

# Estimation of upper limit of AP content in AP/HTPB composite propellant

## — A consideration based on flow characteristics of AP/HTPB suspension —

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Because of the requirements for the preparation of ammonium perchlorate (AP) / hydroxyl - terminated polybutadiene (HTPB) composite propellant, there is an upper limit content,  $\phi_{max}$  (wt%), of AP contained propellant. It is necessary for the design of AP/HTPB composite propellant to estimate  $\phi_{max}$ . In this study, an attempt was made to find out the universal powder property of AP which can determine  $\phi_{max}$ , from the standpoint of the viscosity of AP/HTPB suspension. The results are as follows: 1) Concerning AP/HTPB suspensions prepared in this study, the relationship between the relative viscosity,  $\eta_r$  (-), and the volume fraction,  $\phi_v$  (vol%), can be represented by the Mori-Ototake equation.  $k$  and  $\phi_{rc}$  which are constant in the Mori-Ototake equation are dependent on specific surface area,  $S_{wp}$  (m<sup>2</sup>/kg), measured by the air-permeability method. It is found that  $\eta_r$  can be represented as the function of  $\phi_v$  and  $S_{wp}$ . 2)  $\eta_r$  of AP/HTPB suspension containing  $\phi_{max}$  was almost constant. 3)  $\phi_{max}$  increases with decreasing  $S_{wp}$ . The relationship between  $\phi_{max}$  and  $S_{wp}$  can be represented by the following equation.

$$\phi_{max} = 85 - 0.01S_{wp} \quad (40\text{m}^2/\text{kg} \leq S_{wp} \leq 1200\text{m}^2/\text{kg})$$

It is found that  $\phi_{max}$  can be estimated from  $S_{wp}$ .

### 1. Introduction

Ammonium perchlorate (AP) / hydroxyl - terminated polybutadiene (HTPB) composite propellant was adopted in this study because AP/HTPB composite propellant is the most widely used one at present time. In the preparation of AP/HTPB composite propellant, AP is mixed with HTPB sufficiently. The AP/HTPB suspension is added with a curing agent and mixed again. The AP/HTPB suspension, added with a curing agent, has fluidity for a while after its addition. The AP/HTPB suspension added with a curing agent during this period is called an uncured propellant

in this study. The AP/HTPB suspension added with a curing agent was designated as an uncured propellant. The uncured propellant, mixed sufficiently, is cast into the rocket motor. Therefore it's required that at casting the uncured propellant has the suitable viscosity. On the other hand, when the proportion of bubble contamination in AP/HTPB composite propellant is more than 2.0 vol%, the burning rate of the propellant is influenced by the bubble contamination and consequently the reproducible burning rate can't be obtained<sup>1)</sup>. That is to say, the reliability of the burning rate is poor. In order to prepare the propellant, of which the burning rate isn't influenced by the bubble contamination, it is necessary that the proportion of the bubble contamination in the propellant is less than 2.0 vol%. The uncured propellant is

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mixed and cast under a vacuum in order that the air may not be introduced into the uncured propellant. However, when the volume of HTPB is smaller than that of void in AP powder, it is likely that the proportion of the bubble contamination in the propellant is more than 2.0 vol%. Because of the above two requirements for the preparation of AP/HTPB composite propellant, the upper limit content of AP contained propellant,  $\phi_{max}$  (wt%), exists<sup>2),3)</sup>.

The burning rate and the specific impulse are the very important properties of propellant. When a propellant is prepared from a certain AP sample, the burning rate and the specific impulse increase with increasing the AP content and these values of the propellant containing  $\phi_{max}$  are maximum. If  $\phi_{max}$  could be estimated, the maximum burning rate and specific impulse could be determined easily. It is an important subject for the design of AP/HTPB composite propellant to make the estimation of  $\phi_{max}$ . It is reported that  $\phi_{max}$  can be estimated by a void fraction,  $\varepsilon_{max}$  (-) at the loose packing of powder<sup>2),3)</sup>. The value of  $\varepsilon_{max}$  varies with the change of humidity, temperature, material of cylinder, drop height of powder, etc<sup>4)</sup>. That is to say,  $\varepsilon_{max}$  is dependent greatly on the measuring condition. It is difficult to measure  $\varepsilon_{max}$  under strict constant conditions. Therefore, it is not easy to estimate  $\phi_{max}$  by use of  $\varepsilon_{max}$ . It is necessary to find out the universal powder property which can estimate  $\phi_{max}$ .

Mori and Ototake<sup>5)</sup> reported that this relationship between the relative viscosity,  $\eta_r$  (-), and the volume fraction of solid,  $\phi_v$  (vol%), of concentrated suspension can be expressed by the following equation.

$$\eta_r = 1 + \frac{k}{(100/\phi_v) - (1/\phi_{vc})} \quad (1)$$

where  $k$  and  $\phi_{vc}$  are constant and are dependent on the particle property of the dispersoid. If the relationship of the concentrated AP/HTPB suspension could follow Eq.(1) and the dominating powder property for  $k$  and  $\phi_{vc}$  could be found out, it is expected that  $\phi_{max}$  could be obtained as a function of the particle property. In this study, an attempt was made to find out the universal powder property which can estimate  $\phi_{max}$ , except

$\varepsilon_{max}$ . As a result of this study, it is found that  $\phi_{max}$  can be estimated from specific surface area measured by the air-permeability method. This paper gives the experimental results for the determination.

## 2. Experimental

### 2.1 Samples

AP samples used in this study are shown in Table 1. Samples A-E were prepared by grinding commercial AP for 5, 10, 20, 30 and 40 minutes with a vibration ball mill. The shape of samples A-E was spherical and the width of particle size distribution was wide (5-200  $\mu$ m). Sample F was prepared by shifting ground AP. A sonic sifter (SW-20, Tutui rikagaku) was used and the opening of the sieve was 38  $\mu$ m. The shape of sample F was spherical and the width of particle size distribution was narrow. Samples G-Q were prepared by the spray-drying method<sup>6)-8)</sup>. Samples R and S were prepared by the freeze-drying method<sup>9)</sup>. The shapes of samples G-I, samples J-O and samples P-S were spherical, porous and hollow, respectively. The width of particle size distribution of samples G-S was narrow (0.5-17  $\mu$ m). Mean volume-surface diameter,  $D_{vs}$  ( $\mu$ m), on the number basis of AP used in this study are shown in Table 1.  $D_{vs}$  is in the range of 1.6 to 110  $\mu$ m.

HTPB R-45M (ARCO Co.) was used as a binder. HTPB was cured with isophorone diisocyanate (IPDI). IPDI was added to 8 wt% of HTPB<sup>10)</sup>.

In order to determine  $\phi_{max}$ , a propellant sample should be prepared with the optimum operating condition. The optimum operating condition was studied in Ref.10. In Refs.2 and 3, the preparation of the propellant sample was determined on the basis of Ref.10 and  $\phi_{max}$  was measured. The propellant sample was prepared according to Refs.2 and 3 in this study.

### 2.2 Measurement of specific surface area

Specific surface area was measured by the BET absorption method or air-permeability method. Specific surface area,  $S_{nb}$  (m<sup>2</sup>/kg), measured by the BET absorption method is determined by an amount of gas absorbed on the surface of a particle. Nitrogen was used as the absorption gas for  $S_{nb}$ . Specific surface area,  $S_{nb}$  (m<sup>2</sup>/kg),

Table 1 Particle characteristics of AP used in this study and  $\phi_{max}$ .

Symbol	Sample	Shape	$D_{vs}$ ( $\mu\text{m}$ )	$S_{wb}$ ( $\text{m}^2/\text{kg}$ )	$S_{wp}$ ( $\text{m}^2/\text{kg}$ )	$\phi_{max}$ (wt%)
A	AP ground in vibration mill for 5minutes	Spherical	110	60	40	85
B	AP ground in vibration mill for 10minutes	Spherical	70	80	50	85
C	AP ground in vibration mill for 20minutes	Spherical	50	150	100	84
D	AP ground in vibration mill for 30minutes	Spherical	40	190	150	83
E	AP ground in vibration mill for 40minutes	Spherical	40	210	180	83
F	38 $\mu\text{m}$ passd	Spherical	20	380	260	82
G	AP prepared by the spray-drying method	Spherical	1.6	2000	1060	75
H	AP prepared by the spray-drying method	Spherical	2.0	1800	450	76
I	AP prepared by the spray-drying method	Spherical	2.6	1700	780	77
J	AP prepared by the spray-drying method	Porous	6.3	2100	1010	73
K	AP prepared by the spray-drying method	Porous	7.5	900	840	80
L	AP prepared by the spray-drying method	Porous	7.1	1500	940	77
M	AP prepared by the spray-drying method	Porous	4.1	1900	990	77
N	AP prepared by the spray-drying method	Porous	3.1	2100	900	74
O	AP prepared by the spray-drying method	Porous	3.9	1600	860	76
P	AP prepared by the spray-drying method	Hollow	2.7	3100	1200	72
Q	AP prepared by the spray-drying method	Hollow	2.9	2600	1160	72
R	AP prepared by the freeze-drying method	Hollow	3.9	1400	610	80
S	AP prepared by the freeze-drying method	Hollow	11.2	700	370	81

measured by the air-permeability method is determined by the permeability of air when it passes through a packed bed of powder.  $S_{wp}$  is influenced by the void fraction of a packed bed<sup>11)</sup>. According to a preliminary experiment, it was found that  $S_{wp}$  of AP used isn't influenced by the void fraction when it is in the range of 0.3 to 0.5. In this study,  $S_{wp}$  was measured by use of the AP packed bed, of which the void fraction was in the range of 0.3 to 0.5. The physical meaning of  $S_{wp}$  is different from that of  $S_{wb}$ .

### 2. 3 Measurement of viscosity

A viscosity of AP/HTPB suspension was measured by a flow tester in this study. The measuring temperature was  $333 \pm 1$  K, since this was the temperature used for mixing and casting of AP/HTPB suspension in this study<sup>1),3),10)</sup>. According to the result of a preliminary experiment, the flow curve of the AP/HTPB suspension prepared in this study rises in a concave curve. This indicates that the AP/HTPB suspension is a pseudo-plastic fluid. A viscosity of the pseudo-plastic fluid is dependent on a share rate. In this study, the end correction in the capillary flow<sup>12)</sup> was conducted at the share rate which was  $500s^{-1}$ .

### 2. 4 Measurement of burning rate

The burning rate was measured in a chimney type strand burner which was pressurized with nitrogen at  $288 \pm 1.5$  K. The range of pressure was from 0.5 MPa to 8 MPa. The strand sample was ignited by an electrically heated nichrome wire attached on the top of each strand sample. Two fuse wires were threaded through the strand sample at 25mm distance. The fuse wire was cut as soon as the burning surface passed through the fuse wire. The burning rate was calculated with the cutoff period of two fuses.

## 3. Results and Discussion

### 3. 1 Specific surface area

$S_{wb}$  and  $S_{wp}$  were measured and the results are shown in Table 1. The ranges of  $S_{wb}$  and  $S_{wp}$  are  $60-3100m^2/kg$  and  $40-1200m^2/kg$ , respectively.  $S_{wp}$  is 0.3-0.8 times value as  $S_{wb}$ . The relationship between  $S_{wb}$  and  $S_{wp}$  is shown in Fig.1.  $S_{wp}$  increases with increasing  $S_{wb}$ . Since the physical

meaning of  $S_{wp}$  is different from that of  $S_{wb}$ ,  $S_{wp}$  is not in agreement with  $S_{wb}$ . However,  $S_{wb}$  and  $S_{wp}$  are associated.

### 3. 2 Upper limit content of AP contained in propellant

It's required for the preparation of AP/HTPB composite propellant that uncured propellant has a viscosity required for casting and the proportion of bubble contamination in propellant is less than 2.0 vol%<sup>2),3)</sup>.  $\phi_{max}$  was the largest AP content in the propellants, which satisfy the above two requirements for the preparation of AP/HTPB composite propellant.  $\phi_{max}$  was determined in the following way experimentally according to Refs.2 and 3. First, the uncured propellants are prepared at various AP contents and the largest AP content in the uncured propellants which have the viscosity required for casting is determined. Second three lots of propellants were prepared with the same AP content. The AP content was decreased by 1wt% from the largest AP content in the uncured propellants which have the viscosity required for casting. When the proportion of bubble contamination in the propellant is less than 2.0vol%, the reproducible burning rate is obtained<sup>1)</sup> and the relationship between burning rate and pressure follows Vieille's law according to the experience in this laboratory.

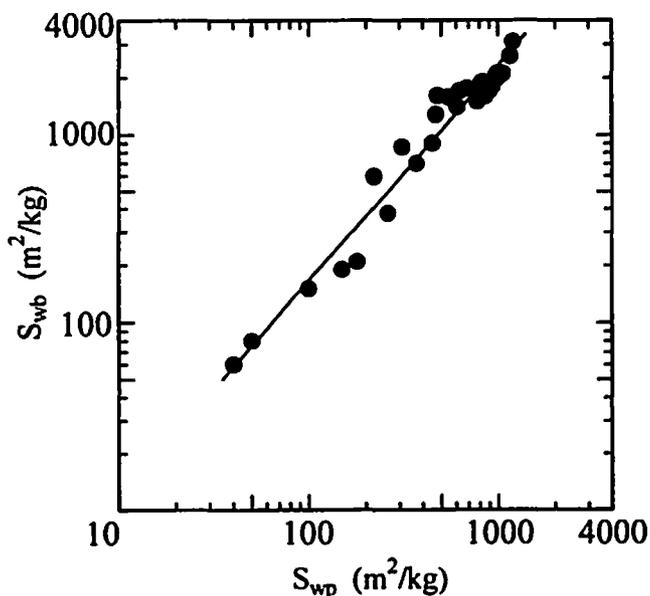


Fig. 1 Relationship between  $S_{wb}$  and  $S_{wp}$

As mentioned above, three propellants were prepared at the same AP content. When the burning rate of three propellants is reproducible and the relationship between the burning rate of their propellants and pressure follows Vieille's law, it's judged that the burning rate of the propellant isn't influenced by the bubble contamination, that is, the proportion of bubble contamination in propellant is less than 2.0vol%. In this study,  $\phi_{max}$  was determined as the largest AP content in the propellants, of which the reproducibility of the burning rate can be obtained and of which relationship between burning rate and pressure follows Vieille's law. The propellants were prepared from AP samples shown in Table 1 and burning rates of their propellants were measured. For example, concerning sample R, the relationships between the burning rate and pressure of propellants containing 79, 80 and 81wt%AP are plotted in Fig.2. When the AP content is less than 80 wt%, the reproducibility of the burning rate can be obtained and the burning rate increases lineally in a plot of  $\ln(\text{burning rate})$  versus  $\ln(\text{pressure})$ , that is, the relationships follow Vieille's law. When the AP content is 81wt%, the reproducibility of the burning rate can't be obtained and the burning rate does not increase lineally in a plot of  $\ln(\text{burning rate})$  versus  $\ln(\text{pressure})$ , that is, the relationship does not follow Vieille's law. These results indicate that  $\phi_{max}$  of the propellant prepared from sample R is 80wt%. With respect to the propellants prepared from AP samples shown in Table 1,  $\phi_{max}$  was determined in the same way as above. The results of  $\phi_{max}$  are shown in Table 1.  $\phi_{max}$  is in the range of 72wt% to 85wt%.  $\phi_{max}$  of samples A and B is maximum, and  $\phi_{max}$  of samples P and Q is minimum.

### 3. 3 Viscosity of AP/HTPB suspension

The AP/HTPB suspension was contained above 50vol%AP in this study. The viscosity of the AP/HTPB suspension was measured and the relationship between  $\eta_r$  and  $\phi_v$  was investigated. AP/HTPB suspension was not added IPDI. Some results are shown in Fig.3.  $\eta_r$  increases with increasing  $\phi_v$ .

Mori and Ototake<sup>5)</sup> reported that this relationship of concentrated suspension can be expressed

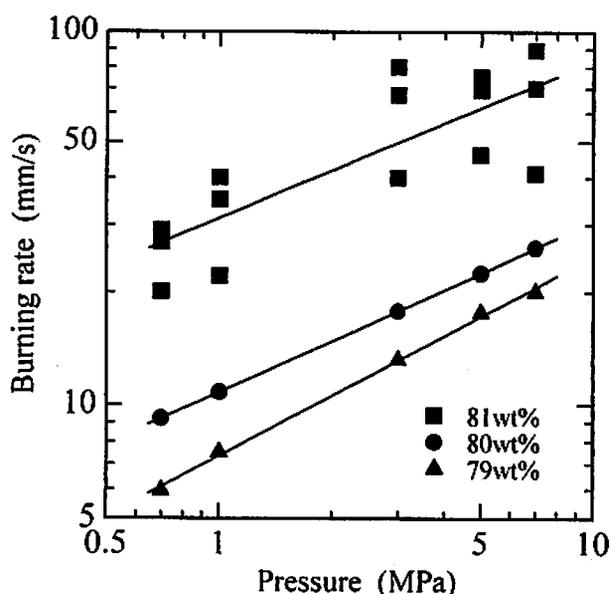


Fig. 2 Burning rate characteristics of experimentally prepared propellants from sample R

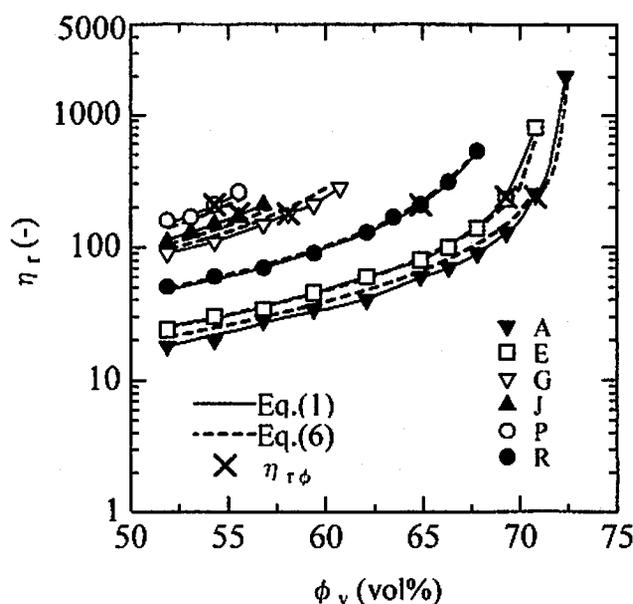


Fig. 3 Relationship between  $\eta_r$  and  $\phi_v$

by Eq.(1).  $k$  and  $\phi_{vc}$  in Eq.(1) are constant and are dependent on the property of the suspended particles. This relationship of the AP/HTPB suspension prepared from spherical AP, of which particle size distribution is narrow, follows Eq.(1)<sup>3),13),14)</sup>. In this study, Some porous or hollow AP samples and some spherical AP samples, of which particle size distribution is wide, were used. It was investigated that the relationship of the AP/HTPB suspension prepared from AP shown in Table 1 could follow Eq.(1).

Eq.(1) can be converted to the following equation.

$$\frac{100}{\phi_v} = \frac{k}{\eta_r - 1} + \frac{1}{\phi_{vc}} \quad (2)$$

According to Eq.(2), the plot of  $1/(\eta_r - 1)$  against  $100/\phi_v$  gives a straight line.  $k$  and  $\phi_{vc}$  can be obtained from the slope and intercept of the straight line, respectively. On the basis of the data in Fig.3, the plot of  $1/(\eta_r - 1)$  against  $100/\phi_v$  is shown in Fig.4. This relationship of the AP/HTPB suspension could be expressed by a straight line.  $k$  and  $\phi_{vc}$  were calculated and the results are shown in Table 2. The relationship between  $\eta_r$  and  $\phi_v$ , obtained by substituting  $k$  and  $\phi_{vc}$  shown in Table 2 into Eq.(1), is represented by a solid line in Fig.3. The measured values can be approximated by the solid line. It is found that the relationship between  $\eta_r$  and  $\phi_v$  of the AP/HTPB suspensions prepared in this study can be represented by Eq.(1).

$\phi_{max}$  is the percentage by weight and  $\phi_v$  in Eq.(1) is the percentage by volume.  $\phi_{max}$  which was converted into the percentage by volume was designated as  $\phi_{v,max}$ . And  $\eta_r$  of AP/HTPB suspension containing  $\phi_{v,max}$  was designated as  $\eta_{r,\phi}$ . As mentioned above, the relationship between  $\eta_r$  and  $\phi_v$  of the AP/HTPB suspension can be represented by Eq.(1). This indicates that the relationship between  $\eta_{r,\phi}$  and  $\phi_{v,max}$  can exist on the curve presented by Eq.(1). Therefore,  $\eta_{r,\phi}$  can be represented by the following equation.

$$\eta_{r,\phi} = 1 + \frac{k}{(100/\phi_{v,max}) - (1/\phi_{vc})} \quad (3)$$

$\phi_{max}$  was determined in section 3.2 already.  $\eta_{r,\phi}$  was displayed with  $\times$  in Fig.3. It is found that  $\eta_{r,\phi}$  is almost constant, approximately 200. Since  $\eta_{r,\phi}$  is almost constant, Eq.(3) indicates that  $\phi_{v,max}$  is dependent on  $k$  and  $\phi_{vc}$ .  $k$  and  $\phi_{vc}$  relate to the particle property of the dispersoid and especially  $k$  is dependent on  $D_{vs}$  and specific surface area<sup>5)</sup>. It is expected that  $k$  and  $\phi_{vc}$  could be obtained as a function of  $D_{vs}$  and/or specific surface area, if  $\phi_{vc}$  would be also dependent on  $D_{vs}$  and/or specific surface area.

The relationship between  $k$  or  $\phi_{vc}$  and  $D_{vs}$  is shown in Fig.5. As  $D_{vs}$  increases,  $k$  decreases and  $\phi_{vc}$  increases. However, these relationships were

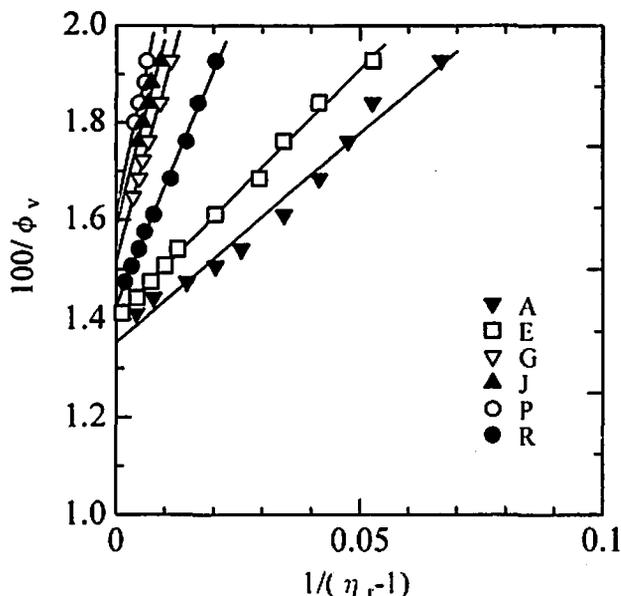


Fig. 4 Relationship between  $100/\phi_v$  and  $1/(\eta_r - 1)$

Table 2 Results of  $k$  and  $\phi_{vc}$

Symbol	$k$ (-)	$\phi_{vc}$ (-)
A	10	0.732
B	11	0.731
C	12	0.728
D	13	0.724
E	14	0.723
F	17	0.712
G	36	0.658
H	35	0.662
I	31	0.674
J	39	0.646
K	18	0.711
L	28	0.683
M	30	0.678
N	38	0.653
O	32	0.670
P	47	0.621
Q	44	0.627
R	23	0.699
S	20	0.704

scattered extensively. The shape of some AP samples used in this study were porous or hollow. It can be supposed that the void in porous or hollow AP particles would influence viscosity.  $D_v$  vs  $k$  can't consider the void in porous or hollow AP particles, because  $D_v$  vs  $k$  is measured on the basis of the projective figure of particle. Therefore, it can be considered that these relationships are scattered.

When the particle diameter is constant, specific surface area of porous or hollow particle is larger than that of spherical particle. It is supposed that the better relationship between  $k$  or  $\phi_{vc}$  and specific surface area could be obtained, compared with  $D_v$ .  $S_{vb}$  and  $S_{vp}$  were adopted as specific surface area in this study.

The relationship between  $k$  or  $\phi_{vc}$  and  $S_{vb}$  is shown in Fig.6. As  $S_{vb}$  increases,  $k$  increases and  $\phi_{vc}$  decreases. These relationships showed some scattering. Porous or hollow AP had very small voids and cracks which could not be charged with HTPB.  $S_{vb}$  is determined on the basis of the area occupied by nitrogen molecule adsorbed on the surface of particle. This supported that  $S_{vb}$  would involve the surface area of the voids and cracks which could not be charged with HTPB. Therefore, it can be considered that these relationships are somewhat scattered.

The relationship between  $k$  or  $\phi_{vc}$  and  $S_{vp}$  is shown in Fig.7. As  $S_{vp}$  increases,  $k$  increases and  $\phi_{vc}$  decreases. These relationships show little scattering.  $S_{vp}$  is determined on the basis of the air permeability of packed bed. It is supposed that the surface area of very small voids and cracks which could not be charged with HTPB could be neglected, since air passes through in the packed bed of AP powder so as to slide on the surface of an AP particle. It can be considered that  $S_{vp}$  would relate to the surface area of an AP particle wetted with HTPB at the mixing of AP and HTPB. Therefore, it was supposed that  $S_{vp}$  was closely related to  $k$  and  $\phi_{vc}$ . When  $S_{vp}$  ranges from 40m<sup>2</sup>/kg to 1200m<sup>2</sup>/kg, the relationship between  $k$  or  $\phi_{vc}$  and  $S_{vp}$  can be represented by the following equation.

$$k = 10 + 0.015S_{vp} + 1.2 \times 10^{-5}S_{vp}^2 \quad (4)$$

$$\phi_{vc} = 0.728 - 1.17 \times 10^{-5}S_{vp} - 6.20 \times 10^{-8}S_{vp}^2 \quad (5)$$

By substituting Eqs.(4) and (5) into Eq.(1), the following equation can be obtained.

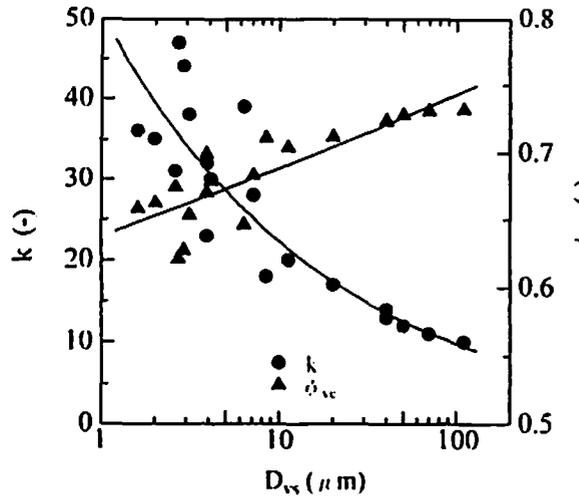


Fig. 5 Relationship between  $k$  or  $\phi_{vc}$  and  $D_v$

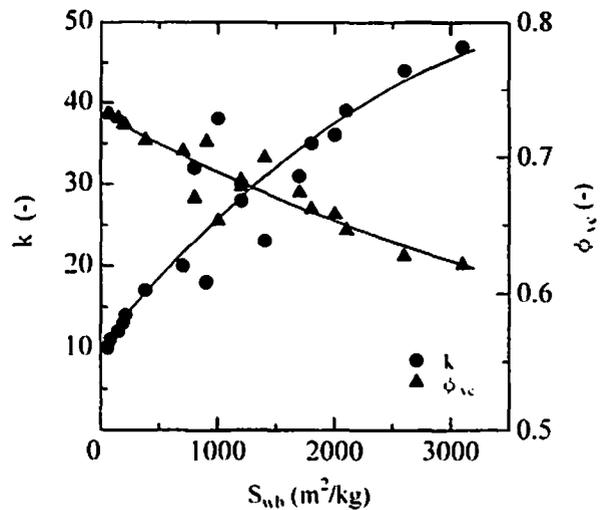


Fig. 6 Relationship between  $k$  or  $\phi_{vc}$  and  $S_{vb}$

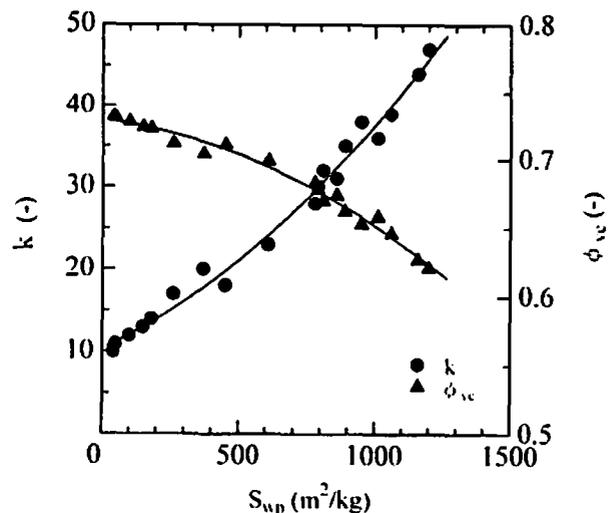


Fig. 7 Relationship between  $k$  or  $\phi_{vc}$  and  $S_{vp}$

$$\eta_r = 1 + \frac{10 + 0.015S_{wp} + 1.2 \times 10^{-5}S_{wp}^2}{(100/\phi_v) - (1/(0.728 - 1.17 \times 10^{-5}S_{wp} - 6.20 \times 10^{-8}S_{wp}^2))} \quad (6)$$

The relationship between  $\eta_r$  and  $\phi_v$  obtained by Eq. (6) is represented by a broken line in Fig. 3. The broken line is in almost agreement with the measured values and the solid line. It is found that  $\eta_r$  can be represented as the function of  $\phi_v$  and  $S_{wp}$  in this study.

It is expected that  $\phi_{max}$  could be represented as the function of  $S_{wp}$ . Because this relationship between  $\eta_r$  and  $\phi_v$  of the AP/HTPB suspension can follow Eq. (1), and  $k$  and  $\phi_{vc}$  can be obtained as a function of  $S_{wp}$ .

### 3.4 Estimation of upper limit content of AP contained propellant

As described in section 3.3, it is expected that  $\phi_{max}$  could be expressed by  $S_{wp}$ . On the basis of Table 1, the relationship between  $\phi_{max}$  and  $S_{wp}$  is plotted in Fig. 8.  $\phi_{max}$  increases with decreasing  $S_{wp}$ . The plot  $\phi_{max}$  against  $S_{wp}$  is found to be linear. AP samples shown in Table 1 were unimodal. In general AP which is mixed the samples with different sizes is used as an oxidizer of AP/HTPB composite propellant. With regard to bimodal AP, the relationship between  $\phi_{max}$  and  $S_{wp}$  was investigated. Bimodal AP samples were prepared by mixing samples A, G, J, P and R. The composition and  $S_{wp}$  of bimodal AP samples are shown in Table 3.  $\phi_{max}$  of the propellant prepared from bimodal AP were examined and the result is shown in Table 3. The relationship between  $\phi_{max}$  and  $S_{wp}$  of bimodal AP samples is also plotted with  $\blacktriangle$  in Fig. 8. The relationship of bimodal AP agrees with that of unimodal AP. When  $S_{wp}$  ranges from 40 m<sup>2</sup>/kg to 1200 m<sup>2</sup>/kg, the relationship between  $\phi_{max}$  and  $S_{wp}$  can be represented by the following equation.

$$\phi_{max} = 85 - 0.01S_{wp} \quad (7)$$

The error between  $\phi_{max}$  calculated from Eq. (7) and the experimental value was within  $\pm 2\%$ . It is found that  $\phi_{max}$  can be estimated from  $S_{wp}$  by use of Eq. (7).

As mentioned at section 2.1, the shape of AP samples used in this study is spherical, porous or hollow. The width of particle size distribution is unimodal or bimodal.  $D_{vs}$  is in the range of

Table 3  $S_{wp}$  and  $\phi_{max}$  of bimodal AP

AP	$S_{wp}$ (m <sup>2</sup> /kg)	$\phi_{max}$ (wt%)
G:A=80wt%:20wt%	480	80
J:A=60wt%:40wt%	470	80
P:A=50wt%:50wt%	550	80
R:A=40wt%:60wt%	220	83
R:A=60wt%:40wt%	310	82
G:R=80wt%:20wt%	820	78
G:R=60wt%:40wt%	690	79
G:R=50wt%:50wt%	630	79

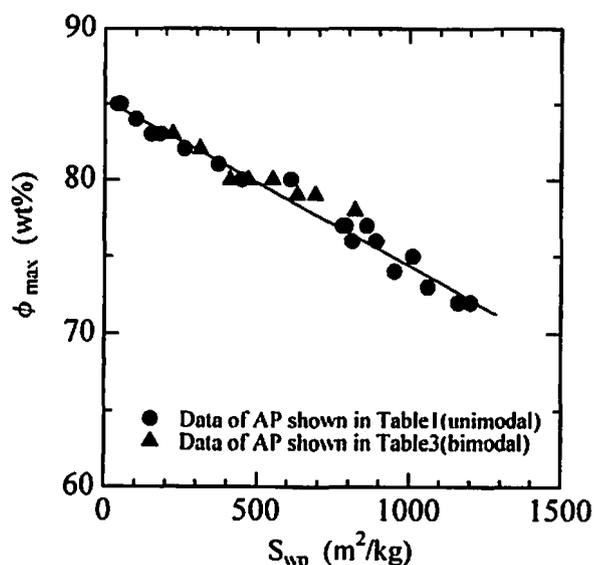


Fig. 8 Relationship between  $\phi_{max}$  and  $S_{wp}$

1.6 to 110  $\mu$ m. The ranges of  $S_{wb}$  and  $S_{wp}$  are 60–3100 m<sup>2</sup>/kg and 40–1200 m<sup>2</sup>/kg, respectively. Therefore, the AP samples used in this study had multiple powder characteristics. In spite of multiple powder characteristics of the AP used,  $\phi_{max}$  can be represented as a function of  $S_{wp}$  as a whole. This indicates that the influences of the multiple powder characteristics of AP on  $\phi_{max}$  could be expressed by  $S_{wp}$  inclusively.

It is reported that  $\phi_{max}$  can be estimated by  $\epsilon_{max}$ <sup>2),3)</sup>.  $\epsilon_{max}$  is dependent greatly on measuring conditions such as humidity, temperature, cylinder material, drop height of powder, etc.<sup>4)</sup>. It is difficult to measure  $\epsilon_{max}$  under strict constant conditions. Therefore, it is not easy to estimate  $\phi_{max}$  by the use of  $\epsilon_{max}$ . On the other hand,  $S_{wp}$  is influenced by the void fraction of a packed

bed<sup>1)</sup>.  $S_{wp}$  of AP is reproducible when the void fraction is in the range of 0.3 to 0.5, as mentioned in section 2.2. It is found that  $\phi_{max}$  can be estimated easily and accurately by the use of  $S_{wp}$ , compared with  $\varepsilon_{max}$ .

In this study HTPB widely used was adopted as a binder. It was predicted for composite propellant prepared from another binder that  $\phi_{max}$  could be represented as a function of  $S_{wp}$ .

#### 4. Conclusions

For the preparation of ammonium perchlorate (AP)/ hydroxyl-terminated polybutadiene (HTPB) composite propellant, it is required that the uncured propellant has the viscosity required for casting and the proportion of the bubble contamination in the propellant is less than 2.0 vol%. Because of these requirements, an upper limit content of AP contained propellant,  $\phi_{max}$  (wt%), exists. It is necessary for the design of AP/HTPB composite propellant to estimate  $\phi_{max}$ . From the standpoint of flow characteristics of AP/HTPB suspension, an attempt was made to find out the dominating powder property of AP for  $\phi_{max}$ .

Concerning AP/HTPB suspensions prepared in this study, the relationship between the relative viscosity,  $\eta_r$  (-), and the volume fraction,  $\phi_v$  (vol%), of AP can be represented by the Mori-Ototake equation.  $k$  and  $\phi_{vc}$  which are constant in the Mori-Ototake equation are dependent on specific surface area,  $S_{wp}$  (m<sup>2</sup>/kg), measured by the air-permeability method. It is found that  $\eta_r$  of AP/HTPB suspension can be represented as the function of  $\phi_v$  and  $S_{wp}$  and  $\eta_r$  of suspension containing  $\phi_{max}$  was almost constant.

The plot of  $\phi_{max}$  against  $S_{wp}$  is found to be linear.  $\phi_{max}$  increases with decreasing  $S_{wp}$ . When  $S_{wp}$  ranges from 40m<sup>2</sup>/kg to 1200m<sup>2</sup>/kg, The relationship between  $\phi_{max}$  and  $S_{wp}$  can be represented by the following equation.

$$\phi_{max} = 85 - 0.01S_{wp}$$

It is found that  $\phi_{max}$  can be estimated by  $S_{wp}$ .

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# AP/HTPB系コンポジット推進薬に混入できるAP含有率の上限界の推算

## — AP/HTPB分散系の流動性に基づく一考察 —

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過塩素酸アンモニウム(AP)/末端水酸基ポリブタジエン(HTPB)系推進薬は製造上の条件があるために、推進薬中に混入可能なAP含有率の上限界、 $\phi_{max}$ (wt%)が存在する。AP/HTPB分散系の流動性の観点から、 $\phi_{max}$ を推算できるようなAPの粒子特性値を見出すことを試みた。なお、本実験では球状、多孔質と中空のAPを用いた。その結果は次のようにまとめられる。

1. 本実験で調製されたAP/HTPB分散系において、相対粘度、 $\eta_r(-)$ と固体容積濃度、 $\phi_v(\text{vol}\%)$ の関係は、森・乙竹の式で表すことができた。森・乙竹の式の定数である $k$ と $\phi_{vc}$ は、APの空気透過法で測定された比表面積、 $S_{sp}(\text{m}^2/\text{kg})$ で表すことができ、 $\eta_r$ は $\phi_v$ と $S_{sp}$ の関数で表せることがわかった。
2.  $\phi_{max}$ で調製されたAP/HTPB分散系の $\eta_r$ はほぼ一定であった。
3.  $\phi_{max}$ は $S_{sp}$ が小さいほど大きかった。 $\phi_{max}$ と $S_{sp}$ の関係は、次式によって表すことができた。

$$\phi_{max} = 85 - 0.01S_{sp} \quad (40\text{m}^2/\text{kg} \leq S_{sp} \leq 1200\text{m}^2/\text{kg})$$

$\phi_{max}$ は用いたAPの $S_{sp}$ を測定すれば上式でほぼ正確に推算できることがわかった。

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