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On the Composite Solid Propellant Impregnated with Metallic Foil

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The control of the burning rate is one of the important aspects in the extension of the capability of a solid propellant. The burning rate of a composite propellant was found to be increased remarkably by the proper impregnation of metallic foil, and upon the addition of a few percent of aluminum, silver and copper (in weight), the burning rate jumped up to several times that of the original one, providing there had been suitable processing. The impregnation of metallic foil was performed by mixing a predetermined amount of finely-cut strips of foil into propellant dough or by laminating the latter with foils. The propellant grain impregnated at random with strips of foil increased the burning rate considerably, but the one laminated with metallic foils which were oriented perpendicular to the burning surface showed the highest burning rate. The increment of the burning rate evidently depended upon the thickness of the foil used; the distance between adjacent foils for lamination showed no remarkable effect. The role of the metallic foil on the augmentation of the burning rate seems to be attributable to the improvement of the heat transfer from the burning zone to the burning surface. However, in the case of copper, the burning rate exceeded that of silver, so the increment of the burning rate can not be explained only in terms of the thermal conductivity of the metallic foil. In the case of copper foil, the effective increase in the burning rate may be due to its catalytic action plus a good thermal conductivity. The mechanical properties of the propellant grain, such as tensile strength and elongation at the yield point, were expected to be improved with the impregnation of metallic foil. However, this coluld not be confirmed in the present tests.

The development of a propellant with an extremely slow burning rate has been successful[1]; the present interest is focussed on one with an unusually rapid burning rate.

During the course of the regression of the solid propellant, it is believed that the amount of heat required for the decomposition is supplied from the flame zone. It is conceivable that the burning rate could be augmented when the end of the impregnated metal protrudes from the matrix of the burning surface, due to its enhanced capability for heat transfer into the subcombustion layer of the condensed phase.

Further, it is plausible that the metal acts as a high-energy ingredient. Recently, modern solid propellants have been metallized with fine aluminum powder in order to improve the specific impulse by taking advantage of its high energy of combustion, and also in order to suppress its acoustic combustion instability.

In this work, we studied a composite propellant impregnated with aluminum by means of laminating or randomly-distributed fine strips cut out from foil; we found that the burning rate was thus boosted remarkably. The metallic ingredients, copper-foil and silver-foil, were tested and compared from the standpoint of the difference in thermal conductivity; contrary to expectations, we found no noticeable inequality between them.

A reinforcing effect of metal impregnation by means of the distribution of strips or lamination may be anticipated, besides its contribution to the burning rate.

Materials

Aluminum Powder. Sprayed powder of 240 mesh under. (Fukuda Kinzoku-Hakufun Kogyo K. K.)

Aluminum Foil. 6, 15 and 17 microns thick. (Nippon Seihaku K. K., Tokai Kinzoku Kogyo K. K., and Mitsubishi-Reynolds Aluminum Co.)

Copper Foil. 10 and 20 microns thick. (Nakajima Kinzoku-Hakufun Kogyo K. K.)

Silver Foil. 13 microns thick. (Tanaka Precious Metals Industry, Ltd.)

Burning Catalyst. Copper chromite catalyst. (Nikki Chemical Co., Ltd.)

Ammonium Perchlorate. Commerical-grade material, used as received. Adjusted to a suitable

1) K. Yamazaki, A. Iwama, S. Aoyagi, T. Sofue, M. Hayashi and K. Kishi, *Kogyo Kagaku Zasshi (J. Chem. Soc. Japan, Ind. Chem. Sect.)*, **66**, 1630 (1963).

particle-size distribution. (Kanto Chemical Co., Ltd.)

Fuel Binder. Carboxy-terminated polybutadiene with suitable curatives and additives.

Experimental

Preparation of the Laminated Grains. The fuel binder and solid ingredients were thoroughly mixed in a jacketed, stainless steel mixer for 40 min at 70°C under a reduced pressure of 2—3 mmHg.

A sheet of foil cleaned with acetone was stretched on a glass plate coated with vaseline grease. A predetermined amount of dough was placed upon the foil, spread manually, and covered with another foil. The formation of cavities inside the grain and at the interface between the foil and the medium were minimized as much as possible. The glass plates were pressed by means of weights until the space between the plates was reduced to the thickness of the spacer, and then cured in an electric oven at 50°C for 24 hr.

After the content had hardened to a certain degree, the glass plate was removed; the foil surface which then appeared was degreased again with acetone for further treatment.

By repeating this process, laminated grains of any number of plies and of any degree of thickness could be prepared. Needless to say, when the bare surface of the propellant was required, a glass plate was used instead of a foil-lined one.

The cured propellant block was cut into strands 6×6×60 mm in size; these served for the measurement

TABLE 1. THE COMPOSITION OF THE SPECIMENS

No.	Metal Impregnation	Propellant*	Thickness of the foil μ	Metal content %	Number of plies Foil	Number of plies Propellant	Remark
1	None	A	—	—	—	—	—
2	Al powder	A	—	2.65	—	—	Powder : 240 mesh under
3	Al powder	A	—	5.00	—	—	Powder : 240 mesh under
4	Al strips	A	14.5	2.65	—	—	Strips : 2×20 mm
5	Al strips	A	14.5	5.00	—	—	Strips : 2×20 mm
6	Al strips	A	14.5	2.65	—	—	Strips : 5×20 mm
7	Al strips	A	6.0	2.65	—	—	Strips : 5×20 mm
8	Al strips	A	6.0	2.65	—	—	Strips : 10×20 mm
9	Al strips	A	17.3	2.65	—	—	Strips : 10×20 mm
10	Al strips	A	17.3	2.65	—	—	Strips : 10×30 mm
11	Al laminate	A	14.5	2.65	6	5	Thickness of the propellant layers: 1.0 mm
12	Al laminate	A	14.5	2.32	6	5	Thickness of the propellant layers: 1.3 mm
13	Al laminate	A	14.5	1.52	4	3	Thickness of the propellant layers: 2.3 mm
14	Al laminate	A	14.5	2.25	6	5	Thickness of the propellant layers**: *cf.* Fig. 1
15	Al laminate	A	14.5, 17.3	3.82	8	7	Thickness of the propellant layers: 0.9 mm
16	Al laminate	A	17.3	1.48	3	2	Thickness of the propellant layers: 3.1 mm
17	Al laminate	A	17.3	0.48	1	2	Thickness of the propellant layers: 3.1 mm
18	None	B	—	—	—	—	—
19	Cu laminate	B	10	3.01	3	2	Thickness of the propelllant layers: 3.1 mm
20	Cu laminate	B	10	1.03	1	2	Thickness of the propellant layers: 3.1 mm
21	Cu laminate	B	20	6.27	3	2	Thickness of the propellant layers: 3.1 mm
22	Cu laminate	B	20	2.00	1	2	Thickness of the propellant layers: 3.1 mm
23	Ag laminate	B	12.7	—	3	2	Thickness of the propellant layers: 3.1 mm
24	Ag laminate	B	12.7	1.49	1	2	Thickness of the propellant layers: 3.1 mm

* The Composition of the propellants A and B

	A	B
The fuel binder	25 parts	25 parts
Ammonium perchlorate	75 parts	75 parts
Copper chromite	3 parts	—

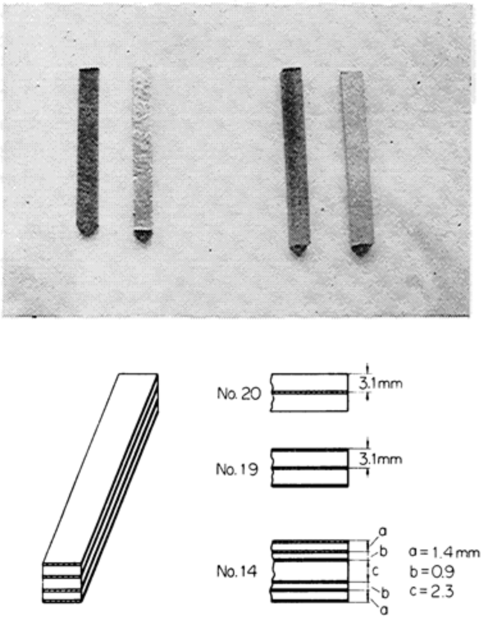


Fig. 1. The view of the laminated strands.

of the burning rate. A prospective view of the laminated strands is shown in Fig. 1.

Preparation of the Non-laminated Grains. The dough was prepared in the way which has been described previously. The fine aluminum powder or strip was mixed together, as in preparing convertionally metallized grains. The dough was then cast into a suitable mold, cured, and cut into standard $6\times6\times60$ mm strands.

The casting device is illustrated in Fig. 2.

Measurement of the Burning Rate. Two fine holes, located at fixed distance of 35 mm apart, were drilled in each strand perpendicular to the surface

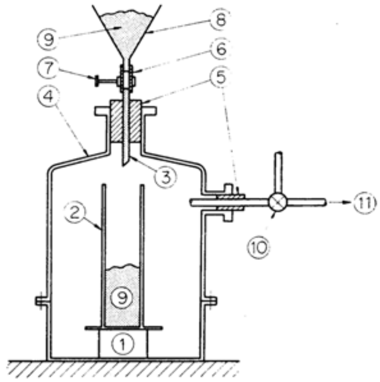


Fig. 2. The casting device for the preparation of the non-laminated propellant grains.

① Vibrator
② Vessel
③ Nozzle
④ Bell-jar
⑤ Rubber stoppers
⑥ Rubber tube
⑦ Screw cock
⑧ Funnel
⑨ The propellant dough
⑩ T-cock
⑪ Vacuum pump

of the laminated foil; two fuse wires 0.8 mm in diameter were then inserted into the holes. The regression rate was obtained from the time required for the burning surface to pass across the fuse wires under the pressure of nitrogen as usual.[2)]

All of the surface except the burning end of the strand was restricted with a coating of an epoxy-polyamide resin.

Measurement of the Mechanical Strength. The tensile properties of the grain were measured by "Tensilon" at 20°C and at a 60% relative humidity with a strain rate of 50 mm/min. Standard $6\times6\times$ 60 mm strands were employed for the experiment.

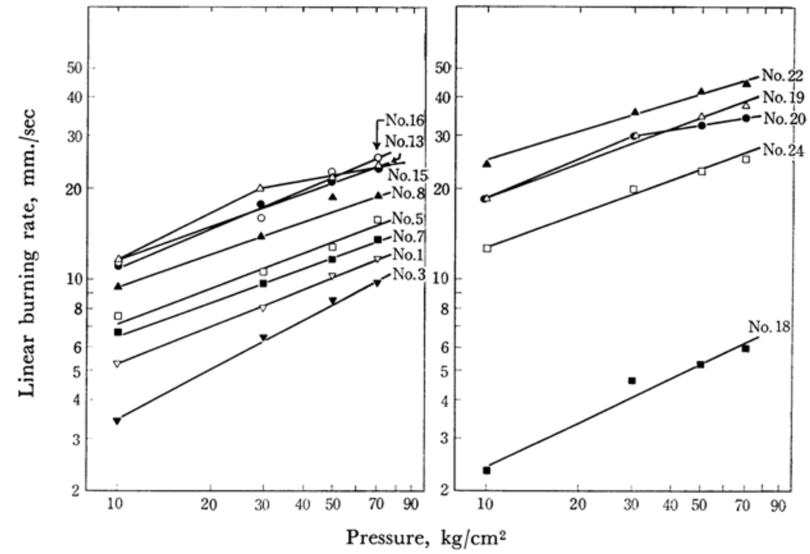


Fig. 3. Linear burning rate of the samples.

2) K. Yamazaki, M. Hayashi and A. Iwama, *ibid.*, **67**, 415 (1964).

The cross heads were initially set 30 mm apart. To protect the specimen from damage by the cross head, that part was strengthened by a coating of an epoxy-polyamide resin.

The measurements were performed for five specimens of each sample; the results are shown, along with the mean value and the standard deviation.

Measurement of the Thickness of the Foil. The thickness of the foil was obtained from the measured weight, the surface area, and the density.

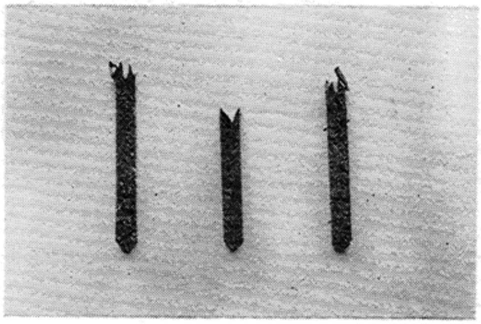
The details on each propellant are shown in Table 1.

Results and Discussion

When the grain was impregnated with metallic foil, the burning rate was remarkably improved; the best results were obtained in the case of foil oriented perpendicularly to the buring surface, though the conventional aluminum powder had no stimulating effect, in some cases, even acting as a retarder, as is shown in Table 2 and Fig. 3.

As far as the laminated grains were concerned, the burning rate became higher with the thickness of the foil. However, the space between two adjacent foils had no remarkable effect upon the burning rate.

It was observed that the laminated strands regressed so rapidly along the foil that the burning surface formed a V-shape or a wedge, as Fig. 4 shows.



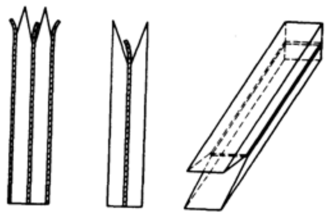


Fig. 4. The view of the V-shaped burning surface of the laminated strands.

Assuming that the burning temperature and the heat conduction along the foil are not affected by

TABLE 2. LINEAR BURNING RATE AND TENSILE PROPERTIES OF THE SPECIMENS

No.	Linear burning rate, mm/sec				n*	Tensile strength kg/cm²	Elongation at the break, %
	70 kg/cm²	50 kg/cm²	30 kg/cm²	10 kg/cm²			
1	11.6	10.3	8.0	5.2	0.41	—	—
2	11.7	10.3	8.3	5.0	0.46	—	—
3	9.6	8.5	6.4	3.4	0.54	—	—
4	14.7	11.5	9.8	6.4	0.41	—	—
5	15.6	12.7	10.5	7.5	0.39	—	—
6	13.2	14.2	10.5	6.2	—	—	—
7	13.5	11.6	9.6	6.6	0.37	—	—
8	18.9	18.6	13.7	9.6	—	—	—
9	14.4	11.8	9.2	5.9	0.44	13.5±0.6	15.2±1.4
10	14.4	11.2	8.7	6.4	0.41	13.7±1.2	16.6±1.4
11	25.0	22.4	21.5	10.3	—	—	—
12	22.8	21.0	16.9	10.2	0.43	—	—
13	23.0	21.0	17.7	11.0	0.39	—	—
14	23.4	19.4	16.0	10.3	0.41	—	—
15	23.5	21.7	19.8	11.4	—	—	—
16	25.2	22.7	15.8	11.2	0.43	—	—
17	26.6	23.5	22.0	12.6	—	—	—
18	5.9	5.2	4.6	2.3	0.48	11.7±1.4	29.9±11.6
19	37.6	34.6	29.8	18.4	0.38	14.5±0.3	11.7±1.7
20	34.2	32.4	29.7	18.4	—	6.7±0.5	44.8±1.3
21	43.6	42.6	36.0	23.9	0.33	15.2±1.1	6.7±1.0
22	44.1	41.9	35.6	24.0	0.34	8.9±0.4	24.7±2.3
23	—	—	—	—	—	14.2±0.6	12.8±1.3
24	25.7	22.8	19.8	12.6	0.37	10.5±0.2	43.3±6.7

*n: Pressure exponent

the number of plies, the burning rate in the close vicinity of the foil should be constant when the thickness of the foil is fixed at a constant value. The increase in number of plies only caused an increase in the number of wedges on the burning surface; it contributed little to the increase in the linear burning rate.

Copper and silver were expected to be preferable for the augmentation of the burning rate because their thermal conductivities are better than that of aluminum, though they should be inferior as fuel ingredients from the point of view of energy.

As Table 2 and Fig. 3 show, they were superior than aluminum to accelerate the burning rate. However, contrary to the expectation, the burning rate of the silver-impregnated grain did not exceed that of copper in spite of its better thermal conductivity. It is conceivable that copper or its burnt products act as a combustion catalyst.

The tensile data are compiled in Table 2. As the table shows, we found no reinforcement effect by the foil impregnation.

Figure 5 shows the stress-strain curves of the samples. In the process of fracture on stretching, a dewetting process occurred and some voids or cavities appeared at the interface between the solid ingredient and the matrix of the binder. These voids or cavities spread rapidly, causing the breaking. For example, the stress-strain curve of the No. 18 sample in Fig. 5 showed that the specimen was already broken at the point of maximum stress. On the contrary, though, some laminated samples showed a certain time lag beyond the point of maximum stress.

Silver foil showed a very poor compatibility in relation to the adhesion of the propellant.

A polybutadiene-type fuel binder has a high ratio of hydrogen to carbon, and there is no oxygen present in the molecule, so it has a tendency to produce soot when burned. Nevertheless, it became exceptionally free from soot when laminated with aluminum, though the same amount of conventional aluminum powder had no effect upon the suppression of the soot formation.

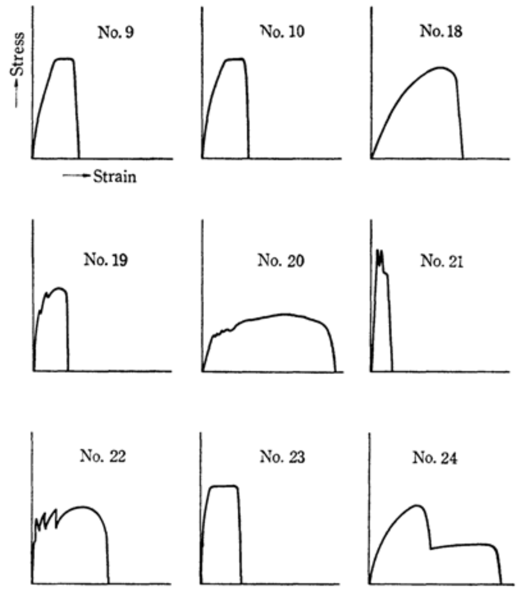


Fig. 5. Stress-strain curves of the samples.

At any rate, aluminum foil seemed to be very effective in augmenting the burning rate.

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