Popping Phenomena with the Hydrazine Nitrogen-Tetroxide Propellant System

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The propellant spray resulting from the impingement of liquid jets of hydrazine and nitrogen tetroxide has been studied at atmospheric pressure by means of streak photography. The streak photographs show periodic small explosions that originate near the impingement point and propagate through the propellant spray at velocities of 3000 to 5000 fps, consuming all propellant droplets over a distance of up to 6 in. Typical streak photographs are presented. The frequency of the explosions or pops ranged up to several hundred cps, and could be controlled by varying the contact time in the liquid phase. Below a minimum threshold contact time, popping did not take place. At high values of contact time, the popping rate was controlled by the transit time of the free jet before impingement. Flashing of the oxidizer prior to impingement prevented popping under certain conditions. It is postulated that popping is initiated by liquid phase reactions. A mechanism for the occurrence of popping and its relation to reactive stream separation is suggested.

I. Introduction

THE propellant combination of nitrogen tetroxide and hydrazine has been widely studied and has shown several unusual characteristics, but a satisfactory phenomenological description of the spray formation and spray combustion process is not yet available. As part of a general program to investigate reactive stream impingement with a single, unlike-doublet injector, it was found that, under certain operating conditions, very fast high-amplitude pressure pulses occurred. Such disturbances have been reported by other investigators and have been labeled "pops." Because of their nature, pops can effect efficiency and may be destructive or instrumental in initiating more destructive combustion instability.

In this paper a pop will be defined as a local small explosion of propellant droplets, resulting in conversion of the propellant drops into gaseous combustion products. The explosion results in a pressure wave that decays fairly rapidly with distance. The reaction behind the wave does not necessarily consume all of the reactants. A pop itself has no relation to a combustion instability wave, although a pop may trigger such instability.

Within this frame of reference, it appears that the first observations of popping-like phenomena with nitrogen tetroxide and hydrazine were made by Elverum and Staudhammer[1] at JPL. They observed periodic explosions at a rate of 100 times/sec with an 80-lb thrust unlike doublet at atmospheric pressure and also at 200 psia. Similar observations were also reported by Burrows, Mills, Campbell, and Zung. Burrows[2] at NASA Lewis Research Center observed periodic variations in atomization distance at a frequency of approximately 1250 cps, using a quadlet injector element. Mills et al.[3] observed explosions near the injector plate, which were classified as ligament shattering and as spray detonations. Campbell et al.[4] made a photographic study of nitrogen tetroxide and hydrazine impingement at atmospheric pressure. Their high-speed films show the impingement region in great detail, particularly the periodic nature of spray formation and explosion. Photographic coverage of popping at elevated pressures has been provided by Zung et al.[5]

The popping phenomena noted above appear to be similar in nature to the phenomena studied in this project in the sense that the initiating mechanism seems to be controlled by chemical reaction parameters. This is in contrast to popping phenomena described by Valentine et al.,[6] where hydraulic flip acts as a trigger for pops.

In JPL's work on the closely related problem of reactive stream separation in impinging jets, popping phenomena, as observed at elevated chamber pressures, were correlated and reported in Ref. 7. This paper presents the results of an investigation of these popping phenomena at atmospheric pressure. Streak photography was used to establish the origin, the propagation velocity, and the extent of the pops. It is shown that popping can be controlled by varying the injection parameters.

II. Streak Photography Experiments

The experiments were conducted with an unlike doublet in a small experimental engine. The combustion chamber was 9 in. in length and 3 in. in inside diameter, with one end open to the atmosphere. Consequently, the chamber pressure was essentially atmospheric. The tests were also run without a chamber to obtain streak films of the burning propellants. Orifice sizes ranged from 0.029 to 0.173 in. diam with a fixed impingement angle of 45°. Free jet length was $4D$ for all tests and orifice L/D was 100 for all orifices tested. Unless otherwise specified, the propellant was conditioned to a temperature of 40° F to prevent flashing of the oxidizer. A mixture ratio (oxidizer/fuel) of 1.2 was used to produce equal stagnation pressures in each jet. This condition also meets the Rupe criteria for optimum mixing of nonreactive jets. The propellant flow rate was decoupled from downstream pressure fluctuations by the use of cavitating venturis in the propellant lines. A Taber gauge on the injector face was used to measure pressure pulses.

Popping was audibly detected as a loud machine-gun-like sound and was given quantitative credence in the form of oscillograph records of a series of high-amplitude pressure pulses. These data were used to determine popping rates and relative amplitudes. Figure 1 is a typical pressure trace taken during a test with the throatless chamber incorporating a set of 0.073 in. diameter injector orifices. The peaks vary in amplitude up to a pressure of 50 psi, while the frequency is of the order of 50 pops/sec. In a few check runs, simultaneous photocon measurements were made for comparison

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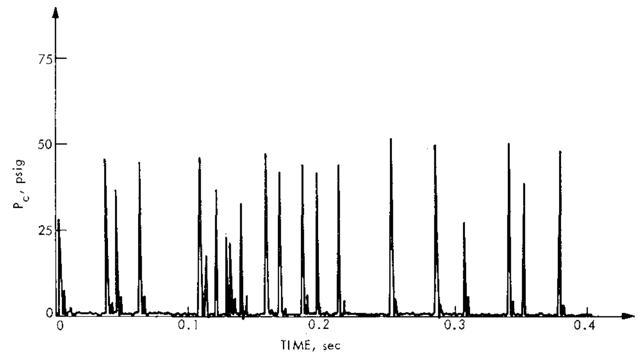

Fig. 1 Typical oscillograph pressure trace of popping.

with the Taber measurements. The two pressure traces looked virtually identical, indicating that the response of the Taber gauges is quite adequate to record popping. With 0.173-in.-diam orifices, pressure spikes with amplitudes of 300 psi were recorded.

Streak films were made of open firings in the atmosphere without a chamber. The streak films are a continuous picture of the combustion process over a 1-in.-wide area from the impingement point to 9 in. downstream, as shown in Fig. 2. The film moves in a direction normal to the direction of the burning propellants, and popping velocities can be determined from their film traces and the film speed. Figure 3 shows some typical results of the streak photography. This figure is a negative, and any luminous particle shows up as a dark trace. Without popping, the film shows the traces of burning droplets traveling at approximately the injection velocity, as shown in Fig. 3a. Figure 3b shows a single pop, while Fig. 3c shows 4 pops in succession. The pop in Fig. 3b shows a well-defined propagation angle which was used to determine that pops propagate within a velocity range of 3000 to 5000 fps. The rate data from the pressure traces checked with the rate data taken from streak films of the same firing, as expected.

The sonic velocity for the adiabatic combustion gases is 3760 fps for the overall mixture ratio of 1.2. The actual sonic velocity is somewhat lower near the injector as the combustion process is incomplete. Therefore, it must be concluded that pop propagation is a supersonic wave phenomena with Mach numbers of the order of 1 to 2. It is apparent that the pop starts close to the injector face, and that immediately downstream of the pop there are no propellant droplets left, as there are no traces on the film (white area on right). However, this area extends only to a distance of approximately 6 in. from the injector face. Immediately after a pop, propellant drops start to show on the film and the spray pattern is being re-established (dark area on left).

Figure 3c shows 4 pops in succession, each one slightly smaller in depth than the previous one. It is apparent that a pop can start long before the complete spray pattern has been re-established.

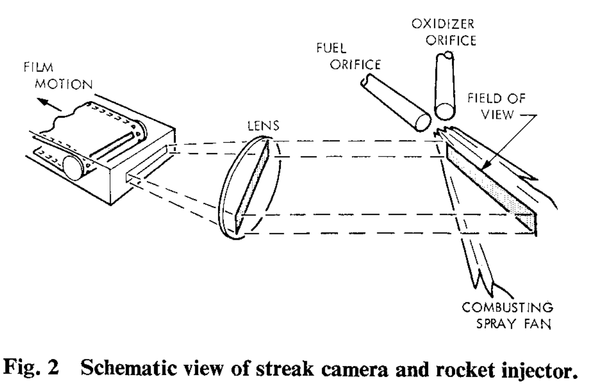

Fig. 2 Schematic view of streak camera and rocket injector.

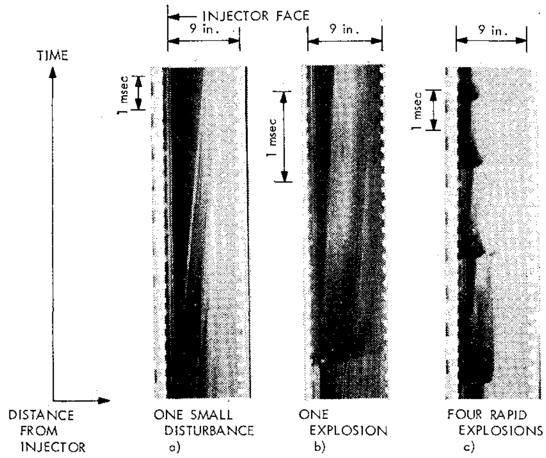

Fig. 3 Streak film segments of burning propellants.

Close examination of Fig. 3a shows a white streak, which indicates an absence of propellant droplets as a result of a small disturbance that consumed droplets over only a very small area. These disturbances are probably similar to the "puffs" observed at Rocketdyne.[4] The puffs appear to be somewhat more cyclic than pops, but no rate data were measured. They were observed with both the 0.073 and 0.173 in. diam orifices and appeared to occur independently of the pops at any time before or after a pop. It was also noted that they seem to originate between the impingement point and approximately 1 in. downstream. The puffs are too small to establish the rate of propagation.

A number of general observations can be made regarding the streak film data. All large pops originated close to the impingement point. A pop never consumed propellants further than approximately 4 to 6 in. downstream of the impingement point, even if propellants were visible across the entire field. Beyond this 4 to 6 in. range the propellant droplet density is probably too low to support propagation of the pop. This would suggest that larger propellant flowrates would result in larger pops. However, the streak photographs did not show a definite trend with either higher injection velocities or larger orifice diameters. On the other hand, the recorded pressure spikes of the 0.173-in.-diam orifices went as high as 300 psig compared to 50 psig for 0.073-in.-diam orifices, indicating a higher propellant density but apparently no appreciably larger propellant field. Very often an intense white spot of very small size and duration was evident on the streak film near the impingement point (see Fig. 3b). It is suggested without verification that these spots mark the location of the initiation of the disturbance. Sometimes, the very intense initial disturbance was followed by two to four similar disturbances within approximately 0.5 msec.

The pop propagation velocity did not change through the field, even though the intensity was attenuated except in cases where a disturbance entered a region where propellants had already been consumed; in such a transition, the velocity radically decreased to another constant value. Very often, reflections of the very intense disturbances appeared at the boundary between regions of very different densities. The time intervals between pops were not exactly the same although long-time average popping rates were very repeatable.

III. Popping Rate Correlation

Generally, the popping rate showed a consistent correlation with the ratio of jet diameter to jet velocity (D/V), almost independent of orifice diameter, as shown in Fig. 4. It has been shown[8] that D/V represents a good approximation for

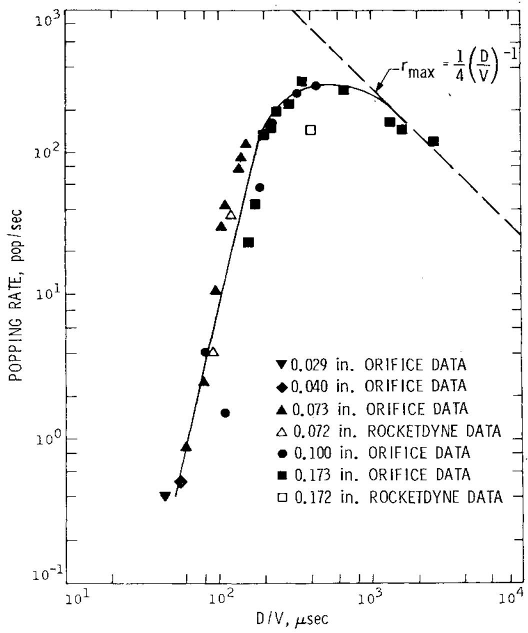

Fig. 4 Effect of contact time on popping rate at atmospheric pressure.

the average residence time of propellants in the impingement zone, and it appears to be a critical controlling parameter for popping. The plot represents data for orifices ranging from 0.029 to 0.173 in. diam, with jet velocities ranging from 5 to 150 fps. Figure 4 shows that the popping rate is very dependent on contact time and varies with approximately the fifth power of D/V. The maximum observed rate of approximately 300 pops/sec is probably associated with hydrodynamic effects. On the other hand the limiting threshold D/V is postulated to be determined by the chemistry of the liquid phase reactions.

The upper rate limit can be accounted for if it is assumed that the limiting factor is the time it takes for the jets to reimpinge after the free jet and the complete flowfield have been destroyed by a pop. For a free jet length of $4D$ and an injection velocity of V, this time is equal to $4D/V$, so the maximum popping rate will be $V/4D$. This relation is represented by the upper diagonal line in Fig. 4. The actual observed maximum popping rate for the 0.100 and 0.173-in.-diam orifices was very close to this theoretical maximum. This suggests that a pop occurs almost immediately after the jets make contact again, an indication of the extremely high chemical reaction rate for the liquid phase reaction. Over-all, the plot may be seen as a smoothing of a step function in going from no popping below the threshold D/V value to the high popping rates of the diagonal ($V/4\ D$) line. It is apparent that a design criteria for essentially pop-free operation of an injector dictates an operating point below the threshold contact time.

IV. Nature of Pop Propagation

As indicated previously, the measurements show that the wave propagation is supersonic, with Mach numbers of the order of 1 to 2, and the wave velocity is fairly constant over distances up to several inches. The latter suggests that pop propagation has a detonation rather than a blast wave mechanism, since the blast wave velocity decreases rapidly with distance. In order to verify this conclusion, blast wave velocities as a function of distance were calculated for this system. It was reasoned that the measured velocities should be compared with the maximum blast velocities that could theoretically arise from a pop. This maximum blast would be a point source with energy equivalent to all the propellant injected between two pops. Actually, this amount of energy will not be available since a considerable fraction of this propellant will burn between pops.

Landau and Lifshitz[9] derived the similarity solution for a spherical blast wave. Their results may be expressed as

$$V = k_1 (E/\rho_1)^{1/2} r^{-3/2}$$

and

$$P_2 = k_2 E r^{-3}$$

where V = propagation velocity, E = blast energy, ρ_1 = density of the undisturbed gas, r = distance from the center to the spherical wave front, P_2 = pressure just behind the front, k_1, k_2 = system constants depending on γ.

The propagation velocity and wave pressure were calculated for the case of 0.073-in.-diam orifices and a popping rate of 100 cps. At distances of 1, 4, and 6 in., the respective velocities are 8020, 1000, and 546 fps, while the peak wave pressures are 7030, 110, and 33 psia. From a comparison of this data with the virtually constant measured velocities of 3000 to 5000 fps, it appears that popping is not a blast-wave phenomena, and behaves more like a detonation wave. The pressure pick-up was located 1.25 in. from the impingement point, where the calculated maximum blast pressure would be 3600 psig. The point source assumption used in the calculation probably invalidates the pressure/distance prediction and hence cannot be verified experimentally.

Let us consider the fairly constant measured propagation velocity and the fact that the droplet concentration falls off rapidly with distance from the impingement point. Williams[10] carried out an analysis of detonations in sprays and derived the corresponding Rankine-Hugoniot equations. He found that, compared with a gaseous detonation with the same total heat release per unit mass, a Chapman-Jouguet detonation in a spray travels at a slightly higher Mach number with a pressure ratio that is roughly 10% larger. Specifically, Williams showed that the Mach number is virtually independent of the mass fraction of the total propellant flux that is present as liquid spray. This provides a possible explanation for the experimental fact that the wave velocity is fairly constant over distances up to 4 or 6 in., while the droplet concentration will drop off sharply over this distance. In other words, pop propagation seems compatible with detonation-like behavior.

V. Mechanism

The unique features of Fig. 4 provide the following conclusions: 1) At low values of D/V, below the threshold value, popping does not take place. 2) At high values of D/V, popping takes place as fast as contact of the free jets can be re-established. 3) In the transition region of intermediate values of D/V, the popping rate is controlled by approximately the fifth power of D/V.

In order to understand the above popping behavior it is necessary to review the phenomenon of reactive jet separation. With very reactive propellant systems, the gas phase reaction rate can be fast enough to maintain a gas film between the impinging jets.[8] Consequently, the jets do not contact each other and do not mix in the normal manner. Instead, the jets are deflected away by the gas film, resulting in a separated spray containing a fuel-rich zone and an oxidizer-rich zone.[11] Such a spray may be explosive in the fuel-rich zone.

We believe that popping is initiated by rapid liquid-phase reactions of the propellants. McLain and Ross[12] report that liquid-phase reactions produce metastable gaseous compounds, such as hydrogen azide and triazine. The presence of these compounds may well play a role in the mechanism of pop initiation. If the residence time D/V in the impingement zone is below the threshold value, not enough chemical reaction has taken place to bring the local conditions to the triggering point of pop formation. This triggering point may be determined by the concentration of metastable compounds leaving the impingement area, or a local temperature rise, or a combination of such conditions. Once this triggering condition is reached, a local explosion takes place at the exit of the liquid phase impingement area, and this explosion travels radially outward at a speed of 3000 to 5000 fps, up to a distance of 6 in. The explosion consumes all liquid propellant droplets within that area.

Previous work[11] with hypergolic propellants like N_2H_4/N_2O_4 has shown that there are two steady impingement configurations for unlike doublets—namely separation or penetration. Although the correlation is not unique for all jet sizes, the association of low D/V with separation is quite clear. As D/V increases, the trend is toward penetration. The most probable mechanism for maintaining separation appears to be the presence of a thin gas film between the jets that prevents the two jets from contacting each other.[8] It is now postulated that popping represents a cyclic deterioration of this gas film. Every time the gas film actually disappears and the two jets contact each other, a pop will occur, provided the contact time D/V is above the required threshold value. It is further postulated that in the transition region of intermediate D/V values this phenomenon is dependent upon the natural turbulent fluctuations in the position of the interface between the jets that are present in each liquid jet impingement situation. These fluctuations of the interface might produce penetration of the gas film with resultant contact of the liquids and a subsequent pop. In between pops, a pseudo steady state exists with a gas film between the jets, as the pressure trace in Fig. 1 suggests. This would explain why pops are not exactly equally spaced in time, although time-averaged popping frequencies are fairly reproducible. All the tests were run with fully developed turbulent jets so that no laminar-turbulent transitions took place.

The gas film separating the liquid jets in the separated mode is believed to be maintained by gas phase reactions of the propellants. Since the rate of reaction is proportional to the pressure, the gas film becomes more stable at higher pressures. Consequently, it should be more and more difficult for the jets to penetrate the gas layer to form a pop as the pressure is increased. Previous work[7] showed that indeed an increase in chamber pressure reduced popping and completely eliminated popping above 150 psia with a set of 0.073 in. diam orifices.

In all of the experiments discussed above, the propellant temperature was kept constant at approximately 40° F to avoid flashing of the oxidizer, which boils at 69° F at the local atmospheric pressure of 14.2 psia. However, in one series of tests when the oxidizer was allowed to flash by increasing the temperature of both propellants to 75° F, no popping took place at all. Apparently the evolution of oxidizer flash vapor prevented popping. To investigate this

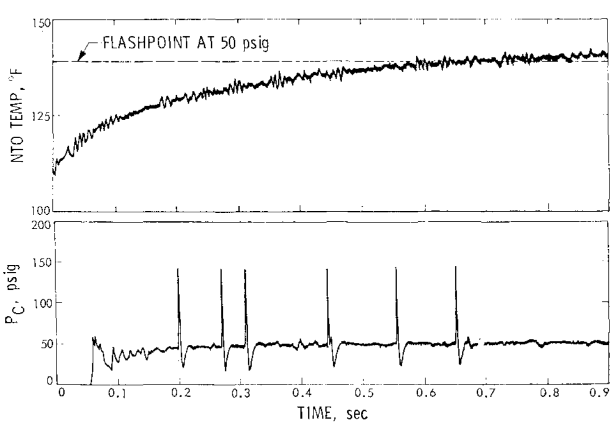

Fig. 5 Effect of flashing on popping.

effect over another temperature range, a test was conducted at elevated pressure by fitting the chamber with a nozzle and allowing the propellant temperature to increase during the run.

The resulting chamber pressure and the oxidizer temperature are plotted in Fig. 5. The chamber pressure reached semisteady state at 50 psig at 0.16 sec, but popping spikes occurred until the oxidizer temperature reached its boiling point at 133° F. No further pops occurred above this temperature. This test confirmed that propellant flashing can prevent pop formation, although not enough evidence is available to make this a general rule. When flashing takes place, the velocity of the oxidizer jet is greatly increased due to its lower average density. For instance, 5° F super heat would increase the velocity by a factor of 3. Consequently, the D/V value falls below the minimum threshold value for popping. A secondary effect might be that the presence of vapor could produce less effective liquid-liquid contact and so interfere with the liquid phase reactions and the production of metastable intermediates.

The preceding observations are confirmed by the experiments of Zung and White[13] with the same propellant system at atmospheric pressure. Interpreting their work in terms of the above mechanism, their experiments show that increasing the oxidizer injection temperature to above its boiling point eliminated popping.

VI. Conclusion

In summary, the observations discussed in this paper indicate that: 1) popping is a detonation-like phenomenon that begins near the impingement point and propagates through the propellant spray at velocities of 3000 to 5000 fps; 2) pops consume propellants over a distance of up to 6 in.; 3) there are other smaller propellant-consuming disturbances, so-called puffs, that do not develop into pops; 4) popping does not take place below a threshold value of contact time (D/V); 5) popping can only occur as rapidly as the jets are able to reimpinge; 6) flashing of propellants prior to impingement can prevent popping under certain conditions.

We postulate that popping is a result of the periodic breakdown of a gas film that separates the highly reactive propellant jets. When the liquid jets contact each other directly, a violent liquid-phase reaction takes place that produces metastable compounds. Upon entering the hot gas zone, these compounds detonate; this detonation triggers the explosion of the propellant spray immediately downstream of the impingement zone. However, not all contacts of the liquid jets result in pops. Popping does not take place unless the contact time is above a minimum threshold value. Also, the local composition of the propellant spray is not always the same, depending on the degree of separation that, in turn, controls the degree of mixing of oxidizer and fuel, and the disturbance may not propagate through the spray. Only weak disturbances (puffs) are observed in that case.

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Analytical Model of the Flash Produced in Aluminum-Aluminum Hypervelocity Impacts

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A theoretical model was developed which represents hypervelocity impact flash as radiation emitted from a high-temperature, optically thin, metallic plasma in local thermodynamic equilibrium. The model employs fundamental physical relationships to arrive at the radiation emitted from an impact plasma. Calculations were carried out for single element (pure aluminum) and multielement metallic plasmas. Two five-element plasma models were used to calculate radiation from type 2024 and type 6061 aluminum alloy impact plasmas. Calculations were carried out for the pressure of 1.0–100 atm and for temperatures of 5000 °K–40,000 °K. The theoretical results indicate that for these pressures and temperatures, the predominant type of radiation is that of spectral-line radiation. In order to obtain qualitative agreement between theory and experiment, it was necessary to include both impurity radiation effects and line shift-line broadening effects. This paper demonstrates that the flash emitted by a transient impact gas cloud can be represented as radiation from a high-pressure metallic plasma.

Introduction

EXPERIMENTAL studies[1–4] have indicated that the radiation emitted from a hypervelocity impact can be used to predict physical parameters associated with the projectile and target materials and to predict physical damage to the target. From only a knowledge of the impact flash, information on projectile mass, velocity,[4] and impact energy can be obtained. In an attempt to correlate the measured impact flash with the physical phenomena present in an impact event, the radiated energy is assumed to arise from high-temperature metallic gas at the projectile-target interface as shown schematically in Fig. 1. At the high temperatures expected, the hot gas will be in the plasma state. This physical model was adopted in order to predict the total radiation emitted from a unit volume of plasma in local thermodynamic equilibrium containing neutral atoms, ions, and electrons. By employing a fundamental approach to the calculation of the radiation, detailed knowledge of the spectral distribution of energy as a function wavelength and impact energy (or temperature of the impact plasma) can be obtained. This model permits quantitative calculations to be made for each of the radiation mechanisms. Such knowledge can assist in the design of diagnostic devices to measure impact flash. It also can assist in the design of devices to enhance radiation (e.g., illumination devices) and to reduce radiation (e.g., radiation and ionization in re-entry devices). Because the fundamental physical model is a general one, the mathematical equations can be applied to many practical situations.

Harwell, Reid, and Hughes[1] made an analytical study of an iron and aluminum plasma and found that such a representation

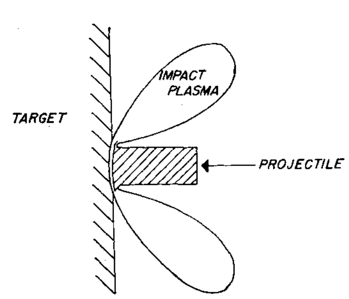

Fig. 1 Schematic representation of an impact plasma.

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