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# A Rubber-Bonded HMX-Based Explosive

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# STORAGE STABILITY OF X-0298:
# A RUBBER-BONDED HMX-BASED EXPLOSIVE

by

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## ABSTRACT

We determined the storage stability at elevated temperatures of X-0298, a rubber-bonded HMX composition. After 34 months in sealed containers at 40, 50, and 60°C, physical properties of tensile specimens exhibited no significant degradation. Gas evolution rates from small cylinders of X-0298 stored in air-filled ampoules at 60, 75, and 90°C were 30, 80, and 1000 $cm^3/kg/yr$. Compared with other energetic explosives, X-0298 is exceptionally stable when stored at temperatures below 60°C.

## I. INTRODUCTION

The search for chemically stable, insensitive, energetic high explosives for military applications is a continuing effort. The requirements for long-term storage in a variety of environments limit the materials that can be used. Explosives such as PBX 9404, acceptable for past applications, do not have the stability and handling safety required for current weapon systems.

In the late 1960s, PBX 9501 was developed at the Los Alamos Scientific Laboratory (LASL) as a replacement for PBX 9404. PBX 9501 had greatly improved handling safety and reasonable chemical stability compared with PBX 9404.[1] However, the physical strength of pressed pieces decreased with time at elevated temperature to about 30% of the initial value[2] because of degradation of the polyurethane binder Estane 5703.* Deterioration of the physical strength of explosive charges does not exclude a material from all weapon systems, but its use in some systems is precluded. Thus, a binder that does not deteriorate at elevated temperatures in a PBX formulation is desirable.

Since 1974, LASL has investigated Kraton G1650,* a thermoplastic rubber, as a binder in high-explosive formulations. Although most of the experimental effort concentrated on developing an HMX formulation, blends containing RDX and TATB were also prepared. This report describes the results obtained from a study of the storage stability of a specific formulation containing 97.5:1.12:1.38 (wt%)—HMX:Kraton G1650:Cenco Hyvac oil,** referred to as X-0298.

## II. BACKGROUND

Kraton G1650 is a thermoplastic rubber, structurally a styrene-ethylenebutylene-styrene (S-EB-S) block copolymer. The average peak molecular weight ($\overline{MW}_p$) of the material used in this work was about $1 \times 10^5$ (polystyrene equivalent), determined by gel permeation chromatography (GPC). Kraton is quite resistant to hydrolytic and oxidative degradation when stabilized with small amounts of antioxidants, such as Irganox or Ionol, and is used in applications that require rubber with these properties.[3]

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*Shell Chemical Company, P. O. Box 1422, Houston, TX 77001.

**Central Scientific Company, Inc., 2600 South Kostner, Chicago, IL 60623.

Cenco Hyvac oil is a low-vapor-pressure, paraffinic, vacuum pump oil with an average molecular weight of about 500 (GPC). No additives were detected in the product used to prepare X-0298.

The X-0298 explosive molding powder is prepared by a standard slurry process, using butyl acetate as the slurry solvent. The resulting powder is quite uniform (1- to 2-mm particle size), with a bulk density of 0.98 g/cm$^3$, and is easy to load and press. Charges attain a density of 1.81-1.82 g/cm$^3$ when pressed at about 120°C. Reference 4 gives a more detailed description of the explosive and material properties of X-0298.

## III. EXPERIMENTAL METHOD

The experimental effort comprised two complementary studies. A number of small pressed cylinders (1.27 cm diam by 3.81 cm long, weighing 8.77 g each) and several 1.0-g samples of molding powder were stored at elevated temperatures for various time intervals to obtain data on the chemical stability of X-0298. The cylinders and molding powder were sealed in Pyrex ampoules containing 53 kPa (400 torr) of either dry air or helium. (Sample preparation is described in Ref. 2.) Larger pressed pieces (1.57 cm diam by 10.1 cm long, machined to a dumbbell shape) were sealed in stainless steel cans (five per can) and stored in ovens to collect information on the change in tensile properties. The cans initially contained 81 kPa (600 torr) of dry air.

All test units were placed in forced-draft temperature-controlled ovens. The small cylinders were stored at 60, 75, and 90°C, and the tensile specimens were stored at 40, 50, and 60°C. The molding powder samples were tested only at 75°C. At specified times, units were removed from the ovens and diagnostic tests were performed, including gas analyses, GPC of the extracted binder, and physical strength measurements on the pressed pieces.[2] In the present study, tetrahydrofuran (THF) was used as the extraction solvent instead of the dichloroethane (DCE) used previously because DCE did not completely extract the Kraton from the aged samples after 2 h on the wrist-action shaker.

## IV. RESULTS AND DISCUSSION

Experimental data were collected on X-0298 small cylinders for 68 weeks at 60, 75, and 90°C, on molding powder for 34 weeks at 75°C, and on tensile specimens for 34 months at 40, 50, and 60°C.

### A. Molding Powder and Small Cylinder Test Results

Tables I and II and Fig. 1 summarize the gas-evolution data obtained from these samples. Table III compares gas-evolution data for cylinders of PBX 9404, PBX 9501, and X-0298. Briefly, X-0298 evolved gas at about 30 cm$^3$/kg/yr at 60°C in air, compared with 300 cm$^3$/kg/yr for 9501, and 800 cm$^3$/kg/yr for 9404. The gas evolved was mainly $CO_2$ and $N_2$ in a ratio that implied HMX decomposition was the predominant gas-producing mechanism. Even at 90°C, the gas-evolution rate from the cylinders in air was only 1000 cm$^3$/kg/yr (equivalent to about 0.2 wt% decomposition).

Molecular weight data obtained on the extracted binder are summarized in Table IV. Very little change occurred during the test period at any temperature. At 90°C, the Kraton began to show some decrease (~15%) in $\overline{MW}_p$ and a larger dispersity, indicating the presence of increasing amounts of lower molecular

TABLE I

GAS EVOLUTION FROM SMALL CYLINDERS OF X-0298 STORED IN SEALED AMPOULES

(8.8-g pieces; ρ = 1.815 g/cm$^3$)

| | | Total Evolved Gas (cm$^3$/kg) | |
|---|---|---|---|
| Weeks | Temp (°C) | Helium Atm | Air Atm |
| 8 | 60 | 1 | 1 |
| | 75 | 11 | 9 |
| | 90 | 40 | 90 |
| 16 | 60 | 2 | 2 |
| | 75 | 22 | 15 |
| | 90 | 110 | 180 |
| 34 | 60 | 7 | 4 |
| | 75 | 76 | 27 |
| | 90 | 440 | 490 |
| 68 | 60 | 17 | 39 |
| | 75 | 100 | 110 |
| | 90 | 2200 | 1400 |

### TABLE II

### GAS EVOLUTION FROM SMALL CYLINDERS OF X-0298 STORED FOR 16 MONTHS

(8.8-g pieces; $\rho = 1.815\ g/cm^3$)

| | Helium | | | | Air | | | |
|---|---|---|---|---|---|---|---|---|
| Temp (°C) | TEG[a] ($cm^3/kg$) | $\%CO_x$ | $\%N_xO_y$ | %Org | TEG ($cm^3/kg$) | $\%CO_x$ | $\%N_xO_y$ | %Org |
| 60 | 17 | 24 | 33 | 43 | 39 | 38 | 23 | 39 |
| 75 | 100 | 17 | 72 | 11 | 110 | 32 | 58 | 10 |
| 90 | 2200 | 29 | 70 | 1 | 1400 | 29 | 70 | 1 |

[a] TEG = Total Evolved Gas

### TABLE III

### GAS EVOLUTION AT 60°C

($cm^3/kg/yr$)

| | X-0298 | 9501 | 9404 |
|---|---|---|---|
| Air | 30 | 300 | 800 |
| Inert | 15 | 75 | 900 |

weight material (Table V and Fig. 2). The HMX peak in the chromatogram interfered somewhat with the oil peak, but the oil peak changed little during the test. Infrared analysis of the extracted binder phase showed no evidence of oxidation products at any time (detection limit is about 0.1%).

## B. Tensile Specimen Test Results

The tensile strength data obtained at intervals during a 34-month period on specimens stored at 40, 50, and 60°C are summarized in Table VI, which also includes the $\overline{MW}_p$ values obtained by GPC analysis on the extracted Kraton polymer. These data show that the physical strength of X-0298 is exceptionally constant. Very little change occurred in the tensile strength of the material during storage at any temperature—indeed, the strength increased slightly with time at 60°C. Of particular interest was the percent elongation at failure for the aged material. After a decrease of about 20% during the early stages of the test, no significant change occurred. The resulting values are 3 or 4 times greater than those of other energetic PBX formulations.

### TABLE IV

### POLYMER MOLECULAR WEIGHT DATA

(Small cylinders; $\rho = 1.815\ g/cm^3$)

| | | $\overline{MW}_p$[a] (thousands) | |
|---|---|---|---|
| Temp (°C) | Weeks | Air Fill | Helium Fill |
| 60 | 8 | 106 | 103 |
| | 16 | 106 | 105 |
| | 34 | 106 | 103 |
| | 68 | 106 | 105 |
| 75 | 8 | 103 | 104 |
| | 16 | 95 | 93 |
| | 34 | 103 | 104 |
| | 68 | 95 | 93 |
| 90 | 8 | 95 | 96 |
| | 16 | 89 | 91 |
| | 34 | 94 | 96 |
| | 68 | 89 | 91 |
| Ambient | 0 | 95 | --- |
| | 16 | 95 | --- |
| | 34 | 101 | --- |
| | 68 | 91 | --- |

[a] Average peak molecular weight (polystyrene equivalent). Differences of 10 000 in $\overline{MW}_p$ are probably insignificant.

## C. Lifetime Predictions

Because very little change occurred in the molecular weight of Kratron polymer or the tensile strength of the

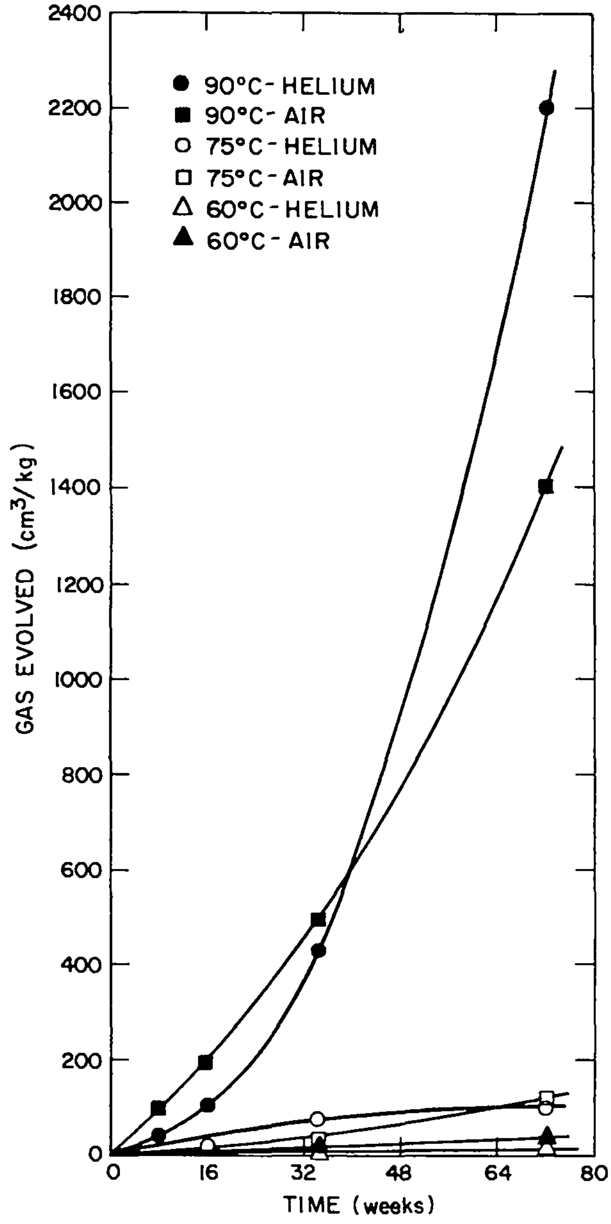

Fig. 1.
Gas evolved from X-0298 small cylinders stored in sealed containers.

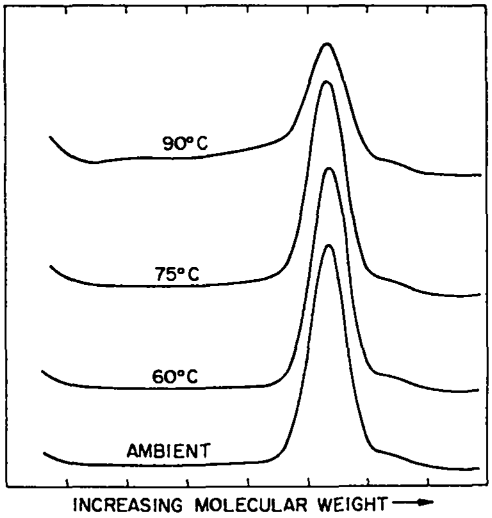

Fig. 2.
Gel permeation chromatograms of Kraton polymer from X-0298 stored for 16 months in air-filled containers.

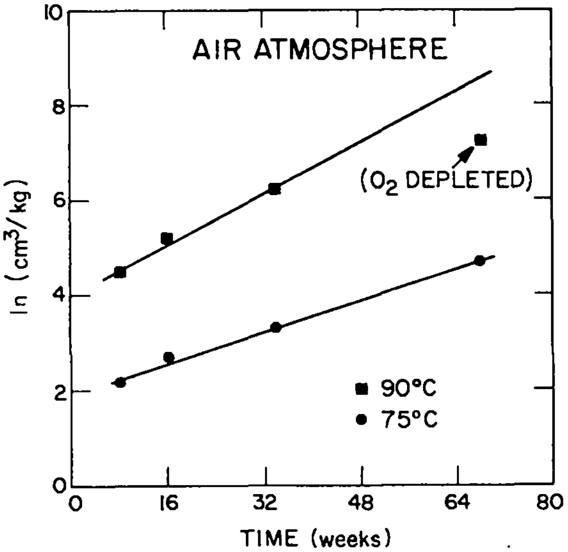

Fig. 3.
Rate constant plot.

pressed pieces, these data were not useful for construction of a model of lifetime predictions. However, we tried to devise a model based on the gas-evolution rates from the small cylinders stored at 75 and 90°C. Rate constants were calculated for the gas-evolution process observed under these test conditions. Figure 3 is a plot of ln(cm$^3$/kg) vs time at 75 and 90°C for samples stored in air. Only four points at each temperature were used because the amount of gas evolved at times less than 8 weeks was insignificant. Even the amounts evolved at longer storage times were small, resulting in considerable scatter of the data points. The slopes of the lines through the points at 75 and 90°C gave rate constants of $6.7 \times 10^{-8}$ and $10.7 \times 10^{-8}$ s$^{-1}$. We calculated an apparent activation energy $E_a$ from the two rate constants for the gas evolution process. A value of about 8 kcal/mole was obtained. Although determined with only two points, an $E_a$ of this magnitude suggests that the rate-determining

## TABLE V

## MOLECULAR WEIGHT OF KRATON POLYMER IN X-0298 AFTER 16 MONTHS STORAGE

| Temp (°C) | $\overline{MW}_P^{(a)}$ | $\overline{M}_N^{(a)}$ | $\overline{M}_W^{(a)}$ | $\overline{M}_W/\overline{M}_N^{(a,b)}$ |
|---|---|---|---|---|
| 60 | 106 | 104 | 112 | 1.077 |
| 75 | 95 | 84 | 95 | 1.131 |
| 90 | 89 | 68 | 86 | 1.265 |
| Ambient | 92 | 91 | 100 | 1.099 |

[a] Values in thousands (polystyrene equivalent).

[b] Dispersity: monodisperse = 1.000; normal 1.5-5; broad >5. $M_w$ = weight average molecular weight; $M_n$ = number average molecular weight.

step in the process resulting in evolved gas is diffusion of decomposition products from the piece. Thus gas-evolution rates from X-0298 cannot be used to derive a model for lifetime predictions because evolution rates depend on configuration and porosity of the explosive charges. Therefore, data obtained in this study do not provide a suitable basis for a model of lifetime predictions, but they do indicate a stability of X-0298 much greater than 9404 and 9501.

## V. CONCLUSION

This study demonstrates that X-0298, a Kraton-bonded HMX formulation, is an exceptionally

## TABLE VI

## X-0298 SURVEILLANCE DATA

4-in. Tensile Specimens: ρ = 1.815

(Stored in Sealed Cans, Air Atmosphere)

| | 40°C | | | 50°C | | | 60°C | | |
|---|---|---|---|---|---|---|---|---|---|
| Time (months) | $MW_p$ ($x10^3$) | Strength (kPa) | % Elong. (100 in./in.) | $MW_p$ ($x10^3$) | Strength (kPa) | % Elong. (100 in./in.) | $MW_p$ ($x10^3$) | Strength (kPa) | % Elong. (100 in./in.) |
| 2 | 109 | 1385 | 0.45 | 111 | 1385 | 0.52 | 109 | 1490 | 0.46 |
| 4 | --- | 1295 | 0.44 | --- | 1450 | 0.45 | --- | 1570 | 0.46 |
| 6 | 94 | 1325 | 0.46 | 95 | 1415 | 0.46 | 86 | 1600 | 0.38 |
| 8 | 101 | 1415 | 0.46 | 107 | 1525 | 0.47 | 106 | 1570 | 0.40 |
| 10 | 93 | 1360 | 0.31 | 109 | 1390 | 0.40 | 106 | 1475 | 0.34 |
| 12 | 110 | 1260 | 0.40 | 106 | 1370 | 0.47 | 107 | 1515 | 0.31 |
| 12 (Ambient) | 97 | 1460 | 0.45 | 97 | 1460 | 0.45 | 97 | 1460 | 0.45 |
| 14 | 106 | 1390 | 0.47 | 104 | 1415 | 0.45 | 100 | 1605 | 0.32 |
| 16 | 94 | 1380 | 0.39 | 99 | 1420 | 0.46 | 113 | 1570 | 0.31 |
| 18 | 86 | 1525 | 0.38 | 89 | 1640 | 0.35 | 104 | 1750 | 0.43 |
| 18 (Ambient) | 94 | 1725 | 0.47 | 94 | 1725 | 0.47 | 94 | 1725 | 0.47 |
| 22 | 97 | 1370 | 0.32 | 97 | 1385 | 0.44 | 100 | 1600 | 0.32 |
| 29 | 101 | 1435 | 0.36 | 95 | 1470 | 0.42 | 100 | 1660 | 0.35 |
| 29 (Ambient) | 95 | --- | --- | 95 | --- | --- | 95 | --- | --- |
| 34 | 79 | 1345 | 0.42 | 60 | 1345 | 0.35 | 80 | 1620 | 0.40 |
| 34 (Ambient) | 94 | 1380 | 0.54 | 94 | 1380 | 0.54 | 94 | 1380 | 0.54 |

stable, energetic explosive when stored in sealed containers at temperatures lower than 60°C. Even at 90°C the material is more stable than other energetic explosives stored at 60°C. The enhanced stability is a consequence of the chemical nature of the binder system (a thermoplastic rubber-type polymer plasticized with a paraffinic oil) combined with a small amount (about 10 ppm) of antioxidant. Increasing the antioxidant content by a factor of 20 will extend the lifetime of X-0298 beyond any present service requirement without detectably affecting the physical characteristics of the X-0298.

## VI. ACKNOWLEDGMENTS

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