



The Response of Propellants to Plasma Radiation

by Richard A. Beyer and Rose A. Pesce-Rodriguez

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14. ABSTRACT Experiments have been done where standard and research propellants were exposed to the visible and infrared (IR) light from a high-energy plasma discharge. Measurements of mass lost by the sample, analysis of the IR spectra of evolved gases, and optical microscopy have been used to characterize the response. In addition, the response for graphite-free JA2 conditioned to hot and cold temperatures has shown that the magnitude of the response for this material is stronger for hot samples. Overall, the samples show strong response when the light can penetrate further into the samples; the response is limited to a thin layer at the surface for composite propellants.					
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Contents

List of Figures	iv
1. Introduction	1
2. Experimental	1
3. Observations and Discussion	4
3.1 Mass Loss	4
3.2 Analysis of Gas Products	4
3.3 Temperature Effects on GF-JA2 Response	7
3.4 Wavelength Dependence and Response	9
4. Photographic Documentation of Propellant Response	9
5. Summary	16
6. References	17
Distribution List	18

List of Figures

Figure 1. Electrical parameters for short (top) and long (bottom) plasma discharge pulses.	2
Figure 2. Schematic of experiment to expose propellant samples to plasma.	3
Figure 3. Transmission curve for 1/8-in polycarbonate adapted from figure 10.16 in <i>Applied Optics</i> , John Wiley & Sons (1).	3
Figure 4. Mass loss of GF-JA2 samples plotted as a function of peak power (top) and pulse energy (bottom).	5
Figure 5. Infrared absorption spectra for gas evolved from propellant samples when exposed to plasma light for (a) GF-JA2, (b) standard JA2, (c) M9, (d) M30, and (e) TGD-033 and TGD-031.	6
Figure 6. Relative mass loss vs. peak plasma pulse power for GF-JA2 at three sample temperatures.	8
Figure 7. The rate of continuing mass loss after the plasma light exposure for three sample temperature conditions.	8
Figure 8. GF-JA2 illuminated from low, medium, and high peak power plasma discharge (top to bottom).	10
Figure 9. Virgin M9 (top) and irradiated M9 propellant irradiated from bottom edge of photo, with damage throughout the sample.	12
Figure 10. Standard JA2, irradiated from bottom edge of photo, with damage limited to region near illuminated surface.	13
Figure 11. M44 propellant before (left) and after (right) exposure to plasma light.	13
Figure 12. Plasma-light exposed M30.	14
Figure 13. St. Marks Powder X5977 and 5978 single-perf grains after plasma irradiation.	14
Figure 14. ETPE propellants after plasma irradiation.	15

1. Introduction

Research to understand the physics and chemistry of the interaction of high-temperature plasmas with gun propellants has been widely discussed for several years. An important difficulty has been in separation of the components: (1) hot, high-velocity flow, (2) highly reactive chemistry in the flow to attack the propellant surface, and (3) a broad spectrum of radiation at intensities unknown in conventional ignition and combustion. While all of these elements are present in most plasma ignition scenarios, the relative importance of the plasma components will depend on many factors from geometry to pulse length and peak power of the discharge.

In the present study, attempts have been made to limit the plasma-propellant interaction to that of radiation only. Because of the particular approaches used here, the spectrum is limited to portions of the visible and infrared (IR) regions. Ultraviolet (UV) light, which is both abundant in the plasma (but not necessarily at the propellant) and chemically important, has not been explicitly addressed.

2. Experimental

The observations described here used a conventional pulse-forming network (PFN) pulsed power source to drive the plasma discharge. It consists of a 1700-mF capacitor and inductors of 12 and 305 mH for short (1 ms) and long (10 ms) pulses, respectively. A clamping diode is used to prevent voltage reversal at the capacitor. A model NL2888A Ignitron switches the current. While the system is rated at 11 kV, the maximum charging voltage of these observations was 7.7 kV. Current in the discharge is measured by integrating the output of a Rogowski coil. The potential across the discharge is measured by recording the voltage at each electrode with a set of high-voltage probes. Typical electrical parameter behavior for the two pulse lengths is shown in figure 1.

A schematic of the apparatus is shown at figure 2. The discharge is between two 4-mm-diameter tungsten (W) electrodes separated by 40 mm. A transparent polycarbonate tube with 15.2-mm inner diameter and 19-mm outer diameter contains the discharge. This tube serves to isolate the propellant sample from nonradiative effects of the discharge as well as enhancing the radiation by increasing the pressure of the contained plasma. A generic optical transmission curve for polycarbonate is shown in figure 3. As is seen there, it has broad transmission across the visible and near IR, and limited transmission in the IR. In particular, it has little absorbance near 1650 cm⁻¹ (~6.1 mm) where strong nitrate absorption is known. Initiation of the discharge is by a 0.13-mm-diameter nickel fuse wire. Explosion of the wire typically consumes ~20 J. The

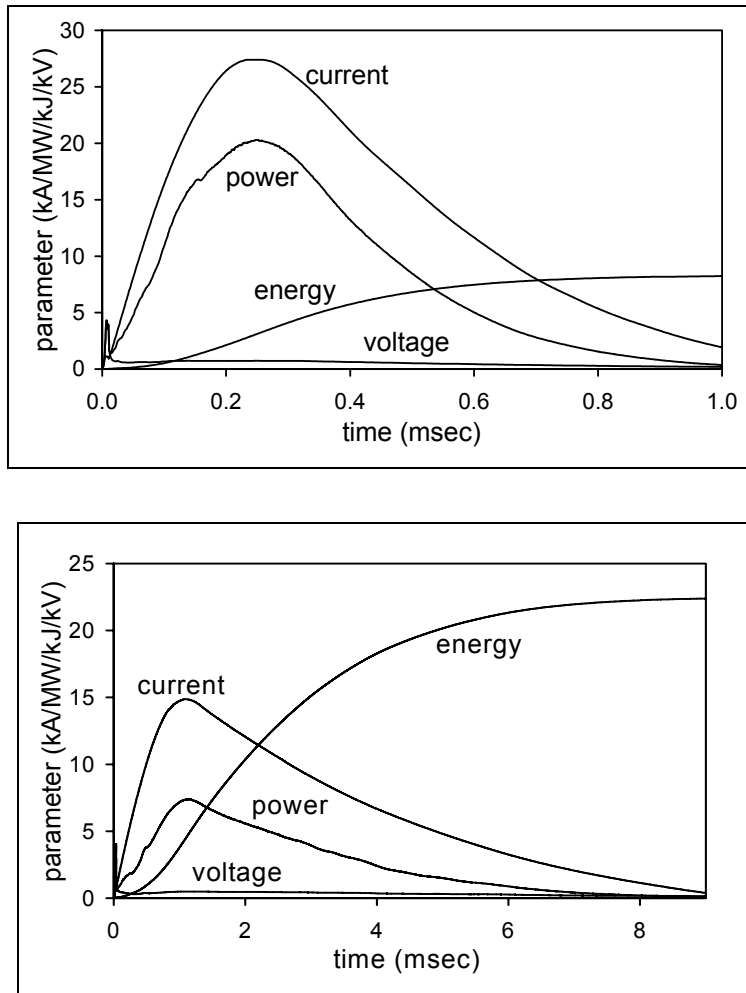


Figure 1. Electrical parameters for short (top) and long (bottom) plasma discharge pulses.

composition of the discharge is unknown. The mass of the wire and the air initially in the tube are about equal.

The primary propellant used here is JA2 sheet, processed without the usual 0.05% graphite, referred to here as graphite-free JA2 or GF-JA2. JA2 is a modified double-base propellant (nitrocellulose [NC] with nitroglycerin [NG] and diethylene glycol dinitrate [DEGDN] plasticizers). The nomenclature for the lot used here is JA2/RPD-445/RAD-PDI-2001-24. Average sheet thickness is 4.2 mm. The rectangular propellant samples were mounted ~2 mm outside (but not touching) the polycarbonate tube as shown in figure 2. Other standard and experimental propellants were also studied in limited quantities. They will be described in more detail as appropriate.

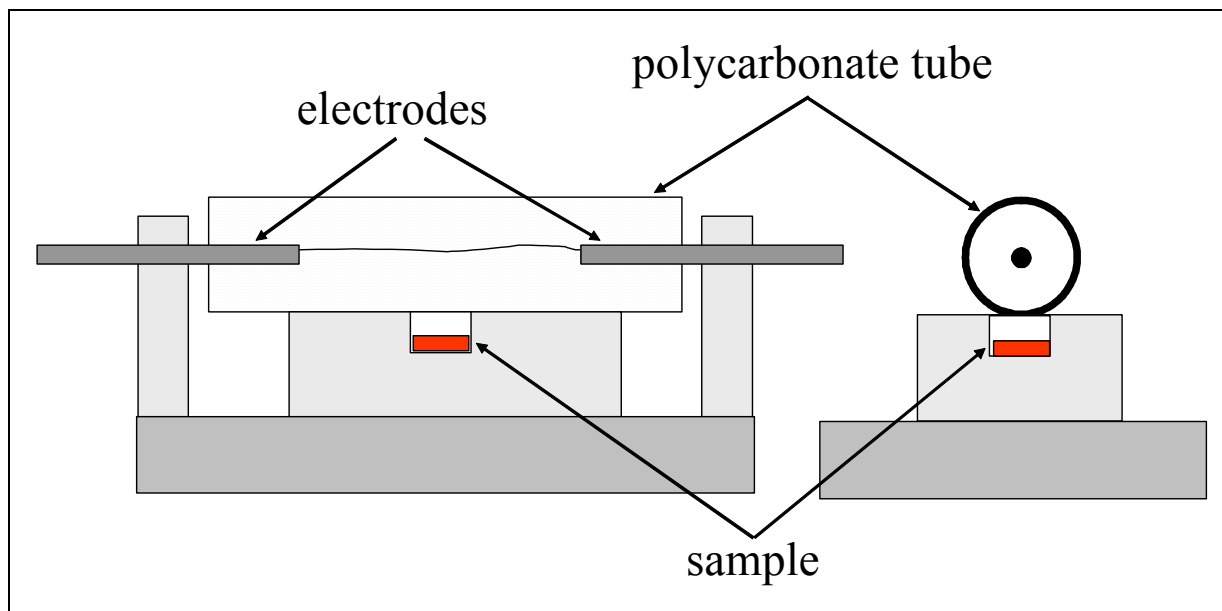


Figure 2. Schematic of experiment to expose propellant samples to plasma.

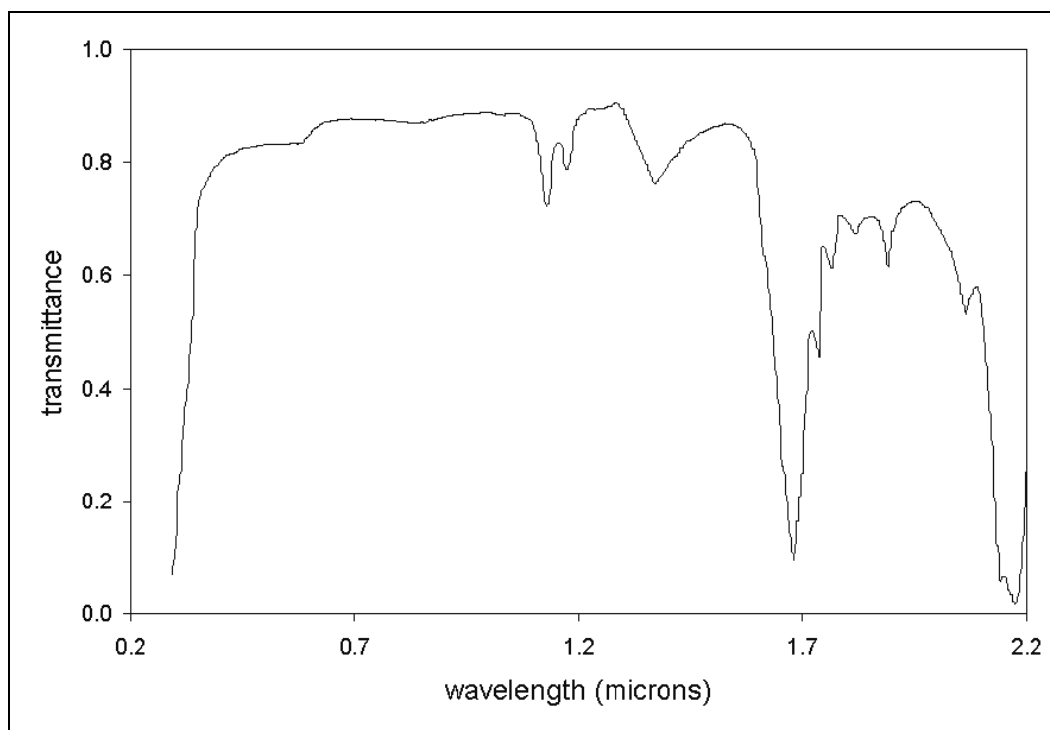


Figure 3. Transmission curve for 1/8-in polycarbonate adapted from figure 10.16 in *Applied Optics*, John Wiley & Sons (1).

3. Observations and Discussion

3.1 Mass Loss

In doing chemical analysis profiling of exposed samples of JA2, it was first noted that the sample thickness was variable (2). This resulted in a systematic study of the changes in sample thickness and sample mass loss, which has been reported elsewhere (3). The key finding of that report was that, under conditions where radiation is important, peak power (or peak current) is more appropriate for characterizing a plasma discharge than is the total energy of the discharge. Figure 4 shows the mass loss data from that study plotted as functions of energy or power. The good correlation with peak power is not unexpected because only the radiation from the plasma is interacting with the propellant and maximum radiation intensity scales with peak power and not energy.

3.2 Analysis of Gas Products

Because of the magnitude of the mass lost from these samples, it was decided to capture and analyze the gases produced. The first level of analysis was with methyl violet indicator paper, which is commonly used to detect propellant decomposition through its interaction with nitrogen oxides. The tests were done by placing a small propellant sample and the indicator paper into vials and then exposing it to the plasma radiation. Although the plasma light was found to bleach the paper, it was found that placing the paper behind the propellant sample was sufficient to preserve the paper color. It was observed that generally the indicator paper did respond to the evolved gases from the propellants (especially JA2) and that more power in the plasma discharge yielded a more rapid response from the paper.

Because of the magnitude of the response with methyl violet paper, a more detailed chemical analysis of the gases was pursued. This was achieved by placing the propellant samples into 5-mL gas-tight syringes and placing that assembly against the outside of the polycarbonate discharge tube. The amount of gas produced in many cases was sufficient to force the plunger from the syringe.

The gas was then injected into a gas sample cell of a Fourier transform infrared (FTIR) analyzer for recording of the IR spectral absorption signature. The results of these analyses are shown in figure 5 for GF JA2, standard JA2, legacy propellants M9 and M30, and two crystal-filled energetic thermoplastic elastomer (ETPE) propellants denoted as TGD-031 (ETPE/RDX/NQ) and TGD-033 (ETPE/RDX/TEX).

Several observations can be made from the absorption spectra. The results of JA2 (figure 5a, 5b) indicate that while the magnitude of the response is quite different with and without graphite present, the gas composition produced is the same. In both cases, there is abundant CO and CO₂

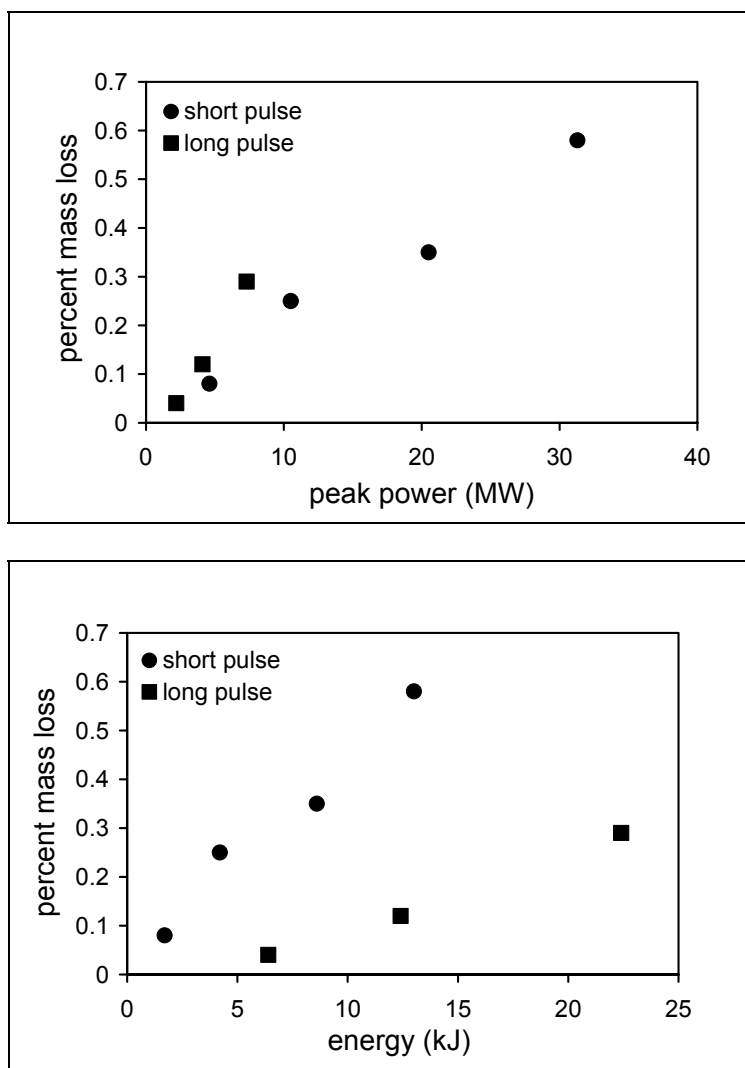


Figure 4. Mass loss of GF-JA2 samples plotted as a function of peak power (top) and pulse energy (bottom).

and CH_4 but no detectable oxides of nitrogen. This result indicates that the graphite in standard JA2 does not change the extent of reaction from the plasma radiation but merely serves to shield the inner portion of the sample from the light energy.

The two legacy propellants, M9 and M30, are expected to have responses which are much different from each other. M9 is transparent and quite similar in physical response to the GF-JA2, with strong physical changes noted deep into the samples. The gas-phase analysis confirms this observation except that there is not as much CH_4 observed from M9. The abundance of N_2O in the M30 gas phase products is probably from nitroguanidine decomposition. Because M30 response to the plasma has been characterized as a strong interaction limited to a thin layer at the surface, this result is consistent with our earlier observations.

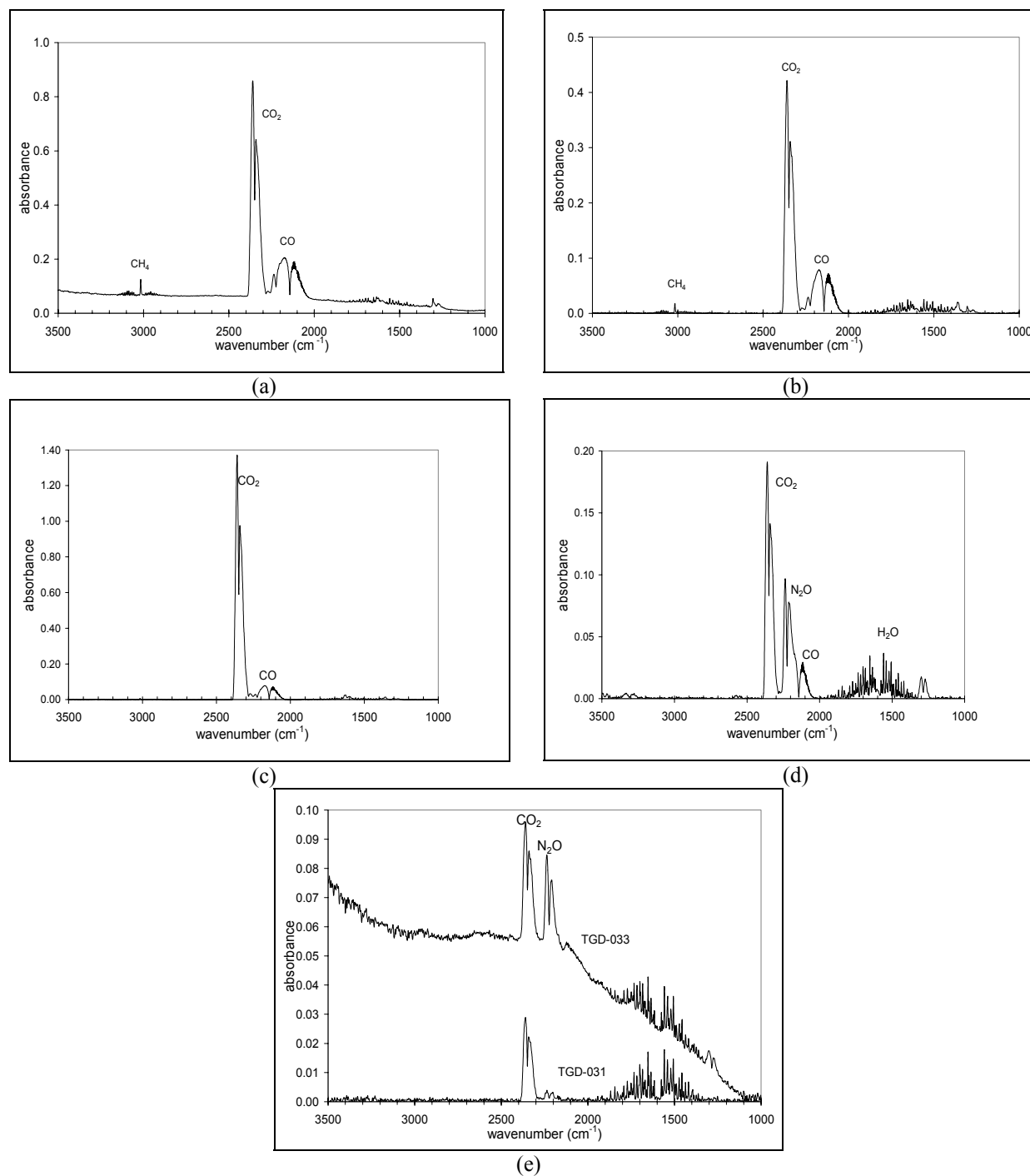


Figure 5. Infrared absorption spectra for gas evolved from propellant samples when exposed to plasma light for (a) GF-JA2, (b) standard JA2, (c) M9, (d) M30, and (e) TGD-033 and TGD-031.

The ETPE propellants show surprisingly differing responses as seen in figure 5e. (Note that the baseline differences are not significant.) Both are filled with RDX, which usually yields significant quantities of N₂O during thermal decomposition. TGD-031 also contains significant amounts of nitroguanidine. However, it would appear that the key observation here is to note the

values of the absorbance values. The three composite propellants (M30 and the TGDs) are showing a small surface response to the plasma light and thus are producing quite small quantities of gas products.

3.3 Temperature Effects on GF-JA2 Response

The mass loss experiments previously mentioned were repeated with the propellant samples carefully temperature conditioned prior to the plasma exposure. Three temperatures were used: $-31\text{ }^{\circ}\text{C}$, “cold,” $60\text{ }^{\circ}\text{C}$, “hot,” and “ambient,” which was typically $22\text{--}24\text{ }^{\circ}\text{C}$. Samples were weighed before conditioning and within minutes after exposure to the plasma. Samples were conditioned for ~ 3 hr. The time between removal of the samples from the conditioning chamber to the plasma discharge was typically 20–30 s.

It was expected (and observed) that the hot-conditioned samples lost mass (plasticizer) during conditioning. To compensate for this effect, double samples were prepared for the hot experiments. For each exposed hot sample, a second control sample was also taken from the conditioning chamber and placed near the experiment but well-shielded from the plasma. Both samples were then weighed after exposure of the one. An average mass loss value was determined from the conditioned but unexposed samples. The variability in the loss from these unexposed samples is an important source of scatter in the measurements.

The results of the averages of two series of experiments are shown in figure 6. The error limits indicated are estimated based on the scatter and uncertainty of the measurements. As can be seen, there is a clear trend toward higher mass loss (propellant response) with increasing sample temperature.

Because of the magnitude of the mass loss from the samples upon plasma light exposure, it was decided to monitor the mass of the samples over an extended period of time to determine if chemical reactions had been initiated that would make the propellant further decompose. The samples were stored in paper envelopes so that evolved gases were allowed to escape and autocatalytic effects would be minimized. The samples did continue to lose mass during the ~ 60 days that they were monitored. This series of observations resulted in a large amount of data that was at first sorted by temperature and power. However, in the final analysis, it seems to make more sense after averaging all the power levels together for each temperature. A plot of the summary of these data is shown in figure 7. This figure indicates that within the scatter of our data after plasma exposure, the rate of loss of mass from the GF-JA2 samples is the same over the next 60 days as if the samples had been temperature conditioned only, i.e., the plasma had no residual effect on the mass loss rate of our samples.

These observations indicate that the porosity of the exposed GF-JA2 is sufficient that no significant amounts of decomposition products are trapped in the propellant. This conclusion is important in the flame spread into the interior of such a propellant following plasma exposure. If

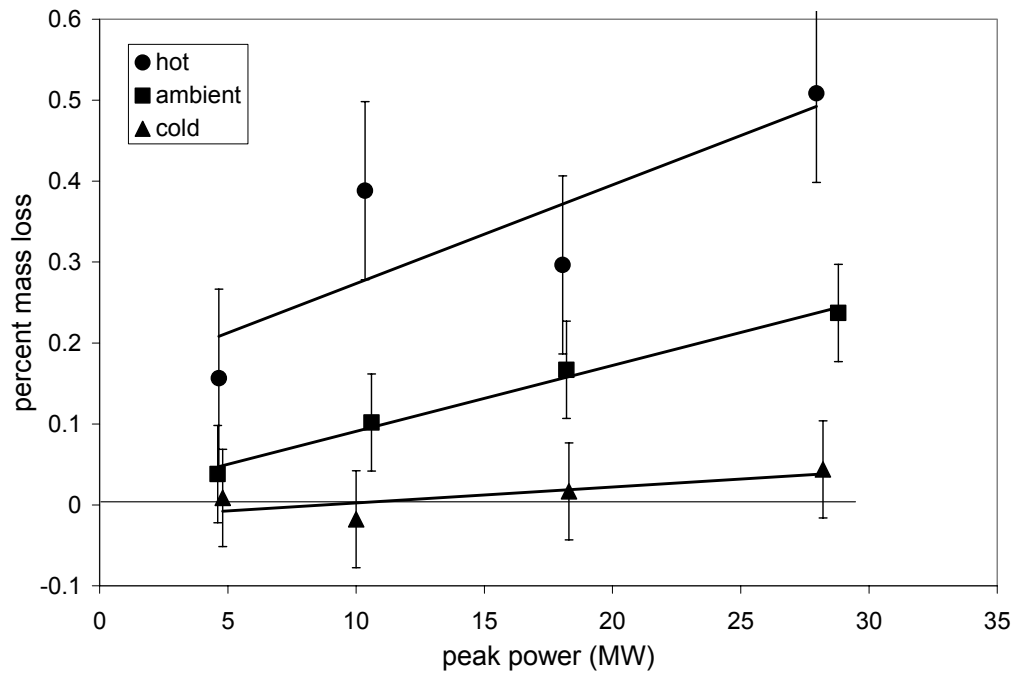


Figure 6. Relative mass loss vs. peak plasma pulse power for GF-JA2 at three sample temperatures.

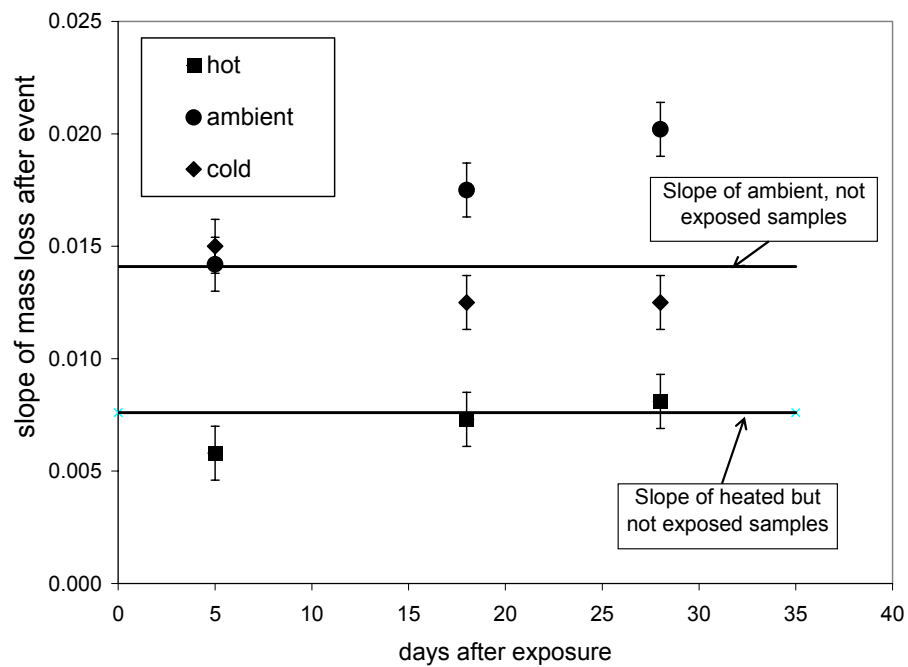


Figure 7. The rate of continuing mass loss after the plasma light exposure for three sample temperature conditions.

the gases flow out freely, then the interior “bubbles” observed are accessible for flames and will be expected to be burning surface area shortly after ignition of the outside surface of the same grain.

3.4 Wavelength Dependence and Response

An often-raised question in this area of research is whether there are specific wavelengths or regions of light that produce stronger responses from the propellant. A series of preliminary experiments was performed with colored glass filters to isolate broad regions of the spectrum in the green to red regions of the visible spectrum. Although there was substantial uncertainty in these experiments, the only clear correspondence was with the energy through the filters and the response of the propellant; wavelength appeared to be of secondary importance.

A specific wavelength that has been mentioned (4) as being of possible interest is at 1650 cm^{-1} . This peak in the JA2 absorption spectrum is at the position of a known absorption of nitrate esters. In order to remove this light from the interaction with the propellant, a suitable filter was sought. It was noted that water also absorbs at this same frequency. The apparatus shown in figure 2 was modified slightly so that the sample located below the discharge was in a cylindrical volume which could be filled with water to above the top of the propellant sample.

The result was that GF-JA2 samples showed approximately the same level of response with and without the water present. There was a major difference however. With the water, the samples were further delaminated or pulled apart along the regions of the usual damage. The result was consistent with pressurization and sudden release of the pressure. Our interpretation of this observation is that the water did absorb enough energy to ablate its surface and send a shock wave through the sample. The energy absorbed by the water did not visibly decrease the response of the GF-JA2.

4. Photographic Documentation of Propellant Response

During the course of this study, much time had been spent documenting propellant response photographically. While a thorough record of this would require a separate report, a limited number of figures are included here to show the difference in response of some legacy and ETPE research propellants. These pictures will perhaps help bring more understanding to the variation in the response of these propellants to the plasma discharge. The field of view in the photos without a scale is $\sim 12 \times 8\text{ mm}$.

The response of GF-JA2 to the plasma has been previously documented in our reports (2, 3) and by others (5). Figure 8 shows propellant response that is moderate, though strong. The “bubble-like” features are very much two-dimensional. The probable explanation of this feature is addressed in detail in another report being prepared concurrently with this one (6). The

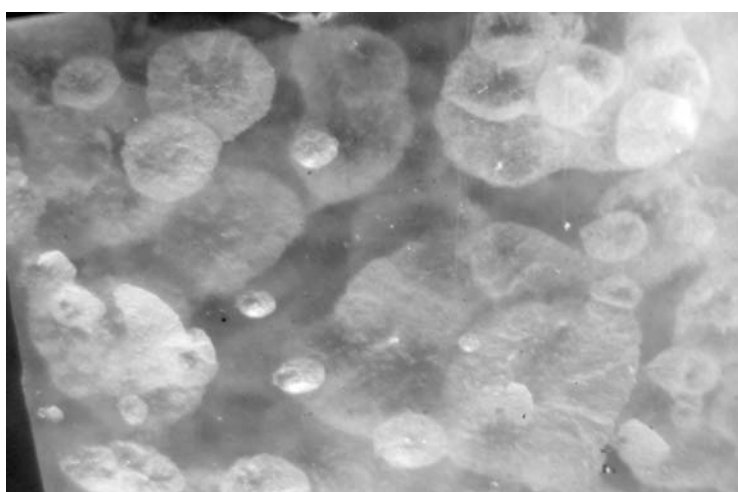
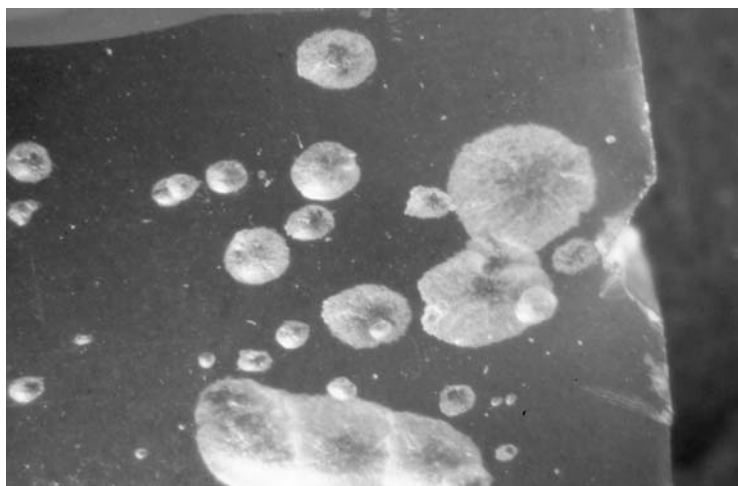


Figure 8. GF-JA2 illuminated from low, medium, and high peak power plasma discharge (top to bottom).

three photos here are taken from the direction of the surface of the flat carpet roll propellant. The amount of “damage” is very much dependent on both the peak power of the plasma pulse and the initial temperature of the sample. The result of increasing plasma power or increasing initial temperature does not appear to be distinguishable using optical inspection. Note that as the response increases, the characteristic size of the bubbles decreases so that there are both more and smaller features.

Figure 9 shows slices taken from a sample of M9 propellant in 0.25-in-diameter stick form before and after plasma irradiation. This propellant is semi-transparent or translucent, somewhat like GF-JA2. As can be seen, the light has affected the samples throughout the volume. The stick was cut axially through the diameter, and the flat surface faced the plasma discharge (lower edge of the photo). Figure 10 shows a section of a similarly treated sample of (standard) stick JA2. In this typical sample, the damage is isolated to the lower edge, which faced the incident light from the discharge.

Figure 11 shows M44 propellant that is virgin (left) and plasma-light exposed (right). This propellant contains sufficient carbon black that it is strongly light absorbing. All the features shown are at the surface with no indication of penetration to any significant depth. Figure 12 similarly shows M30 propellant that has been exposed. It shows no effects at all under detailed optical microscope inspection. The samples for both M44 and M30 are 0.25-in solid stick.

Figure 13 shows grains of double base propellants X5977 and X5978 from General Dynamics St. Marks Powder. They were chosen because of their optical transparency. They are similar in appearance to M9 except that they are quite rigid. This rigidity is the result of reduced plasticizer, which may also result in a greater number of bare NC fibers as potential optical absorption points. More discussion of this interpretation is presented in a separate article (2). As can be seen in the figure the response is similar to M9 and GF-JA2 but the hardness of the material results in physical fracture of the grains. This feature could possibly be exploited in an ignition design.

Research ETPE propellants from Thiokol are shown in figure 14. Both TGD-031 and TGD-009 show the characteristic surface blisters that are observed with these propellants when exposed to the light from the plasma discharge. Although the samples are typically on the order of 2.5 mm thick, the damage is usually confined to the front surface; however, an occasional blister will appear on the rear surface. Whether this is indicative of random light transmission through the material or another process has not been determined.

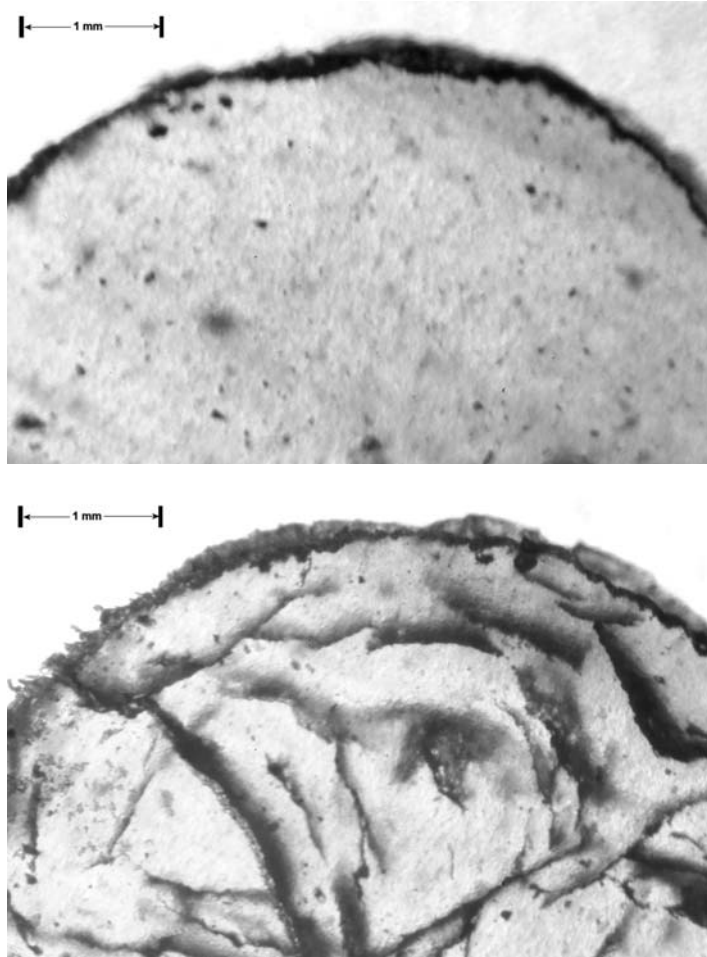


Figure 9. Virgin M9 (top) and irradiated M9 propellant irradiated from bottom edge of photo, with damage throughout the sample.

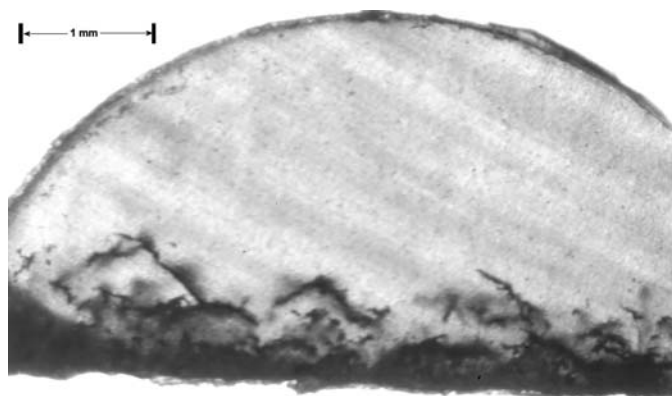


Figure 10. Standard JA2, irradiated from bottom edge of photo, with damage limited to region near illuminated surface.

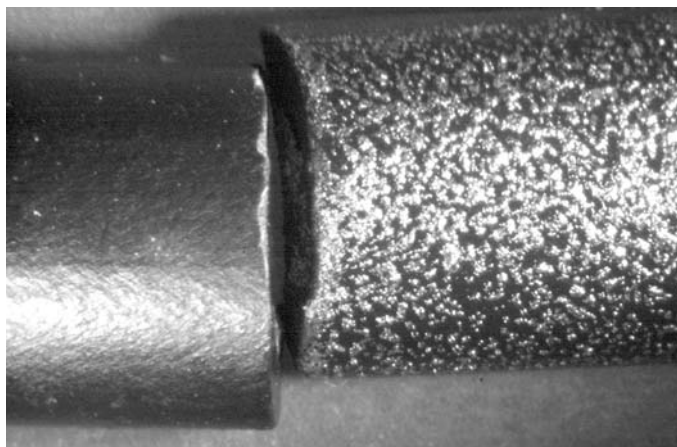


Figure 11. M44 propellant before (left) and after (right) exposure to plasma light.

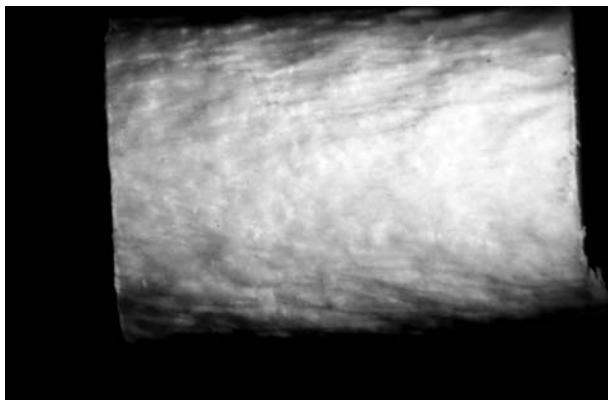
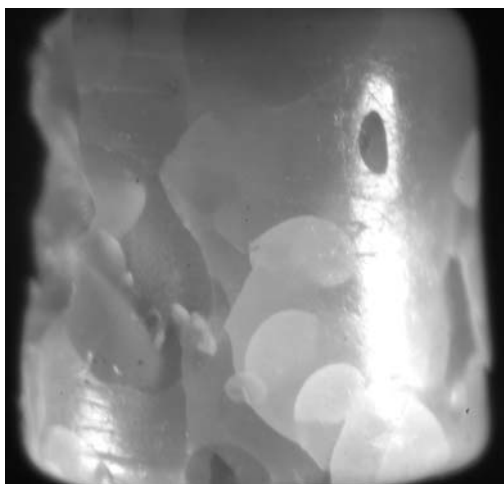
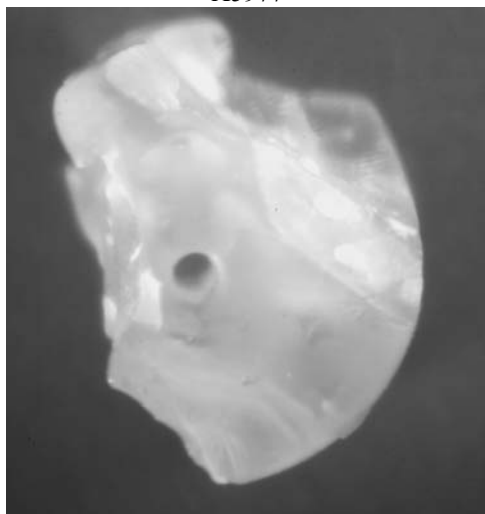


Figure 12. Plasma-light exposed M30.

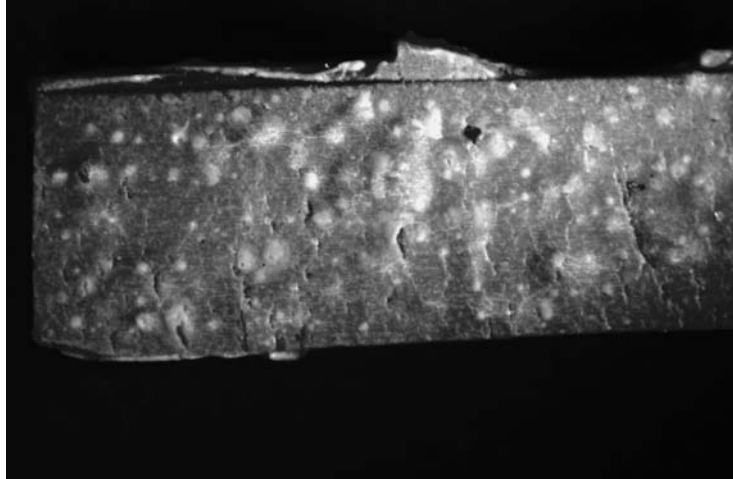


X5977

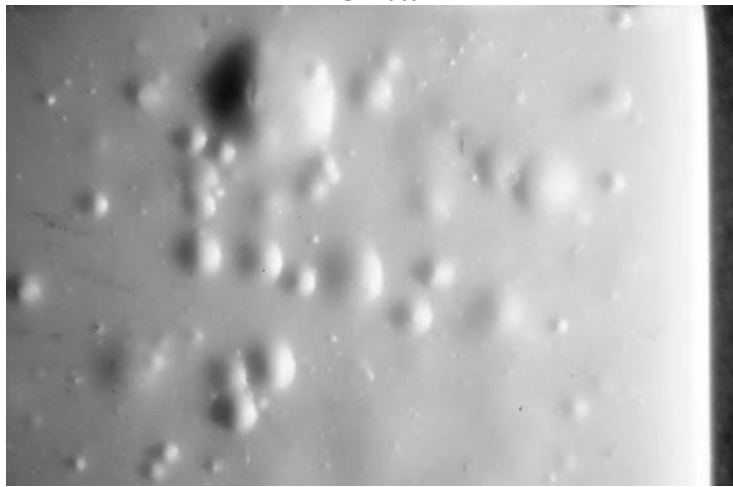


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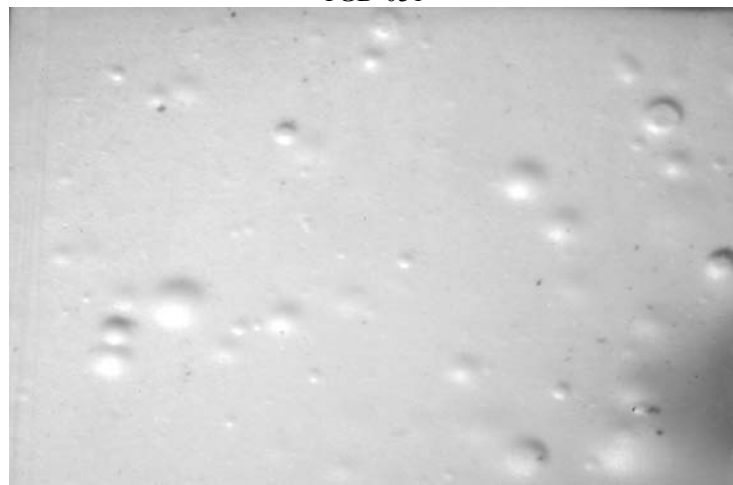
Figure 13. St. Marks Powder X5977 and 5978 single-perf grains after plasma irradiation.



TGD-009



TGD-031



TGD-033

Figure 14. ETPE propellants after plasma irradiation.

5. Summary

A portion of the radiant spectrum from a high-energy plasma discharge has been isolated for interaction with a variety of gun propellants. In those propellants with sufficient optical transparency, chemical activity and physical damage are observed at depth in the samples. It is expected that this type of interaction would produce significantly enhanced surface area for burning of exposed propellants. Composite propellants addressed here are uniformly quite opaque and produce localized surface interactions. The gas phase products which result from these interactions are normal for propellant reactions; optical transparency and stronger response produce greater amounts of reaction and gas-phase products.

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R A BEYER
A BIRK
A L BRANT
S W BUNTE
C F CHABALOWSKI
L M CHANG
T P COFFEE
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C LEVERITT
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