

BALLISTIC EFFECTIVENESS OF SUPERDENSE SOLID COMPOSITE PROPELLANTS WITH ZIRCONIUM OR ZIRCONIUM HYDRIDE

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1. ABSTARCT

The ballistic effectiveness of propellants depends not only on specific impulse value but also on many other performances, primarily on density. Nevertheless the density itself does not enter in the expression of the rocket velocity ($W = I_{sp} \cdot \ln(M_{launch}/M_{finish})$, where M_{launch} and M_{finish} – launch mass of the rocket and the mass after the propellant had been burnt accordingly) the density influences on the value M_{launch}/M_{finish} of the concrete rocket, thus if one charges the construction having fixed volume for propellant with more dense propellant, the ratio M_{launch}/M_{finish} rises and, consequently, W rises too. In this paper we are considering the ability to create solid composite propellants with zirconium (density 6.49 g/cm³) and zirconium hydride (density 5.61 g/cm³) as energetic compounds instead of aluminum. It was found for what kinds of engines these propellants have to be more effective than propellants basing on aluminum.

2. INTRODUCTION

Creating new kinds of propellant one has to consider in what kinds of engines these propellants would be used. Besides specific impulse (I_{sp}) the density is one of the most important factors defining ballistic effectiveness. The higher is the ratio M_{launch}/M_{finish} , the higher is the density input.

It is possible to fulfill the same engine (that is the same propellant volume) with a propellant having lower I_{sp} , but higher density; therefore it is possible to achieve the velocity growth for engines with the ratio M_{finish}/V_{prop} (V_{prop} = propellant volume) higher than the definite value. Replacing aluminum with zirconium or its hydride we can create new propellants with I_{sp} rather lower (at least 20 sec down), but with a density considerably higher (2.3 g/cm³ and higher) than formulations with aluminum commonly have (~1.8g /cm³).

3. PROBLEM STATEMENT AND RESULTS

If we replace Al with Zr or ZrH₂ I_{sp} value always falls, but in the same time the density growths considerably. In what cases the I_{sp} lost may be recompensed with density growth. It is widely known that the higher is the value Z ($Z = M_{launch}/M_{finish}$, where M_{finish} is the total mass with no propellant), the higher is the density input in achievement of the maximal rocket velocity. For multistage space vehicles – the higher is the stage, the lower is the density input.

In this investigation a comparative estimation of the ballistic effectiveness of the solid composite propellants (SCP) with Zr or ZrH₂ in comparison with SCP with Al have been carried out. Many different oxidizers and two different binders have been considered as compounds of SCP (Tabl.1). I_{sp} values have been calculated using the standard codes ASTRA and TERRA [1] at the pressure in combustion chamber 40 atm and 1 atm at the exit nozzle section. For the estimation of the mutual influence of the I_{sp} and density (d) on the ballistic effectiveness a new value I_{ef} (effective impulse) has been entered by the following manner: let one has a rocket with the definite ratio $F = V_{prop}/M_{finish}$. Being fulfilled with the propellant with $I_{sp0} = 251$ and $d_0 = 1.85$ (the basic formulation 20%Al+9% standard hydrocarbon binder + ammonium perchlorate, $Z_0 = M_{finish} + 1.85 \cdot F$) this engine would attain the value $W_0 = g_0 \cdot 251 \cdot \ln(Z_0)$. Being fulfilled with another propellant with I_{sp2} and density d_2 , the value Z changes ($Z_2 = 1 + d_2 \cdot (Z_0 - 1) / 1.85$), and the resulting $W_2 = g_0 \cdot I_{sp2} \cdot \ln(Z)$. Then we calculate what I_{sp} value has to have a formulation with the density d_0 for reach the velocity W_2 being used in this engine. Namely this I_{sp} value is so called I_{ef} .

Different Z_0 values have been considered. Below the Z_0 values under consideration are represented together with the corresponding F values. $F = (Z_0 - 1)/1.85$.

Z_0	1,5	2	2,5	3	3,5	4	5	6	8	10
F , l/kg	0,27	0,54	0,81	1,08	1,35	1,62	2,16	2,70	3,78	4,86

Table 1. Main properties of compounds.

Functional purpose	Component	$\Delta^\circ H_f$, kcal/kg	d , g/cm ³	$\alpha = O/[2C + 0.5(H - Cl)]$
Oxidizer	Hydroxylammonium perchlorate (HAP)	-493	2.07	3.33
	Ammonium Perchlorate (AP)	-593	1.95	2.7
	Ammonium dinitramine (ADN)	-270	1.82	2.0
	Hyrazinium perchlorate semihydrate (SHHP). $N_2H_5ClO_4 \cdot 1/2 H_2O$	-552	1.94	2.0
	Hydrazonium nitroformate (HNF)	-380	1.91	1.33
	HMX	77	1.92	0.67
Binder	Standard Hydrocarbon binder (SHCB) $C_{73.17}H_{120.9}$	-93	0.92	0
	Active binder (AB) 20% polyvinyltetrazol, plasticized with the mixture trinitroglycerole and diazapentane. $C_{18.96}H_{34.64}N_{19.16}O_{29.32}$, [2]	-181	1.49	0.53
Energetic compound	Al	0	2.7	0
	Zr	0	6.49	0
	ZrH ₂	-455	5.61	0

Formulations with all six oxidizers and two binders have been calculated. In each formulation there was 20 vol.% of the binder (either SHCB or AB), because if the binder volume percentage is less it is too hard to form a propellant with acceptable rheological and physico-mechanical properties. Aluminum content was varied from 16 up to 28 mass%, Zr and ZrH₂ – from 28 up to 55 mass%.

Fig. 1-5 represent calculated data of all formulations containing various combinations of oxidizers and binders at Z_0 values from 1.5 up to 3.5 (the most diffused values of real rocket engines for different purposes). There are values of lef growth (dlef) in comparison with the basic formulation (20% Al + 20 vol.% SHCB + AP, $I_{sp} = 250,9$; $d = 1.85$) on the y-coordinate while there are values of condensed phase amount in combustion products (Al₂O₃, ZrO₂) on the x-coordinate. Digits on the figures: 1) Al+SHCB + oxidizer; 2) Al+AB+oxidizer; 3) Zr+SHCB+oxidizer; 4) Zr+AB+oxidizer; 5) ZrH₂+SHCB+oxidizer; 6) ZrH₂+AB+oxidizer. The name of the oxidizer is written under each graph.

It is evident that the most of formulations with Zr or ZrH₂ are more effective than the similar formulation with Al, especially at low Z_0 values. First we are considering how this advantage depends on Z_0 if the oxidizer is the same.

Formulations with HAP. This oxidizer is the most rich with oxygen ($\alpha = 3.33$). If $Z_0 = 1.5$ there is no considerable difference using either SHCB or AB for both Zr and ZrH₂. dlef achieves 30-35 s only because replacing Al with Zr or ZrH₂. If $Z_0 = 2.0$ dlef falls to 20-25 s. If Z_0 increases further the advantage of ZrH₂ in comparison with Zr increases (I_{sp} becomes more important than the density). dlef achieves 10-12 s at $Z_0 = 2.5$ and 5-8 at $Z_0 = 3.0$. At $Z_0 = 3.0$ dlef is already a few sec only. In formulations Zr+SHCB +HAP dlef rises to higher levels at Zr or ZrH₂ content is about 50 mass % Zr (that is about 67% of condensed phase in combustion products)

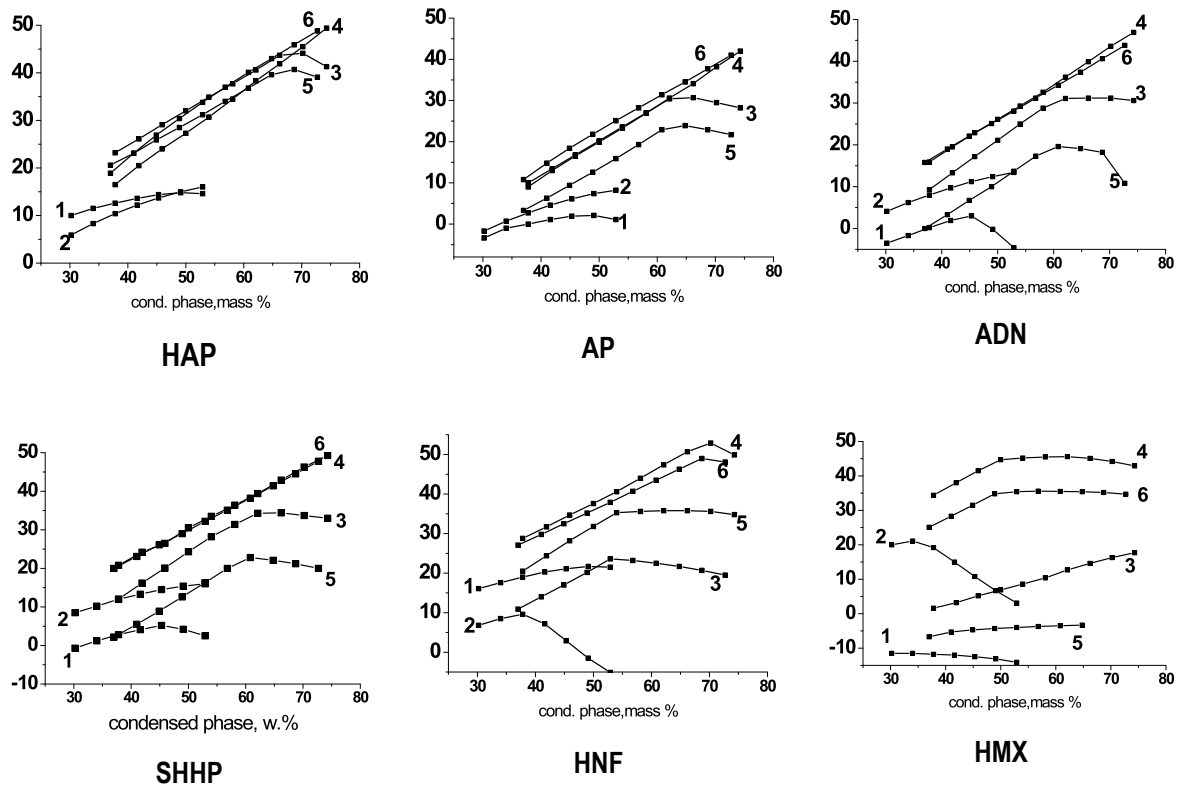


Fig.1. dlef values for $Z_0=1.5$ ($F=0.27$).

Formulations with AP have a bit lower dlef values (in comparison with formulations basing on HAP) if they contain Zr or ZrH_2 instead of Al. The reason is the oxygen deficit in AP in comparison with HAP. Anyway dlef values are rather high (25-30; 20; 13-15; 10 and 4-5 sec at $Z_0=1.5$; 2; 2.5; 3 and 3.5 correspondingly). In the system Zr+SHCB+AP the maximal lef values are achieved at Zr or ZrH_2 content $\sim 44-45\%$ Zr (that is $\sim 60\%$ of condensed phase in combustion products.)

Formulations with ADN. The advantage of AB over SHCB becomes more distinct. If AB is used there is no difference between dlef values of formulations with Zr and ZrH_2 while in formulations basing on HAP the advantage of ZrH_2 over Zr rises with the Z_0 growth. On the other hand in formulations with ADN the effectiveness of the aluminum replacement for Zr or ZrH_2 is almost the same than in formulations with HAP and AP (30; 20; 12-13; 7-8 and 4-5 sec at $Z_0=1.5$; 2; 2.5; 3 and 3.5 correspondingly). In the system Zr + SHCB+ ADN maximal lef values are achieved at Zr content 45-47% (that is 60-62% condensed phase in combustion products)

Formulations with SHHP. Here the lef gain is almost the same as with ADN. Only formulations with ZrH_2 together with SHCB are not enough effective. If $Z_0=1.5$ the lsp gain is 30 sec (Zr and AB), and 20 sec (Zr and SHCB); if $Z_0=2$ the winning is ~ 20 sec (Zr and AB) and 15 sec (Zr and SHCB); if $Z_0=2.5$ the winning is ~ 15 sec (Zr and AB) and 5 sec (Zr and SHCB); if $Z_0=3$ the winning is $\sim 7-8$ sec (Zr and AB), if $Z_0=3.5$ there is only a small winning for formulations with Zr and AB.

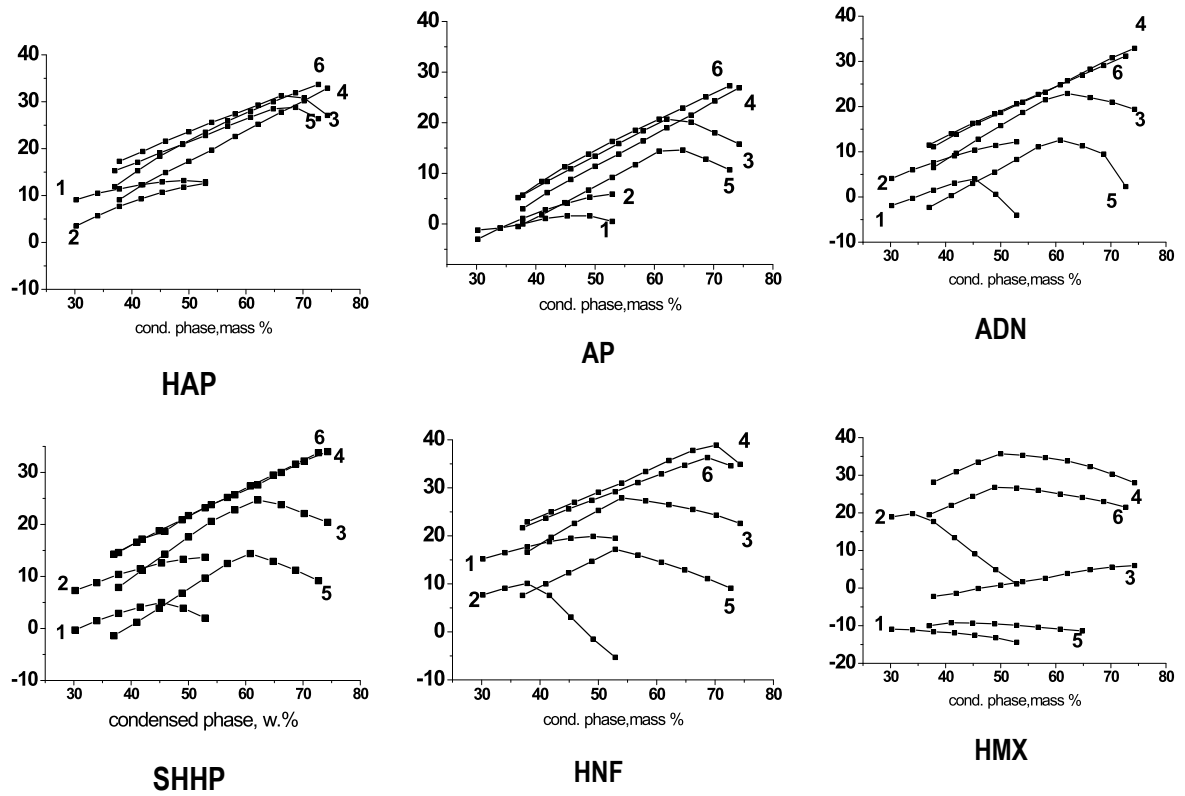


Fig.2. dlef values for $Z_0=2.0$ ($F=0.54$).

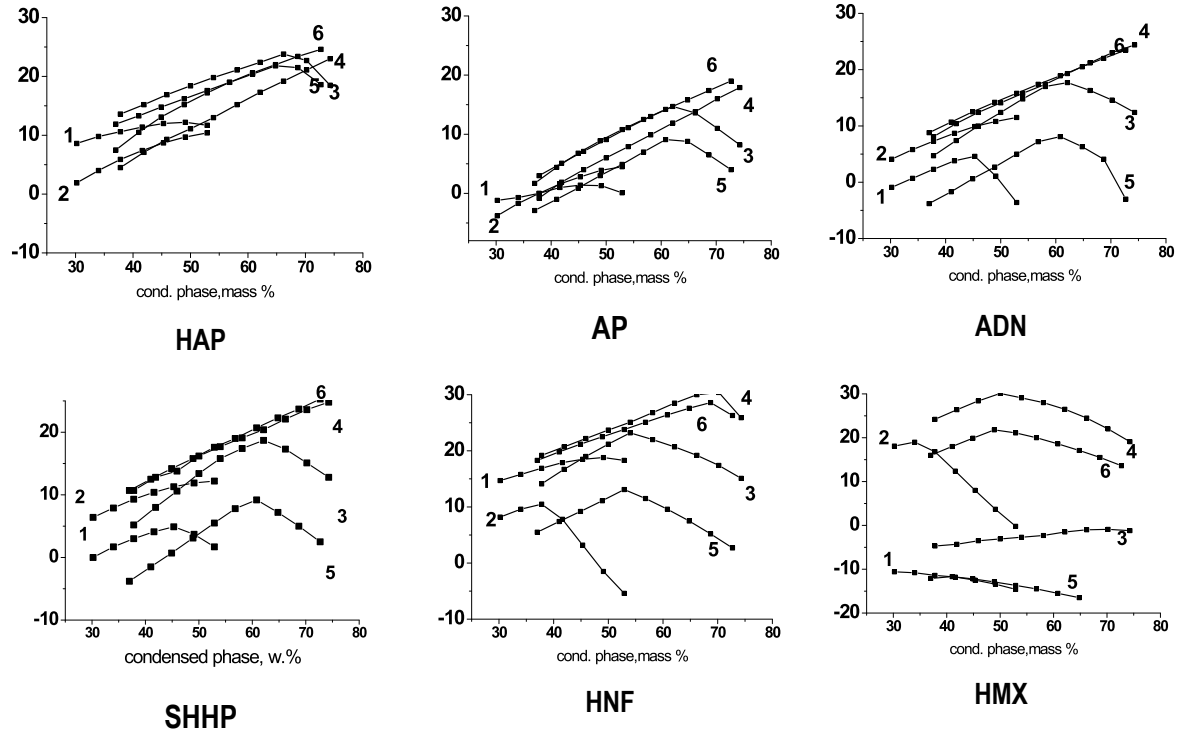


Fig.3. dlef values for $Z_0=2.5$ ($F=0.81$).

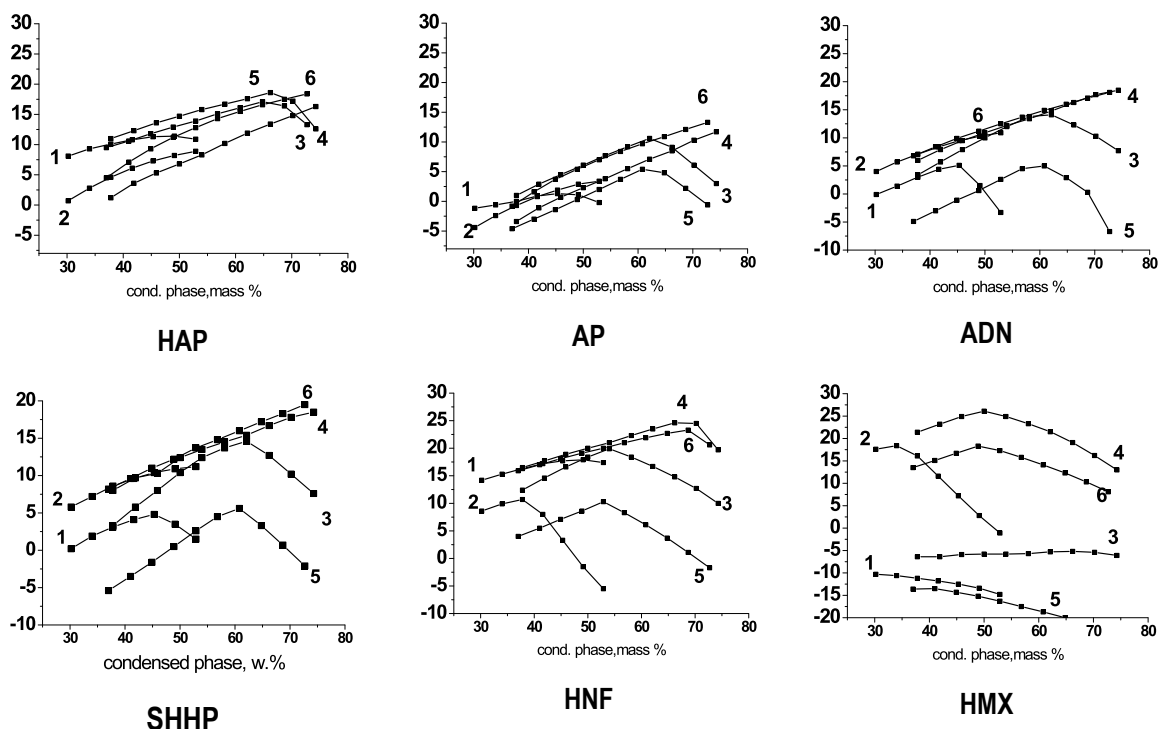


Fig.4. dlef values for $Z_0=3.0$ ($F=1.08$).

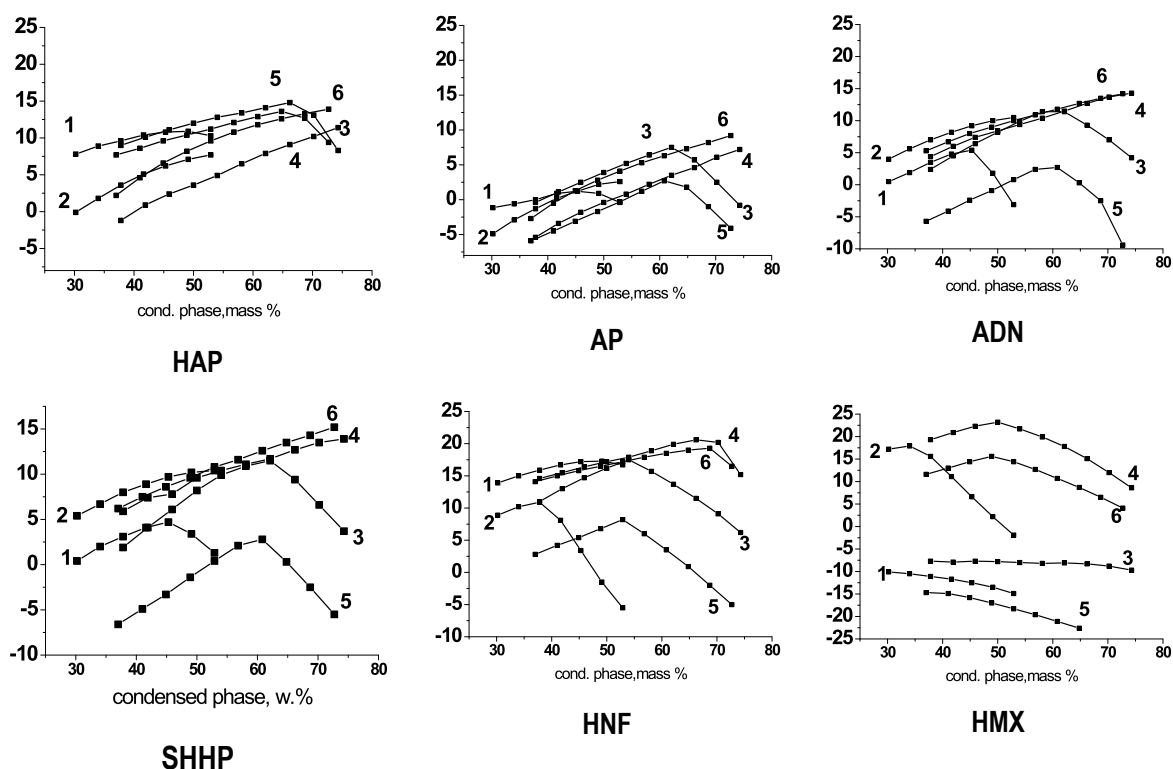


Fig.5. dlef values for $Z_0=3.5$ ($F=1.35$).

Formulations Zr+SHCB+SHHP achieve the maximal lef values at 45-47% Zr or ZrH_2 (that is 60-62% condensed phase in combustion products). In formulations with Zr and AB lef continues to increase with the Zr content growth up to 70% ZrO_2 . The same effect showed similar formulations basing on oxidizers HAP, AP, and ADN.

Formulations with HNF. If $Z_0=1.5$ the winning is 30 sec (Zr and AB), If $Z_0=2.0$ - ~20 sec, if $Z_0=2.5$ - 10-12 sec, if $Z_0=3.0$ - 5-8 sec. All formulations with AB are better than with SHCB.

Formulations with HMX. This oxidizer differs considerably from all other ones because HMX has the lowest value of α . Therefore formulations with SHCB are not interesting entirely- they are far worse than formulations with AB. Formulations with AB and Zr are considerably better than formulations with AB and ZrH_2 for all values of Z_0 . If $Z_0=1.5$ the lef gain is 15-20 (Zr and ZrH_2), if $Z_0=2.0$ formulations with AB and Zr win 15 sec while formulations with AB and ZrH_2 win 7 sec. If $Z_0=2.5$ the lef gain is 10 sec (Zr) and a few sec only for ZrH_2 . If $Z_0=3$ there is a winning 7 sec for formulations with Zr and AB and no winning for ZrH_2 . In the system Zr+SHCB+HMX the maximal lef values achieve at Zr or ZrH_2 content 38-41 % (that is 52-55% ZrO_2 in combustion products).

Comparing formulations with different oxidizers we are remarking a regularity – for oxidizers with rather high oxygen content ($\alpha \geq 2.0$: HAP, AP, ADN, SHHP) the lef increases with the growth of Zr or ZrH_2 content in formulations with AB, while in similar formulations basing on oxidizers with lower α (HNF or HMX) there is a maximum at Zr or ZrH_2 content, corresponding ZrO_2 percentage ~70 and 50% for HNF and HMX accordingly.

Therefore oxidizers with lower α values require replacing SHCB with AB and the winning due to using Zr-containing energetic compound instead of Al is higher if Zr is used, not ZrH_2 . All said above may be illustrated on Fig.6..

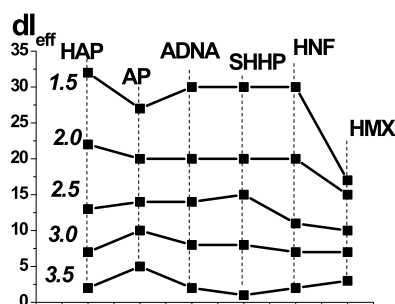


Fig.6. Maximal dlef values due to replacing Al for Zr or ZrH_2 in formulations basing on different oxidizers.

We have to notice additionally that formulations with Zr and AB show too high combustion temperatures (T_c). If ZrO_2 content in combustion products reaches 50% T_c is already higher than 3700-3800 K, if ZrO_2 content in combustion products reaches 65-70% (that is when lef is maximal) T_c values become 4000 K and even higher (Fig.7). The fact that formulations with Al show less T_c values than formulations with Zr (though the heat of formation of 1 g Al_2O_3 is about two times higher than the heat of formation of 1 g ZrO_2) may be explained simply. First, compositions with high content of condensed phase in combustion products (in Zr-containing compositions this value is about two times higher than in Al-containing compositions) show higher T_c values because the specific (per 1 g) heat capacity of condensed products are considerably less than the specific heat capacity of gases (particularly, H_2O and H_2). Besides, specific heat capacity of solid ZrO_2 is almost two times less than the heat capacity of solid Al_2O_3 . Second, at temperature ≥ 3600 K ZrO_2 dissociates in considerably less level than Al_2O_3 does, so for Al_2O_3 dissociation rather high amount of heat is spending, for example, for heating the system of the gross formula ZrO_2 from 3600 up to 4000 K it is needed the heat amount three times less than to heat Al_2O_3 from the same 3600 up to 4000 K.

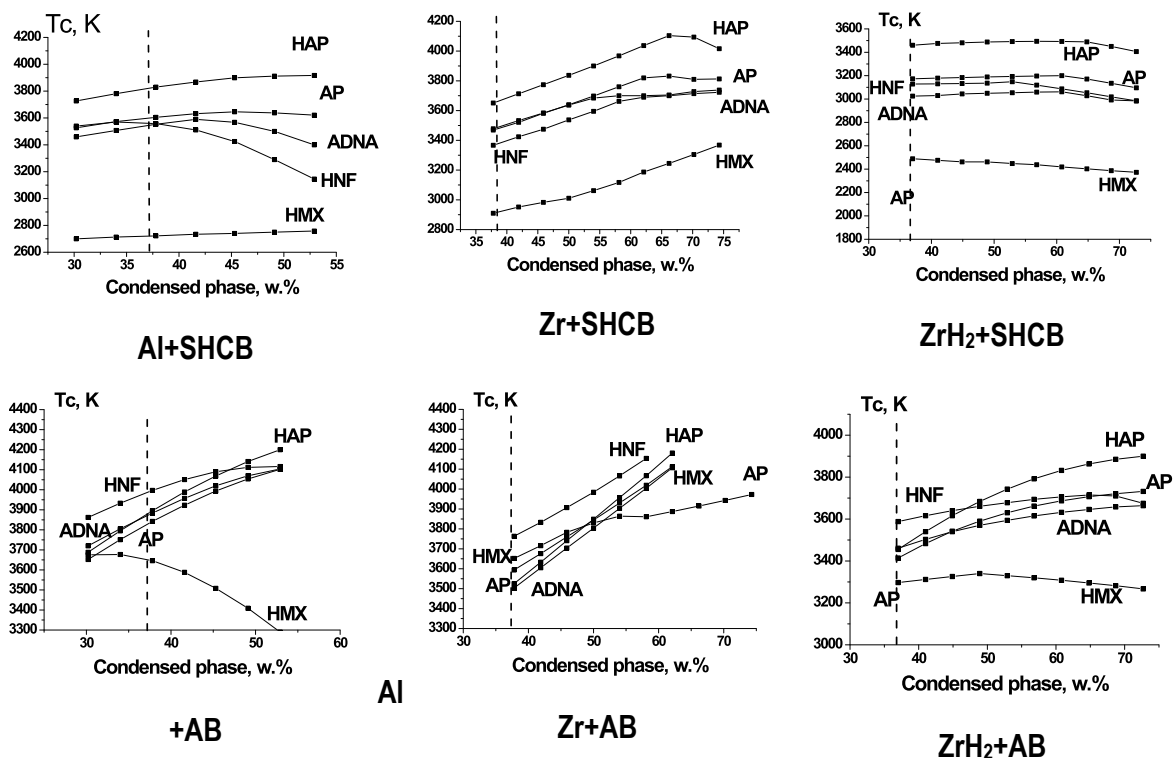


Fig.7. Tc values for formulations with different oxidizers, energetic components and binder (SHCB or AB, both 20 vol.%)

Unlike compositions with Zr and AB, formulations with ZrH₂ and AB have rather acceptable Tc values (not higher than 3800 K for all oxidizers except HAP) because these formulations contain higher hydrogen amount and a bit lower $\Delta^{\circ}H_f$. Formulations with Zr and SHCB have Tc values considerably less, than with Zr and AB, and these Tc values are already quite acceptable. Compositions with HMX, ZrH₂ and SHCB show too small Tc values.

Let us end a scientific analysis of main regularities while replacing Al for Zr and ZrH₂ and begin to compare ballistic properties of compositions with all oxidizers under consideration at different values Z_0 . So we are trying to find the best formulations for different engines with different Z_0 or F values. As the basic formulation we are still considering above-mentioned one: 20% Al + 20vol% SHCB + AP.

Below there are brief conclusions on relative effectiveness of formulations under investigation in comparison with the basic one at different Z_0 or F values.

$Z_0=1.5$; (F=0.27). Maximal winning show formulations with HAP, dlef = 40-47 s for Zr and ZrH₂, ZrH₂ is a bit better than Zr. Formulations with AB are a bit better than with SHCB (if we do not consider Tc values). The lower is the value α of the oxidizer, the higher is the difference between effectiveness of formulations basing on AB and SHCB, that is the lower is α , the worse are formulations with SHCB. Then the lower is α , the higher is the winning of Zr over ZrH₂. Formulations with Zr and AB may achieve dlef in comparison with the basic formulation about 40-45 sec. Using formulation with Zr and SHCB the winning depends much on oxidizer nature: 40-43 for HAP, 22-30 for AP, 25-31 for ADN, 28- for SHHP, 30-35 for HNF, and only for HMX the winning is at least ~ 15 sec.

$Z_0=2$; (F=0.54). All oxidizers in formulations with AB show the lef gain up to 35 sec. Formulations with SHCB continue to loose the effectiveness, particularly with ZrH₂.

$Z_0=2.5$; (F=0.81). The lef gain is up to 21-23 sec with HAP, 15-18 with AP, 30 with HNF, 23-24 with ADN, 24-25 with SHHP, 30 (Zr) and 20 (ZrH₂) with HMX.

$Z_0=3$; (F=1.08). In formulations with AB the lef gain is up to 30 with HNF, 22-23 with ADN, 15-20 with HAP, 25 with SHHP, 22 with AP, and 30 (Zr) or 22 (ZrH₂) with HMX.

$Z_0=3.5$; ($F=1.35$). The lef gain is up to 17-20 with ADN, 17-18 with HAP, 17-19 with SHHP, 12-13 with AP, 23-25 with HNF, 25 (AB+Zr) и 17 (AB+ ZrH₂) with HMX.

So, replacing Al with Zr or ZrH₂ only (if we are considering the best formulation in each kind of composition) with the best binder (for Al-containing as well as for Zr-containing formulations) permits to obtain lef gain 30 sec for $Z_0=1.5$; ~ 20 sec for $Z_0=2$; 13-14 for $Z_0=2.5$; 8 sec for $Z_0=3$ and ~5 sec for $Z_0=3.5$.

However, for further development of compositions containing Zr and ZrH₂ we have not to follow only lef values growth, we also must not increase Tc value very much because if Tc is too high (≥ 3800 K) one should weight considerably the nozzle section with heat shield. In Tables 2-4 we are representing a part of obtained data where the most acceptable formulations for further investigation are including. In these Tables only formulations basing on oxidizers AP, ADN and HMX are included, because these oxidizers are rather developed while other ones are not used in practice yet.

Table 2. The best formulations for engines with Z_0 values 1.5 and 2.0 ($F=0.27$ and 0.54)

Formulation	d	Tc	Condensed phase, %		Isp	$Z_0=1.5$	$Z_0=1.5$	$Z_0=2.0$	$Z_0=2.0$
			mass	vol		lef	dlef	lef	dlef
20% Al +AP+SHCB	1.847	3605	37	17.5	250.9	250.9	0.0		0.0
46%Zr+AP+SHCB	2.571	3820	62.1	27.9	215.9	281.2	30.4	271.6	20.7
43%Zr+AP+SHCB	2.494	3760	58.1	25.3	218.4	-	-	269.3	18.4
46%Zr+ADN+SHCB	2.451	3690	62.1	26.6	224.7	282.0	31.1	273.8	22.9
37%Zr+AP+AB	2.507	3804	50.0	21.9	212.0	270.8	19.9	262.3	11.4
34%Zr+ADN+AB	2.322	3760	45.9	18.6	227.6	273.8	22.9	267.6	16.7
37%Zr+ADN+AB	2.390	3844	50.0	20.9	225.1	277.0	26.1	269.6	18.7
34%Zr+HMX+AB	2.411	3783	45.9	19.3	236.0	292.4	41.5	284.4	33.5
37%Zr+HMX+AB	2.480	3830	50.0	21.6	233.3	295.6	44.7	286.6	35.7
46%ZrH ₂ +AP+AB	2.654	3687	60.8	28.2	211.4	282.2	31.4	271.6	20.7
49%ZrH ₂ +AP+AB	2.730	3707	64.8	30.9	209.2	285.4	34.5	273.8	22.9
46%ZrH ₂ +ADN+AB	2.544	3632	60.8	27.0	220.7	285.1	34.3	275.7	24.8
49%ZrH ₂ +ADN+AB	2.621	3646	64.8	29.6	218.0	288.3	37.4	277.9	27.0
37%ZrH ₂ + HMX+AB	2.423	3340	48.9	20.7	229.7	285.7	34.8	277.7	26.8

Table 3. The best formulations for engines with Z_0 values 2.5 and 3.0 ($F=0.81$ and 1.08)

Formulation	d	Tc	Condensed phase, %		Isp	$Z_0=2.5$	$Z_0=2.5$	$Z_0=3.0$	$Z_0=3.0$
			mass	vol.		lef	dlef	lef	dlef
20% Al +AP+SHCB	1.847	3605	37		250.9	250.9	0.0		0.0
46%Zr+AP+SHCB	2.571	3820	62.1	27.9	215.9	265.6	14.7	261.5	10.6
43%Zr+ADN+SHCB	2.375	3662	62.1	25.7	228.4	267.9	17	264.7	13.8
37%Zr+ADN+AB	2.390	3844	50.0	20.9	225.1	265	14.1	261.8	10.9
34%Zr+HMX+AB	2.411	3783	45.9	19.3	236.0	279.3	28.4		
37%Zr+HMX+AB	2.480	3830	50.0	21.6	233.3	281	30.1	276.9	26.0

49%ZrH ₂ +AP+AB	2.730	3707	64.8	30.9	209.2	265.1	14.2	261.9	11.0
46%ZrH ₂ +ADN+AB	2.544	3632	60.8	27.0	220.7	269.8	18.9	265.8	14.9
49%ZrH ₂ +ADN+AB	2.621	3646	64.8	29.6	218.0	271.4	20.5		
37%ZrH ₂ + HMX+AB	2.423	3340	48.9	20.7	229.7	272.7	21.8	269.2	18.3

Table 4. The best formulations for engines with Z_0 value 3.5 ($F = 1.35$)

Formulation	d	Tc	Condensed phase, .. %		Isp	$Z_0=3.5$ lef	$Z_0=3.5$ dlef
			mass	vol.			
20% Al +AP+SHCB	1.847	3605	37	17.5	250.9		0.0
43%Zr+ADN+SHCB	2.375	3662	62.1	25.7	228.4	262.3	11.4
37%Zr+ADN+AB	2.390	3844	50.0	20.9	225.1	259.3	8.4
37%Zr+HMX+AB	2.480	3830	50.0	21.6	233.3	274.1	23.2
49% ZrH ₂ +AP+AB	2.730	3707	64.8	30.9	209.2	261.9	11.0
46% ZrH ₂ +ADN+AB	2.544	3632	60.8	27.0	220.7	262.7	11.8
37% ZrH ₂ + HMX+AB	2.423	3340	48.9	20.7	229.7	266.5	15.6

Analysis of the results shows that there is a considerable reserve to rise ballistic effectiveness for engines with $Z_0=1.5-2.0$ (there are many missiles with such Z_0 values) only due to replacing Al with Zr or ZrH₂.

All said above have considered a comparative analysis of formulations (on one hand with Al, and with Zr or ZrH₂ on the other hand) in accord with achieved values lef and Tc only. However the replacement of Al for Zr or ZrH₂ may have other consequences, both positive and negative. Among possible negative consequences: Zr powder is rather pyrophoric, it may complicate the propellant producing and exploitation. ZrH₂ is less pyrophoric than Zr, and therefore ZrH₂ may be a good alternative to Zr, moreover it was shown above that in many formulations ZrH₂ may create compositions with higher ballistic effectiveness than individual Zr may.

Then we have to discuss the problem of the quantitative level of the Isp loss because of the presence of condensed phase in combustion products, so called biphasic loss. Usually compositions with 20% Al lose ~0.22% Isp per each Al percent. Sure this value depends on particles size (the smaller are solid particles, the less is the level of loss), on the heat capacity (the higher is specific heat capacity, the higher is Isp loss), on mass percentage of solid particles in combustion products (the higher is percentage, the higher is Isp loss). We see in Tables 2-4 that formulations with 46-49% Zr or ZrH₂ (that is with 60-65 mass% ZrO₂ in combustion products) show the most optimal ballistic properties. Common compositions containing 20% Al have 37% condensed Al₂O₃ in combustion products. As the specific heat capacity of ZrO₂ is more than two times less (0.49 against 1.05 J/(g·K)) than specific heat capacity of Al₂O₃, we may estimate to a first approximation that in Zr-containing compositions biphasic loss is a bit less than in Al-containing (other conditions being equal, that is if the particles size is the same).

There is another problem by replacing of Al with Zr or ZrH₂ – the possibility of slag-formation during the combustion process. Usually one does not increase Al percentage higher than 21-22% for not to achieve Al₂O₃ percentage in combustion products 38-40%, otherwise some slag forms during the combustion. Compositions with 46-49% Zr or ZrH₂ have higher mass percentage (1.7 times) of solid metal oxide in combustion products than compositions with 20% Al have, but because of higher ZrO₂

density the volume percentage of condensed metal oxides in combustion products are almost equal. As slag-formation is the consequence of partial coagulation of metal and its oxide at initial combustion phase in the combustion chamber, the probability to slag-formation in Zr-containing compositions may be less than in Al-containing ones because ZrO_2 melting point is by 700-800 K higher than Al_2O_3 melting point.

Similar study of formulations with titanium and its hydride (instead of aluminum) has been carried out. It was shown that unlike formulations with Zr and ZrH_2 formulations with Ti or TiH_2 are not so perspective because the densities of Ti (4.5) and TiH_2 (3.9) are rather less than ones of Zr and ZrH_2 and in formulations with Ti and TiH_2 (instead of Al) the growth of propellant density does not compensate the loss of Isp value.

4. CONCLUSIONS

Replacing aluminum with zirconium and its hydride in practically all solid composite propellants may increase missile velocity for engines with the ratio (propellant volume)/(construction mass) less than 1.0 – 1.4 l/kg.

Optimal ballistic effectiveness of propellants with zirconium or its hydride actualizes at 35-40% Zr or ZrH_2 in formulation, that is considerably higher than the optimal content of aluminum in Al-containing compositions.

Compositions with ZrH_2 are practically equivalent to compositions with individual Zr. ZrH_2 is better together with oxygen-rich oxidizers while Zr is better with oxidizers containing less oxygen.

For all oxidizers under investigation (except hydroxylammonium perchlorate) the most optimal formulations are those with Zr or ZrH_2 with an active binder.

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