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微小重力環境を用いたポリプロピレンの燃焼速度定数の決定法

Determination strategy of the burning rate constant of Polypropylene (PP) in a microgravity environment

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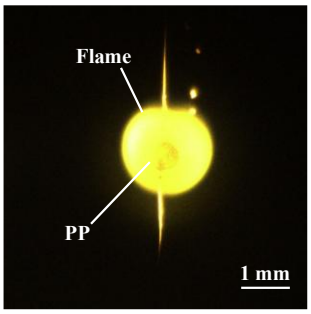
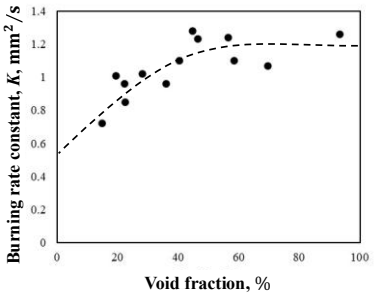
Abstract: This study investigates the burning characteristics of polypropylene (PP) spheres under microgravity conditions, focusing on the effect of the initial void fraction inside the sample on the burning rate constant. Combustion experiments were conducted using a 50-meter droptower, where spherical samples with various void fractions were prepared by coating ceramic beads with PP film. A total of 13 samples with different void fractions were tested. The burning process was initiated using a small amount of ignition gel, ensuring quasi-steady spherical flame formation within the short duration of microgravity. Results revealed a positive correlation between the burning rate constant and void fraction in the range of 0–40%, while the value plateaued beyond 40%. This behavior contrasts with previous studies on PMMA, which showed a continuous increase in burning rate with bubble content. The differences are attributed to PP's higher thermal decomposition temperature and lower surface tension, which suppress both bubble retention and the impact of bubble rupture on combustion dynamics. These findings provide new insight into polymer combustion mechanisms in microgravity and offer a practical method for determining burning rate constants of non-PMMA polymers.





Keywords: **Polypropylene, Microgravity, d-square law, Effect of void fraction, Droptower experiment**

1. Introduction

In recent years, space development has expanded beyond large-scale national projects, with an increasing number of private companies entering the field[1)]. Along with this trend, attention has been directed toward reducing costs in space applications while ensuring safety. Polymers, with their advantageous properties such as low cost, light weight, high durability, and excellent insulating performance, are considered a promising means to significantly reduce costs in space utilization[2)]. However, polymers are inherently flammable, and

eliminating concerns regarding their fire hazards is essential. Since the behavior of solid materials may alter significantly under various gravitational conditions[3], in general, it is crucial to evaluate their combustion characteristics under microgravity.

Previous studies[3-6] have reported the features of numerous microgravity combustion using spherical polymethyl methacrylate (PMMA). These results have clearly shown complex dynamic behaviors during combustion process, such as shell rupture and the formation and bursting of bubbles. Nevertheless, the burning rate constant, defined by the d^2-law and known as one of important burning character of the specimen, remains nearly constant. Yet, some studies have also reported that the burning rate constant increases due to the presence of initial bubbles within the sample[6], suggesting that the in-situ bubble generation process inside the specimen during the burning may affect the obtaining the burning rate constant. This fact implies the difficulty to "define" the burning rate constant of the tested polymer sample as material constant unless we have certain way to control the bubble generation during the burning process. Recently, interesting methodology to estimate the "pure-material-defined burning rate constant (without any dynamic effect likely bubble generation inside the specimen)" using extrapolation of set of burning data of various initial voids specimen for PMMA and found its effectiveness[7,8]. However, as is well-known, PMMA is a particularly unique and straightforward sample because its main pyrolysis product is consistently the monomer (MMA), even under a wide range of heating conditions[9]. Hence, it is important to verify whether the previously proposed method is valid for other type of polymer beyond PMMA. This is main objective of this study.

In this study, polypropylene (PP), which is commonly used on the International Space Station as packaging material, was selected as a representative example of a different polymer. Spherical PP samples with varying internal bubble contents were prepared, and the effect of these bubbles on the burning rate constant under microgravity was comparatively investigated.

2. Experimental method

2.1. Experimental facilities and equipment

The combustion experiments in this study were conducted under microgravity using the 50-meter drop tower "COSMOTORRE," operated by the Hokkaido Space Science Creation Center. A combustion chamber with dimensions of 250 mm (width) × 245 mm (depth) × 250 mm (height) was installed inside the drop capsule, where the combustion experiments were performed. An overview of the experimental apparatus is mostly the same as one used in our previous study[6] so that the brief explanation is made here.

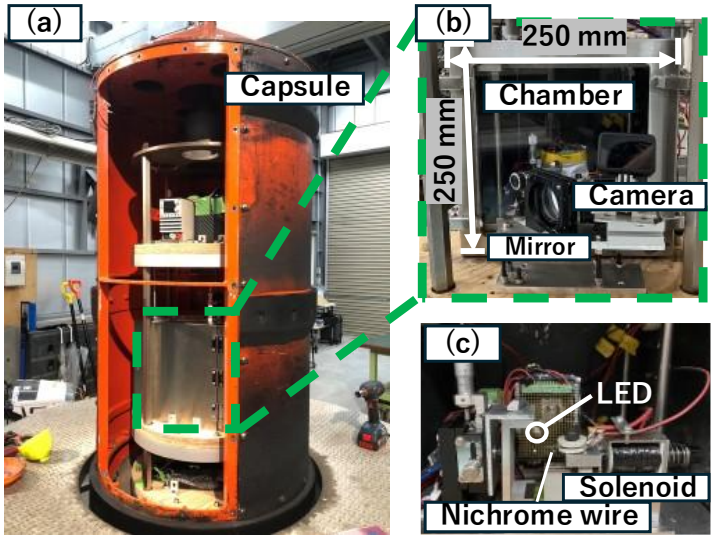


Figure 1. Experimental apparatus, (a) Drop tower capsule, (b) Chamber and visualization devices, (c) Inside the combustion chamber.

A photograph of the inside of the chamber are shown in **Fig. 1**.

The combustion process was recorded through a front-facing observation window using a CMOS camera (DFK33UX273, 1440×1080 pixels, 25 fps) equipped with a micro lens (VS-LDA25). An LED was placed behind the sample and flashed at 12.5 Hz, allowing the camera to alternately capture images of the sample and direct

flame in each frame during combustion. Ignition was achieved using a nichrome wire. To reduce ignition delay, a small amount of ignition aid was applied to the surface of the sample. The atmosphere inside the chamber was maintained at ambient pressure (101.0 kPa), room temperature, and an oxygen concentration of 21 vol%.

A timeline of the experimental procedure is shown in **Fig. 2**, which is especially re-designated for PP burning test. Upon separation of the drop capsule from the top of the tower, it enters a 2.8-second free fall. Heating of the nichrome wire, LED flashing, and camera recording were all automatically triggered by the signal indicating the start of separation and descent. Upon receiving the signal, the nichrome wire was preheated while positioned away from the sample. Simultaneously, a solenoid actuator mounted beneath the heater base was activated to rapidly move the wire toward the sample, thereby igniting it.

Once the descent signal was received, power to both the nichrome wire and the solenoid was cut off, and the ignition system was retracted from the sample. This ensured that the subsequent combustion under microgravity proceeded with minimal external interference. The amount of ignition aid applied was kept below the sample mass and did not affect the combustion behavior of the PP.

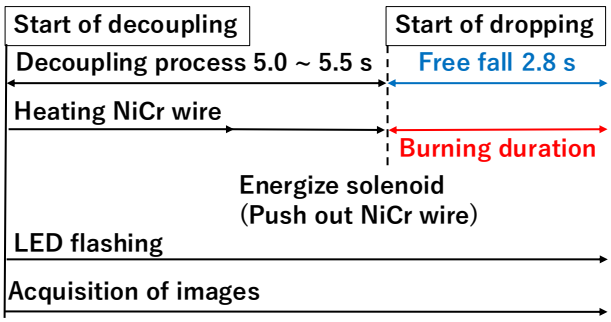


Figure 2. System time sequence

2.2. Sample for combustion test (PP)

The combustion samples used in this study are shown in **Fig. 3**. Each spherical sample was constructed by placing a ceramic bead with a diameter of less than 1 mm on a 14 μm-diameter SiC fiber (Hi-Nicalon™, Nippon Carbon Co., Ltd.), and layering PP over it. The PP layer was fabricated by cutting 0.2 mm-thick PP craft film (Acrylic Sunday Co., Ltd.) into 2 mm × 2 mm pieces, which were then melted and thermally bonded together to form the spherical shape.

While this fabrication method allows for a qualitative trend in which a shorter distance between the sample and the heater leads to higher bubble content, it does not enable precise and reproducible control of the bubble conditions in a quantitative manner. This limitation is likely due to the fact that the final solidification process is based on natural cooling. However, although clear classification is not achievable, this method still allows for the preparation of samples with various levels of bubble content.

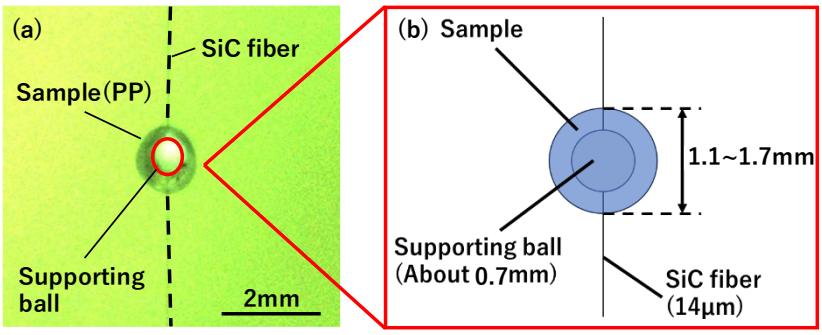


Figure 3. Burning sample, (a) Actual sample, (b) Image of sample

To quantitatively evaluate the influence of bubble rupture on the burning rate during PP sphere combustion, the initial bubble content within the sample was expressed as a void fraction, defined in **Eq. 1**.

$$\text{Void fraction} = \frac{V_e - V_t}{V_t} \times 100 \quad (1)$$

where V_t was calculated as the theoretical volume from the sample mass, and V_e was calculated as the experimental volume from the apparent diameter. Note that this value can exceed 100% when the initial void exceed the total volume of solid under the present definition.

Figure 4 summarizes the void fraction in the sample (i.e., the volume fraction of voids) observed within the samples prepared using this method. As shown in the figure, when the void fraction is grouped in 20% intervals, at least one sample is available within each range. Therefore, this approach enables the evaluation of the burning rate constant as a function of void fraction.

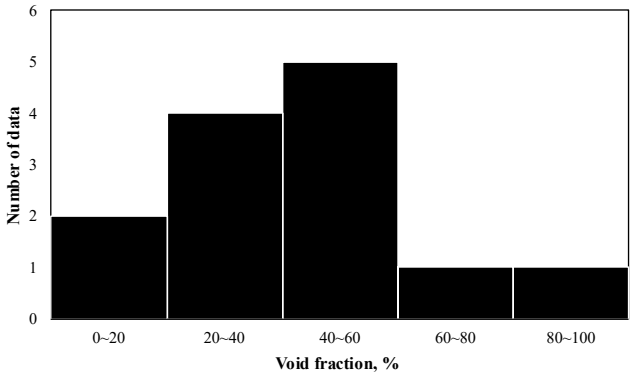


Figure 4. Void fraction distribution of fabricated samples

3. Results and discussion

3.1. Appearance of 1-D flame

Figure 5 presents the typical flame structure and combustion behavior of the polypropylene (PP) sample. In the second frame, a weak flame generated by the igniter gel envelops the sample, promoting vaporization of the PP. Approximately 0.16 seconds later, sustained combustion is initiated. In the third frame, a distinct spherical flame structure is observed, indicating a transition from initial combustion under normal gravity to the microgravity-specific spherical flame regime. **Figure 6** shows two types of dynamic flame behavior previously observed during combustion of polymethyl methacrylate (PMMA) spheres: rupture of the soot shell and bubble rupture. A similar phenomenon is observed during the combustion of the PP sample in **Fig. 5**, where a transient brightening of the flame occurs locally. This is presumed to result from soot shell rupture, as reported in previous studies on PMMA combustion[3-5,7]. Despite these local events, the overall flame maintains a spherical shape throughout the combustion process, indicating stable burning. In contrast, the pronounced flame surface deformation associated with bubble rupture, which is characteristic of PMMA combustion, is not clearly observed during PP combustion. No rapid flame expansion or distortion, such as that shown in **Fig. 6**, was identified in the case of PP. These observations suggest that in the combustion of PP spheres, bubble rupture either does not occur, or occurs with such limited intensity that it does not significantly affect the flame surface.

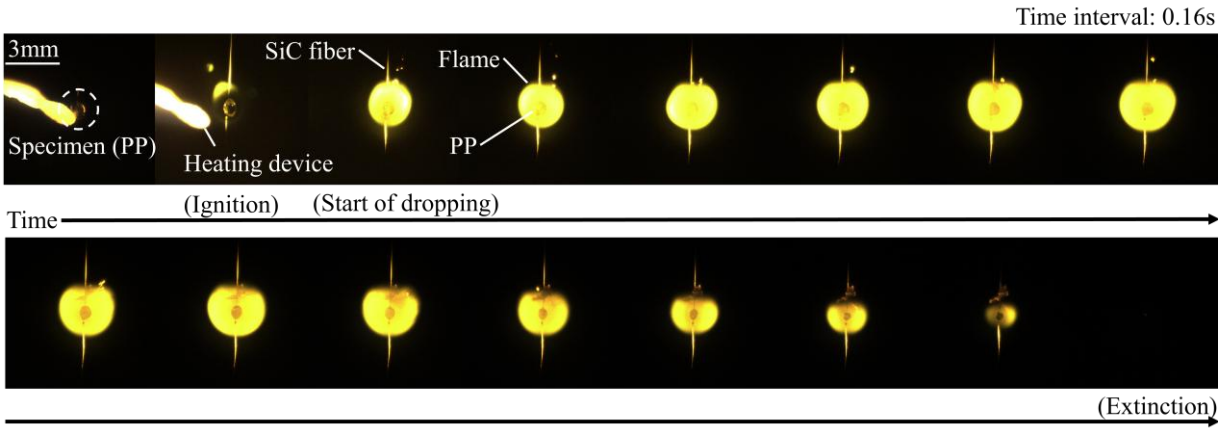


Figure 5. Timely variation of PP burning behavior from ignition to extinction in the present microgravity experiment. Time interval for each image is 0.16 sec (from top-left to bottom-right).

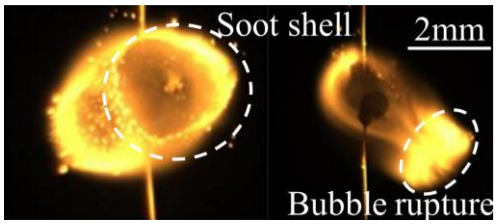


Figure 6. Dynamic behaviors observed during PMMA combustion[6] (Soot shell and bubble rupture)

3.2. Measured burning rate constant

According to the droplet combustion theory, when a one-dimensional combustion structure is established, the well-known "d²-law (**Eq. 2**)" should be applied.

$$d(t)^2 = d_0^2 - Kt \tag{2}$$

Here, t is the elapsed time [s], $d(t)$ is the diameter of the burning sample at time t [mm], and K represents the burning rate constant [mm²/s]. **Figure 7** shows the d²–t plots for both PMMA spheres from a previous study[5] and PP spheres from the present experiments. The burning rate constants were determined by plotting the measured d² values over time, as illustrated in **Fig 7**. In both cases, the initial stage of burning exhibits random fluctuations in d², whereas in the later stage, a linear decrease in d² with time is observed. The early stage is referred to as "Stage I " and the later stage as "Stage II " The boundary between these stages is defined by a sharp transition in the moving average. Changes in the sample diameter during combustion are attributed to the competing effects of shrinkage due to material consumption and volumetric expansion caused by internal bubbles within the polymer. However, during Stage I, the shrinkage due to combustion proceeds relatively steadily without sudden changes, and thus is unlikely to be the main cause of the observed fluctuations. These findings indicate that Stage I is also present in PP combustion. The reason why the flame surface disturbances described earlier were not prominent in PP combustion is presumably because the impact of bubble rupture is smaller compared to PMMA, resulting in less influence on the overall combustion behavior.

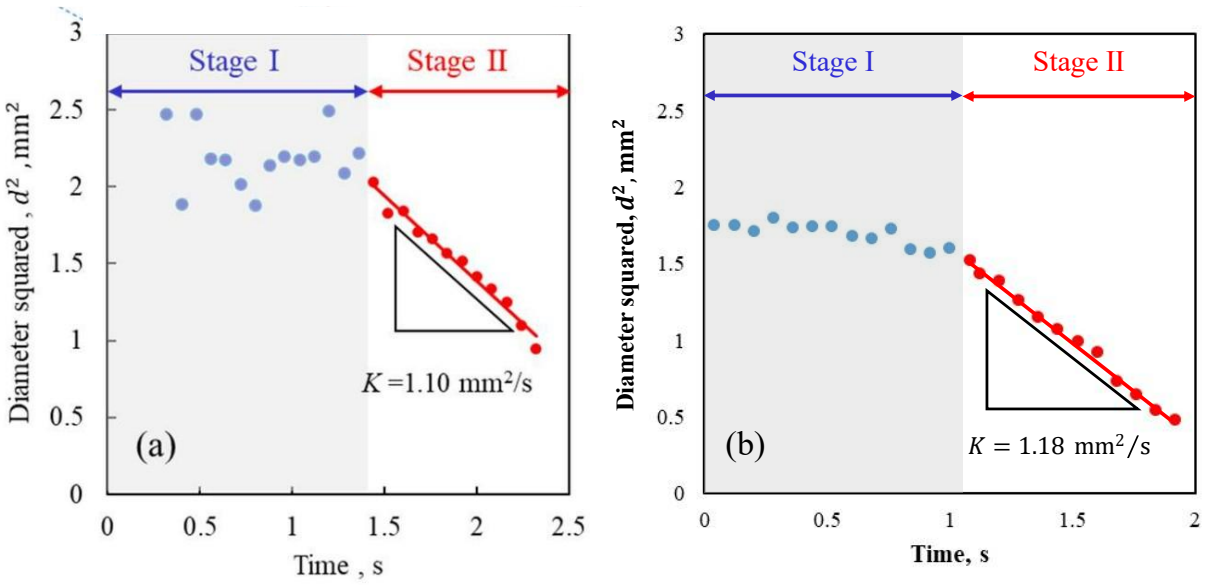


Figure 7. Changes of the diameter squared in time: (a) Changes of the diameter squared in time for PMMA[6] (K = 1.10 mm²/s), (b) Changes of the diameter squared in time for PP (K = 1.18 mm²/s).

3.3. Relationship between burning rate constant and (initial) void fraction

A relationship between initial void fraction and with the burning rate constant was shown in **Fig. 8**. For comparison, **Fig. 9** presents the relationship between bubble content and burning rate constant for PMMA samples based on a previous study[6]. In the previous study[5], the bubble content was estimated based on the sample thickness of PMMA, whereas in the present study, the void fraction was calculated using triaxial

diameter measurements[7]. As shown in **Fig. 8**, the burning rate constant for PP spheres increases with void fraction in the range of 0-40%, indicating a positive correlation. However, beyond 40% void fraction, the burning rate constant plateaus and no further increase is observed. In contrast, the PMMA samples shown in **Fig. 9** exhibit a continuous increase in the burning rate constant as bubble content increases.

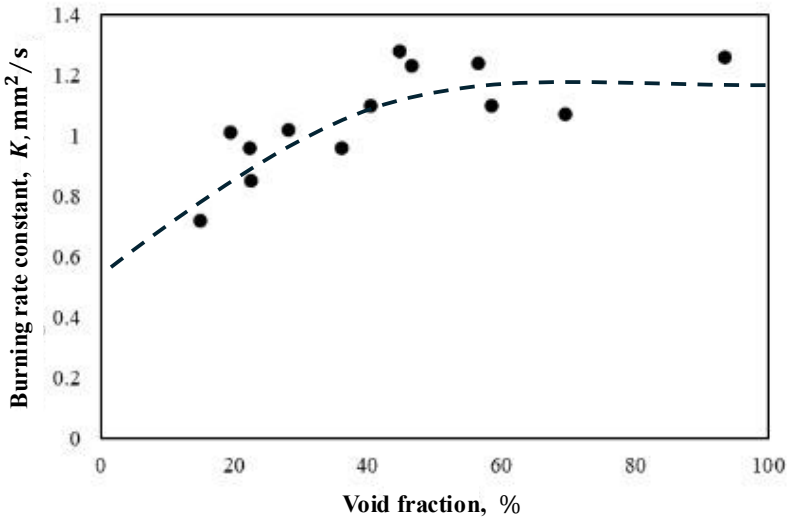


Figure 8. Void fraction and burning rate constant for PP under microgravity.

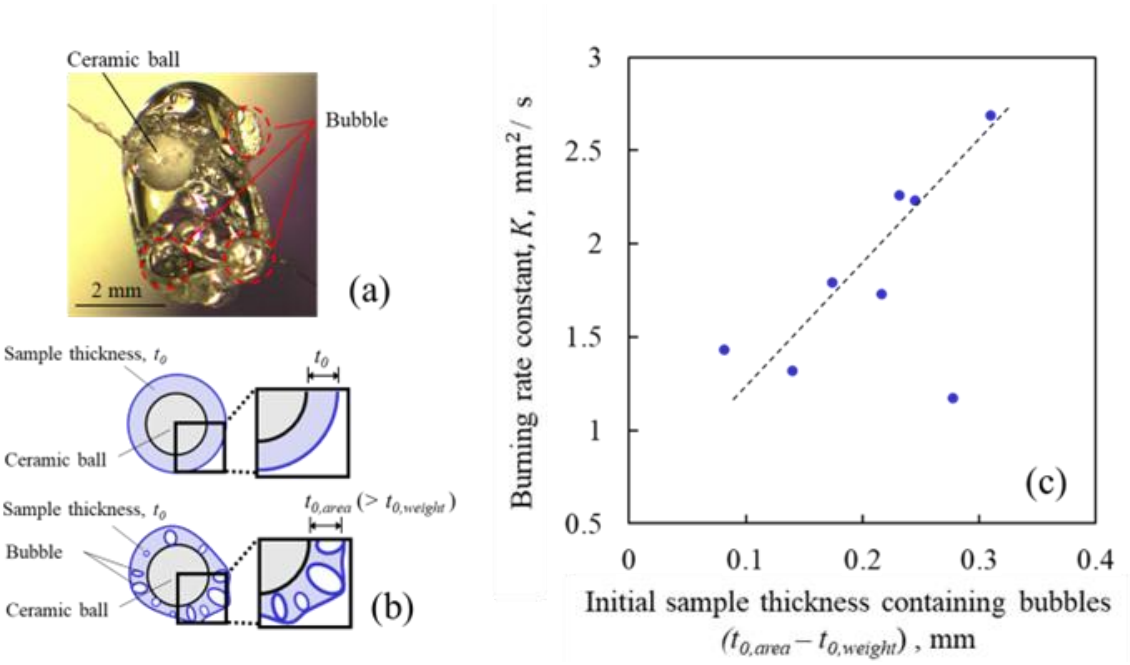


Figure 9. Expected initial void fraction effect on the burning character for PMMA sample[6]: (a) direct pic of the burning sample, (b) schematic illustration to define thicknesses ($t_{0,area}$, t_0) considered in this study and (c) burning rate constant (K) vs ratio of Stage I for successful nine microgravity test data.

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Table 1. Physical Properties of PMMA and PP

	PMMA (Polymethyl methacrylate)	PP (Polypropylene)
Glass-transition temperature, K	373	273
Thermal decomposition temperature, K	650	710
Density, kg/m^3	1190	910
Specific heat kJ/Kg/K	1.4	1.7
Thermal conductivity, W/m/K	0.17	0.12
Surface tension, mN/m	41.1	29.4

Let us consider the bubble formation and rupture processes in polymers. According to a previous study[10], heating induces the formation and growth of void fraction in polymers, which eventually rupture upon reaching the surface, or become trapped inside the solidified matrix. The progression from bubble growth to rupture is considered to be significantly influenced by the material's thermal decomposition temperature and surface tension. As shown in **Table 1**.

PMMA shall start to decompose at approximately 650 K, releasing volatile products even at relatively low temperatures. This facilitates early bubble formation and growth during heating. In contrast, PP has a higher decomposition temperature of about 710 K, implying that bubble generation timing is delayed under the same heating conditions (assuming that heat capacity is nearly the same). Moreover, by the time bubbles appears inside the PP sample to from "active" void, the sample has already reached a high temperature. Hence the viscosity is reduced yet internal pressure of the "active" void is increased, resulting that the bubbles can escape easily before sufficient bubble growth occurs.

Note that there is not-negligible difference in surface tension for PMMA and PP (see **Table 1**), suggesting that PP tends to weak "retaining" feature of bubbles inside the material as compared to PMMA.

Considering altogether, PP can be characterized by both a lower tendency for bubble formation and a lower ability to retain bubbles. Consequently, the influence of bubble rupture on flame dynamics during PP combustion is expected to be limited, although it is clearly presented (namely, un-avoidable).

The overall trend observed in the K–Void fraction relationship for PP is then summarized in **Fig. 10**. In the void fraction range of 0–40%, bubble formation and growth are promoted during heating, and despite the relatively low surface tension, the bubbles are retained within the sample, leading to an increase in the burning rate constant. On the other hand, when the void fraction exceeds 40%, even if new bubbles are formed during heating, the low surface tension prevents them from being retained within the sample, and they are instead released to the outside. This explains the observed plateau in the burning rate constant beyond 40% void fraction. To verify the validity on this process, we plan to test with other kind of polymers (e.g. PE) under microgravity in near future.

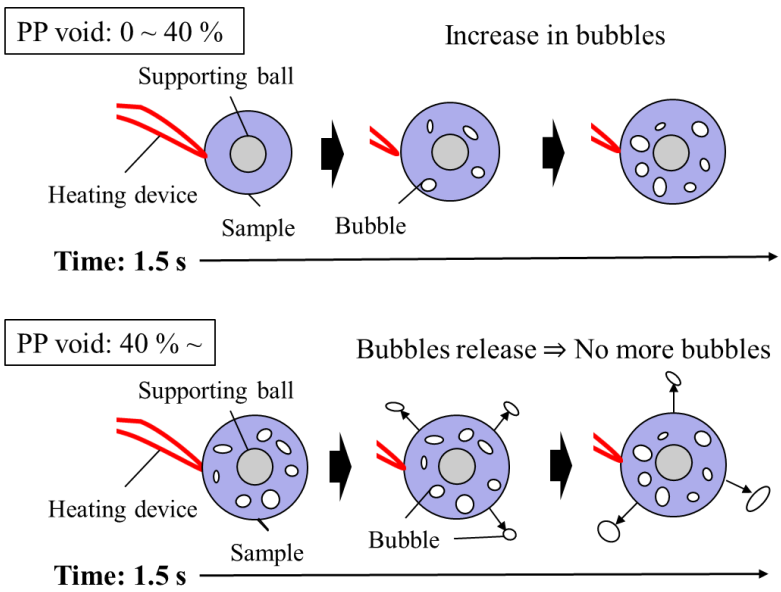

Figure 10. Expected bubble dynamics for PP with various initial void fraction under microgravity.

4. Remarks

The microgravity combustion experiments of Polypropylene spheres were conducted to quantitatively evaluate the effect of internal void fraction on the burning rate by preparing samples with varying void fraction. 13 experiments were conducted under identical conditions, varying only the void fraction. The results showed a positive correlation between void fraction and the burning rate constant in the range of 0–40%, while the burning rate constant tended to remain constant when void fraction exceeded 40%. This trend suggests that surface tension, which plays a key role in the process from void fraction growth to rupture, has a significant influence on the relationship between void fraction and the burning behavior.

Acknowledgments

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