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Performance of composite solid rocket propellants for rocket assisted projectiles (RAP)

Mohamed S. Nawwar [1,2], Tamer Z. Wafy[2] and Hossam E. Mustafa [3]

Abstract

Evidence suggests that rocket assisted projectiles (RAP) is among the most important factors to extend the range of the large caliber ammunition over standard gun systems. To date, there are few studies that have investigated the association between the mechanical properties of solid rocket propellant and the high acceleration forces which experienced by the rocket assisted projectiles (RAP) when it is launched. In the present paper, the effects of binder percentages and the stoichiometric ratio of isocyanate and hydroxyl groups (NCO/OH ratio) were used for investigating the mechanical and ballistic performance of composite solid rocket propellants (CSRP). Theoretical thermodynamic combustion properties of propellants formulations of the base binder hydroxyl terminated polybutadiene with solid loading above 60% have been calculated using the ICT-Thermodynamic Code v7.00, in order to assess the theoretical energetic performances of composite propellants as a function of their compositions.

Keywords: Rocket Assisted Projectiles, Composite Solid Rocket Propellant, and Hydroxyl Terminated Polybutadiene

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[1, 2, 3] Egyptian Armed Force, Cairo, Egypt

1. Introduction

Most studies in the field of solid propellants have focused on how to design the solid propellant rocket motor rather than how to extend the range of the large caliber ammunition gun system[1, 2] In recent years, a few authors have begun to design solid propellants for rocket assisted (both fin and spin-stabilized) projectiles (RAP) to extend the range of the large caliber ammunition over standard gun systems In his interesting analysis of the forces experienced by the spin-stabilized vehicles when it is launched from a gun system [3] identifies the high acceleration forces and the centrifugal forces. This makes it necessary to uses solid rocket propellants with a high tensile strength and must therefore have a high flexibility and ultimate tensile strain limit. It has been demonstrated that a crack in the solid propellant results in miss function of the motor and also cause explosion of the whole projectile[4].

The main objective of rocket assisted (both fin and spin-stabilized)) projectiles (RAP) designer is to provide the artillery projectile with a propellant grain that is consistent with thrust-time schedule required for range increasing and consequently the mechanical properties[5, 6].

Polyurethanes (PU) are a versatile class of polymer considered as a binding ingredient in solid propellants due to the possibility of tailoring properties according to the application [7, 8]. However the molecular architectures for the polyurethane (PU) backbone has a primary effect on motor reliability, mechanical properties, propellant processing complexity, storability, aging, and costs, several authors have explored the propellant grain design in order to achieve specific characteristics such as flexibility stability and ballistic performance [9].

These polymers are obtained by reacting a polyol with an isocyanate. The specific isocyanate and polyol used in the synthesis process determine the properties of the final product [10].

Isocyanates are characterized by the NCO chemical group and are related to hard segments on polyurethane polymer molecules. Polyols are OH containing groups and account for the soft segments of the polymer molecule. The performance of polyurethane based solid propellants depends on the type of isocyanate and on the NCO/OH molar ratio which called isocyanate index. The polyurethane molecule is composed of long, low-melting, flexible polyol joined to high-melting, rigid, concentrated urethane area. Increasing the NCO/OH ratio will increase the concentration of high- melting, rigid area of the chain, and thereby affects the physical properties of elastomer [11, 12].

It has conclusively been shown that mechanical characteristics of solid propellant mainly depend on the binder, the particle size and on the adhesion between particles and binder. They vary with temperature and stress rate or strain in such a way that time temperature equivalence has been determined for each type of binder[13, 14].

The mechanical properties are affected by binder variation including bonding agent, curing agent, oxidizer distribution, burning rate catalyst. All these parameters were varied to assure selection of the formulation with the highest possible strain capability and optimum processing characteristics. Also the viscosity is an important to the processibility and castability. It is affected not only by the binder, but also by size, content, shape and surface properties of solid fillers in propellant [10, 13, 15].

The bonding between the binder and the fillers, it is the structural properties of the binder and that of the bonding that govern the mechanical behavior of the propellant. The total solids content, their shape, and particle size distribution influence the propellant behavior by affecting the bonding properties[16, 17].

The mechanical characteristics of the solid propellants have a significant effect on the ballistic performance requirements that satisfy the mission objectives of the rocket motor. Static and dynamic loads and stresses are imposed on the propellant grains during manufacture, transportation, storage, and operation. In their introduction to solid propellant rocket fundamentals, George P. Sutton [18] identify the most common failure modes in solid propellants such as cracks, large areas of unbonding, air bubbles, porosity, or uneven density, an excessively high ambient grain temperature, excessive deformations of the grain and weakness of the adhesion between individual solid particles and the binder in the propellant. They demonstrated that the pervious failure modes may cause the vehicle to fly a different trajectory and this may cause the mission objective to be missed.

It has conclusively been shown that the burning rate of solid propellant is a function of propellant composition and the motor manufacturing conditions[4, 19].Up to now previous studies have highlighted factors that are associated with the content of propellant mixtures such as addition of catalyst materials or new burning rate enhancer, reduction of oxidizer particle size, increase of the percentage of oxidizer agents, increase of the amount of binder or oxidizer agent enhancing burning rate and addition of metal rods or metal fibers into the metallic fuel. Also the effects of motor manufacturing conditions are combustion chamber pressure, initial temperature of the propellant before the burning, temperature of burning gas, the speed of gas flowing parallel to the burning surface, the motor movement and effects of spinning on the burning rate [20, 21].

In spite of the relevance of range increasing of rocket assisted projectiles (RAP) to the performance of solid propellants, it has been hardly investigated for composite solid propellant systems. Furthermore, the correlation of mechanical characteristics with the ballistic performance is not encountered in the literature. This work aims to fulfil this gap by investigating the mechanical characteristics with the ballistic performance and ballistic behaviour of polyurethane-based composite solid propellants synthesized using HMDI as diisocyanate and with different NCO/OH molar ratios.

A simple uniaxial tensile test at constant strain rate was used to investigate failure criteria of polyurethane-based composite solid propellants. The solid propellants were further characterized by hardness and density, the linear burning rate, specific impulse and characteristic exhaust velocity of polyurethane-based composite solid propellants in order to give a detailed assessment of their ballistic performance

2. Experimental

Materials

All materials were used as purchased. Hydroxyl Terminated Polybutadiene (HTPB, number-average molecular weight of 2500, Brazil),Hexa-methylene-diisocyanate (HMDI, Fluka AG, Leverkusen, Germany),Tris(2-methyl-1-aziridinyl) phosphine oxide(MAPO, Orion ChemPvt Ltd), (AA, Morgan company, Cairo, Egypt), (TA, Morgan company, Cairo, Egypt), Di (2-ethylhexyl) Azelate (DOZ,China), Ammonium perchlorate (AP having particle size 400 µm , 200 µm and 7-11 µm, Abozabal, Egypt), Aluminium (Al having particle size 40 µm , and 8-22 µm particle diameter, Abozabal, Egypt) and Copper chromites ($Cu_2Cr_2O_5$, Morgan company, Cairo, Egypt) were used as purchased

Thermochemical calculations

The thermodynamic calculations were performed on polyurethane-based composite solid propellant formulations using ICT-Thermodynamic Code so that the effect of NCO/OH molar ratios and the binder content on condensed phase products, flame temperature, oxygen balance, and the specific impulse could be determined. The code computes chemical equilibrium by solving the non-linear equations derived from the mass action and mass balance expressions. The calculations were performed for isobaric adiabatic combustion at 7.0 MPa, assuming an adiabatic expansion through a nozzle in one-dimensional flow at chemical equilibrium and an expansion ratio of 70:1. All propellant calculations were compared with standard one containing 0.85 NCO/OH molar ratio propellant containing, 0.30 wt. % bonding agent, 14 wt. % binder, 69 wt. % oxidizer and 17 wt. %metallic fuel.

To study the effect of NCO/OH molar ratios in propellant containing 21wt.% binder, 0.45 wt.% bonding agent, 65 wt.% oxidizer and 13 wt.% metallic fuel., the weight percent of cross-linking (HMDI) and the weight percent of pre-polymer(HTPB) to achieve the polyurethane-based composite solid propellant formulations with NCO/OH molar ratios of 0.90, 0.95 and 1.00.To study the effect of binder content in 0.95 NCO/OH molar ratio propellant containing, 0.45 wt. % bonding agent, 19, 21, 23 wt. % binder.66, 65, 64 wt. % oxidizer and 14, 13, 12 wt. % metallic fuel respectively.

Composite propellants fabrication

All propellant formulations have been manufactured at AboZa3bal Factory, Egypt. Details of the formulations are shown in Table 1.For convenience, the polyurethane- based propellants are called F_X Formulations were prepared in a vertical kneader having a 3.7L volume and cured in an electrical oven cabinet . Ammonium perchlorate (AP) was dried at 65°C for 2 days. Initially, a weight of binder ingredients (HTPB-DOZ-MAPO-AA-TA) were premixed and stirred 150 rpm at 65 ºC using a heavy duty-mixing unit (200, 150, 100 rpm) for 15 min , followed by addition of metallic fuel (Al) and was mixed for 10 min. Then the oxidizer (three stages of addition (AP) every stage is 1/3 of the all oxidizer amount and every stage was added and the mixing was containing for 15 min. Then the slurry was left in a thermostat vacuum oven for 20 min. The curing agent were added, after the slurry was cooled to room temperature, and mechanically stirred for 15 min. The mixture was put into a vacuum oven to de-gas for a further 60min. After the prepared propellant formulations were cured at 65 °C for 84 h, they were removed immediately from the oven and left to cool at room temperature. The tensile testing samples were cast into a dumbbell mould, as shown in Figure 1. For density, samples were prepared of30 mmX30 mmX10 mm as shown in Figure 2

Table 1 the polyurethane- based Composite solid propellants

Ingredients		Formula	F0	F1	F2	F3	F4	F5	F6
Binder%	Pre-polymer	HTPB	10.45	13.89	15.39	16.89	15.44	15.39	15.34
	Cross-linking	HMDI	0.63	0.95	1.05	1.15	1	1.05	1.1
	Bonding agent (MAT4)	MAPO	0.225	0.34	0.3٤	0.34	0.3٤	0.3٤	0.3٤
		AA	0.056	0.08	0.08	0.08	0.08	0.08	0.08
		TA	0.019	0.03	0.03	0.03	0.03	0.03	0.03
	Plasticizer	DOZ	2.62	3.71	4.11	4.51	4.11	4.11	4.11
Oxidizer %	AP	400 µm	38	36.62	35.80	35.25	35.80	35.80	35.80
		200 µm	16	14.46	14.13	13.91	14.13	14.13	14.13
		7-11 µm	15	15.42	15.07	14.84	15.07	15.07	15.07
Metallic fuel%	Al	40 µm	11	9.38	8.41	7.78	7.78	8.41	8.41
		8-22 µm	6	5.12	4.59	4.22	4.22	4.59	4.59
Burning rate modifier%		Copper Chromites	--	1	1	1	1	1	1

Characterisation

The mechanical properties of the Composite propellants such as tensile strength, Young´s modulus, and elongation were investigated on a tensile test machine. Tensile tests of dumb bell shaped specimens were performed at 25°C and at a constant cross-head rate of 50 mm/min on an Instron electro-mechanical testing machine. The test load is 10 kN, the strain was measured with an extensometer. The tensile test was carried out for five specimens for each prepared formulation and then the mean value of the obtained results was recorded. The propellant formulations samples were prepared with the specific dimension.

Shore A was tested every day for every sample. Samples were taken out from the curing oven for 5 minutes, and then shore A was measured for all samples by using the hardness tester ZWICK (model 3102) by immersing the apparatus needle slowly in the sample and reading the numerical indication shown on the apparatus screen, and the mean value was taken. This study aimed to ensure the development of curing measured by the increase of the sample hardness value and when shore A is constant for successive three days, Curing is completed and samples were taken from the curing oven. [13, 22, 23]

The propellant density was measured at 20 °C using Archimedes rule fig (8) shows the samples of propellant used to measure density[24]. Rectangular aped samples of 30 mmX30 mmX10 mm, which were steeped in liquid paraffin at a temperature) 20 -2٥°C). The quality of the cured sample sheets was tested via X-ray unit to assess the inner homogeneity, cracks, air bubbles, porosity and foreign matter.[25]

The ballistic properties of prepared propellant formulations such as linear burning rate, specific impulse and characteristic exhaust velocity were measured by using standard two inches rocket motor.
The burning takes place using 7, 7.2, 7.5 mm nozzle which secure certain operating pressure and burning rate for each examined formulation. The samples casted in steel cylinders then casted samples loaded in the testing motor.

The combustion parameters were studied as follows: three values of burning rate were calculated per formulation, and then the linear burning rate values versus proposed operating pressure at normal temperature (20°C) were analysed to determine the ballistic parameters like pressure exponent (n) and burning rate constant (a) by using C language computer program. The pressure - time history was recorded for each test and the ballistic performance calculated [4, 26-28].

3. Results and Discussion

The effect of NCO/OH molar ratios

The mean score for the propellant density was 1.66 ± 0.04 Kg/m^3, investigated the differential effect of the molar ratio NCO/OH of polyurethane-based composite solid propellant formulations on their mechanical properties. In order to obtain solid rocket propellants with a high tensile strength and have a high tensile strain limit, it is preferable to use NCO/OH ratio to about 0.9:1 to 1:1 [10, 29]. In this major study, identify performance characteristics of CSRP for RAP projectiles. It has been observed that the Specific impulse, flame temperature and the Characteristic exhaust velocity (m / s) of CSRP for RAP projectiles are typically in the range of 210 -260 s, 2300 – 3200 K, and 400–650 °C respectively.The comparative analyses of the calculated theoretical performances and ballistic experimental test of composite propellant AP/Al/Binder/ burning rate modifier 65/13/21/1 with NCO/OH molar ratios of 0.90, 0.95 and 1.00are shown in Table 2 and Figure 3. It is observed that the ballistic performance does not affected by the changing of the NCO/OH molar ratios of 0.90 to 1.00.

Also these performance parameters are in the ranges which were identified. However all theoretical analyses are only approximations of what really occurs in the combustion chamber and nozzle flow, and they all require some simplifying assumptions[2, 5, 30].The ballistic performance values from ballistic experimental test of composite propellant are almost the same and constant as shown in Figure 3.

Table 2 The mean calculated theoretical performances and ballistic experimental test of composite propellant AP/Al/Binder/burning rate modifier65/13/21/1with NCO/OH molar ratios of 0.90, 0.95 and 1.00

Ballistic Performances	ICT-Code	Experimental
Specific impulse, (I_{sp}),[s]	248.8±0	240.8±0
Characteristic exhaust velocity (C*) [m/sec]	1469.1±0	1458±4

As expected, mechanical testing of the propellant prepared according to Figure 4(a) indicated an increase in tensile strength, tensile modulus and hardness and decrease in tensile elongation with the NCO/OH ratio. However the tensile strength of the elastomeric matrix increases with an increasing NCO/OH ratio up to 0.95 and then starts to decrease. This result may be explained by the fact that an increase in the NCO/OH ratio obviously leads to an increase in the crosslink density of the matrix. In Figure 4(b), there is no significant difference among the burning rate values. On average the burning rate values were shown to have 9.4±0.26 mm/sec.

The effect of binder content in 0.95 NCO/OH molar ratio propellants

Figure 5 shows Comparative analyses the theoretical performances and ballistic experimental test - The specific impulse (I_{SP}), The Characteristic Exhaust Velocity (C*) of the effect of binder content in 0.95 NCO/OH molar ratio propellants polyurethane-based composite solid propellant formulations AP/Al/Binder/ burning rate modifier with binder content of 19, 21 and 23% . Figure 4 clearly shows The Characteristic Exhaust Velocity (C*) and The specific impulse (I_{SP}), delivered in the motor are less than theoretical values by a significant amount The finding is consistent with findings of past studies by [9, 14, 31-33], which suggest that the reductions in values are the result of fluid flow losses including two-phase flow in which particles fail to achieve kinetic and thermal equilibrium, heat losses to motor hardware, and combustion inefficiency.

As shown in Figure 6 the specific impulse of 0.95 NCO/OH molar ratio polyurethane-based composite solid propellant formulations AP/Al/Binder/ burning rate modifier with binder content of 19, 21 and 23% are decreased as the mass fraction of the binder is increased and are the highest at 19%.The characteristic exhaust velocity is a function of the propellant combustion process; c* is proportional to $\sqrt{\frac{T_C}{M}}$, where T_C is the propellant flame temperature and M is the average molecular weight of the gas, and therefore has a slight dependence on chamber pressure. As well as the later formulation offer the advantages of high flame temperature and low molecular mass combustion products.

In Figure 7 (a) there is a clear trend of increasing tensile strength, tensile modulus and hardness and decreasing in tensile elongation with the increasing of weight fraction of solid fillers. This behaviour in the tensile elongation may be caused by different dewetting nature of large and small particles. Figure 7(b) provides the burning rate values of 0.95 NCO/OH molar ratios of composite propellant formulations. It is apparent from this Figure that high burning rate values as increase in solid content (decreases in binder content).

4. Conclusions

Composite solid propellant formulations with different NCO/OH molar ratio have been prepared and then characterized. The mechanical and ballistic properties, as determined by tensile measurements, increased with the NCO/OH ratio. Moreover, the NCO/OH molar ratio strongly not affected the burning behavior of the Composite solid propellant formulations. The mechanical measurements revealed that the Composite solid propellant formulations with a NCO/OH of 0.95have a high tensile strength with high tensile elongation at break, indicating that the NCO/OH ratio of 0.95 provided the best mechanical performance .The effect of solid loading in composite solid propellant formulations with constant NCO/OH molar ratio (0.95). The 78% solid loadings (21% binder) Composite solid propellant formulations grained micro structural features resulted in high strength and hardness. The results reveal that 78% solid loadings (21% binder) Composite solid propellant formulations with the NCO/OH ratio of 0.95 provided the best mechanical and ballistic results for RAP (CSRP) performance. Future recommendation is the aim of studying the particle size of the solid fillers and its distribution to achieves better results

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Figure 1 the tensile testing specimen of the polyurethane- based composite solid propellant

Figure 2 density specimen of the polyurethane- based composite solid propellant

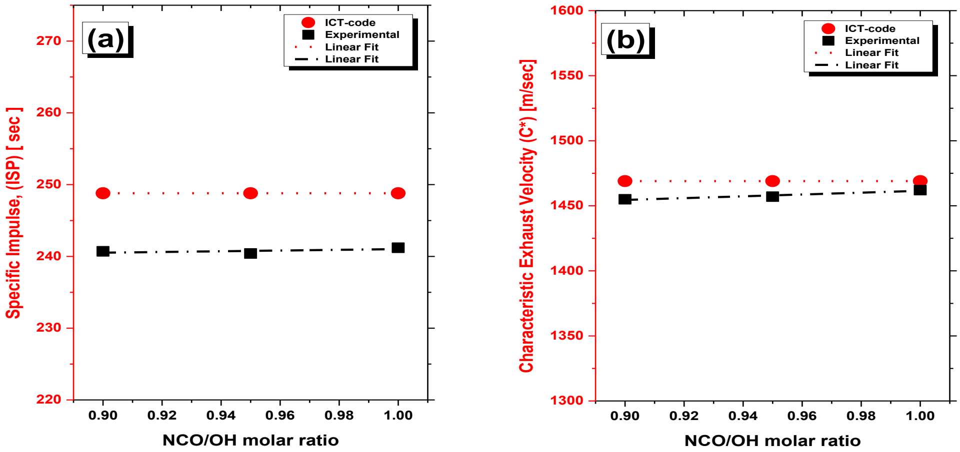

Figure 3 Comparative analyses of the calculated theoretical performances and ballistic experimental test of composite propellant AP/Al/Binder/ burning rate modifier 65/13/21/19 with NCO/OH molar ratios of 0.90, 0.95 and 1.00, (a) The specific impulse (I_{SP}), (b) The Characteristic Exhaust Velocity (C*)

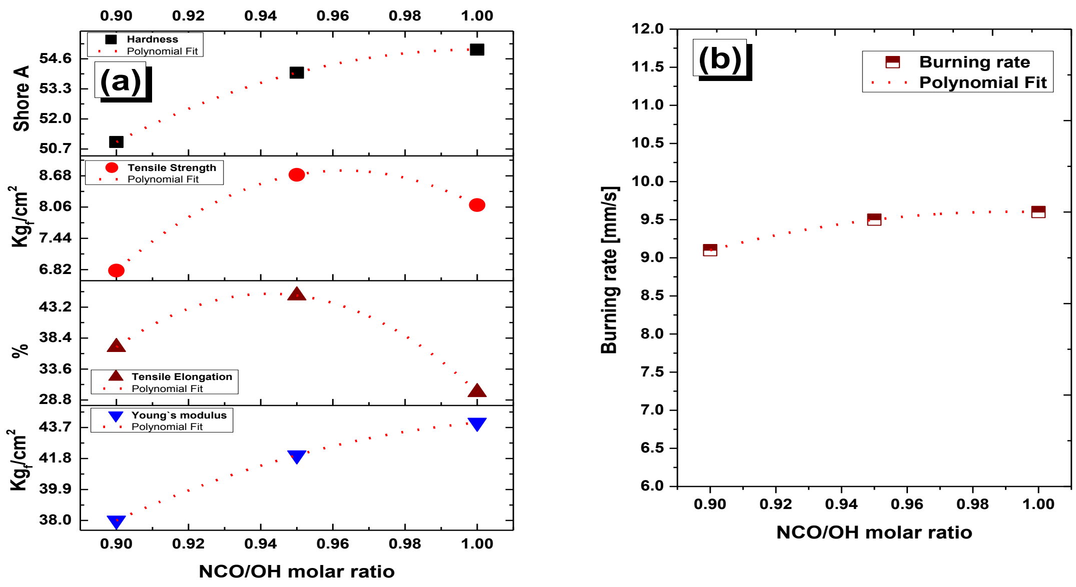

Figure 4 Effect NCO/OH molar ratio of composite propellant AP/Al/Binder/ burning rate modifier 65/13/21/1 with NCO/OH molar ratios of 0.90, 0.95 and 1.00 (a) on the mechanical properties (i) the hardness, (ii) the tensile strength, (iii) tensile elongation at break, and (v)the young`s modulus and on (b) the burning rate

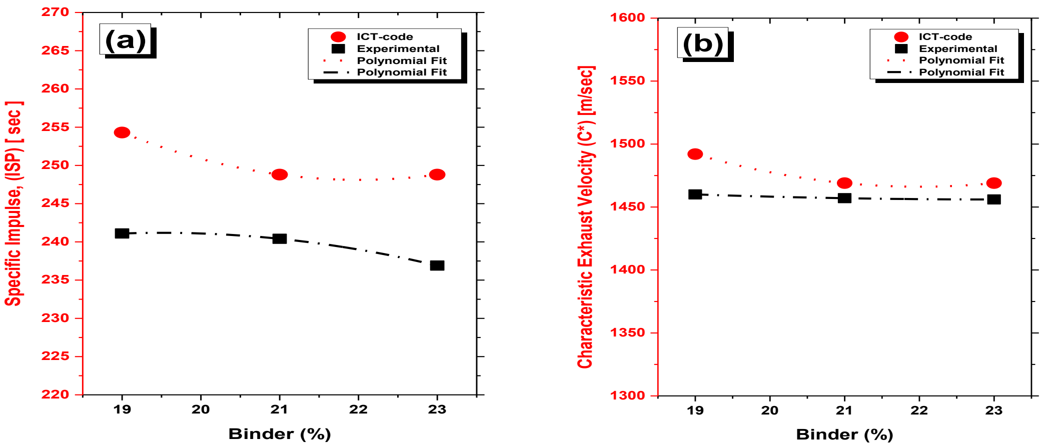

Figure 5 Comparative analyses of the Effect of binder content in 0.95 NCO/OH molar ratio propellants polyurethane-based composite solid propellant formulations AP/Al/Binder/ burning rate modifier with binder content of 19, 21 and 23% (theoretical performances and ballistic experimental test), (a) The specific impulse (I_{SP}), (b) The Characteristic Exhaust Velocity (C*)

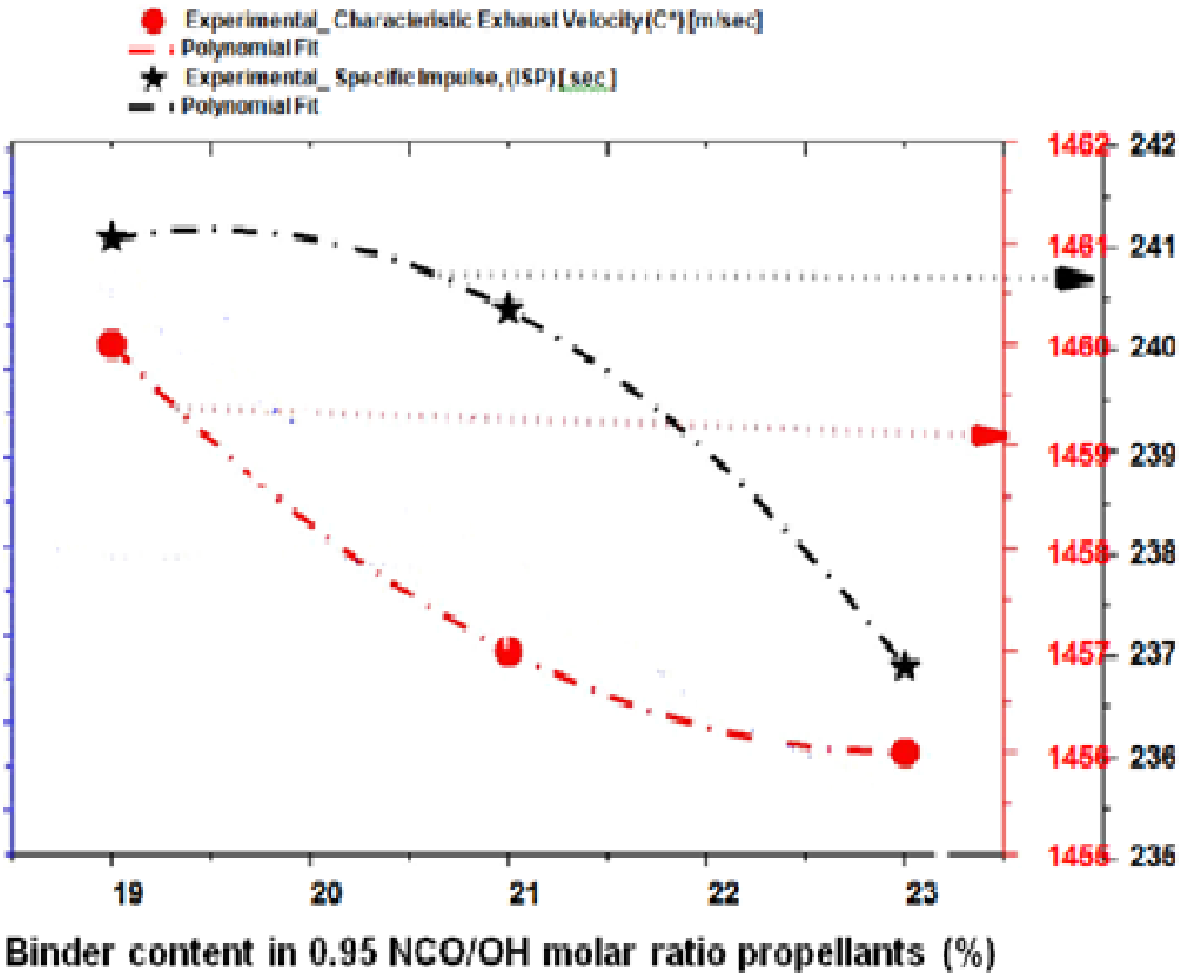

Figure 6 Experimental analysis the effect of binder content in 0.95 NCO/OH molar ratio propellant polyurethane-based composite solid propellant formulations AP/Al/Binder/ burning rate modifier with binder content of 19, 21 and 23% (The specific impulse, black solid star symbol, the characteristic exhaust velocity, the red circle symbol

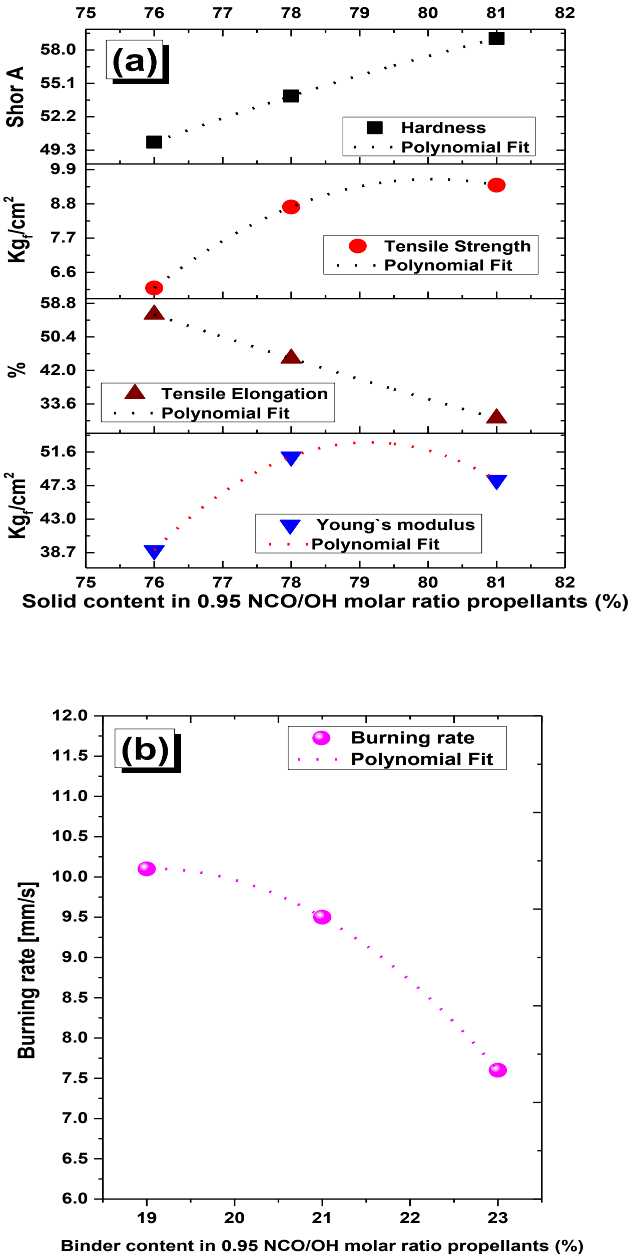

Figure 7 Effect Solid content in 0.95 NCO/OH molar ratio of composite propellant AP/Al/Binder/ burning rate modifier (a) on the mechanical properties (i) the hardness, (ii) the tensile strength, (iii) tensile elongation at break, and (v) the young`s modulus and on (b) the burning rate