Ammonium Nitrate as an Oxidizer in Solid Composite Propellants

George Manelis[®], David Lempert Institute of Problems of Chemical Physics, Russian Acad. of Sciences, acad.Semenov av. 1. Chernogolovka, 142432 Russia.

1. Introduction

Ammonium nitrate (AN) is the cheapest and most available oxidizer for solid composite propellants (SCP). It was AN that the early solid propellants were based on. AN combines a favorable element composition (5% hydrogen, 60% oxygen, absence of halides), high thermal stability, sufficient compatibility with a number of other constituents; however, a too low enthalpy of formation (-4514 kJ/kg) and insufficiently high density (1.725 g/cm³) result in AN loosing competition in energy to a majority of other known oxidizers.

Still, the attractive features of AN (environmentally benign combustion products, low cost, a very low sensitivity to mechanical loading, high thermal stability) can revive the interest to this oxidizer for applications with special requirements to environmental performance, cost, thermal stability, etc.

2. The objects and criteria for the comparison.

In this work, we compare the energetic potential of AN-based SCP against analogous formulations based on the better developed oxidizers: ammonium perchlorate (AP) and ammonium dinitramide (ADN). All thermodynamic calculations for the specific impulse (Isp) were performed for the pressure within combustion chamber (Pc) of 40 bar and that at the nozzle exit section (Pa), 1 bar. Since these three oxidizers have different density (AP – 1.95; ADN – 1.80-1.82; AN – 1.725 g/cm³), and density of propellant (ρ) substantially affects the ballistics, the formulations were compared not only by Isp but also by so called ballistic efficiency. In this paper we consider the Efficiency of i-th stage (Ei) defined as E1= Isp* $\rho^{0.6}$; E2= Isp* $\rho^{0.2}$. Naturally, the exponents for density depend on the volume-to-mass ratio for each stage and can differ substantially. Since AN has lower density than ADN and AP, its efficiency suffers worst for the first stage.

The choice of a binder is important to materialize the potential of oxidizers; four different binders were used in the calculations, namely:

- Hydrocarbon binder (HB), $\Delta H_f = -390 \text{ kJ/kg}$, $\rho = 0.91 \text{ g/cm}^3$, hydrogen content ~12%,
- Poly(methylvinyltetrazole) (PMVT) (C₄N₄H₆)_n, a high-enthalpy polymer with high content of nitrogen (~46%), ΔH_{f=}+1255 kJ/kg, ρ= 1.28 g/cm³
- Active binder (AB), a model binder analogous to practical ones, 20% poly(vinylmethyltetrazole), extended with a mixture of trinitroglycerol with 2,4-dinitro-2,4-diazo-pentane; its gross formula is close to C₁₉H_{34,5}N₁₉O_{29,5}; ΔH_{f=}-757 kJ/kg, ρ= 1,49 g/cm³, oxygen content ~47%
- Poly(vinylmethoxydiazen-N-oxide) (PVMDO) (C₃H₆N₂O₂)_n, ΔH_f=0 kJ/kg, ρ= 1.31 g/cm³, a model binder rich in hydrogen (~6%) and oxygen (~31%).

3. Energy potential of the formulations

Both formulations containing energetic components (aluminum, beryllium, or aluminum hydride) and simple pairs of oxidizer + binder were considered. The lower is formation enthalpy of an oxidizer, the more the oxidizer needs an additive energetic component¹. To save the space of this publication, here we present the calculated data for formulations with a constant content of energetic component, 20% Al, 13% Be, or 25% AlH₃ only. For these concentrations, the formulations are close to optima in energy, although it is known that, owing to two-phase loss, the higher stages require a somewhat lower content of metal. Tables 1-5 and Figs. 1 - 8 present major results of the calculations.

Table.1. Energy potential of metal-free formulations.

Oxidizer	bnd, wt.%	bnd, vol.%	Isp	ρ	Тс	Та	E1	E2	E3
AB + AN	25	27.8	219.3	1.660	2440	1297	297.2	268.6	242.7
	26	28.9	221.2	1.657	2476	1327	299.5	270.7	244.7
	27	30.0	223	1.655	2511	1358	301.7	272.8	246.6
HB + AN	10,5	18.2	209.8	1.577	2017	998	275.7	251.7	229.8
	11	19.0	208.5	1.570	1976	974	273.3	249.7	228.2
PMVT + AN	13	16.8	218.3	1.650	2348	1200	294.9	266.7	241.3
	14	18.0	217.1	1.645	2309	1172	292.7	264.9	239.8
	15	19.2	215.8	1.640	2268	1143	290.3	263.0	238.2
PVMDO + AN	14	17.7	221.5	1.652	2408	1276	299.3	270.7	244.9
	15	18.9	224.9	1.647	2460	1329	303.4	274.6	248.5
	16	20.1	226.3	1.642	2490	1338	304.7	275.9	249.9
AB + AP	25	30.4	214.8	1.810	2663	1457	306.7	272.4	241.9
	26	31.5	216.7	1.805	2697	1490	308.9	274.4	243.9
	27	32.6	218.5	1.800	2730	1525	310.9	276.4	245.8
	28	33.7	220.4	1.795	2762	1560	313.1	278.5	247.8
HB + AP	9,5	18.4	239.8	1.759	2989	1953	336.5	300.6	268.5
	10	19.2	240.6	1.750	3003	1910	336.6	301.0	269.1
	11	20.9	240.5	1.732	3007	1793	334.4	299.6	268.4
PMVT + AP	20	27.6	238.2	1.765	3004	1770	335.0	299.0	266.9
	21	28.8	234.7	1.757	2994	1709	329.1	294.0	262.7
	22	30.1	236.5	1.749	2975	1651	330.7	295.7	264.5
PVMDO + AP	22	29.6	239.7	1.761	3005	1927	336.6	300.6	268.4
	23	30.8	241.8	1.753	3025	1983	338.6	302.7	270.5
AB + ADN	26	30.0	239.3	1.721	2930	1714	331.4	297.3	266.7
	27	31.1	240.6	1.717	2950	1743	332.8	298.7	268.1
HB + ADN	10	18.2	250.9	1.655	3017	1660	339.4	306.9	277.5
	11	19.8	249	1.640	2967	1580	335.0	303.5	274.9
PMVT + ADN	14	17.5	250	1.596	3025	1894	331.0	301.4	274.5
	15	18.5	250.4	1.583	3038	1833	329.8	300.9	274.5
	16	19.6	250	1.569	3043	1774	327.6	299.4	273.6
PVMDO + ADN	20	25.8	254	1.689	3073	1945	347.8	313.2	282.1
	21	27.0	254.3	1.682	3082	1904	347.5	313.1	282.2
	22	28.2	254.3	1.676	3088	1865	346.7	312.7	282.0

Table.2. Energy potential of the formulations with 20%Al

Formulation	bnd, wt.%	bnd, vol.%	Isp	ρ	Tc	Та	E1	E2	E3
Al 20% + AB +	20	24.1	247.3	1.798	3498	2429	351.7	312.7	278.1
AN	21	25.3	247.5	1.795	3500	2426	351.6	312.8	278.2
	22	26.5	247.7	1.792	3503	2421	351.5	312.8	278.4
Al 20% + HB +	10	18.6	245.7	1.696	2965	1881	337.3	303.5	273.1
AN	11	20.3	245.1	1.681	2896	1804	334.7	301.7	271.9
Al 20% + PMVT +	10	14.0	245.2	1.792	3295	2300	348.0	309.6	275.5
AN	11	15.3	245.4	1.786	3271	2260	347.5	309.5	275.6
	12	16.7	245.5	1.779	3245	2216	346.9	309.1	275.5

Al 20% +	16	21.5	248.7	1.763	3334	2326	349.5	312.0	278.6
PVMDO+ AN	17	22.8	249.1	1.757	3322	2304	349.4	312.1	278.8
	18	24.1	249.4	1.752	3309	2282	349.1	312.1	279.0
Al 20% + AB + AP	24	30.8	245.5	1.915	3859	2901	362.5	318.3	279.6
	25	32.0	246	1.909	3860	2897	362.6	318.6	280.0
	26	33.2	246.5	1.903	3860	2893	362.6	318.9	280.4
Al 20% + HB + AP	8	16.5	249.2	1.882	3716	2681	364.2	321.0	282.8
	9	18.4	250.1	1.862	3670	2604	363.2	320.7	283.2
Al 20% + PMVT +	10	15.3	250.8	1.956	3810	2840	375.1	328.0	286.8
AP	11	16.7	251.1	1.946	3800	2814	374.4	327.7	286.9
	12	18.1	251.2	1.936	3787	2784	373.4	327.2	286.7
Al 20% +	20	28.6	251.7	1.871	3762	2721	366.6	323.4	285.3
PVMDO+ AP	21	29.9	252.3	1.862	3748	2694	366.4	323.6	285.7
	22	31.1	252.8	1.854	3733	2666	366.1	323.6	286.0
Al 20% + AB +	15	18.9	255.3	1.880	3884	2888	372.9	328.6	289.7
ADN	16	20.1	255.6	1.876	3884	2885	372.8	328.7	289.9
	17	21.4	255.9	1.872	3884	2881	372.7	328.8	290.1
	18	22.6	256.1	1.867	3884	2877	372.5	328.8	290.2
	19	23.8	256.4	1.863	3884	2873	372.4	328.9	290.4
	20	25.0	256.6	1.859	3884	2867	372.2	328.8	290.5
	21	26.1	256.9	1.855	3884	2861	372.2	328.9	290.7
Al 20% + HB +	8	15.8	262	1.793	3695	2507	372.0	331.0	294.5
ADN	9	17.6	262.1	1.776	3642	2420	369.9	329.8	294.0
	10	19.3	262	1.759	3584	2331	367.6	328.4	293.3
Al 20% + PMVT +	12	17.3	259.7	1.847	3785	2640	375.3	331.9	293.6
ADN	13	18.7	259.9	1.839	3763	2596	374.6	331.6	293.6
	14	20.0	260	1.831	3738	2550	373.8	331.2	293.4
Al 20% + PVMDO	13	18.3	260.2	1.847	3763	2596	376.0	332.6	294.2
+ ADN	14	19.7	260.7	1.840	3738	2550	375.8	332.7	294.5
	15	21.0	261.1	1.832	3710	2503	375.5	332.7	294.7
	16	22.3	261.5	1.825	3679	2455	375.2	332.7	294.9
	17	23.6	261.9	1.818	3645	2405	374.9	332.7	295.2
	18	24.9	262.2	1.811	3608	2358	374.5	332.5	295.3

Table 3. Energy potential of the formulations with 13%Be

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Formulation	bnd, wt.%	bnd, vol.%	Isp	ρ	Тс	Та	E1	E2	E3
Be 13%+ AB + AN	23	25.9	262.2	1.679	3469	2789	357.8	322.6	290.8
	24	27.0	262.5	1.676	3471	2788	357.9	322.8	291.1
	25	28.1	262.8	1.674	3473	2787	358.0	322.9	291.3
	26	29.2	263.1	1.671	3475	2785	358.0	323.1	291.6
Be 13%+ AB + AP	25	30.2	254.8	1.799	3893	2961	362.4	322.2	286.5
	26	31.3	255.3	1.793	3891	2954	362.5	322.5	286.9
	27	26.8	255.8	1.788	3889	2949	362.1	321.4	285.3

Be 13%+AB+	15	17.8	265.7	1.765	3899	2949	373.6	333.5	297.7
ADN	16	18.9	265.9	1.761	3898	2945	373.4	333.5	297.8
	17	20.1	266.2	1.758	3897	2941	373.4	333.6	298.0
	18	21.2	266.5	1.754	3896	2937	373.3	333.6	298.2
	19	22.3	266.7	1.750	3895	2933	373.1	333.6	298.3
	20	23.4	267	1.746	3893	2927	373.1	333.7	298.5
	21	24.6	267.2	1.743	3891	2921	372.9	333.7	298.6
	22	25.7	267.4	1.739	3889	2916	372.7	333.6	298.7
Be 13% + HB +	10	17.5	270.5	1.596	3038	2260	358.1	326.1	297.0
AN	11	19.1	271.2	1.583	2990	2190	357.2	325.9	297.3
	12	20.7	271.8	1.570	2941	2120	356.3	325.5	297.5
Be 13%+ HB + AP	12	22.5	266.2	1.704	3554	2787	366.6	329.5	296.2
	13	24.1	267.8	1.687	3500	2727	366.6	330.1	297.3
	14	25.7	269.3	1.671	3441	2661	366.4	330.7	298.4
	15	27.3	270.4	1.655	3374	2581	365.8	330.7	299.1
	16	28.8	270.7	1.639	3290	2477	364.1	329.8	298.8
Be 13% + HB +	14	24.6	269.3	1.599	3295	2426	357.0	325.0	295.8
ADN	15	26.1	270.4	1.586	3153	2344	356.5	325.1	296.5
Be 13%+ PMVT +	17	21.8	268.4	1.642	3145	2378	361.5	327.3	296.4
AN	18	23.0	268.9	1.637	3124	2345	361.4	327.5	296.8
	19	24.2	269.1	1.632	3102	2311	361.0	327.3	296.8
	20	25.4	269.7	1.626	3080	2276	361.1	327.6	297.2
	21	26.6	270	1.621	3057	2240	360.8	327.5	297.4
	22	27.8	270.3	1.616	3034	2205	360.5	327.5	297.5
Be 13%+ PMVT +	20	27.4	263	1.754	3630	2850	368.5	329.3	294.3
AP	21	28.6	264.3	1.746	3598	2827	369.2	330.3	295.5
	22	29.9	264.7	1.738	3561	2791	368.8	330.2	295.6
	23	31.1	265.3	1.730	3518	2747	368.6	330.3	296.0
Be 13%+ PMVT +	19	25.1	276	1.688	3603	2812	377.9	340.3	306.5
ADN	20	26.3	276.4	1.682	3559	2772	377.6	340.3	306.7
	21	27.5	276.1	1.675	3489	2666	376.3	339.4	306.1
Be 13%+ PVMDO	21	26.1	270.1	1.631	3269	2531	362.2	328.5	297.9
+ AN	22	27.3	270.7	1.626	3259	2514	362.4	328.8	298.3
	23	28.5	271.2	1.621	3249	2497	362.4	329.0	298.7
Be 13%+ PVMDO	20	26.9	261.1	1.765	3786	2851	367.2	327.7	292.5
+ AP	21	28.2	261.9	1.757	3773	2851	367.3	328.2	293.2
	22	29.4	262.7	1.750	3760	2851	367.5	328.6	293.8
Be 13%+ PVMDO	19	25.7	274.1	1.773	3765	2851	386.5	344.7	307.4
+ ADN	20	26.9	274.6	1.765	3751	2850	386.2	344.7	307.6
	21	28.2	275.2	1.757	3736	2850	386.0	344.8	308.0
	22	29.4	275.8	1.750	3721	2850	385.8	345.0	308.4
	23	30.6	276.4	1.742	3706	2850	385.6	345.1	308.9







Figure 2: Ballistic efficiency of compositions HB + oxidizer.



Figure 3: Ballistic efficiency of compositions PMVT + oxidizer



Figure 4: Ballistic efficiency of compositions PVMDO + oxidizer.



Figure 5: Ballistic efficiency of compositions AB + oxidizer + 20%Al



Figure 6: Ballistic efficiency of compositions HB + oxidizer + 20%Al.



Figure 7: Ballistic efficiency of compositions PMVT + oxidizer + 20%Al.



Figure 8: Ballistic efficiency of compositions PVMDO + oxidizer + 20%Al.



Figure 9: Ballistic efficiency of compositions HB + oxidizer + 13% Be.







Figure 10: Ballistic efficiency of compositions AB + oxidizer + 13% Be.





The calculations were performed for widely varied content of binders. Tables 1-3 show the formulations distinguished by maximum Ei under constraint of volume fraction of a binder exceeding 18-19% (for lower content of binders, a formulation with satisfactory mechanical properties cannot be made).

Figures 1-12 show that for the active binder and PVMDO, having relatively high oxygen content, the optima in Isp and Ei are achieved for higher volume content of binders, compared to the hydrocarbon binder and PMVT. Table 4 summarizes the maximum attainable Ei, provided the volume fraction of a binder is not less than 19%.

		bi	nder					binder			
oxidizer	HB	AB	PMVT	PVMDO	oxidizer	HB	AB	PMVT	PVMDO		
	Meta	al-free – I	E1			20% Al – E2					
AN	272	300	293	303	AN	303	312,5	308	312		
AP	335	310	338	335	AP	321	315,5	326	321		
ADN	335	335	331	345	ADN	328	327	331	332		
	Meta	al-free – I	E2				20% A	Al – <i>E3</i>			
AN	250	275	262	275	AN	272	278	275	278		
AP	300	275	299	300	AP	283	275	286	280		
ADN	305	298	301	311	ADN	293	291	293	294		
	Meta	al-free – I	E3		13% Be – <i>E2</i>						
AN	224	248	238	250	AN	326	322	327	329		
AP	264	245	268	270	AP	331	321	330	328		
ADN	278	268	274	283	ADN	323	333	340	344		
	209	% Al – E	1				13% B	e – <i>E3</i>			
AN	337	354	345	350	AN	297,5	291	297	295		
AP	363	362	372	366	AP	299	287	296	298		
ADN	368	372,5	374	375	ADN	296,5	299	307	308		

Table.4: Maximum attainable *Ei* for different compositions.

An accurate analysis of above results shows that in metal-free compositions AN is inferior to AP and ADN for all stages. Introduction of aluminum, and even more so beryllium, substantially decreases the loss. Important is that the higher is the stage, the better AN performs compared to AP and ADN, for 3rd stage AN is no worse than AP and but a little worse than ADN.

Use of aluminum hydride in formulations with the active binder (the other binders are substantially less efficient with the hydride) results in enhancement of E2 and E3 over those for formulations based on aluminum (with any binder), these values become close to the Be-containing formulations. Table 5 shows the calculated data. The content of a binder in the formulations with aluminum hydride must be substantially higher compared to the metal-based formulations, since due to the small size of the hydride particles having high surface area, the formulations with the binder content less than 28-33% do not have satisfactory physico-mechanical and rheological properties.

Table.5. Energy potential of the formulations with anaminan hydrade (25%) and the active binder											
Formulation	bnd, wt.%	bnd, vol.%	Isp	ρ	Tc	Та	E1	E2	E3		
$AIH_3 25\% + AB + AN$	25	26.8	267.4	1.596	3208	2125	354.0	322.4	293.6		
	27	28.8	267.6	1.591	3206	2117	353.6	322.3	293.7		
	29	30.9	267.9	1.587	3204	2109	353.4	322.2	293.8		
$AIH_3 25\% + AB + ADN$	25	27.4	277.6	1.636	3564	2345	372.9	338.0	306.3		
	27	29.5	277.4	1.629	3548	2327	371.8	337.2	305.8		
	28	30.6	277.4	1.626	3540	2327	371.3	336.9	305.7		
AlH ₃ 25% + AB + AP	25	28.3	270.3	1.686	3589	2466	369.8	333.1	300.1		
	27	30.4	270.5	1.677	3573	2438	368.9	332.6	300.0		
	29	32.5	270.8	1.668	3557	2410	368.1	332.3	300.0		

Table.5. Energy potential of the formulations with aluminum hydride (25%) and the active binder

Table 6 summarizes the calculated maximum attainable E2 and E3. These data are also presented in Fig. 13.

Table 6: The calculated maximum attainable E2 and E3 for the compositions with the active binder, oxidizer, and energetic component: Al (not higher than 20%), or Be (not higher than 14%), or AlH₃ (not higher than 25%).

oxidizer	AN				AP			ADN			
Energy. comp	Al	Be	AlH ₃	Al	Be	AlH ₃	Al	Be	AlH ₃		
E2	312	329	322	321	331	333	332	344	338		
E3	278	297	294	286	299	300	294	308	306		



The above data show that upon replacement of Al with AlH_3 or Be the maximum attainable E2 and E3 become less sensitive to the nature of oxidizer, although the order of relative efficiency of the oxidizers remains the same ADN > AP > AN.

Use of ammonium nitrate instead of AP and ADN can be justified in the systems where overall content of oxidizers is low, e.g., in formulations based on the active binder with a high content of HMX (usually 50-55%, whereas AP content amounts to ~10%). Since further increase of HMX content deteriorates combustion properties, one should replace AP with AN at the expense of a minor loss in energy potential (Table 7). Such formulation becomes environmentally safe, compared to AP-based one or much more thermally stable than ADN-based one. This type of AN formulations surpass AP in energy potential and are virtually same as ADN-based.

Oxidizer	Isp	ρ	Tc	E1	E2	E3
AP	259.6	1.924	3790	384.4	337.3	295.9
ADN	261.9	1.905	3780	385.5	338.9	297.9
AN	262.3	1.895	3760	384.9	338.7	298.1

Table 7: Energy potential of compositions containing 18% active binder, 20% Al, 50% HMX, and 12% oxidizer.

4. Stability of ammonium nitrate, its compatibility with other components of SCP, and the problem of phase stability.

Ammonium nitrate is a sufficiently stable compound. The rate $constant^2$ for its solid-phase deformulation (i.e.under 166°C) is k=2.2*10¹² exp(-45000/RT), s⁻¹; so, even at 140°C, k=3.4*10⁻¹² s⁻¹. This stability by orders of magnitude exceeds general requirements. The thermal decomposition proceeds via dissociation of the salt with formation of free nitric acid. Therefore, a compound having acidic nature e.g., oxalic acid, catalyzes AN decomposition. This catalysis depends upon various properties, in particular, upon acidity of a component, e.g. organic acids that are considerably weaker than oxalic do not affect AN decomposition³. To date, formulation of stable AN-based formulations is a solved problem.

Ammonium nitrate has a specific drawback, i.e., occurrence of three phase transitions accompanied by change in density in the temperature range of normal exploitation of solid propellant (V-IV transition at -17°C, IV-III at +32°C, and III-II at +82°C). These transitions cause mechanical degradation of propellant during storage. There is a known method to suppress these phase transitions by adding over 8% potassium nitrate⁴. We performed a number of studies aimed at phase stabilization of AN with additives that contain neither metals nor halides. It seemed possible to achieve stabilization by introduction of anions of close dimensions into AN lattice. However, the choice of anions commensurate with nitrate is but limited; NO_2^{-2} and CO_3^{-2} do not fit due to poor stability of ammonium nitrite and carbonate. Azide, cyanate, and oxalate anions could not be introduced into AN lattice neither by alloying nor by crystallization from solution; this is apparently due to substantial difference in anion structure. Only formate anion proved capable of forming isomorphous alloys in AN lattice in both cocrystallization and alloying⁵. Unfortunately, the phase stabilization could be achieved only at high content of formate, so the resulting compound makes little sense as an oxidizer. Our studies showed that a number of compounds, namely derivatives of five- or six-member nitrogen-containing heterocycles with carbonyl, amine, or imine groups, have stabilizing effect on ammonium nitrate. In some cases, phase structure is already stabilized at additive content of 3-4%. Some of these stabilizers are given below.



However, this stabilization is not complete, one phase transition, IV-II at +50°C or so persists. Nevertheless, this incomplete stabilization dramatically increases mechanical stability of AN items in cyclic shock heating. Fig. 15 presents pressed AN tablets after such tests.



Figure 14: Samples of pressed AN tablets after multiple cyclic shock heating from -40 to $+90^{\circ}$ C. Left to right: neat AN after two cycles, stabilized AN with uric acid after ten cycles, the latter after forty cycles. Initial dimensions of the tablets: height ~8-10 mm, diameter 6.0 mm

Note that the cracks after 40 heating cycles (Fig. 15, right) are due to very fast (in 20-30 min) wide range temperature sweep in small samples. The tests were aimed at application of stabilized AN in gas generating compositions for airbag inflators. The temperature conditions for SCP are substantially milder. Additionally to that, the remaining IV-II phase transition occurs at ~50, not at 32°C, in neat AN. Therefore, the gap between the average storage temperature and phase transition is many times increased, and the number of cases when the temperature could reach phase transition point during years of storage is reduced by orders of magnitude. Still, despite this optimistic assessment, a search for further phase stabilization of AN is necessary to get rid of any transition within -40 to +70°C. The work is underway at the Institute of Problems of Chemical Physics of the Russian Academy of Sciences.

5 5. Conclusion

1. Despite the fact that ammonium nitrate is less energetic oxidizer compared to AP and ADN, it may compete with those in cases when the cost plays an important role, when environmental impact of combustion products is limiting (advantage over AP), and when storage in a wider temperature range is necessary (advantage over ADN).

2. The ballistic efficiency of AN-based formulations, upon addition of energetic components (Al, Be, hydrides thereof), grows faster than for other oxidizers.

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