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Assessment of Poisson's Ratio for Hydroxy-terminated Polybutadine-based Solid Rocket Propellants

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ABSTRACT

Poisson's ratio of hydroxy-terminated polybutadine (HTPB)-based composite propellant is estimated from uni-axial tensile testing. Double dumbbell specimens as per ASTM D638 type IV standard were used and Poisson's ratio at break, obtained by change in volume of specimen, was calculated as approximately 0.25. It was also observed that Poisson's ratio is different along different lateral directions of the propellant specimen. Poisson's ratios in two orthogonal directions perpendicular to longitudinal axis were calculated as 0.17 and 0.30. As ASTM specimen has rectangular cross-section of approximate size 6 mm x 4 mm, the directional behaviour of Poisson's ratio may be attributed to initial dimensions. Prismatic propellant specimen with square cross-section and of 115 mm x 6 mm x 6 mm dimension do not show any variation wrt Young's modulus, tensile strength, and percentage elongation as compared to ASTM specimen. Directional behaviour of Poisson's ratio with almost similar numerical value was again observed, thus ruling out dependence of this behaviour on different initial dimensions of propellant cross-section. Further, Poisson's ratio varies linearly with strain even in linear portion of stress-strain curve in uni-axial tensile testing. The rate of reduction of Poisson's ratio with increase in strain is slower in linear region and it accelerates after dewetting due to formation of vacuoles. Variation of Poisson's ratio with strain has two different slopes in linear (slope = 0.3165) and nonlinear regions (slope = 0.61364). Numerical value of slope for variation of Poisson's ratio with strain almost doubles after dewetting. It must be noted that no change in volume does not necessarily indicate constant Poisson's ratio equal to 0.5. Composite propellants behave as compressible material in most of the regions and near-failure region or at higher strains; Poisson's ratio is not anywhere near to 0.5, instead it is near 0.25.

Keyword: Composite propellants, Poisson's ratio, tensile test, incompressible materials, filled polymers, solid rocket-propellants

NOMENCLATURE

- v Poisson's ratio=lateral strain/longitudinal strain
- ε , ε , Longitudinal strain
- ε, Lateral strain
- \vec{V} Instantaneous volume
- *Vo* Initial volume

1. INTRODUCTION

Solid propellants are used in rockets due to their simple design, compactness, safe transportation and quick operation. Hydroxy terminated polybutadine (HTPB)-based composite solid propellants are used in many operational propulsion systems due to better performance, repeatability of performance, ease of tailoring, and developed processing methodology. These propellants are reported to show incompressibility and Poisson's ratio is reported to be very near to 0.5 for most of the operating range of strain and temperature envelop. However, for homogenous class of NC/NG-based propellants, concept of incompressibility may be applicable but extrapolation of this concept to heterogeneous composite propellants are developed

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based on theory of physics and although binder constitute only 12-16 per cent part by volume of total propellant volume, mechanical properties are reported to be following those of the binder. It is also reported that measurement of Poisson's ratio in a single-shot experiment is difficult; however, invariably measurement of volume change is considered a valid method for determination and indication of Poisson's ratio. Poisson's ratio gained importance because of modern structural integrity analysis tool, where Poisson's ratio is an input parameter. In addition, variation of Poisson's ratio is also treated as one of the measurable controlling degradation parameters for propellants and can be used for prediction of the shelf-life¹. Since Poisson's ratio is an important parameter, which is difficult to determine directly, many indirect methods have been developed using extensometers in uni-axial tensile testing.

2. ASSESSMENT OF POISSON'S RATIO

Poisson's ratio is defined as negative of ratio of lateral strain and longitudinal strain. It is one of the significant mechanical properties parameters for solid rocket propellants. Initial attempts were made to understand propellants as polymeric masses exhibiting rubber like incompressible behaviour. Landel², *et al.* presented interpretation and assessment of Poisson's ratio for HTPB-based solid propellants. In general, filler particles adhere to binder matrix at low strains and material remains incompressible, exhibiting a Poisson's ratio of 0.5. At high deformation, vacuoles form around the filler particles and dewetting starts resulting in reduction of Poisson's ratio. For an isotropic incompressible material, Poisson's ratio depends on extension, but Poisson's ratio defined as logarithmic measure of strain is independent of lateral strain. The correlation with change in volume is given as Eqn (1).

$$\nu = -\frac{d \ln(\varepsilon_2)}{d \ln(\varepsilon_1)} = \frac{1}{2} \times \left[1 - \frac{d \ln(V/V_o)}{d \ln \varepsilon_1} \right]$$
(1)

where, v is the Poisson's ratio, ε_1 is the longitudinal strain, ε_2 is the lateral strain, V/V_0 is the ratio of instantaneous to original volume.

It is also reported that Poisson's ratio is 0.5 till volume of composite solid propellants is not changing, and after that, it attains a constant value independent of strain and temperature. In fact, rigid fillers in composite propellant formulations (oxidisers and metallic powder) hinder lateral contraction and reduce Poisson's ratio³. For single isotropic material, material properties in elastic region are characterised by Poisson's ratio⁴. Poisson's ratio is defined as dependent on longitudinal strain, solely as depicted by Eqn (2).

$$\nu = \frac{1}{\varepsilon_{l}} \times \left[1 - \sqrt{\frac{V/V_{o}}{1 + \varepsilon_{l}}} \right]$$
(2)

where, v is the Poisson's ratio, ε_1 is the longitudinal strain, ε_2 is the lateral strain, V/V_0 is the ratio of instantaneous to original volume.

It is also stated that reduction in Poisson's ratio due to stretching of specimen is higher for higher solid loading in a polymer. For 45 per cent solid loading, Poisson's ratio reduces to as low as 0.3 from initial value of 0.5. Poisson's ratio is attempted by putting strain gauges in longitudinal and transverse directions in a uniaxial tensile testing experiment and value of Poisson's ratio for composite propellant is reported⁵ in the range of 0.45-0.48.

Poisson's ratio is revisited for thermoviscoelastic solid propellant grains and using finite element simulations, effect of Poisson's ratio on maximum principal thermal strains and stresses are simulated⁶. When Poisson's ratio changes from 0.4999 to 0.47, maximum principal thermal strain changes by 68.7 per cent. Maximum principal thermal stresses and maximum shear thermal stresses changes by 73.5 per cent and 74.8 per cent, respectively for the same change in Poisson's ratio. Although Poisson's ratio is a very important factor, it is not ascertained properly and convincingly yet.

Poisson's ratio is correlated to velocity of

longitudinal and transverse ultrasonic waves in plasticbonded explosives and anomalies are expressed in the paper in reported and observed values⁷. In one of the recent papers⁸, Poisson's ratio is stated to be independent of binder concentration for plastic-bonded explosives, which resembles composite solid propellants in structure, crosslinking, and properties.

To ascertain Poisson's ratio correctly, tensile testing experiments were conducted for case-bonded solid propellants' specimens.

3. EXPERIMENTAL OBSERVATIONS

Double dumbbell specimens conforming to ASTM D638 type IV were prepared from HTPB-based case-bonded aluminised composite propellant formulations. The propellant composition contains 15 per cent binder (HTPB+DOA), 67 per cent oxidiser (Ammonium perchlorate) and 17 per cent (metallic fuel). After incorporation and mixing of all ingredients, obtained propellant slurry is cured using toluene diisocyanate (TDI) at 50 °C for 5 days. Specimen length and thickness were approximately 115 mm and 4 mm, respectively. Central gauge section had a width of 6 mm. Uni-axial tensile testing of 5 dumbbells was carried out at strain rate of 0.0185 per second with grip distance of 60 mm and gauge length of 45 mm. Five specimen were tested and since stress-strain curves are reproducible as well as superimposing, only three stress-strain curves have been depicted in Fig. 1. Initial modulus is obtained as 3.1 MPa and linearity continues upto around 13.5 per cent of strain. Lateral compressions were measured at break. For a typical propellant specimen, initial dimension of 3.73 mm became 3.41 mm at break, where other lateral side in gauge section reduced from 6.06 mm to 5.17 mm at break. Longitudinal extension at break was recorded as 47 per cent.

Since failure region in double dumbbell specimen as per ASTM 638 type IV had an approximate cross-section of 6 mm x 4 mm and different lateral compressions were recorded in both the directions, directional properties can



Figure 1. Stress-strain curve for propellant specimen as per ASTM 638 type IV.

be attributed to different initial lateral dimensions. To check the directional properties of propellant specimens, prismatic propellant specimens with square cross-section were prepared. Propellant sticks with square cross-section of 6 mm \times 6 mm and length 115 mm were taken and stretched at 0.0185 per second strain rate in constant strain rate of loading machine. Square cross-section ensures same lateral compression on both transverse directions and measurements can be homogenised. If Poisson's ratio using prismatic specimen gives the same values of lateral Poisson's ratios in both directions, directional properties can be attributed to initial dimensions.

As stick specimen had no shoulder region and uniform cross-section was subjected to strain during uni-axial tensile test, gauge length in this case was taken equal to grip distance. Five

specimens were tested and repeatability was ensured. Stressstrain curves for three specimens have been plotted in Fig. 2 for prismatic propellant specimens. Elastic modulus of propellant specimens tested using double dumbbell as well prismatic specimens were the same. Tensile strength and percentage elongation was also not varying much between dumbbell and prismatic specimen. For a typical prismatic specimen, initial dimensions of $6.22 \text{ mm} \times 5.81 \text{ mm}$ were reduced to $5.44 \text{ mm} \times 5.36 \text{ mm}$ for a longitudinal strain of 45.9 per cent at break. The directional variation of lateral compression persists even in prismatic specimen with square cross-section. So this directional variation cannot be attributed to different initial lateral dimensions and this aspect needs further investigation in respect of specimen orientation in propellant block.

4. ANALYSIS AND DISCUSSION

During linear region of stress-strain curve, propellant is assumed to be incompressible and its volume remains constant during extension. For ASTM specimen,

constant during extension. For ASTM specimen, propellant is incompressible till longitudinal strain reaches 13.5 per cent. Using relation given in Eqn (2), variation of Poisson's ratio is plotted against strain. It is observed that variation is linear as shown in Fig. 3. This variation is valid for strain up to dewetting only and beyond dewetting strain; Poisson's ratio at a given strain is lower than predicted in the Fig. 3. It should be noted that although linearity holds till 13.5 per cent strain but Poisson's ratio is not 0.5 for the entire region. Even if stress-strain curve in uni-axial tensile test is linear, Poisson's ratio does not remain constant. It reduces with increase in strain, linearly.

Extension measured at break for ASTM specimen indicates that for longitudinal strain of 47 per cent, lateral strains are 8.58 per cent



Figure 2. Stress-strain curve for propellant prismatic specimen.

and 14.68 per cent, respectively in two lateral directions. It is clear that propellant show directional properties and value of Poisson's ratio is dependent on direction, strongly. This behaviour cannot be observed by any of the uniaxial tensile test properties like tensile strength, elastic modulus or elongation. Poisson's ratio in lateral directions is calculated to be 0.1825 and 0.3125, respectively, using ratio of lateral strain to longitudinal strain.

As far as values of Poisson's ratio are considered, it is difficult to use these directional values for structural analysis. For true and unique value of Poisson's ratio for solid rocket propellant based on HTPB/AP/Al formulation, Eqn (2) is again exploited and change in volume of propellant specimen within grip distance is calculated. Initial volume of 1356.23 mm³ becomes 1554.94 mm³ at break. Using Eqn (2), value of Poisson's ratio is obtained as 0.2489 at break corresponding to longitudinal strain of 47 per cent.

Prismatic propellant specimen was also subjected to similar calculations. Longitudinal strain was reported to



Figure 3. Variation of Poisson's ratio for incompressible material.

be 45.9 per cent. Lateral strains in mutually perpendicular directions were calculated to be 0.125 and 0.0774. Corresponding Poisson's ratios were 0.2732 and 0.1687, respectively, based of basic definition of Poisson's ratio as ratio of lateral and longitudinal strains. Directional properties of Poison's ratio are again observed as are observed with ASTM specimens. Change in volume was calculated and using Eqn (2), value of Poisson's ratio was found to be 0.2371 at break. This value of Poisson's ratio matches well with calculated value of Poisson's ratio with ASTM double dumbbell.

Propellants are so called incompressible for a very small region during their stretching. As per Figs 1 and 2, linear portion is stretching to less than 30 per cent (only 0.135 out of maximum strain of 0.47) of total strain up to break. It is also clear that even in linear region, propellant's Poisson's ratio is not 0.5. Additionally in the failure region, propellant is always a compressible material and Poisson's ratio is much lower than 0.5.

Propellant is a highly filled elastomeric material with solid loading of the order of 85 per cent. This clearly

indicates that in a properly mixed homogeneous propellant, any cross-section has 85 per cent area filled by solid fillers, which will never deform on stretching. In the initial portion of extension, 15 per cent cross-linked polymeric part of a crosssection is stretched and lateral compression is observed. This lateral compression is purely due to polymeric part but is observed as a bulk property. When propellant specimen is stretched, propellant specimens behave as rigid solid fillers adhering to polymeric matrix getting rubbed against each other. This is equivalent to rubbing two emery papers against each other. To observe this effect, scanning electron microscope is used to study the fractured surface of the propellant (Fig. 4).

At break, significant propellant dewetting is expected and tensile strength is governed by strength of polymeric matrix alone. From SEM snap, it is clear that severe dewetting is observed around smaller filler particles, preferably of aluminum

powder, which are of 15-20 μ m size or fine ammonium perchlorate of size 10 μ m. Dewetting is less severe for coarse ammonium perchlorate of 200-300 μ m size. The emery paper concept is allowing smaller particles to leave binder matrix faster than bigger particles, resulting in excessive creation of vacuoles around them. Naturally, lateral compression is greatly obstructed by these particles and propellant no longer remains incompressible even in small deformation zones.

Since using Eqn (2) also a linear variation of Poisson's ratio is observed with increase in strain as shown in Fig. 3, a further linear variation with different slope is assumed for later part of stress-strain curve. It is clear that for compressible part of stress-strain curve, Poisson's ratio reduces with strain at a faster pace and slope of variation will be higher numerically. The variation is plotted in Fig. 5.



Figure 4. SEM of fractured surface of composite propellant.

Figure 5. Variation of Poisson's ratio with strain for composite propellants.

Slope of variation of Poisson's ratio with longitudinal strain doubles when propellant becomes compressible from incompressible. Overall propellant behaves as compressible material at or near break with Poisson's ratio approximately equal to 0.25.

5. CONCLUSIONS

Poisson's ratio of solid propellant is determined by direct measurement of lateral and longitudinal strains. It is observed that propellant is incompressible for small strain and even in this region Poisson's ratio steadily reduces from 0.5 to up to 0.45 for the tested propellant specimen with around 85 per cent solid loading in HTPB/ AP/Al system. ASTM and prismatic propellant specimen are tested and almost matching values of mechanical properties like tensile strength, elastic modulus, and percentage elongation is observed. However, Poisson's ratio at breaks showed different values in different directions. In one direction value of Poisson's ratio was 0.17, while in other direction, it reached around 0.3 for both types of specimens. It is also clear that considering compressibility, based on volume change of propellant specimen, for both types of specimens, value of Poisson's ratio at break is calculated to be around 0.24. Linear variation of Poisson's ratio is plotted against strain till break and variation is found to have two different slopes. In incompressible linear region, slope is -0.3165, while in compressible region, slope of variation of Poisson's ratio with strain is -0.6136. Poisson's ratio reduces twice as fast in compressible region as in incompressible zone. Value of Poisson's ratio for propellant is highly dependent on strain and in failure region, it reaches around 0.25.

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