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Study on Comparative Performance of CL-20/RDX-based CMDB Propellants

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Abstract: The energy, combustion and combustion residues properties of composite modified double-base (CMDB)propellants with CL-20 were compared to those propellants with RDX. The energy characteristic of CL-20/RDX-CMDB propellants had also been calculated theoretically based on the principle of minimum free energy. The energy property of propellants with CL-20 was found to be evidently enhanced in comparison to those propellants containing RDX. The findings on combustion properties revealed that the combustion properties of CL-20-CMDB propellants were contrary to the of RDX-CMDB propellants. With the mass fraction of CL-20 increasing in the propellants, the burning rates of propellants can be enhanced significantly, but the burning rates of propellants containing RDX decreased. Analysis of the combustion residues for CL-20/RDX-CMDB propellants revealed that the C, Cu and Pb elements aggregated on combustion surface, which may be useful for guiding the regulation of combustion performance of high-performance CMDB propellants containing CL-20.

Keywords: CL-20/RDX-CMDB propellant · energy · combustion properties

1 Introduction

Propellants is the obligatory energy delivering systems of weapons. The development of propellants technology is focused on the improvement of the range of missiles [1]. The conventional nitramines 1,3,5-trinitroperhydro-1,3,5-triazine (RDX) and octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (HMX) are the commonly used explosives. However, such insensitive explosives are much less energetic and therefore cannot be widely used in long-range weapons. Research in this direction brought into focus the concept of high energy dense materials (HEDMs) [2–4]. The materials have a prospect using as a primary component in the high-performance propellant for the booster rocket motor and the kinetic energy ammunition [5]. Hexanitro hexaazaisowurtzitane (HNIW or CL-20) is superior to hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) and cyclo-tetra-methylene-tetranitramine (HMX) with respect to density, enthalpy of formation, and oxygen balance [6–9]. CL-20 has evoked interest as a promising candidate, replacing various energetic compounds such as RDX, HMX and ammonium perchlorate (AP), for realizing the eco-friendly, high-performance, high-specific impulse(Isp), minimum signature propellants for futuristic missiles and space missions [10–14].

Ramesh Kurva et al. [15] reported that a successful attempt had been made to incorporate RDX, HMX and CL-20 at 10% level partly replacing ammonium perchlorate to enhance the performance of HTPB/AP/Al based composition having 86% solid loading and studied in detail for viscosity build up, mechanical, thermal and ballistic properties. In another study, the thermal decomposition behavior of CL-20-NEPE propellant and the interaction among the components had been studied by Li Ding et al. [16]. Joseph Kalman et al. [17] investigated the effect of CL-20 particle size on the combustion of CL-20/HTPB propellants down to submicrometer sizes, the burning rate and combustion mechanism were discussed in detail. A large number of papers have been concerned with the combustion and energetic properties of HTPB and NEPE propellants [18–24]. However, there are very few publications on the effect of CL-20 particles on the combustion and energetic properties of composite modified double-base propellants (CMDB) [25–27], especially on its combustion residues properties.

The objective of this paper was to compare the effects of CL-20 and RDX on the basic properties of CMDB propellants, such as the energy, combustion and combustion residues properties. Comprehensive information on the composition of combustion surface catalysts for CL-20-CMDB and RDX-CMDB propellants had been obtained, which may be able to guide the regulation of combustion performance of CMDB propellants. The main emphasis was placed on the formulation, design, energy and combustion characterization of high-performance CMDB propellants containing CL-20 particles, which may be used for booster rocket motor.

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2 Experimental Section

2.1 Materials and Sample Preparation

The sample (CL-20/RDX-CMDB propellant) used in the experiment was a composite modified double-base propellant, composed of nitrocellulose (NC) and nitroglycerine (NG), CL-20, combustion catalyst and other assistant reagent. The combination of NC/NG (NC/NG = 1:1 by mass percentage) offered a conventional energetic binder and plasticizer system for advanced propellants for achieving higher performance, superior structural integrity, and low vulnerability. RDX and CL-20 were from industrial products. The structural formulas and the characteristics of RDX and CL-20 were showed in Figure 1, 2 and in Table 1. RDX and CL-20 as the energetic additive oxidants, were added to the formulation of the solid rocket propellant. Six different propellant formulations with different content of RDX and CL-20 were prepared. Except where otherwise stated, all propellants were manufactured, processed, and tested at Xi'an Modern Chemistry Research Institute under identical conditions and using identical procedures. Table 2 listed the percentages, by mass content of the ingredients, used in six different propellant formulations. The propellant samples were prepared by solventless extrusion technology, which included mixing, rolling, extrusion, and cutting. The colloid of NC/NG and CL-20 or RDX were mixed with warm water (55 °C–60 °C) to form fine slurry. Then the catalysts and oth-

Table 2. CL-20 and RDX-CMDB propellant formulations.

No.	NC + NG binder + plasticizer/%	RDX/%	CL-20/%	Catalyst and other assistant reagent/%
C-1	77.52	–	17.65	4.83
C-2	66.15	–	–	3.85
C-3	55.12	–	41.67	3.21
R-1	77.52	17.65	–	4.83
R-2	66.15	30	–	3.85
R-3	55.12	41.67	–	3.21

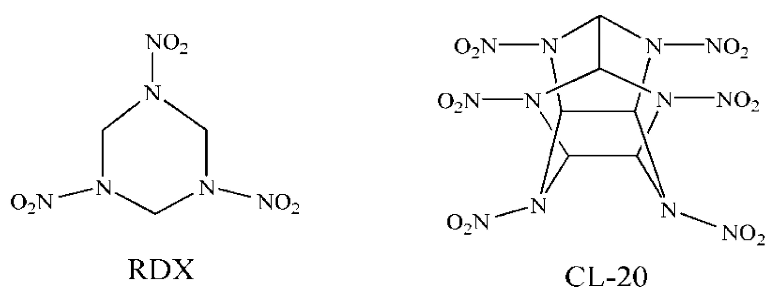

Figure 1. Structural formula of RDX and CL-20

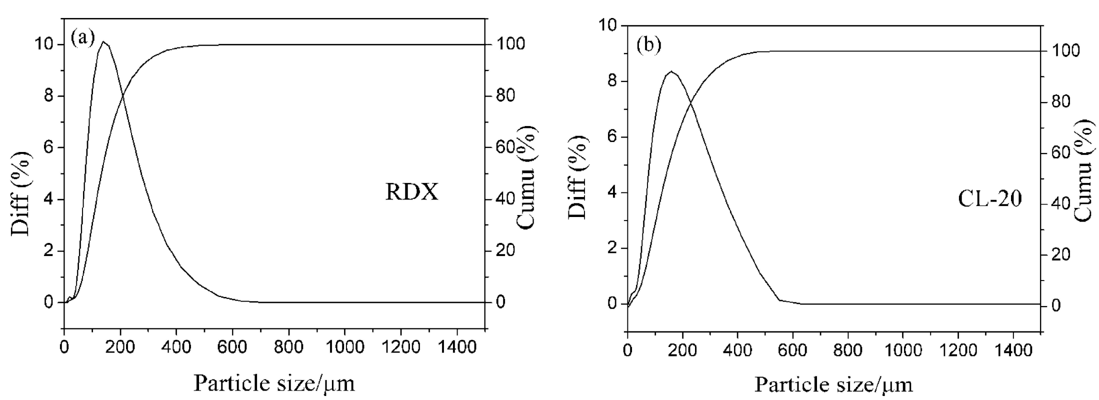

Figure 2. Grain size distribution of (a) RDX and (b) CL-20

Table 1. The characteristics of RDX and CL-20.

No.	formula	molecular mass	density/g cm^{-3}	OB	$\Delta_f H_m^\theta$/kJ mol^{-1}	D0.5/µm	Specific surface area/m^2 g^{-1}
CL-20	$C_6H_6N_{12}O_{12}$	222.12	2.04	−10.95 %	415.5	136.13	0.0332
RDX	$C_3H_6N_6O_6$	438.19	1.81	−21.61 %	61.55	132.292	0.0559

*OB is the oxygen balance, $_f H_m^\theta$ is the enthalpy of formation, and $D_{0.5}$ is the median size.

er assistant reagent were continuously added to the slurry. Thereafter the slurry was stored for at least one day at room temperature. The moisture content of the slurry will be reduced to around 10% by further centrifugal drying. Finally, the sheets of propellant were shaped in even-speed smooth rollers with temperature 95 °C and cut into 5 mm × 5 mm × 150 mm strands.

2.2 Apparatus and Measurements

The ideal properties of CL-20/RDX-CMDB propellant were calculated by means of "Energy Calculation Star (ECS)" software (5.0 edition), which was developed by Xi'an Modern Chemistry Research Institute, based on the principle of minimum free energy [28]. The operating conditions were as follows: (1) the combustion chamber pressure equaled 6.9 MPa; (2) environment pressure equaled 0.1 MPa; (3) nozzle expansion ratio was optimal, that exit pressure of nozzle equaled environment pressure; (4) nozzle exit spread angle equaled 0°; (5) the initial temperature was 298 K. The combustion products of the compositions got balance instantly in expanding process, which was equilibrium flow hypothesis.

The burning rate was measured by a chimney-type strand burner system. The propellant samples were cut to strands as 5 mm × 5 mm × 150 mm and coated by polyvinyl alcohol and aired for six times. A fine metal wire was threaded through the top of the samples with an alternating voltage of 100 V to ignite the propellant at the initial temperature of 20 °C in a chamber, which was filled with nitrogen atmosphere. In order to get the burning rate, two other low-melting-point fine fuse wires were threaded through the strand at a separate distance of 100 mm which recorded the start and end time signal of combustion. The real-time data were recorded by a computer which processed and calculated the burning rate. Five replicate experiments were conducted at each test pressure, and the average experimental results were obtained with a standard deviation of 0.13–0.25. The pressure exponent was obtained by means of Vieille's law ($u = aP^n$), where a is the pre-exponential factor.

Experiments were carried out in four observation windows type transparent combustion chamber to get the combustion residue. The propellant samples were cut to strands as 1.5 mm × 4 mm × 25 mm and fixed vertically on the combustion rack and sealed in the chamber, which was filled nitrogen. Pressure in the chamber was maintained at the desired levels to ignite the propellant strands. The combustion process was terminated to the surface of copper pillar due to the rapid heat conduction. Combustion residue was obtained representing the original appearance of the combustion surface. The component and relative quantity of elements in the residue on the combustion surface were analyzed from X-ray photoelectron spectrometer (PHI Quantera II). All samples were performed under the identical conditions.

The specific surface area refers to the particle size distribution. Particle size and size distribution of samples were performed through laser scattering (Malvern Mastersizer 2000) using a dry dispersion unit.

3 Results and Discussion

3.1 Energetic Properties of CL-20/RDX-CMDB Propellants

Energetic properties are the much important factors to evaluate the propellant performances. Propellant decomposes into high temperature working fluid in the combustion chamber, which is heat balance and chemical equilibrium process (energy conservation and mass conservation). According to the isenthalpic principle, the equilibrium composition, chamber temperature (Tc) and other thermodynamic functions in the combustion chamber can be further calculated. Through the expansion process of combustion products (isentropic expansion) in the nozzle, we can get the outlet temperature (Te), outlet species composition, etc. Standard theoretical specific impulse (Isp), equals to units of thrust per unit weight of propellant consumed per unit time, is the most important parameter to evaluate energy characteristics of propellants. Isp has great influence on the range of missiles, the higher Isp, representing the farther range of missiles.

In order to compare the effects of CL-20/RDX on the energetic properties of propellants, several propellant formulations were designed. The energetic characteristics of containing CL-20/RDX-CMDB propellants at 6.9 MPa were calculated and listed in Table 3. The comparative analysis of energetic characteristics was showed in Figure 3. It could be seen from Figure 3 that the values of Isp, C_V, Tc, Te, oxygen coefficient and relative molecular mass of gaseous products for CL-20-CMDB propellants were higher than for RDX-CMDB propellants at the same oxidizer mass content. It could be seen that with the mass fraction of oxidants increasing, the energetic characteristics values of CL-20/RDX-

Table 3. The energetic characteristics of CL-20/RDX-CMDB propellants.

No.	Isp/N s kg^{-1}	Cv/m s^{-1}	ϕ	Mc	Te/K	Tc/K
C-1	2413.35	1505.13	0.73	27.37	1523.68	3111.34
C-2	2463.43	1529.17	0.74	27.65	1610.53	3213.72
C-3	2503.41	1547.91	0.75	27.90	1687.76	3291.07
R-1	2393.94	1498.71	0.71	26.57	1443.57	3017.72
R-2	2431.77	1521.09	0.71	26.28	1469.75	3070.84
R-3	2461.47	1539.59	0.70	26.00	1486.29	3110.45

*Isp is ideal specific impulse, Cv is characteristic velocity, ϕ is oxygen coefficient, Mc is relative molecular mass of gaseous products, Te is outlet temperature, and Tc is adiabatic flame temperature.

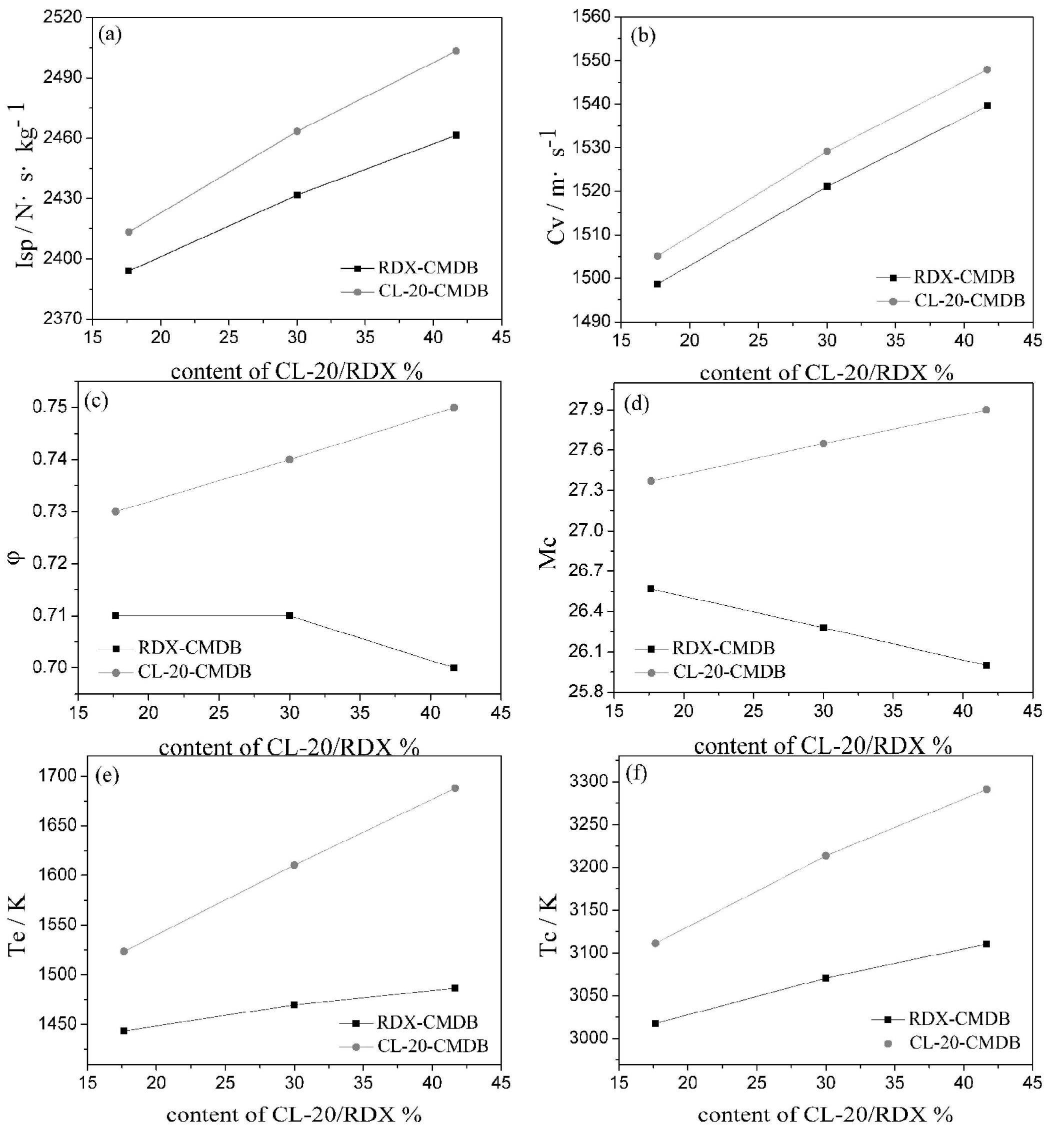

Figure 3. The energetic characteristics of CL-20/RDX-CMDB propellants

CMDB propellants were increased. The properties of CL-20-CMDB and RDX-CMDB propellants regarding density, enthalpy of formation, and oxygen balance of CL-20 were superior to RDX, which resulted in more complete combustion. Thus, increasing the amount of high density materials in the propellant leads to an increase of the energetic properties of the propellant.

3.2 Combustion Characteristics of CL-20/RDX-CMDB Propellants

3.2.1 Burning Rate and Pressure Exponent

Propellant burning rates determine the rate of gas generation, which determines the pressure inside the motor and the overall thrust. Burning rates herein are obtained experimentally by burning small propellant strands and measuring the surface regression versus time.

In order to compare the effect of mass fraction of RDX/CL-20 in the propellants on combustion properties, the strand burning rate experiments were conducted in the pressure range of 2–22 MPa. Figure 4 showed the burning rate curves versus pressures of CL-20/RDX-CMDB propellants. It could be seen from Figure 4 that with increasing the pressure, the burning rates of six samples increased from almost 4 mm/s to over 48 mm/s. The burning rates of CL-20-CMDB propellants were higher than of RDX-CMDB propellants at same mass content, it suggested that the combustion behavior of CL-20-CMDB propellants was superior to that of the RDX-CMDB system. From the energetic point of view, the heat release and heat feedback to the

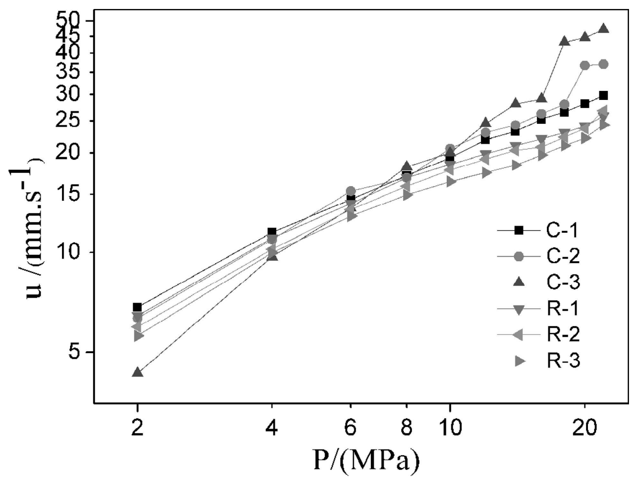

Figure 4. Burning rates of CL-20/RDX-CMDB propellants at different pressures

combustion surface for CL-20 particles are higher than those of RDX, and its promoting effect on the propellant combustion process is the main function. From a heat transfer point of view, the addition of CL-20 to the propellant formulation can increase the heat adsorption effectively during the combustion process. The six curves almost overlapped in the pressure range of 2–8 MPa, but there were different over the higher pressure range (10–22 MPa). The combustion properties of CL-20-CMDB propellants were contrary to them of RDX-CMDB propellants. With the increase of mass fraction of RDX in propellants, the burning rates over pressure range dropped less. However, with the increase of mass content of CL-20 in propellants, the burning rates over pressure range increased. Especially in the high pressure range (higher than 12 MPa) the increase of the burning rate (12–59%) is observably. Various factors affect the burning rate of propellants, such as the particle diameter, oxidizer species, pressure, and temperature etc. [29]. This might be related to the increase of burning rate of propellants with CL-20 in the high pressure range. The burning rate pressure exponents of propellants were calculated between 2 and 22 MPa and listed in Table 4. The burning rate pressure exponent of propellant with CL-20 in the whole pressure range was higher than that pressure exponent of propellant with RDX, it should be decreased in future experimental works.

Table 4. Burning rate pressure exponents of CL-20/RDX-CMDB propellants.

No.	Burning rate pressure exponents			
	2–10 MPa	10–16 MPa	16–22 MPa	2–22 MPa
C-1	0.642	0.553	0.511	0.597
C-2	0.722	0.499	1.232	0.702
C-3	0.967	0.817	1.429	0.967
R-1	0.658	0.380	0.488	0.546
R-2	0.682	0.347	0.763	0.577
R-3	0.671	0.384	0.640	0.561

3.2.2 Combustion Residue Analysis

Burning of propellant in a booster rocket motor may leave some residue [30–32]. In order to get better comparative analysis of the chemical components of the CL-20/RDX-CMDB propellant surface before and after combustion, we firstly calculated the percentage of C, O, Al, Cu and Pb elements in the formulations, and the results were shown in Table 5. X-ray photoelectron spectrometer (XPS) is an effective method to provide information regarding the component and relative quantity of elements in the sample. Sampling and analysis of combustion residue were made by XPS, the results were shown in Table 6. The comparative analysis of element content before and after combustion of propellants were shown in Figure 5.

Table 5. Mass percentage of element in the propellant formulation.

No.	C/%	O/%	Al/%	Cu/%	Pb/%
C-1	36.42	62.82	0.65	0.050	0.052
C-2	36.09	63.26	0.56	0.045	0.046
C-3	35.68	63.75	0.50	0.038	0.040
R-1	36.43	62.81	0.65	0.050	0.052
R-2	36.08	63.28	0.56	0.045	0.046
R-3	35.69	63.73	0.50	0.038	0.040

Table 6. Mass percentage of element in combustion residue.

No.	C/%	O/%	Al/%	Cu/%	Pb/%
C-1	51.14	33.59	12.59	2.14	0.55
C-2	53.95	32.57	11.16	1.90	0.54
C-3	56.28	30.35	11.08	1.75	0.42
R-1	46.11	37.55	13.40	2.23	0.71
R-2	44.35	39.01	13.65	2.43	0.56
R-3	36.04	45.18	15.91	2.45	0.43

Table 5 indicated that the content of C, O, Al, Cu, Pb elements was nearly same, when the content of CL-20/RDX in the propellant formulation was the same. The content of C element in the formulation dropped less, the content of O element increased slightly, and content of Al, Cu and Pb elements decreased with the increase of CL-20/RDX content. The results in Table 6 indicated that the change of content of C, O, Al, Cu, Pb elements in combustion residue for CL-20-CMDB propellant was almost opposite in comparison to those propellants containing RDX. In the CL-20-CMDB propellant formulation the mass percentage of C element in the combustion residue was increased, the mass percentage of O element was decreased, and the mass percentage of Al and Cu elements was reduced slightly with the in-

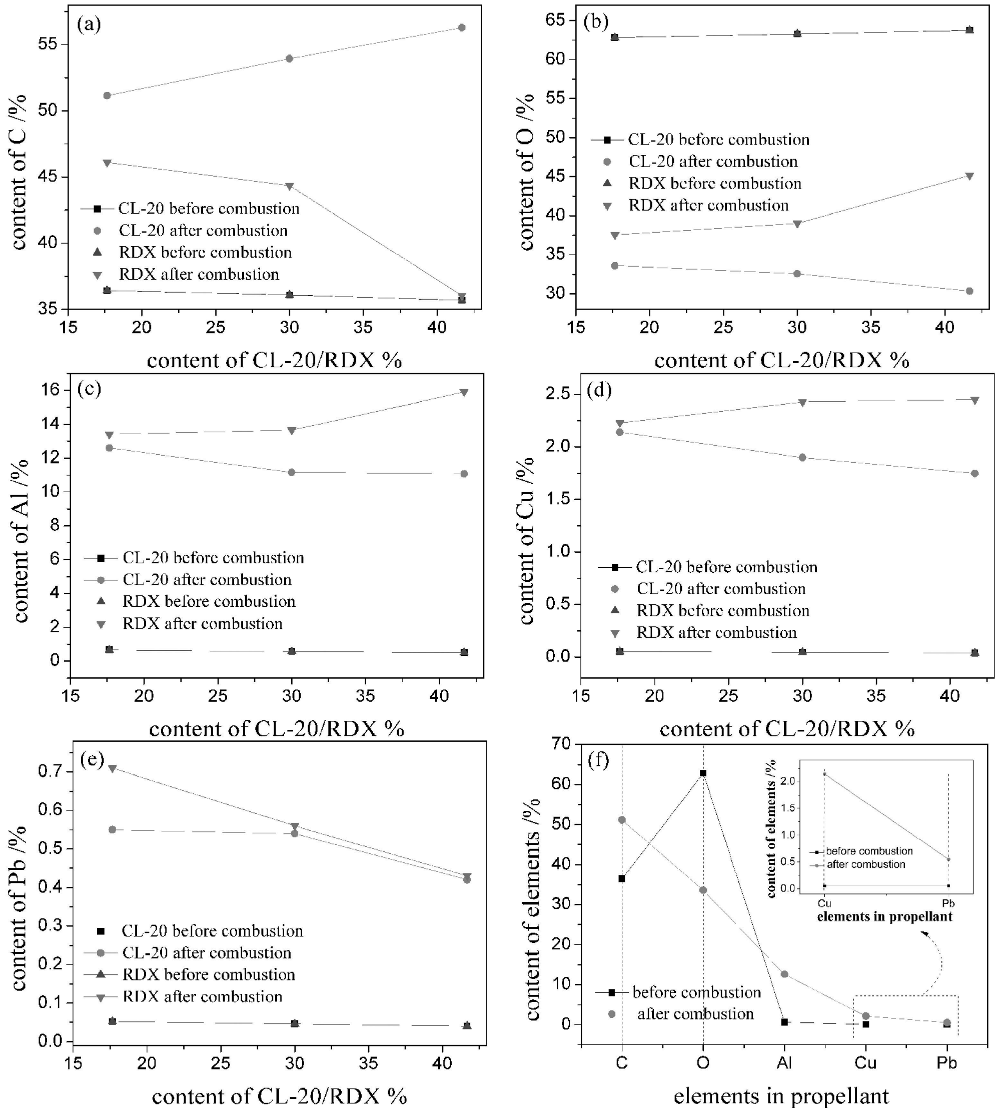

Figure 5. Element content before and after combustion of CL-20/RDX-CMDB propellant

crease of CL-20 content, and the mass percentage of Pb element was almost unchanged. Whereas in the RDX-CMDB propellant formulation the content of C was decreased, the content of O was increased, the content of Al was increased slightly, the content of Pb was reduced with the increase of RDX content, and the content of Cu was less changed.

According to the combustion theory, the burning rate of the propellant increases and combustion platform moves to the higher pressure zone with rising content of C element on the combustion surface. In the CL-20/RDX-CMDB propellant formulations the mass percentage of C element in the combustion residue was increased/decreased with the increase of CL-20/RDX content. This was consistent with the variation trend of burning rate. The role of oxygen is to oxidize carbon, so an increase of the carbon content led to a decrease of the oxygen content on the combustion surface. Al_2O_3 was added to the propellant formulation as internal ballistic stabilizer. The amount of Al element in combustion residue was more than 20 times that of the formulation, indicating that the enrichment of Al_2O_3 on the combustion surface. And it had a certain effect on the combustion performance. The most notable observation was that a large amount of Cu and Pb accumulated on the combustion surface compared to before combustion. Cu and Pb metal salts were added to the propellant formulation as burning catalyst and improved the combustion properties of propellant. According to the catalytic mechanism of condensed phase, the catalyst acts on the condensed phase of the combustion surface layer, and the exothermic decomposition reaction of condensed phase is accelerated, thus increases the surface heat results in the enhancement of the burning rate of propellants [33–34]. The amount of Cu and Pb elements in combustion residue was more than 42.2 ~ 64.5 and 10.5 ~ 13.7 times that of the formulation, and the catalytic effect was enhanced notable. Pb metal salt was used as main catalyst for improvement of combustion property. Cu metal salt was used as auxiliary catalyst for catalytic decomposition, which might be lead to broaden the combustion platform in the lower pressure range and increase burning rate little. In the CL-20-CMDB propellant formulation the mass percentage of Cu element was reduced slightly with the increase of CL-20 content. This might be one of the reasons for the higher burning rate pressure exponent of propellant with CL-20. Thus, decrease pressure exponent of propellant with CL-20 by increasing the mass percentage or changing kind of Cu metal salt is emphasis in the future work.

4 Conclusion

In this study, we compared energy and combustion properties of CMDB propellants with CL-20 and RDX, respectively. The effects of CL-20 and RDX on basic properties of CMDB propellants were investigated, the major findings could be summarized as follows:

1) The calculations on energetic properties for CL-20/RDX-CMDB propellants showed that energetic properties of propellant with CL-20 were superior to of propellant with RDX.
2) The data on burning rate revealed that with the increase of RDX/CL-20 mass fraction in the propellants, the burning rates at low pressure (2–8 MPa) remained almost the same but differ observably at high pressure (10–22 MPa). With the increase of CL-20 mass fraction in the propellants, the burning rates of propellants could be enhanced significantly, but the burning rates of propellants containing RDX decreased.
3) The results of residue components on combustion surface showed that considerable amount of Cu and Pb elements was aggregated on the combustion surface, which indicated the catalytic reactions of CL-20/RDX-CMDB propellants occured in condensed phase of combustion surface.

In this study we compared the effects of CL-20 and RDX on the energy, combustion and combustion residues properties of CMDB propellants, which may be useful for the regulation of combustion performance of high-performance CMDB propellants with CL-20.

Symbols and Abbreviations

CMDB	composite modified double-base
HEDMs	high energy dense materials
HNIW or CL-20	Hexanitro hexaazaisowurtzitane
RDX	hexahydro-1,3,5-trinitro-1,3,5-triazine
HMX	cyclo-tetra-methylene-tetranitramine
AP	ammonium perchlorate
Isp	specific impulse
HTPB	hydroxyl-terminated polybutadiene
NEPE	Nitrateester plasticized polyether propellant
NC	nitrocellulose
NG	nitroglycerine
OB	oxygen balance
XPS	X-ray photoelectron spectrometer

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