Reaction wave front propagation of tri-n-butyl phosphate and fuming nitric acid (TBP/FNA) mixture under weak shock loading

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1 Introduction

In the PUREX method of a spent nuclear fuel reprocessing process, tri-n-butyl phosphate (TBP) and nitric acid are generally used. However, several explosion accidents caused by the deteriorated mixture of TBP and nitric acid were reported in the past. Studies have been conducted regarding the explosion risk of the mixture of TBP and fuming nitric acid (FNA), supposing 'the worst conditions' by considering those accidents as teachings [1][2][3]. When the card gap tests were done, it was found that three kinds of different modification appeared in the witness plate by the difference in incident shock pressure [4][5]. In this paper, the results of having measured the detonation velocities of the TBP and FNA mixture by optical and electrical methods, in order to consider the difference in the reaction by incident shock pressure, are reported.

2 Experiments

2-1 Measurement by the optical method

TBP/FNA = 22/78 wt% of chemical equivalence ratio was used for the sample. The assembly is shown in Figure 1. The sample container is a transparent polyvinyl chloride (PVC) tube (inside diameter of 20 mm) or a glass tube (inside diameter of 20 mm), and length is 200 mm. A precision detonator (Nippon Kayaku Co., Ltd.) was used for the ignition of a donor explosive, Pentolite of 34 g, (Chugoku Kayaku Co., Ltd.). In the case of the PVC tube container, four kinds of PMMA gaps (thickness of 15, 20, 23, 25 mm) were installed between the donor explosive and the container. And with the glass tube container, the experiments were conducted by installing five kinds of PMMA gaps (thickness of 10, 15, 20, 23, 25 mm) between the donor explosive and the container. In case of the gap length of 25 mm, it was considered that no detonation occured from the card gap test result.

The streak camera (CORDIN, MODEL-116) was used for photography. Since the spontaneous light of the sample was weak, a krypton flash was used as a back light source. The pulse generator (Berkeley Nucleonics Corp., BNC-555) was used for the synchronization of an explosion signal and the trigger signal for measurement.

2-2 Measurement by the electrical method

The outline of the assembly is shown in Figure2. The kind of the sample and container used here is the same as that of the previous section. Twisted polyester enameled copper wire with a diameter of 0.2 mm was used as

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the ion gap to detect the location of the reaction front. The ion gap was protected by winding fluoride tape around the portion in contact with the sample. Pulse generating equipment (Yatoro Denshi, Model-223HV) was used, and the signals were recorded by oscilloscope (Tektronix, TDS3014B).

The experiments were conducted using the PMMA gap material. For the PVC tude container, a gap thickness of 15, 20 mm was used, and for the glass tube container, a gap thickness of 5, 15 mm was used.

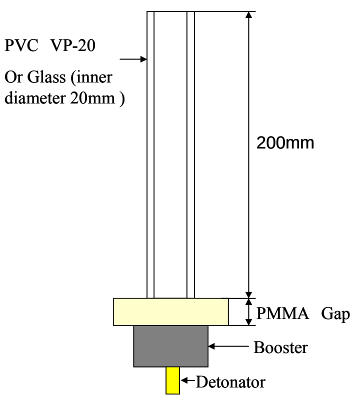

Fig.1 Assembly for optical method

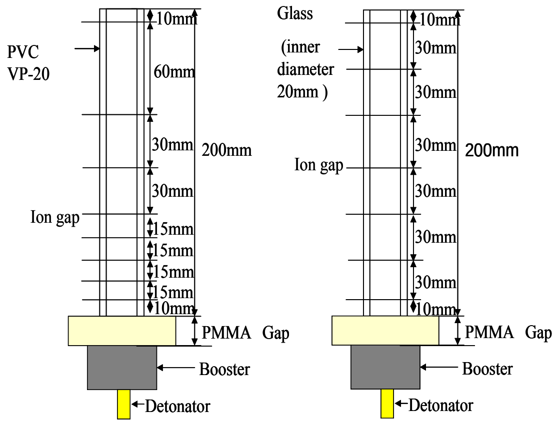

Fig.2 Assembly for electrical method

3 Results and discussion

3-1 Measurement by the optical method

As an example of streak photography, the result which was obtained for a gap length of 15 mm and the PVC tube container is shown in Figure 3. Inclination of the straight line in the figure represents the propagation velocity of the wave front. Two velocities were observed at this experimental condition. All experimental results are summarized in Table 1. The velocity of approximately 6.5 km/s is the C-J detonation velocity of the TBP/FNA mixture from the past research and calculation result.

Since instead of the luminescence of the detonation of a sample, the situation that back light was interrupted by the phenomenon was observed in this experiment, the measured low velocity has a high possibility of being the destruction speed of the sample container. This can be guessed also from it being mostly the same speed with the result of the gap length of 25 mm, a condition which will not detonate.

Moreover, as listed in table 1, the phenomenon in which speed changed on the way was observed only for the PVC tube container, and it was not recordable for the glass tube container. If it is the same gap length, a similar phenomenon should be observed even if the quality of the material of a container is different. This may be a result peculiar to the optical measurement method which uses a back light. Thus, in the present study, measurements by the electrical method were tried for the same sample container.

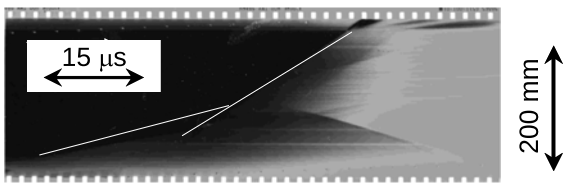

Fig.3 An example of a streak photograph (PVC tube, 15 mm gap)

Table 1 Experimantal results by optical method

Material	Thickness (mm)	Velocity (km/s)
PVC	15	1.83 → 6.53
	20	1.83 → 6.53
	23	1.83 → 6.53
	25	1.83
Glass	10	6.44
	15	4.81
	20	4.66
	23	4.80
	25	4.81

3-2 Measurement by the electrical method

Figure 4 shows the result of plotting the short circuit time of the ion gap and the relation to the distance, at the conditions of glass tube container and gap length of 15 mm. Table 2 summarizes the measurement results by the electrical method. Unlike the result of the optical method, also for the glass tube container, the two velocities were clearly observed, that is, the low velocity (1.46 km/s) state up to 60 μs after initiation and the C-J detonation velocity (6.57 km/s). This shows that when a weak shock occurs which does not result in explosion directly, the shock wave that propagates inside the sample may transfer to the C-J detonation state after a certain time delay. This is very important knowledge for the issue of safety. And this low velocity propagation phenomenon of the reaction wave front might be not the deflagration but the low velocity detonation. On the other hand, in the case of the PVC tube container, velocities of less than 1 km/s were observed. In these cases, it seems that the advance of the deflagration wave front was observed.

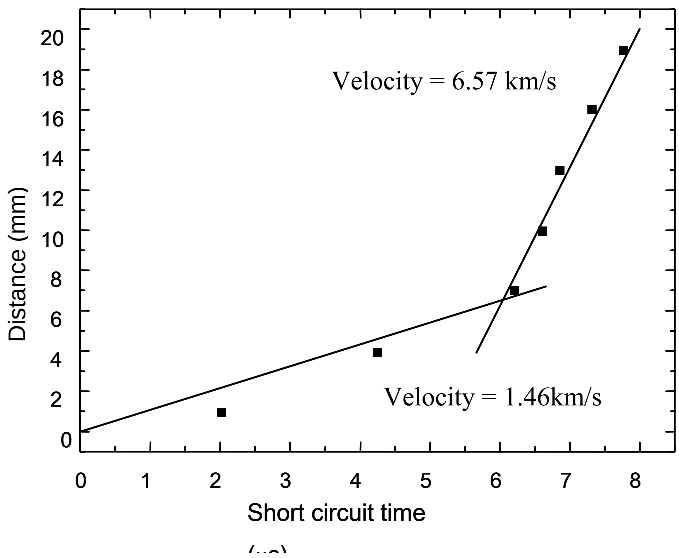

Fig.4 Relation of time and distance (glass tube, 15 mm gap)

Table 2 Experimantal results by electrical method

Material	Thickness (mm)	Velocity (km/s)
PVC	15	0.78 → 6.58
	20	0.49 → 6.58
Glass	5	6.59
	15	1.46 → 6.57

4 Conclusion

It was found that the TBP/FNA mixture explodes by different mechanisms according to the strength of the incident shock pressure. That is, for a strong incident shock pressure, the reaction propagates in the state of C-J detonation from the beginning, and for a weak incident shock pressure which does not result in direct explosion, the low velocity reaction wave front propagates inside the sample for a certain amount of time, and the phenomenon transferred to the C-J detonation state after that.

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