MEASUREMENT OF THE LIQUID PHASE MASS IN GAS-LIQUID SPRAYS BY X-RAY ATTENUATION

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Abstract--An optical method for measuring the mass of liquid phase in a spray has been developed into an X-ray method. An X-ray of about 6 keV is used to measure the mass distribution of liquid phase in the spray formed by a concentric injector with water and gaseous nitrogen at one atmosphere of pressure. The possibility of measuring the mass of gaseous oxygen and hydrogen is discussed.

INTRODUCTION

Most liquid fuels are atomized into a great number of small droplets in order to promote the combustion process in combustion chambers of engines and furnaces. The sprays of liquid fuels are described by velocities, temperatures and pressures of the two phases and fuel-air ratio and interface shape between the two phases. Among these parameters droplet diameters are often measured to evaluate injectors because liquid fuels become spheres in sprays due to surface tension and the smaller droplets vaporize more quickly. Droplet diameters can be measured more easily than other parameters; however, dense sprays near injectors, which set the initial condition of combustion process, impede the measurement of droplet diameters, e.g. due to multiple scattering in optical methods.

Fuel-air ratio is one of the most important factors in combustion. There are few reports about the measurement of fuel-air ratio. Mehegan *et al.* (1970) used water/gaseous nitrogen as simulants of liquid oxygen/gaseous hydrogen and determined the distribution of mass flux fraction in gas/liquid sprays by two-phase impact probes. The two-phase impact probe technique is based on several assumptions and cannot be applied to a spray with combustion.

In nuclear engineering, void fraction, which is the volume fraction of gas phase, is measured in relation to a loss of coolant accident in a nuclear reacter (Jones & Delhaye 1976). The attenuation method of X-rays and gamma rays make it possible to measure the void fraction through the coolant pipe wall. Penetration of the wall requires high energy X-ray and gamma rays. Ball *et al.* (1958) used approximately monoenergetic X-ray of about the 17.5 keY molybdenum/~ energy. Pike *et al.* (1965) operated an X-ray tube with a tungusten target at a peak voltage of 45 kV. Schrock & Selph (1963) used tungusten characteristic X-ray K_{α} and K_{β} at 58.5 and 68.0 keV.

In this study the attenuation method with a low energy X-ray of about 6 keV was used to measure the mass of liquid phase in a spray under the atmospheric condition. The spray was formed with water and gaseous nitrogen by a concentric injector of the type which is frequently used in a liquid oxygen/liquid hydrogen propellant rocket engine.

THEORY

In this section the measurement of liquid phase mass by an optical method is discussed. This is then developed into an X-ray method.

As the absorption of Visible light by water in the air is negligible, the attenuation of light passing through a spray results from the scattering of light by liquid droplets. When the multiple scattering of light is negligible, the attenuation rate of light is calculated by

$$
\Delta I = \frac{I_0 - I}{I_0} = \int_0^\infty \frac{\pi}{4} Q_{\text{ex}} D_p^2 N(D_p) \, \mathrm{d}D_p
$$

where I_0 is the intensity of incident light, I is the intensity of transmitted light, Q_{sea} is the efficiency for scattering, D_p is the diameter of droplets and N is the number of droplets.

The efficiency for scattering of a plane electromagnetic wave by an isotropic, homogeneous sphere can be calculated by a rigorous theory. The efficiency for scattering increases to a maximum value and undergoes a damped oscillation about the limiting value of 2 (e.g. Kerker 1969). So $Q_{\text{max}} = 2$ is a good approximation for the dimensionless size parameter, $\pi D_n/\lambda$, more than 50 which is equivalent to a droplet diameter of 10 μ m with He-Ne laser ($\lambda = 0.633~\mu$ m). This simplifies the calculation of the light attenuation rate as follows:

$$
\Delta I = \int_0^\infty \frac{\pi}{2} D_p^2 N(D_p) \, \mathrm{d}D_p. \tag{1}
$$

With the assumption that the density of liquid phase is constant, the mass of liquid phase in a unit volume is

$$
m_L = \rho_L \int_0^\infty \frac{\pi}{6} D_p^3 N(D_p) \, \mathrm{d}D_p. \tag{2}
$$

Subetituting [1] into [2] yields

$$
m_L = \frac{1}{3} \rho_L \text{SMD } \Delta I \tag{3}
$$

where

$$
SMD = \frac{\int_0^\infty D_p^3 N(D_p) \, \mathrm{d}D_p}{\int_0^\infty D_p^2 N(D_p) \, \mathrm{d}D_p}.
$$

The Sauter mean diameter (SMD) is the representative length which is equal to the total volume divided by the total area of droplets. If *AI* and SMD are measured, the mass of liquid phase can be calculated by [3].

In most of actual sprays the attenuation of light is high, which means that multiple scattering cannot be negiected, especially in dense sprays near injectors. And the measurement of SMD by an optical method is also affected by multiple scattering. Therefore the optical method to make the measurements necessary to calculate the mass of liquid phase by $[3]$ is only applicable to spray regions where the droplet concentration is low.

The scattering of light may be divided into classical diffraction, geometrical reflection and refraction. When the wavelength of light λ is much shorter than the diameters of droplets, diffracted light is concentrated forwards and cannot be distinguished from the undiffracted light. Reflection and refraction occur at an interface between a droplet and the surrounding air which have different refractive indices. If the relative refractive index m equals one, there is neither reflection nor refraction.

Under the condition that $\lambda \ll D_p$, and $m = 1$, there is no multiple scattering although droplets may not be visible. There is, however, a possibility of measuring the liquid phase mass by absorption of light. The effect of absorption is related to the imaginary part in the complex refractive index. Therefore if $\lambda \ll D_n$ and $m = 1 + ix$, the mass of liquid phase can be measured with no multiple scattering. The refractive index depends on the liquid and the wave length of the light. X-rays are electromagnetic waves which satisfy most of these conditions in the measurement of sprays, where the X-ray diffraction by crystal need not be considered.

Absorption method by X-ray

The attenuation of an X-ray passing through matter is due to photoelectric absorption, Thomson scattering, Compton scattering and pair creation. The photoelectric effect is predominant in a low energy X-ray which can be used to measure the mass of liquid phase in a spray.

The absorption of a monochromatic X-ray passing through matter is expressed as

$$
I = I_0 \exp(-\mu l) \tag{4}
$$

where I_0 is the intensity of the incident X-ray, I is the intensity of the transmitted X-ray, μ is the linear absorption coefficient of the matter and *l* is the path length of the X-ray in the matter. As the absorption by gas phase is small in a spray, the absorption coefficient is that of liquid phase and the path length is the total length or equivalent thickness of liquid phase through which the X-ray passes as shown in figure 1.

The mass absorption coefficient, μ/ρ where ρ is the density, is presented in handbooks (e.g. Clark 1963) instead of the linear absorption coefficient because the former is usually considered as independent of the physical state or the chemical state of the matter. Therefore the mass absorption coefficient of a mixture or a chemical compound can be calculated with the elements which compose the matter as follows:

$$
\mu/\rho = \Sigma(\mu_i/\rho_i)Y_i \tag{5}
$$

where μ/ρ_i is the mass absorption coefficient and Y_i is the mass fraction of element i.

The measurement of the liquid phase mass by X-ray needs no calibration because the attenuation rate of an X-ray is the intensity ratio of the transmitted X-ray to the incident X-ray and the mass absorption coefficient can be calculated by [5].

Selection of X-ray energy

The mass absorption coefficient of an X-ray is inversely proportional to approximately the third power of the X-ray energy. The most suitable energy for measurement of objects must be selected to get a reasonable attenuation of the X-ray.

Figure 1. Equivalent thickness of liquid phase.

Energy of X-ray	Thickness of water			
	mm	$0.1 \, \text{mm}$	0.01 mm	0.001 mm
5 keV	0.017	0.66	0.960	0.9959
6 keV	0.087	0.78	0.976	0.9976
8 keV	0.34	0.90	0.989	0.9989
10 keV	0.56	0.94	0.994	0.9994

Table 1. Transmittance of X-ray

The variation of the X-ray relative intensity is given from [4] by

$$
\Delta(I/I_0)=-\exp(-\mu I)\Delta(\mu I).
$$

The smaller μl gives the larger variation of the relative intensity. When $\mu l \ll 1$,

$$
\Delta(I/I_0)=-\Delta(\mu I)
$$

and the variation of the relative intensity is proportional to μ . There is little problem of "dynamic bias" (Harms & Laratta 1973).

On the contrary, an accurate measurement demands the condition that $I_0 - I \ge \epsilon$ because the measured intensity of an X-ray contains the error ϵ . The measurement error of X-ray intensity in the present experiment was about 0.5% . The energy of the X-ray was selected to get an attenuation of about 50%.

The object measured in the experiment was a spray formed by a concentric injector shown in figure 2. Water was injected from a centre nozzle at a velocity of 6 m/s and gaseous nitrogca was injected at a velocity of 50-200 m/s parallel to the water jet. As the droplets of water spread conically, the equivalent thickness of water passed by X-ray is a maximum at the water nozzle, decreases monotonously downstream and depends on the injected mass of water and the injection velocity of gas.

The transmittance of an X-ray through water is shown in table 1. An X-ray of 10 keV may be suitable for the measurement of a water jet length and an X-ray of 5 keV may be too high for downstream measurements in sprays.

EXPERIMENTAL EQUIPMENT

Figure 2 shows the experimental arrangement used to measure the mass of liquid phase where the X-ray from a tube travels through a slit and a spray to a detector. The alignment

Figure 2. X-ray system for liquid phase mass measurement in a spray.

of the experimental equipment was made by measuring the intensity of the X-ray. The X-ray tube was set in a metal box with a shutter. The slit cut the X-ray beam in order to obtain an X-ray fan of fiat intensity distribution without a spray. The intensity of the X-ray was adjusted by the width of the slit and the anode current and voltage of the X-ray tube. The zero filament voltage gave a sufficient intensity of X-ray.

Detector

A one-dimensional position-sensitive proportional counter (PSPC) was used to measure the X-ray intensity (Yoshioka *et al.* 1983). X-Rays induce pairs of electrons and ions in the detector. The electrons increase in number due to avalanche effect (gas amplification) with nearly constant ratio (order of $10⁴$) under the suitable voltage between an anode and a cathode. Therefore the height of each output pulse from the detector is proportional to the energy of each incident X-rayand the detector is called a proportional counter.

Both ends of the cathode were connected to charge-sensitive preamplifiers. The charge division method gives the pulse whose height has the linear relation with the incident location of X-rays. The histogram of the pulse height obtained by a pulse height analyzer is the one-dimensional intensity distribution of X-rays. There is no moving part in the measurement system. The space resolution was about 0.5 mm. The maximum allowable counting rate is about 40,000 counts per second (cps).

Accuracy of measurement

The time average intensity of X-rays depends on the number and energy of X-rays or photons in a unit time. For simplicity a constant energy is assumed. The number of incident photons $n(t)$ in a detector changes in accordance with the Poisson distribution due to statistical fluctuations. The standard deviation of the number is the square root of the number, \sqrt{n} . The uncertainty of the X-ray intensity measurement is in the order of $1/\sqrt{n}$. The smaller uncertainty requires the longer time for measurement.

The intensity of the X-ray was adjusted to about 5000 cps in order to avoid counting losses and was measured for 100 sec. As the distribution of X-ray intensity was measured by one detector and the distribution was indicated by about ten points, the average counts at each point was about 50,000. Therefore the smallest change of X-ray intensity which could be distinguished from statistical fluctuations was about 0.5%.

Energy spectrum of X-rays

The energy spectrum of X-rays depends on the metal of the X-ray tube anode and its voltage. A chromium anode X-ray tube was operated at a peak voltage of 10 kV obtained by half wave rectification. The spectrum was measured by the above-mentioned PSPC with a

Figure 3. Energy spectrum of X-ray.

single anode wire. The peak of characteristic X-ray K_a at 5.4 keV is seen in figure 3. As Schrock & Selph (1963) and Pike et al. (1965) used data of spectra by other researchers, they had to calibrate their measurement systems with mock-ups made of plastics which had nearly the same absorption coefficients of X-rays as water.

Measurement of transmittance

The transmittance is defined by the ratio of n to n_0 where n and n_0 are counts of X-rays in a fixed time with and without a measurenent object, respectively. The transmittance is related to the thickness of measured objects by

$$
n/n_0 = \int_0^\infty W(E) \exp(-\mu(E)l) dE
$$
 [6]

where $\mu(E)$ is the linear absorption coefficient and $W(E)$ is the normalized count at an X-ray energy of E. If n/n_0 and $W(E)$ are measured and $\mu(E)$ is calculated by [5], the thickness l can be obtained by [6].

The count n_0 had been measured for one hundred seconds just before the count n was measured for the same period. The intensity varied with time. The counts were adjusted to make the counts n and n_0 equal at a position where the attenuation of the X-ray was negligible.

For the verification of this method, an aluminum foil of 17 μ m was used. The transmittance of the foil was 0.71. The thickness of the foil calculated by [6] was 12 μ m.

MEASUREMENT OF SPRAY

A spray was formed by the concentric injector shown in figure 2. Water was injected from a centre nozzle at a speed of 6 m/s. Gaseous nitrogen was injected at a speed of 50-200 m/s from an annular orifice around the water jet. The diameter of the centre nozzle was 1.8 mm. The outer and inner diameters of the annular orifice were 6.0 mm and 3.0 mm respectively.

The equivalent thicknesses of water measured by X-ray are shown in figure 4. The distribution of the thickness was nearly axial symmetrical. The assumption of axial symmetry makes it much easier to calculate the local fraction of liquid phase. The simultaneous equations equivalent to the Abel integral inversion were solved to obtain the local fraction

Figure 4. Equivalent thickness of liquid phase in sprays.

Figure 5. Local volume fraction of liquid phase in sprays.

of fiquid phase. The radial distribution of the fraction became less steep with an increase of the gas injection velocity, as shown in figure 5. When the injection velocity of gas was 80 m/s, the local volume fraction of liquid phase had a small peak at the periphery of the spray, which was not so clear in the distribution of equivalent thickness in figure 4.

These data measured by X-ray cannot be compared with other data because other methods cannot be applied to the measurement of dense sprays. The validity of the X-ray method depends on the theory and experimental equipment which are described at length in this report.

The X-ray method was applied to the downstream region in the spray where droplet concentration was low. The result at a distance of 150 mm from the injector with a water injection velocity of 6 m/s and a gas injection velocity of 200 m/s is shown in figure 6 and is less reliable because the wide distribution of the spray made the counting rate low at each point and made it impossible to correct the time fluctuation of X-ray intensity with the present measurement system.

In figures 4 and 6 the equivalent thickness at the centre of the spray is about inversely proportional to the injection gas velocity and the distance from the injector.

The diffraction method (Yule *et al.* 1981) was also used to measure the thickness of liquid phase at the centre of the spray. The attenuation of Ne-He laser was 0.7 and SMD was 38 μ m (Gomi 1983). Substitution of these values into [3] yeilds an equivalent water thickness of 9 μ m. Multiple scattering was serious at the attenuation of 0.7. Correcting for multiple scattering gives a thickness nearer to that obtained by the X-ray method (18 μ m).

With the nearly same injector and injection conditions as those of the present study shown in table 2, Yatsuyanagi (1982) measured the mass flux of liquid phase and the

Figure 6. Equivalent thickness by X-ray and by two-phase impact probe (Yatsuyanagi 1982).

	Symbols	Injection velocity		Axial distance	
		Water	Gaseous nitrogen	from injector	
Present study	Δ.	6 m/s	50 m/s	50 mm	
	o		80 m/s		
	٥		200 m/s		
				150 mm	
Yatsuyanagi (1982)	×	4 m/s			

Table 2. Experimental conditions

with the concentric injector;
the outer diameter of annular orifice = 6.0 mm,
the inner diameter of annular orifice = 3.0 mm,
the diameter of center nozzle = 1.8 mm (present study),
the diameter of center nozzle = 2.2 mm

velocity of gas phase with two-phase impact probes. The velocity of liquid phase is needed in order to convert the mass flux to the equivalent thickness which is the spatial average of the liquid phase thickness in an X-ray. For simplicity the velocity of droplets was assumed equal to the gas velocity as the velocity difference between the two phase becomes small with the distance from the injector. The result shown in figure 6 is consistent with data obtained by the X-ray method.

Possibility of measurement of the gas phase mass by X-ray

Hitherto the absorption of X-ray by the injected gaseous nitrogen and by the air around the spray was omitted in the measurement of liquid phase mass, because the attenuation of X-ray by the gas which was substituted for water was too small to be detected. An X-ray of 6 keV attenuates by 32% when passing through gaseous oxygen of 100 mm thickness and by 21% through gaseous nitrogen of the same thickness at 1 atm and 273 K. As the present measurement system is sensitive to an intensity variation of 0.5%, it can measure the gas thickness with about 2 *mm* resolution. The attenuation by gaseous hydrogen and helium of 100 mm thickness is less than 0.1% . Therefore the X-ray attenuation method can measure the mass of gaseous oxygen or nitrogen in gaseous hydrogen or helium.

It is an aim of the authors to measure sprays with combustion formed by concentric injectors in a liquid oxygen/liquid hydrogen propellant rocket engine. The X-ray attenuation method can measure the mass of oxygen atoms although it cannot distinguish whether they may compose oxygen elements or water elements in gas or liquid states. Nuclear magnetic resonance (NMR) is a promising method to measure the mass of hydrogen without being affected by other species (Lauterbur 1973). These methods by X-ray and NMR will help us to understand the distribution oxygen and hydrogen in the sprays with combustion.

In future hydrogen will be more widely used as a fuel; however, at present most fuels are hydrocarbons. The absorption coefficient of carbon is nearly proportional to that of oxygen at a suitable energy for a spray measurement. Di-chromatic X-ray absorptiometry cannot thus distinguish between oxygen and carbon.

CONCLUSIONS

The mass distribution of liquid phase in a water/gaseous nitrogen spray produced by a concentric injector was measured by the attenuation method using a 6 keV X-ray.

(I) The equivalent thickness of water was inversely proportional to the injection gas velocity and the distance from the injector.

(2) The local volume fractions of water were calculated on the assumption of an axial symmetrical spray. The radial distributions of the fraction became less steep with the increase of gas injection velocity and had a small peak at the periphery of the spray at a gas injection velocity of 80 m/s.

Although X-rays are potentially hazardous, the advantages of the X-ray attenuation method are

- (1) no need for calibration when the energy spectrum of the X-ray is measured,
- (2) the suitability for application in a dense spray, and

(3) the applicability to the mass measurement of gaseous oxygen in gaseous hydrogen with and without combustion.

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