , cm^{-1}) for an elemental medium is given by

$$\mu = \sigma n = \sigma \frac{N\rho}{A} \quad (2)$$

where

σ total interaction cross section per atom, cm^2/atom

N Avogadro's number on physical scale, 6.025×10^{23} atoms/mole

ρ density of the medium, g/cm^3

A atomic or molecular weight, g

Since σ is a function of gamma ray energy, μ is clearly energy dependent. The expression for μ can be generalized for any medium containing different kinds of atoms. It is the basis of the conventional nuclear radiation attenuation (NRA) monitors.

A more useful variation of equation (2) is

$$\frac{\mu}{\rho} = \sigma \frac{N}{A} \quad (3)$$

where μ/ρ is in units of centimeters squared per gram and called the *mass attenuation coefficient* of the medium for the incident gamma rays. Mass attenuation coefficient is a *more* fundamental parameter than the linear attenuation coefficient since it is independent of the actual density and physical state of the medium. If the medium is a mixture of several different compounds or elements, its mass attenuation coefficient is given by

$$\left(\frac{\mu}{\rho}\right)_{\text{mixture}} = \sum_i w_i \left(\frac{\mu}{\rho}\right)_i \quad (4)$$

where

w_i fraction by weight of the i th component

$\left(\frac{\mu}{\rho}\right)_i$ mass attenuation coefficient of the i th component

For a multiphase mixture of any given chemical compound, equation (4) can be written as follows:

$$\begin{aligned}
\left(\frac{\mu}{\rho}\right)_{\text{multiphase}} &= \sum_{i=1}^3 w_i \left(\frac{\mu}{\rho}\right)_i \\
&= w_1 \left(\frac{\mu}{\rho}\right)_{\text{solid}} + w_2 \left(\frac{\mu}{\rho}\right)_{\text{liquid}} + w_3 \left(\frac{\mu}{\rho}\right)_{\text{gas}} \\
&= \left(\frac{\mu}{\rho}\right)_{\text{single phase}} (w_1 + w_2 + w_3) \\
&= \left(\frac{\mu}{\rho}\right)_{\text{single phase}} \quad (5)
\end{aligned}$$

It is therefore expected that a measurement of linear attenuation coefficient μ_{mixture} for a multiphase mixture of a given compound should provide its instantaneous density ρ_{mixture} , since μ/ρ for any of its single phases is presumably already known. Exploitation of this principle in the NASP program will require simultaneous measurement of fuel volume by judicious locations of several counting systems in the fuel tank (ref. 3). If total fuel volume measurements are made every second, the rate of decrease of the remaining fuel in the tank should equal its rate of consumption (or flow rate out of the fuel tank).

Experimental Procedure and Results

Five multiphase systems were selected to verify the principle of phase mixtures. They were water, acetic acid, bromobenzene, α^2 -chloroisodurene, and cetyl bromide. These systems were selected for convenience of handling and appropriateness of their physical properties, as summarized in table I.

Table I. Summary of Chemicals Studied

Chemical species	Melting point, °C	Boiling point, °C	Chemical purity, percent (a)
Water	0.0	100.0	100
Acetic acid	16.6	118.5	99.9
α^2 -chloroisodurene	33.4	122.5	98
Bromobenzene	-31.0	156.0	99
Cetyl bromide	18.0	149.0	97

^aExcept for acetic acid, all chemicals were obtained from Aldrich Chemical Company. Acetic acid was supplied by J. T. Baker Chemical Company.

All systems exist in liquid phase at room temperature (25 °C) except α^2 -chloroisodurene. They can be solidified and vaporized rather easily. The Cd ¹⁰⁹/Ag ¹⁰⁹ radionuclide was selected as the source of probing photons. It has a reasonable half-life (454 days)

and emits appropriate energy photons. The X- and gamma radiation photons were counted with a NaI(Tl) counter. The attenuation coefficient measurements were made in standard narrow-beam geometry (ref. 4). All measurements were made at the temperatures necessary for the appropriate phases of the test chemicals. For instance, in the case of water, μ_{gas} was measured at 100 °C, μ_{liquid} was measured at room temperature (25 °C), and μ_{solid} was measured at 0 °C. Figure 1 shows the schematic diagram of the experimental system. The test cell for liquid, solid, and liquid plus solid mixtures of phases of water and acetic acid consisted of a multicomponent 51- by 51- by 51-mm Dupont Teflon cube, shown in figure 2. The other three chemicals were tested in a specially designed quartz cell, shown in figures 3 and 4. An appropriately heated 102-mm-long quartz cylindrical cell, through which vapors from the boiling liquids were passed, was used to provide a selected gas medium for X-ray attenuation measurements. (See fig. 5.) The gas-phase densities were calculated from the ideal gas law (i.e., $\rho_{\text{gas}} = PM/RT$, where P is the pressure, R is the gas constant, T is the vapor temperature, and M is the molecular weight of the chemical compound). The liquid-phase densities were measured with a precision specific gravity chain balance, accurate to 0.0001. The solid-phase density values were determined by extending the graph of ρ_{liquid} versus T for the test chemical to a temperature 5° below its solidification temperature.¹ The slush densities, on the other hand, were determined from the graph of ρ_{liquid} versus T exactly at the melting point of the solid phase of the chemical. The liquid-phase densities were measured at a minimum of six temperatures for a good least-squares linear fit to the data for ρ_{liquid} versus T .

The results are summarized in table II and illustrated in figure 6. The last column in table II(c) lists the theoretical value of mass attenuation coefficient of the species tested (ref. 5). It should be noted that we have included two other chemical compounds (nitrogen and dibromomethane) in table II and figure 6, even though they required higher energy gamma rays for measuring their μ/ρ values. It is apparent from the data

¹ In the case of water, ice density ρ_{solid} was taken from the Handbook of Chemistry and Physics and the slush density ρ_{slush} was calculated for several percentages of ice in water as follows:

$$\rho_{\text{slush}} = \frac{M_1 + M_2}{(M_1/\rho_1) + (M_2/\rho_2)}$$

where M_i and ρ_i are the mass and density of the i th phase (ice or water).

that the values of the mass attenuation coefficients for all the systems are independent of their physical states.

As a further test of the validity of equation (5), the experimental and calculated values of the densities of multiphase mixtures of α^2 -chloroisodurene and acetic acid were compared. The experimental values of the density of the mixture ρ_{exp} were determined from $\mu_{\text{exp}}/(\mu/\rho)_{\text{theory}}$ ratios for the test chemicals. The calculated values of the density ρ_{cal} were determined from the following relation:

$$\rho_{\text{cal}} = \frac{l_1\rho_1 + l_2\rho_2 + l_3\rho_3}{l_1 + l_2 + l_3}$$

where l_i and ρ_i represent the path length and density of the individual phases (solid, liquid, and gas) of the test chemicals.

The results for α^2 -chloroisodurene are $\rho_{\text{exp}} = 1.0331 \pm 0.0124$ g/cm³ and $\rho_{\text{cal}} = 1.0392$ g/cm³. For acetic acid, the results are $\rho_{\text{exp}} = 1.0619 \pm 0.0127$ g/cm³ and $\rho_{\text{cal}} = 1.0590$ g/cm³.

Discussion of the Proposed Slush Hydrogen Monitoring System

Figure 7 shows a schematic diagram of the proposed slush hydrogen monitoring system. It is as-

sumed that the interface of gaseous hydrogen and slush hydrogen will remain a well-defined plane without the use of an appropriate restraining diaphragm.² It is also assumed that the slush hydrogen mixture is well stirred to ensure its steady flow and to avoid sedimentation of solid hydrogen.³

For clarity only a single pair of detectors are shown. Obviously, several such pairs will be needed to determine the volume of the fuel in the tanks (ref. 3).

The horizontal detector system in figure 4 can provide the slush density ρ_{slush} as shown below. Let $I(d_2)$ be the counting rate at detector 2:

$$I(d_2) = I_0 e^{-\mu_{\text{slush}} d_2} \quad (6)$$

where

² Operation in microgravity environment might dictate the necessity for a restraining diaphragm.

³ Active steps will have to be taken to ensure the validity of these assumptions if slush hydrogen is to be used as a propellant for NASP.

d_2	distance between the source and the detector in horizontal system
I_o	incident photon flux in the horizontal system
μ_{slush}	linear attenuation coefficient for slush hydrogen in the horizontal path

With $\mu_{\text{slush}}/\rho_{\text{slush}} = \mu_{\text{liquid}}/\rho_{\text{liquid}}$ known from system calibration data, equation (6) can easily provide ρ_{slush} from a measurement of μ_{slush} .

The hydrogen slush level at any time—and hence the total slush volume in the tank—can be determined with the vertical detector system. Suppose the instantaneous height of the gaseous hydrogen–slush hydrogen interface is x . Then the counting rate $I(x)$ at detector 1 is given by the following expression:

$$I(x) = I'_o \exp[-\mu_{\text{slush}} x - \mu_{\text{gas}}(d_1 - x)] \quad (7)$$

where

I'_o	incident photon flux in the vertical system (it can be arranged to be equal to I_o)
d_1	distance between the source and the detector in the vertical system (it can be arranged to equal d_2)
μ_{gas}	linear attenuation coefficient in gaseous hydrogen

The only unknown in equation (7) is x , since μ_{slush} is already known from equation (6) and μ_{gas} is easily calculable from the following equation:

$$\frac{\mu_{\text{gas}}}{\rho_{\text{gas}}} = \frac{\mu_{\text{liquid}}}{\rho_{\text{liquid}}} = \frac{\mu}{\rho} \quad (8)$$

where μ/ρ is the calibration constant for hydrogen. A simultaneous measurement of temperature and pressure of the pressurizing hydrogen gas can provide a value of ρ_{gas} and thus enable a calculation of μ_{gas} .

Since linear attenuation coefficients are strong functions of the photon energy and the test medium atomic number, it will be necessary to use much lower energy gamma ray sources for slush hydrogen measurements. A 5.9-keV $\text{Fe}^{55}/\text{Mn}^{55}$ source should serve the purpose well, as indicated by typical calculations

for slush hydrogen (50 percent solid):

$$\begin{aligned} I(x) &= I_o e^{-\mu x} \\ &= 0.52 I_o \quad (x = 25 \text{ cm}) \\ &= 0.26 I_o \quad (x = 50 \text{ cm}) \end{aligned}$$

Clearly there is a reasonably strong attenuation, concomitant with a statistically strong survival rate, for the 5.9 keV X rays through appropriate path lengths in slush hydrogen. An additional advantage of using a low-energy radiation source is the convenience of shielding against it.

Concluding Remarks

Mass attenuation coefficients for Cd^{109} radiation have been measured in a number of mixed-phase fluids. These measurements have clearly confirmed, as expected, that the mass attenuation coefficients for X and gamma rays in fluids are independent of the physical phases of the fluids. It can therefore be concluded that a similar system based on an appropriate photon source—such as $\text{Fe}^{55}/\text{Mn}^{55}$ (5.9 keV)—can serve as the basis for slush hydrogen monitoring on-board the National Aero-Space Plane.

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February 20, 1992

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REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project (0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE April 1992	3. REPORT TYPE AND DATES COVERED Technical Paper		
4. TITLE AND SUBTITLE Feasibility Study of a Low-Energy Gamma Ray System for Measuring Quantity and Flow Rate of Slush Hydrogen			5. FUNDING NUMBERS WU 307-50-10-02	
6. AUTHOR(S) Jag J. Singh, Chih-Ping Shen, and Danny R. Sprinkle				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) NASA Langley Research Center Hampton, VA 23665-5225			8. PERFORMING ORGANIZATION REPORT NUMBER L-16980	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration Washington, DC 20546-0001			10. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA TP-3150	
11. SUPPLEMENTARY NOTES Singh and Sprinkle: Langley Research Center, Hampton, VA; Shen: Old Dominion University, Norfolk, VA.				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Unclassified-Unlimited Subject Category 19			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) As part of a study to demonstrate the suitability of an X-ray or gamma ray probe for monitoring the quantity and flow rate of slush hydrogen, mass attenuation coefficients for Cd ¹⁰⁹ X- and gamma radiation in five chemical compounds have been measured. The Ag ¹⁰⁹ K X rays were used for water and acetic acid, whereas E3 transition from the first excited state at 87.7 keV in Ag ¹⁰⁹ provided the probe radiation for bromobenzene, α ² -chloroisodurene, and cetyl bromide. Measurements were made for a single phase (gas, liquid, and solid) as well as mixed phases (liquid plus solid) in all cases. It has been shown that the mass attenuation coefficient for the selected radiations is independent of the phase of the test fluids or phase ratios in the case of mixed-phase fluids. This report describes the procedure and results for the five fluid systems investigated.				
14. SUBJECT TERMS Slush hydrogen; Multiphase chemical systems; Gamma rays; Mass attenuation coefficient; National Aero-Space Plane			15. NUMBER OF PAGES 12	
			16. PRICE CODE A03	
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT	