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EXPLOSIVE BEHAVIOR OF A SIMPLE COMPOSITE PROPELLANT MODEL

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EXPLOSIVE BEHAVIOR OF A SIMPLE COMPOSITE PROPELLANT MODEL

D. Price, J. O. Erkman, A. R. Clairmont, Jr.,
and D. J. Edwards

ABSTRACT: AP/Wax mixtures were used to model a simple composite propellant. Addition of wax to the AP increased its infinite diameter detonation velocity, increased its shock sensitivity, decreased its critical diameter and its reaction zone length. The maximum of these effects occurred at about 20% wax. At this composition, the model is an explosive comparable to TNT.

Approved by: Carl Boyars

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U. S. NAVAL ORDNANCE LABORATORY
White Oak, Silver Spring, Maryland

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EXPLOSIVE BEHAVIOR OF A SIMPLE COMPOSITE PROPELLANT MODEL

This work was carried out under the tasks MAT 03L 000/R011 01 01 FR 59 and ORDTASK 033 102 F009 06 01. It is part of a continuing program on the systematic investigation of the explosive behavior of composite propellant models.

E. F. SCHREITER
Captain, USN
Commander

ALBERT LIGHTBODY
By direction

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EXPLOSIVE BEHAVIOR OF A SIMPLE COMPOSITE PROPELLANT MODEL

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INTRODUCTION

This work is part of a systematic study of models of composite propellants. It is concerned with the detonability, shock sensitivity, and detonation behavior of the model, ammonium perchlorate (AP)/Wax, as a function of physical conditions (charge diameter and porosity) and of chemical composition. The present study covers the range of 5 - 31.5% wax. The composition of commercially produced, non-aluminized composite propellants (AP/organic matrix) is typically 18-26% matrix material. The model duplicates the usual propellant behavior of difficult detonability (large critical diameter) near voidless density, but is much easier to study as a granular charge, i.e., in regions where it is easily detonable.

Earlier work on this program dealt with the explosive behavior of pure AP[1-3] which provided necessary information for interpreting the present results. Most of the literature information on the oxidizer AP concerns its behavior as a propellant rather than as an explosive. This is also true of the system AP/organic fuel for which there are very few references, and those, in turn, provide only a small amount of information about its explosive behavior.

Waxed AP provides an interesting contrast to waxed organic H.E., e.g., waxed RDX. Addition of wax to RDX decreases its sensitivity and the size of its detonation parameters, (velocity, pressure and temperature designated D, P_j and T_j, respectively); addition of wax to AP increases its sensitivity and the size of its detonation parameters. This opposite effect must be explained on the basis that AP is a better oxidizer and a poorer explosive than RDX or other organic H.E.

EXPERIMENTAL

Materials

All AP used was propellant grade and contained 0.15 - 0.25% tricalcium phosphate (TCP). Lots N119 and N126 with average particle sizes of 200 and 25μ, respectively were described in References 2 and 3. Lot N127 was supplied by the same manufacturer (American Potash & Chemical Corporation) as Lot N126. The manufacturer's average particle size determination by Micromerograph was 44 and 43μ for N126 and N127, respectively. Ro-Tap sieve analysis of N127 was quite similar to **that** of N126.* Moreover, a newly available set of screens extending to smaller mesh sizes (Cenco Sieve Shaker) confirmed the Ro-Tap analysis of N127 in the region of overlap and indicated essentially the same weight mean particle size (34.4μ and 36.5μ, respectively by Ro-Tap and Cenco S.S.). If the Ro-Tap data for N126 and N127 are treated the same way, i.e., fitted to a linear log - log plot of accumulated percentage vs screen mesh size, the means are respectively 30.4μ and 34.4μ. The mean size of N126 was previously assigned[2] on the basis of a number of different examinations (e.g., microscopic and micromerograph determinations); the

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* Sieve Analyses

Wt. % Retained on Sieve No.

AP	100	140	200	230	270	325	Pan
N126[2]	0	1.0	2.5	3.8	6.0	10.3	74.6
	0	1.1	2.4	3.0	6.1	10.1	74.8
N127	0	0	1.7	3.6	8.3	12.3	74.0
				4.2[a]		20.0[a]	a

a. By Cenco Sieve Shaker with finer screens of 30μ and 20μ which retained 54.0% and 21.3%, respectively.

assigned value was 25μ. Consequently the present data were used to assign a value of (34.4/30.4) 25 = 28.3μ to N127 relative to N126 at 25μ. The fact the N127 is a slightly coarser material than N126 is further confirmed by its D vs ρ_o curve given later in the text.

The wax chosen for this work was carnauba, and a refined, powdered, grade No. 1 yellow was supplied by Frank B. Ross Co., Inc. According to the Ross literature, this wax has a melting point of 81.5-84°C and density of 1.00 g/cc at 15°C, 0.999 g/cc at 25°C. In its powdered form it is easily dry-mixed with AP to produce apparently uniform mixtures. By Ro-Tap, the average particle size was about 140μ but since the wax stuck to the sieves, this is too large a value. Microscopic examination showed that most particles were in the size range of 75-178μ with an average of about 125μ.

Carnauba wax[5] is a natural product (a tree exudate) and consists chiefly of fatty acids and alcohols (roughly, 30-carbon chains) in the form of esters. There are also small percentages of free acids and paraffin hydrocarbons. For computational purposes, the composition was approximated by $C_{30}H_{61}OH$ or $(CH_2)_{30}\cdot H_2O$ and also by $(CH_2)_n$. The grade 1 material is 96.7%-99.3% wax, melts at about 83°C, boils at 320°C, has a flash point of 270-327°C and a fire point of about 330°C. It is consequently a relatively volatile fuel for a composite propellant.

Charge Preparation and Experimental Procedure

AP/Wax mixtures were prepared by tumbling the dry components; they were stored in moisture vapor proof bags until used. Charge preparation and experimental procedures were those reported in the previous work[3] except that waxed mixtures were not heated above 35°C. All charges were either unconfined or supported in 0.08 mm-thick cellulose acetate envelopes. Boosters were of the same diameter as the test charge and 5.08 cm long*; they were of 50/50 pentolite (1.56 g/cc). Charges were 0.64-7.62 cm in diameter, 20.3 cm long*, and were frequently followed by a pentolite witness. The shock induced disturbance was recorded by a 70 mm smear camera at writing speeds of 1 to 4 mm/μsec. Smear camera records were similar to those

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* Near the end of this work, booster length was changed to 2.54 cm and charge length to 22.8 cm. This is noted in the appropriate tables.

of the previous work[1-3] with a tendency for waxed AP to produce greater luminosity of both the detonation front and the gas products for $\rho_0 \leq 1.2$ g/cc.

Record Reduction and Data Obtained

Record reduction was carried out as in the earlier work[1-3] but, in addition, all detonation velocities have been corrected for the effect of using a non-planar initiating shock[6,7] and for the difference in detonation velocity between the test charge and the booster[7]. Both corrections are made for an axial point 3.8 cm from the free end of the charge, i.e., at the midpoint of that part of the smear camera trace read for the velocity determination.

In the first correction, the 5.08 cm long booster is treated as part of the charge length. This correction amounts to -0.1% to -1.5% for length/diameter (ℓ/d) ratio values of 10 to 2.9. The second correction is positive and ranges from <0.1% to 0.5%. Hence the net correction amounts to 0 to -1.1% of the velocity read from the smear trace, and its absolute value decreases with decreasing charge diameter.

Eight sets of data included replications (seven pairs and one set of three). These showed an average precision of 0.64% with a range of 0.14 to 2.25%. However, seven of the eight were $\leq$0.78% and the average precision of these was 0.41%

RESULTS AND DISCUSSION

Detonation Velocity as a Function of % TMD

Diameter (d) = 5.08 cm. A general picture of the effect of wax is given by the D vs ρ_0 curves for 5.08 cm (2 in.) diameter charges. Unwaxed AP, N126 (25µ), had been studied earlier[2,3]; for completeness, these data are repeated in Table 1 and the D values have been corrected according to our present procedures. Tables 2 and 3 contain comparable data for AP/WAX, 90/10 and 80/20. All of these data are plotted in Figure 1 which shows the large increase in D (up to 60%) effected by adding wax to AP. The AP/Wax D vs ρ_0 curves are similar in kind to those of AP, i.e., AP/Wax shows group 2 explosive behavior. Addition of the wax, although it has not changed the explosive classification, has extended the range (raised the critical %TMD)

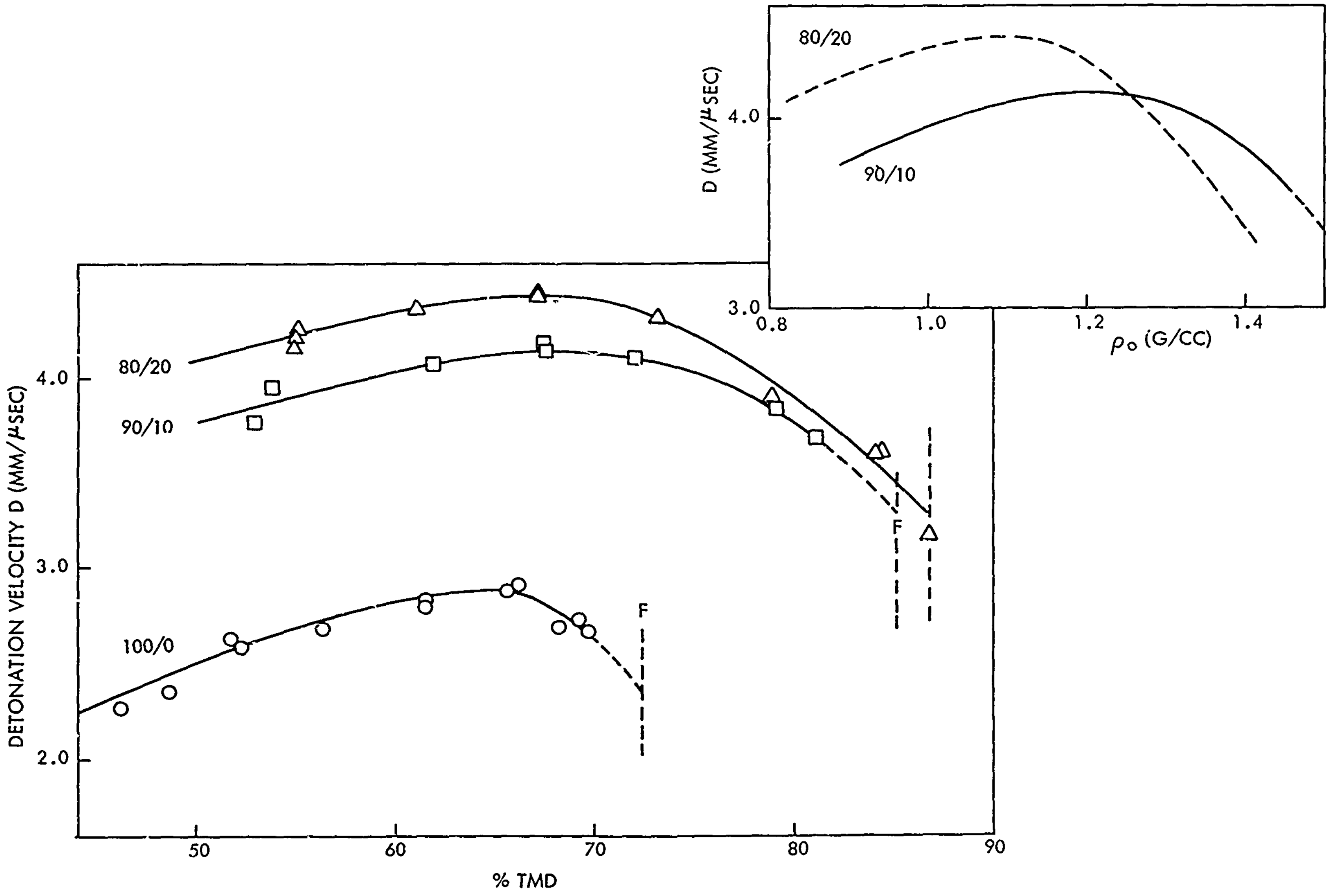

FIG. 1 EFFECT OF WAX ON DETONATION VELOCITY OF 25μ AP(N126) AT d = 5.08 CM

TABLE 1 - Detonation Velocity vs Density for 25µ AP (N-126)

Shot No.	ρ_o g/cc	%TMD	D(mm/µsec) Read	D(mm/µsec) Corrected once	D(mm/µsec) Corrected twice
169	0.90h	46.2	2.28	2.26	2.27
194	0.95h	48.7	2.36	2.34	2.35
179	1.01h	51.8	2.64	2.62	2.63
178	1.02h	52.3	2.60	2.58	2.59
172	1.10h	56.4	2.69	2.67	2.68
181	1.20H	61.5	2.81	2.79	2.80
180	1.20H	61.5	2.84	2.82	2.83
183	1.28i	65.6	2.89	2.87	2.88
182	1.29i	66.2	2.92	2.90	2.91
171	1.33i	68.2	2.70	2.68	2.69
201	1.35i	69.2	2.74	2.72	2.73
189	1.36i	69.7	2.67	2.65	2.66
188	1.41i	72.3	F(2.73)	δ = 18.5 cm	
177	1.43i	73.3	F(2.68)	δ = 16.5 cm	
170	1.47i	75.4	F(2.63)	δ = 8.4 cm	

ρ_v = 1.95 g/cc d = 5.08 cm

TABLE 2 - Detonation Velocity vs Density for AP (N126)/Wax, 90/10

Shot No.	ρ_o g/cc	% TMD	D(mm/µsec) Read	D(mm/µsec) Corrected Once	D(mm/µsec) Corrected Twice
319	0.941h	52.9	3.784	3.758	3.766
274	0.958h	53.8	3.973	3.946	3.955
278	1.101h	61.8	4.091	4.063	4.071
287	1.200H	67.4	4.179	4.152	4.159
322	1.201h	67.5	4.139	4.112	4.119
284	1.281i	72.0	4.118	4.090	4.098
285	1.408i	79.1	3.846	3.820	3.828
286	1.447i	81.3	3.694	3.669	3.677
283	1.516i	85.2	F(3.23) $\delta > 20.3$ cm		
272	1.749i	98.3	F $\delta \leq 7.3$ cm		

ρ_v = 1.78 g/cc, d = 5.08 cm, and 25µ AP

TABLE 3 - Detonation Velocity vs Density of AP (N126)/Wax, 80/20

Shot No.	ρo g/cc	% TMD	D(mm/μsec) Read	D(mm/μsec) Corrected Once	D(mm/μsec) Corrected Twice
333	0.901h	54.9	4.176	4.148	4.155
378	0.902h	55.0	4.231	4.203	4.210
354	0.903h	55.1	4.273	4.244	4.252
334	1.001h	61.0	4.381	4.352	4.359
335	1.101h	67.1	4.447	4.417	4.424
356	1.101h	67.1	4.459	4.429	4.436
336	1.201H	73.2	4.340	4.311	4.318
332	1.294i	78.9	3.904	3.878	3.886
337	1.379i	84.1	3.605	3.581	3.589
375	1.384i	84.4	3.613	3.589	3.597
381	1.424i	86.8	3.181	3.160	3.168

ρ_v = 1.64 g/cc, d = 5.08 cm, and 25μ AP considerably. This effect on detonability will be discussed in greater detail below.

Pure AP presents difficulties in the extrapolation of the D vs d^{-1} curve to the infinite diameter value (D_i) for charges at higher %TMD[1-3]. It is quite possible that similar difficulties will arise with AP/Wax. To minimize these, we worked with mixtures at 50-67% TMD which lie at, or to the left of, the maximum in the D vs ρ_o curves of Figure 1. The curves at 10 and 20% wax parallel each other (and roughly parallel the 0% wax curve) up to about 72% TMD; after that they tend to converge in a manner that indicates that the greater amount of wax is less effective in the less porous charges.

Comparisons are made here, as in all work of this project, at equal %TMD or equal % porosity (100 - %TMD) to assure equal void space, interior surface, and volume of materials being compared. As Figure 1 shows, on this basis, the curves for the two waxed mixtures show some convergence, but do not cross. The inset of Figure 1 shows the same data, plotted on an absolute density basis instead of on a porosity basis. In this case the curves do cross to show that the 80/20 mix exhibits higher velocities than the 90/100 at $\rho_o \leq 1.25$ g/cc and lower velocities at $\rho_o > 1.25$ g/cc.

The supply of AP N126 was exhausted before the planned work had been completed. Since particle size does not affect the infinite diameter results, a new lot of AP (N127) was used to obtain the D_i vs ρ_o curves of mixtures at 5 and 31.5% wax. For comparison, data from this second lot of AP are given in Table 4 and plotted in Figure 2 (analogous to Figure 1). Although the two lots of AP show distinct differences, the addition of wax almost obliterates them. For example, a plot of D (5.08 cm, 55% TMD) vs % wax shows the waxed AP (N126) and waxed AP (N127) on essentially the same curve. However, a valid comparison can be made only with infinite diameter values (D_i), and that is done in the next section.

<u>Diameter Effect</u>. Because there was some information in the literature on AP/Wax, 90/10, this mixture was first studied for the effect of diameter and porosity on detonation velocity. The data obtained are given in Table 5 and plotted in Figures 3 and 4. The former displays D vs d for two porosities (52.8 and 67.5% TMD) of the 90/10

TABLE 4 - Detonation Velocity vs Density for 28μ AP (NJ.27) and Its Wax Mixtures

			D(mm/μsec)			
Shot No.	ρ_o g/cc	% TMD	Read	Corrected Once	Corrected Twice	ρ_v g/cc
		100/0, AP/Wax				1.95
432	0.901h	46.2	1.993	1.979	1.988	
431	1.001h	51.3	2.290	2.274	2.283	
445	1.001h	51.3	2.361	2.345	2.354	
433	1.101h	56.5	2.511	2.494	2.503	
434	1.222i	62.7	2.628	2.664	2.673	
442	1.267i	65.0	2.635	2.617	2.626	
443	1.316i	67.5	F(2.48)	$\delta > 20.3$ cm		
444	1.338i	68.6	F(2.47)	$\delta > 20.3$ cm		
435	1.362i	69.8	F	$\delta = 15.7$ cm		
		95/5, AP/Wax				1.86
439	1.025h	55.1	3.419	3.396	3.[illegible]	
482	1.236i	66.4	3.689	3.664	3.670	
		68.5/31.5, AP/Wax				1.50
477	0.826h	55.0	3.802	3.776	3.782	
490	1.006h	67.0	4.036	4.008	4.014	

d = 5.08 cm, 28μ AP

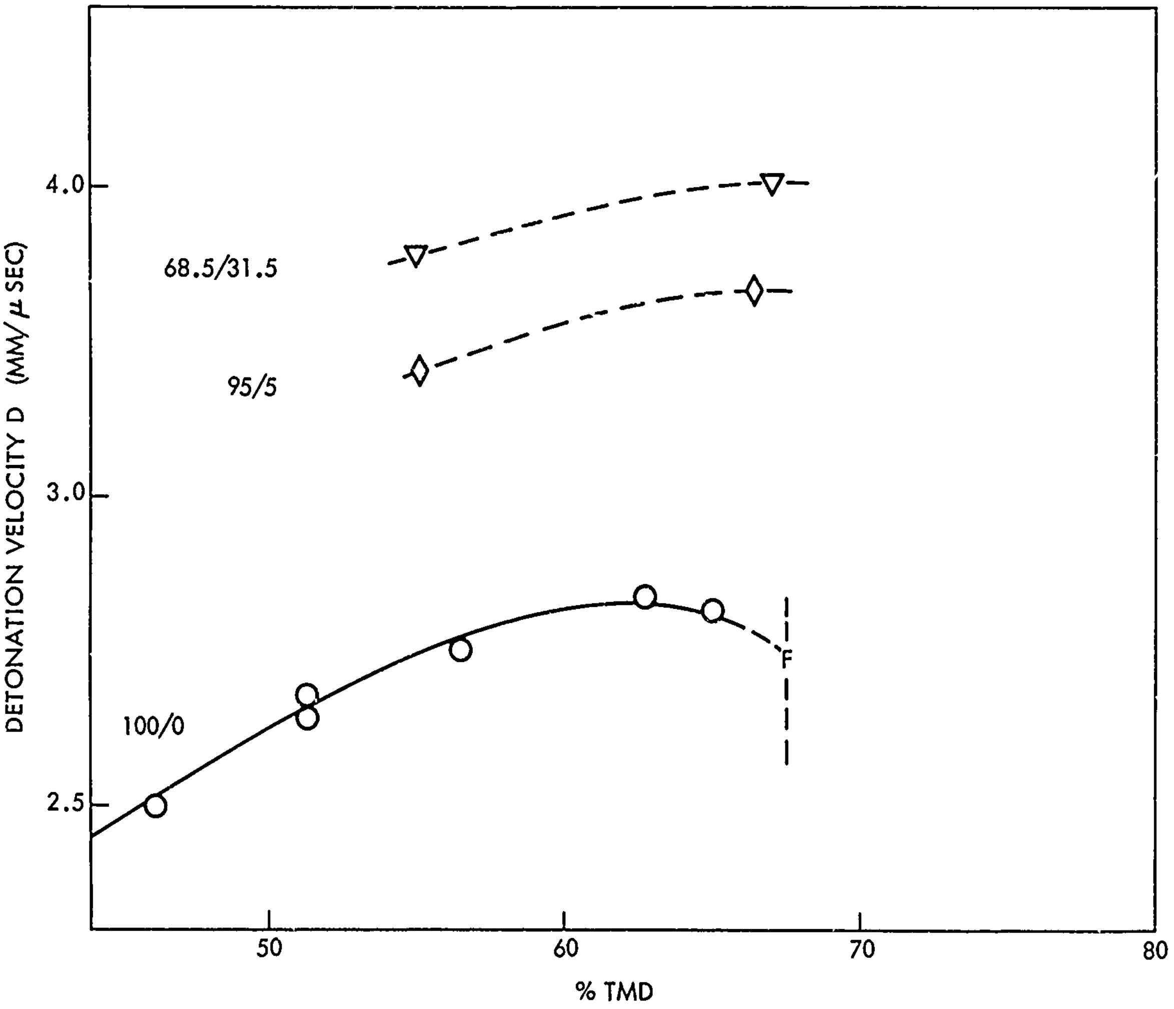

FIG. 2 EFFECT OF WAX ON DETONATION VELOCITY OF 28μ AP(N127) AT d = 5.08 CM

mixture. The curves are typical of those found for the other three mixtures in that they show a regular, smooth increase of D with increasing d. They can be compared with the analogous curve for a 10μ AP at 51.3% TMD[3,9], also shown in Figure 3. It is evident that the curves for the 53% TMD mixture and the 51% TMD AP are essentially parallel although the former extends to much lower d values before it reaches its failure limit, as shown in Figure 3. Over the same range in d, extrapolation of D vs d^{-1} should be as satisfactory for the mixture as for the pure AP. There is no comparable guidance for the 67% TMD mixture because the curve for the 65% TMD AP[1] is parallel to other AP curves at $d \geq 6.35$ cm, but goes through a point of inflection at about d = 5.08 cm. Extrapolation of data for pure AP at 65% TMD gives questionable results[1,3] and so too may extrapolation of the higher density 90/10 mixture.

Finally, Figure 3 contains a dashed curve (from Reference 8) for 10μ 90/10, AP/paraffin, at 56.1% TMD. This curve cannot be explained from our present data since none of our mixtures exhibited an S-type D vs d curve. Moreover, the smaller particle size mixture would be expected to exhibit higher detonation velocity than the N126 mixtures at all values of the diameter, not just at $d > 5$ cm. The Reference (8) curve appears to be approaching such expected behavior at $d > 6$ cm, but the curve at lower diameters must result from measured velocities which arose from metastable reactions rather than from true detonations. Certainly we observed no constant velocity (independent of diameter for d small) that could be attributed to decomposition of AP only, and no transition to another constant (higher) velocity, independent of diameter for d large, which could be attributed to reaction of AP and wax. Hence our observations conflict with those of Reference (8) and with the explanation of them proposed there.

Figure 4 displays the D data for the 90/10 mixtures plotted as a function of reciprocal diameter. In contrast to pure AP, the smaller diameter D values lie above the extension of the upper linear section of the D vs d^{-1} curve. Of course, the pure AP is not detonable at most of the smaller diameters, but where a velocity can be measured off the linear section of the curve, it lies below the

TABLE 5 - Diameter Effect in AP/Wax, 90/10

Shot No.	Diameter d		ρ_0	D	D(0.940)*	D_{c1}	D_{c2}
	in.	cm	g/cc	mm/μsec			
			a AP(N126), 25μ				
					52.8% TMD		
367	0.25	0.635	0.901h	F	δ < 7.6 cm	--	--
365	0.375	0.953	0.907h	2.254	2.242	2.242	2.242
347	0.50	1.27	0.901h	2.556	2.560	2.560	2.560
346	0.75	1.905	0.901h	3.050	3.060	3.060	3.060
338	1.00	2.54	0.918h	3.201	3.220	3.215	3.217
319	2.00	5.08	0.941h	3.784		3.759	3.766
274	2.00	5.08	0.958h	3.973	3.959	3.933	3.940
320	2.50	6.35	0.937h	4.002		3.962	3.974
321	3.00	7.62	0.937h	4.066		4.005	4.024
					67.5% TMD		
368	0.25	0.635	1.212h	F	δ < 4.4 cm	--	--
366	0.375	0.953	1.202h	2.149		2.149	2.149
348	0.50	1.27	1.204h	2.586		2.586	2.586
349	0.75	1.905	1.202h	3.127		3.127	3.127
339	1.00	2.54	1.204H	3.457		3.450	3.453
322	2.00	5.08	1.201h	4.139		4.112	4.119
287	2.00	5.08	1.200H	4.179		4.152	4.159
323	2.50	6.35	1.201H	4.369		4.325	4.335
324	3.00	7.62	1.202H	4.538		4.470	4.485
			b AP(N119), 200μ				
					60.3% TMD		
401	1.375	3.49	1.071h	F(2.77)		δ > 20.3 cm	
402	2.00	5.08	1.075h	3.906		3.879	3.888
403	2.50	6.35	1.071h	4.365		4.318	4.331
394	3.00	7.62	1.074h	4.473		4.405	4.423
					67.4% TMD		
398	3.00	7.62	1.201h	F(4.15)		δ > 20.3 cm	

* Small correction to D assuming linear variation with density

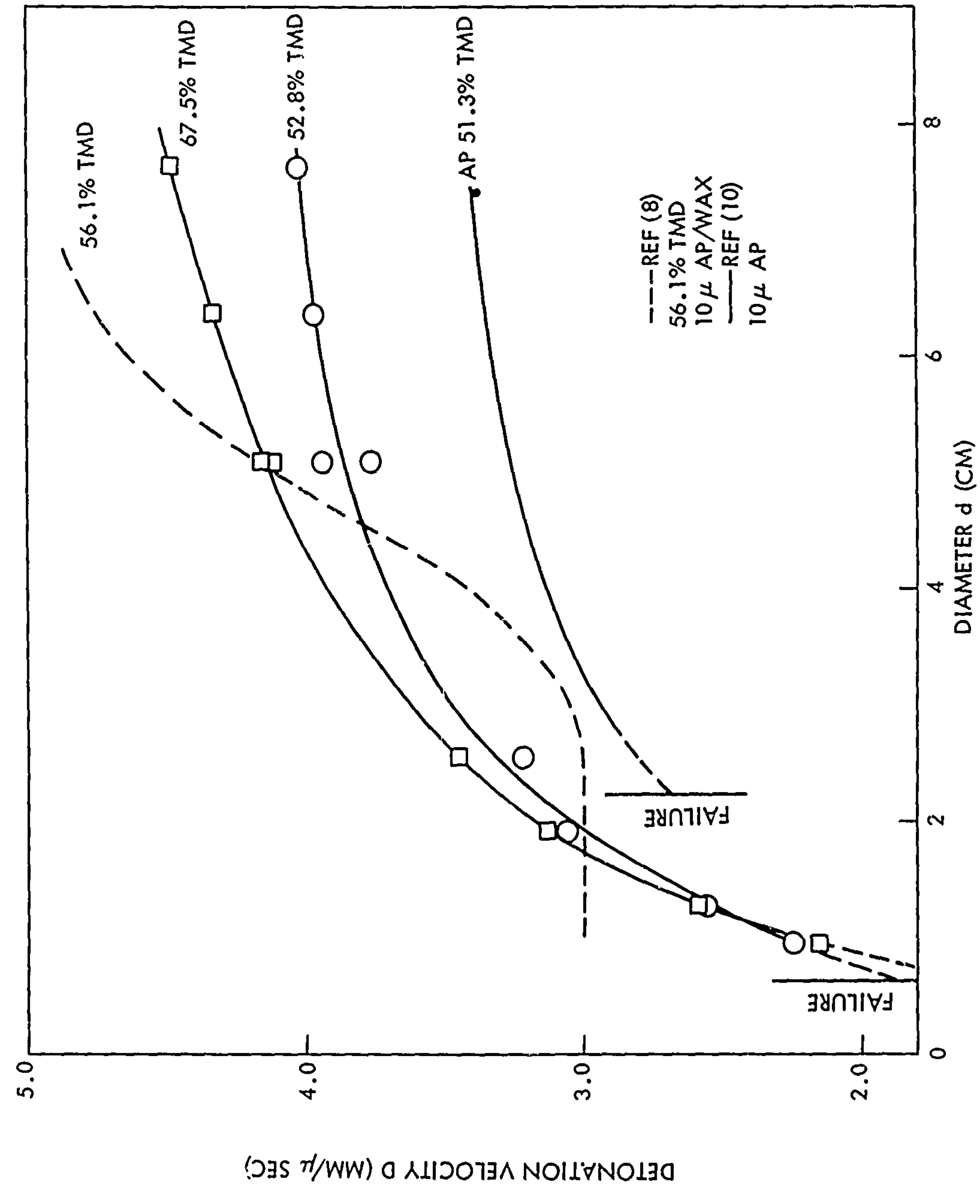

FIG. 3 DIAMETER EFFECT IN AP(N126)/WAX, 90/10

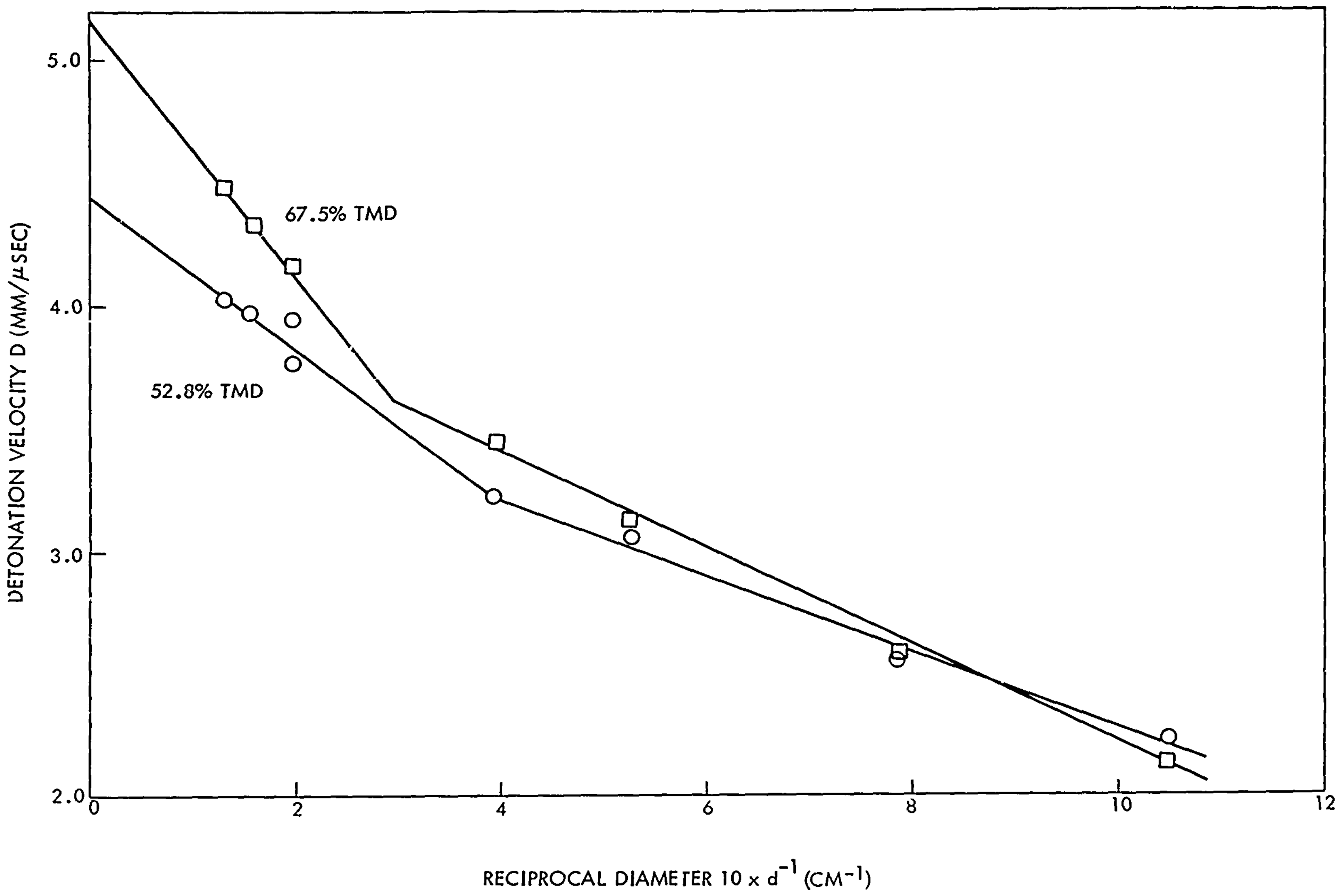

FIG. 4 EXTRAPOLATION OF D VS d^{-1} DATA FOR AP(N126)/WAX, 90/10

extension of the linear section.

Tables 6, 7, and 8 contain, respectively, the data obtained for the AP/Wax mixtures, 80(N126)/20, 95(N127)/5 and 68.5(N127)/31.5. Figure 5 illustrates the diameter effect in the 80/20 mixture and confirms the fact, suggested in Figure 3, that the D vs d curves for mixtures of different porosities can cross. Figures 6, 7, and 8 show the extrapolation curves for the mixtures of 20, 5, and 31.5% wax, respectively.

Infinite Diameter Detonation Velocities, D_i. All of the curves D vs d showed perfectly continuous variation (e.g., Figures 3 and 5). Consequently D vs d^{-1} curves should also be smooth. However, if D approaches D_i asymptotically as d increases, a portion of the D vs d^{-1} curve generally approximates a straight line. Figures 4, 6, 7, and 8 show how the D vs d^{-1} curves were approximated either by one straight line over the whole range of d^{-1} or, as was more generally the case, by two. The range showing linearity at the larger diameters was selected from these graphs. The data for these ranges were then treated by least squares to find the best linear fit, and the resulting straight line extrapolated to D_i at $d^{-1} = 0$*.

The D_i values so obtained are listed in Table 9 and plotted as linear curves of D_i vs %TMD in Figure 9. The slopes for the 90/10 and 80/20 mixtures were so nearly equal that of the pure AP[9], also shown in Figure 9, that the value for the 95/5 mixture at 55% TMD was selected to give the same slope. As Table 9 shows for this set of data, dropping the lowest diameter point from the data set results in a much lower standard deviation for the fit; that was our first choice for the extrapolation. However, with only 5% wax it is harder to get reproducible charges; hence greater scatter would be expected for the 95/5 than for the other three mixtures. That consideration added to the way the D_i of the 95/5 mixture (at 55% TMD) fit into the D_i - ρ_o - % wax patterns of Figure 9 led to the final choice made.

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*The D vs d data were also fitted to the curve $D = D_i \left[1 - (\alpha/d)^K\right]$. This manner of determining D_i was unsuccessful for reasons discussed in the appendix.

TABLE 6 - Diameter Effect in AP (N126)/Wax, 80/20

25μ AP

Shot No.	Diameter d in.	cm	ρ_o g/cc	D mm/μsec	D(0.901)*	D_{c1}	D_{c2}
						54.9% TMD	
382	0.25	0.635	0.916h	F(1.67)	δ = 5 cm		
373	0.375	0.953	0.934h	2.359	2.408	2.408	2.408
383	0.50	1.27	0.916h	2.843	2.867	2.867	2.867
384	0.75	1.905	0.901h	3.381		3.381	3.381
385	1.00	2.54	0.901h	3.716		3.710	3.711
351	1.00	2.54	0.901h	3.758		3.752	3.754
377	1.375	3.495	0.902h	3.949		3.937	3.939
378	2.00	5.08	0.902h	4.231		4.203	4.210
354	2.00	5.08	0.903h	4.273	4.271	4.243	4.250
333	2.00	5.08	0.901h	4.176		4.149	4.156
352	2.50	6.35	0.906h	4.384	4.377	4.330	4.343
379	2.50	6.35	0.908h	4.390	4.381	4.337	4.347
353	3.00	7.62	0.901h	4.510		4.442	4.458
380	3.00	7.62	0.905h	4.480	4.473	4.405	4.423
					D(1.101)*		
						67.1% TMD	
374	0.375	0.953	1.105h	F	δ < 7.6 cm		
386	0.50	1.27	1.103h	2.542		2.542	2.542
387	0.75	1.905	1.102h	3.288		3.288	3.288
388	1.00	2.54	1.101h	3.665		3.659	3.661
355	1.00	2.54	1.101h	3.676		3.670	3.672
335	2.00	5.08	1.101h	4.447		4.418	4.424
356	2.00	5.08	1.101h	4.459		4.430	4.437
357	2.50	6.35	1.101h	4.651		4.605	4.613
358	3.00	7.62	1.094h	4.791	4.802	4.729	4.746

* Small correction to D assuming linear variation with density

TABLE 7 - Diameter Effect in AP (N127)/Wax, 95/5

28µ AP

Shot No.	Diameter d in.	Diameter d cm	ρ_o g/cc	D mm/µsec	D(1.250)	D_{c1}	D_{c2}
						55.1% TMD	
437	0.75	1.905	1.025h	1.871		1.871	1.871
436	1.00	2.54	1.026h	2.688		2.683	2.686
438	1.375	3.495	1.025h	3.089		3.078	3.083
439	2.00	5.08	1.025h	3.419		3.396	3.404
440	2.50	6.35	1.025h	3.602		3.564	3.578
441	3.00	7.62	1.025h	3.705		3.649	3.669
						67.2% TMD	
480*	1.00	2.54	1.2351	2.735	2.738	2.733	2.735
481	1.375	3.495	1.2621	3.169	3.166	3.155	3.158
482	2.00	5.08	1.2361	3.689	3.692	3.667	3.673
483	2.50	6.35	1.2741	3.948	3.943	3.902	3.911
484	3.00	7.62	1.2501	4.157	4.157	4.094	4.107

ρ_V = 1.86 g/cc

* Note that all shots of number >480 were made with boosters 2.54 cm long (instead of 5.08 cm) and acceptors 22.8 cm long (instead of 20.3 cm)

TABLE 8 - Diameter Effect in AP (N127)/Wax, 68.5/31.5
28µ AP

Shot No.*	Diameter d in.	Diameter d cm.	ρ_o g/cc	D mm/µsec	D_{c1}	D_{c2}
					55.1% TMD	
475	1.00	2.54	0.825h	3.187	3.181	3.183
476	1.375	3.495	0.826h	3.542	3.530	3.533
477	2.00	5.08	↓	3.802	3.776	3.781
478	2.50	6.35		3.936	3.895	3.904
479	3.00	7.62		4.078	4.016	4.029
					67.1% TMD	
488	1.00	2.54	1.006h	3.137	3.131	3.133
489	1.375	3.495	1.007h	3.629	3.617	3.619
490	2.00	5.08	1.006h	4.036	4.009	4.014
491	3.00	7.62	1.006h	4.362	4.296	4.307

ρ_v = 1.50 g/cc

* All shots made with booster 2.54 cm long

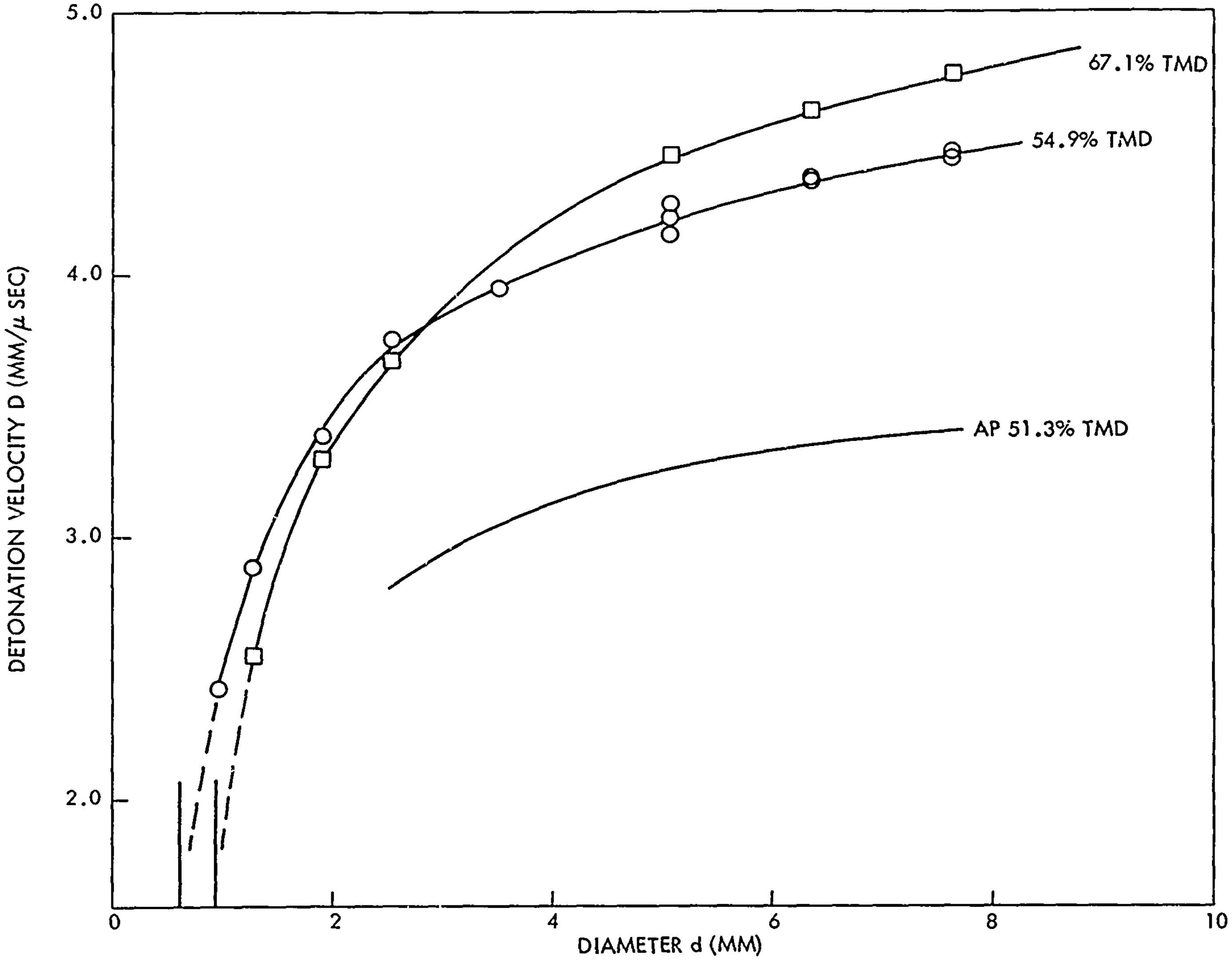

FIG. 5 DIAMETER EFFECT IN AP(N126)/WAX, 80/20

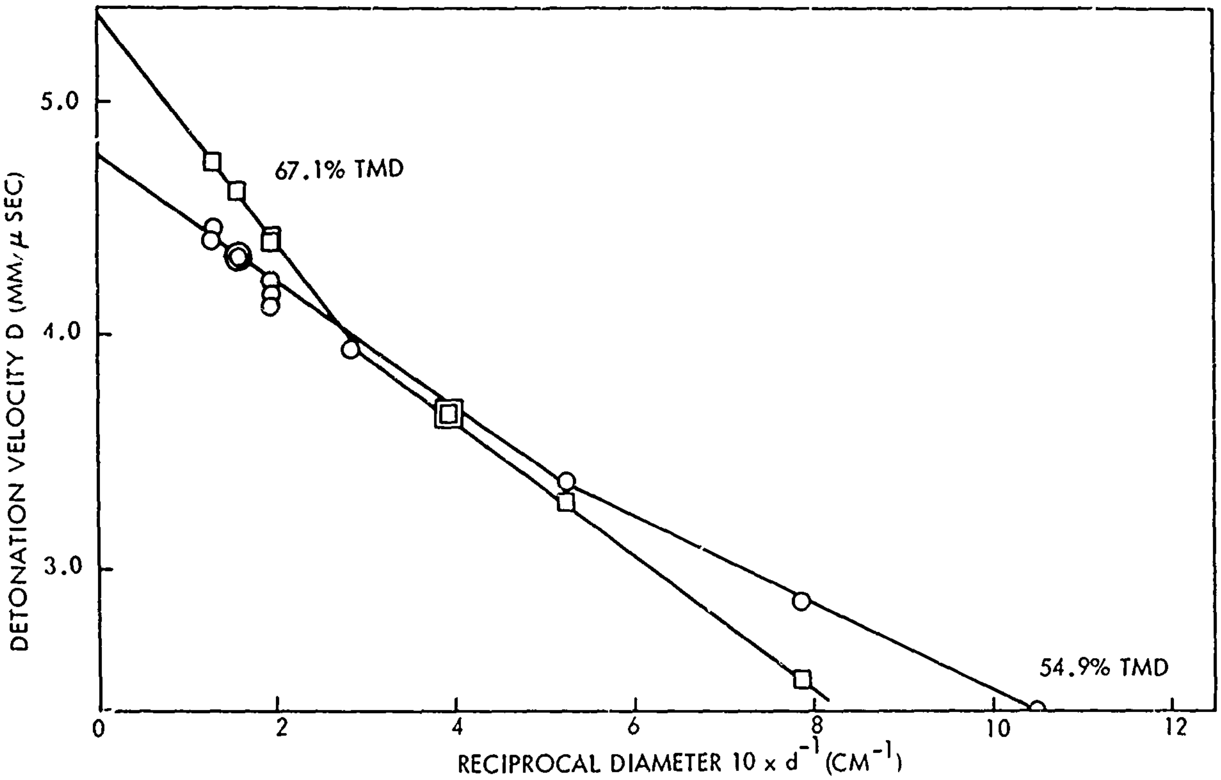

FIG. 6 EXTRAPOLATION OF D VS d^{-1} DATA FOR AP(N126)/WAX, 80/20

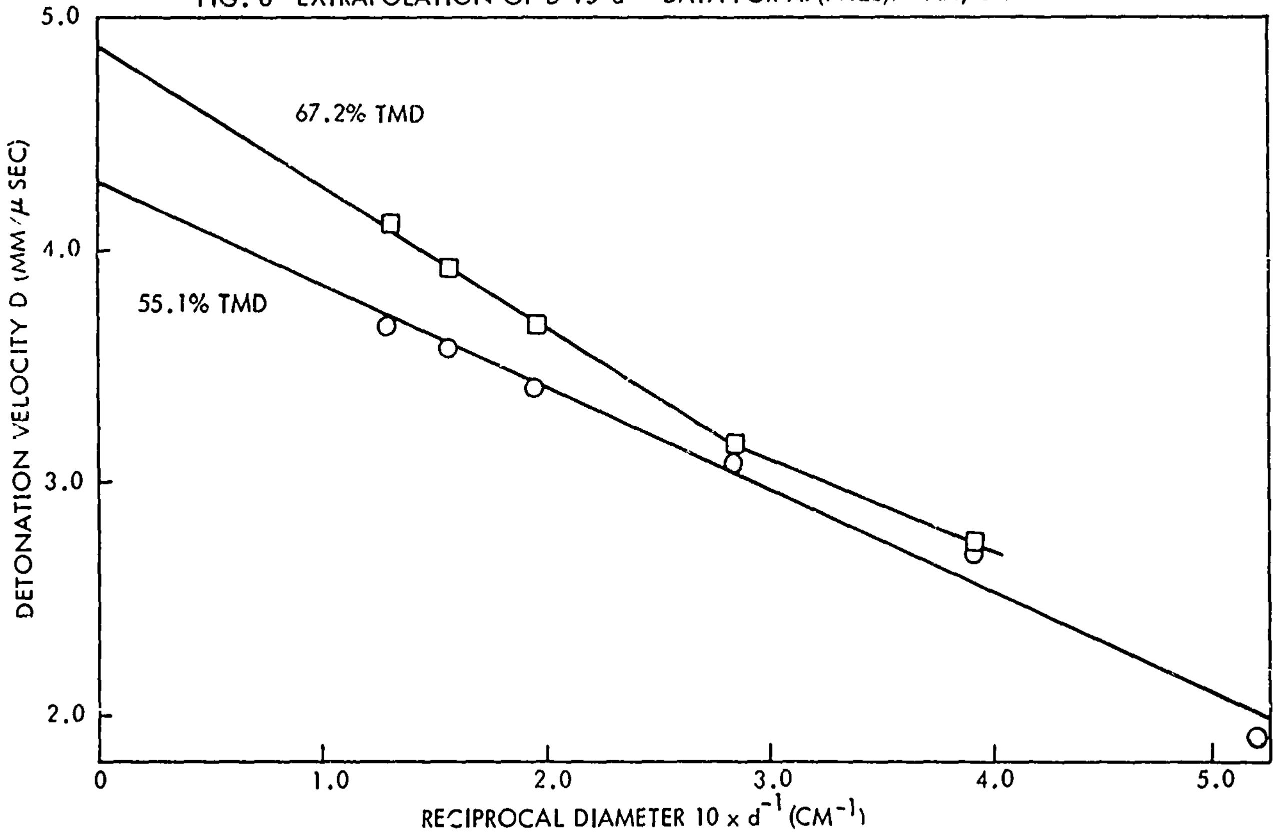

FIG. 7 EXTRAPOLATION OF D VS. d^{-1} DATA FOR AP(N127)/WAX, 95/5

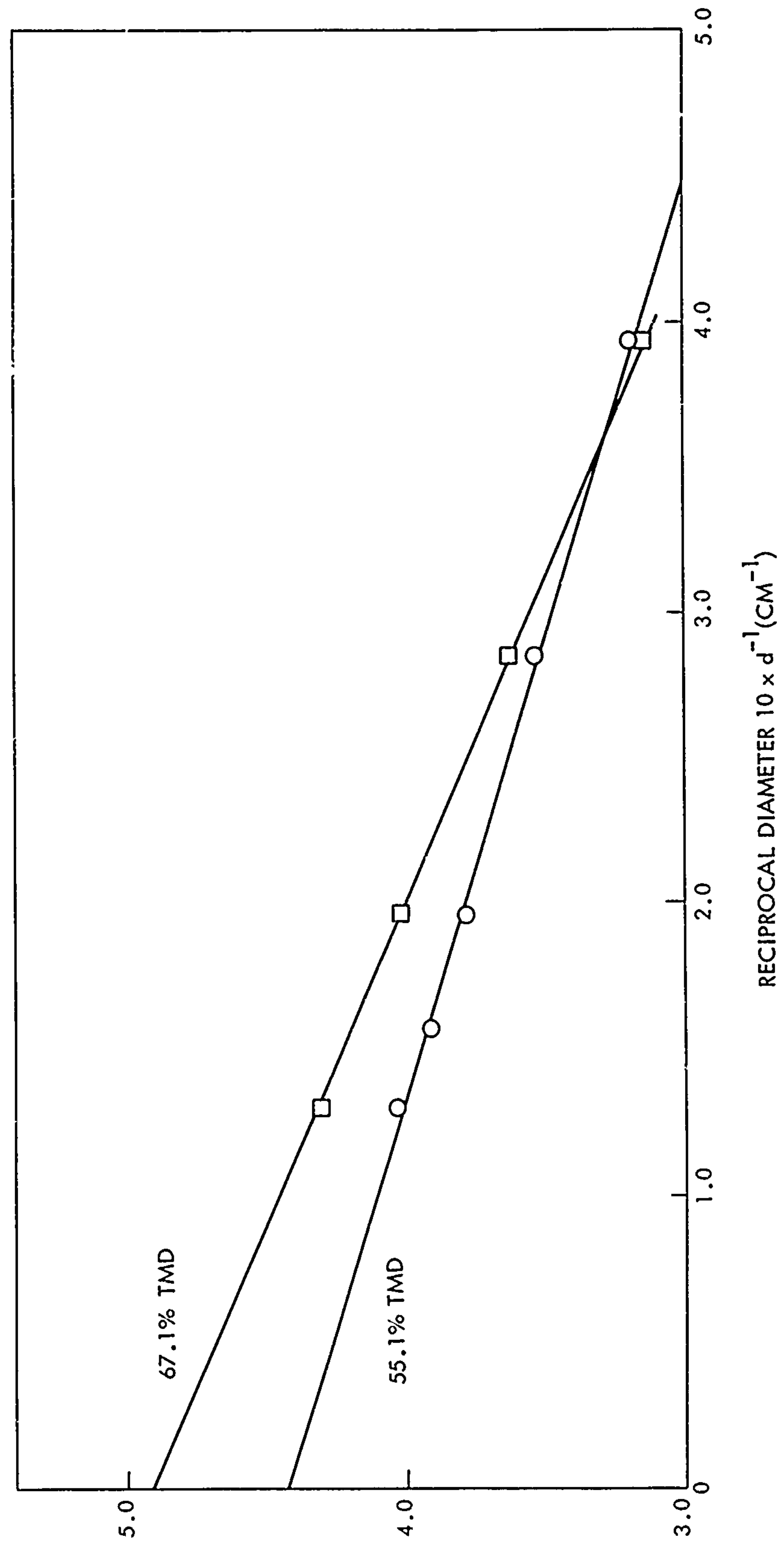

FIG. 8 EXTRAPOLATION OF D VS. d^{-1} DATA FOR AP(N127)/WAX, 68.5/31.5

With the use of smoothed values from the D_1 vs ρ_0 curves of Figure 9, the two upper curves of D_1 vs % wax at 55 and 67% TMD were constructed. They show D_1 increasing up to a broad maximum at about 20% wax, and decreasing thereafter. The curves should be smooth, but not necessarily symmetrical, because as long as the wax increases the energy release, it should increase D_1, but beyond that it would act as a diluent. The upper curves of Figure 9 indicate that the diluent effect is much more evident at 67% TMD than at 55% TMD, but the concentration at maximum D_1 and the effect at lower concentrations seem much the same for both porosities.

Reaction of Wax with AP

It was shown in earlier work[9] that the decomposition of AP in detonation of very low density charges approximates the reaction

$$2NH_4ClO_4 \longrightarrow N_2 + 2HCl + 3H_2O + 2.5O_2 \qquad (1)$$

But at practical charge densities, the arbitrary decomposition

$$2NH_4ClO_4 \longrightarrow N_2 + Cl_2 + 4H_2O + 2O_2 \qquad (2)$$

seems to be a better approximation. If we now assume that the AP in an AP/Wax charge first decomposes according to Eq. (2) we then have corresponding simple arbitrary equations for reactions with the wax: (a) all Cl appears as Cl_2 in the products, O goes to form H_2O, CO_2 and CO in sequence, and (b) all Cl appears as HCl in the products, O goes to form H_2O, CO, and CO_2 in sequence. These lead to the stoichiometric amount of wax and a constant volume heat of reaction of (a) 7.66% and 1120 cal/g and (2) 9.40% and 1234 cal/g. Although the stoichiometric concentrations are less than half that found at maximum D_1, the arbitrary mechanisms have some value in approximating computed equilibrium products.

For a kilogram of the mixtures, the molecular compositions of 90/10 and 80/20 are represented, respectively, as

$$7.6596\ NH_4ClO_4 + 0.2279\ C_{30}H_{61}OH \quad \text{and}$$

$$6.8085\ NH_4ClO_4 + 0.4558\ C_{30}H_{61}OH.$$

With a heat of formation of $-\Delta H_f^0$ (298°K) = 223 kcal/mole for $C_{30}H_{61}OH$, computations were made with the arbitrary mechanisms (a) and (b) and

TABLE 9 - D_1 Values for AP/Wax Mixtures

AP/Wax	AP	ρ_o g/cc	%TMD	Range used in d, cm	D_1 mm/µsec	q.m.e. mm/µsec	a mm
95/5	N127	1.025	55.1	1.9-7.6 2.5-7.6	4.29 (2.0%) 4.16 (0.3%)	0.093 0.012	10.3 (4.5%) 9.0 (1.1%)
90/10	N126	0.940	52.8	2.5-7.6	4.46 (1.9%)	0.073	7.0 (9.6%)
80/20	N126	0.901	54.9	1.9-7.6	4.75 (0.6%)	0.048	5.5 (3.2%)
68.5/31.5	N127	0.826	55.1	2.5-7.6	4.41 (0.6%)	0.022	7.1 (2.8%)
90/10	N119	1.073	60.3	6.4-7.6*	4.88	--	7.2
95/5	N127	1.250	67.2	3.5-7.6	4.88 (0.8%)	0.023	12.4 (2.4%)
90/10	N126	1.200	67.5	5.1-7.6	5.17 (1.3%)	0.021	10.1 (6.0%)
80/20	N126	1.100	67.1	5.1-7.6	5.41 (0.7%)	0.012	9.2 (3.6%)
68.5/31.5	N127	1.006	67.1	2.5-7.6	4.89 (0.1%)	0.003	9.1 (0.3%)

* Two point extrapolation

Notes: The quadratic mean error (q.m.e.) is defined as $\sigma = \left[\Sigma(y_i - \hat{y}_i)^2/n-k\right]^{\frac{1}{2}}$ (See Reference 9). It measures the extent to which the derived straight line fits the data. Values in parentheses are corresponding standard deviations (converted to percentages) for D_1 and a. The latter is defined by $D = D_1\ (1 - ad^{-1})$.

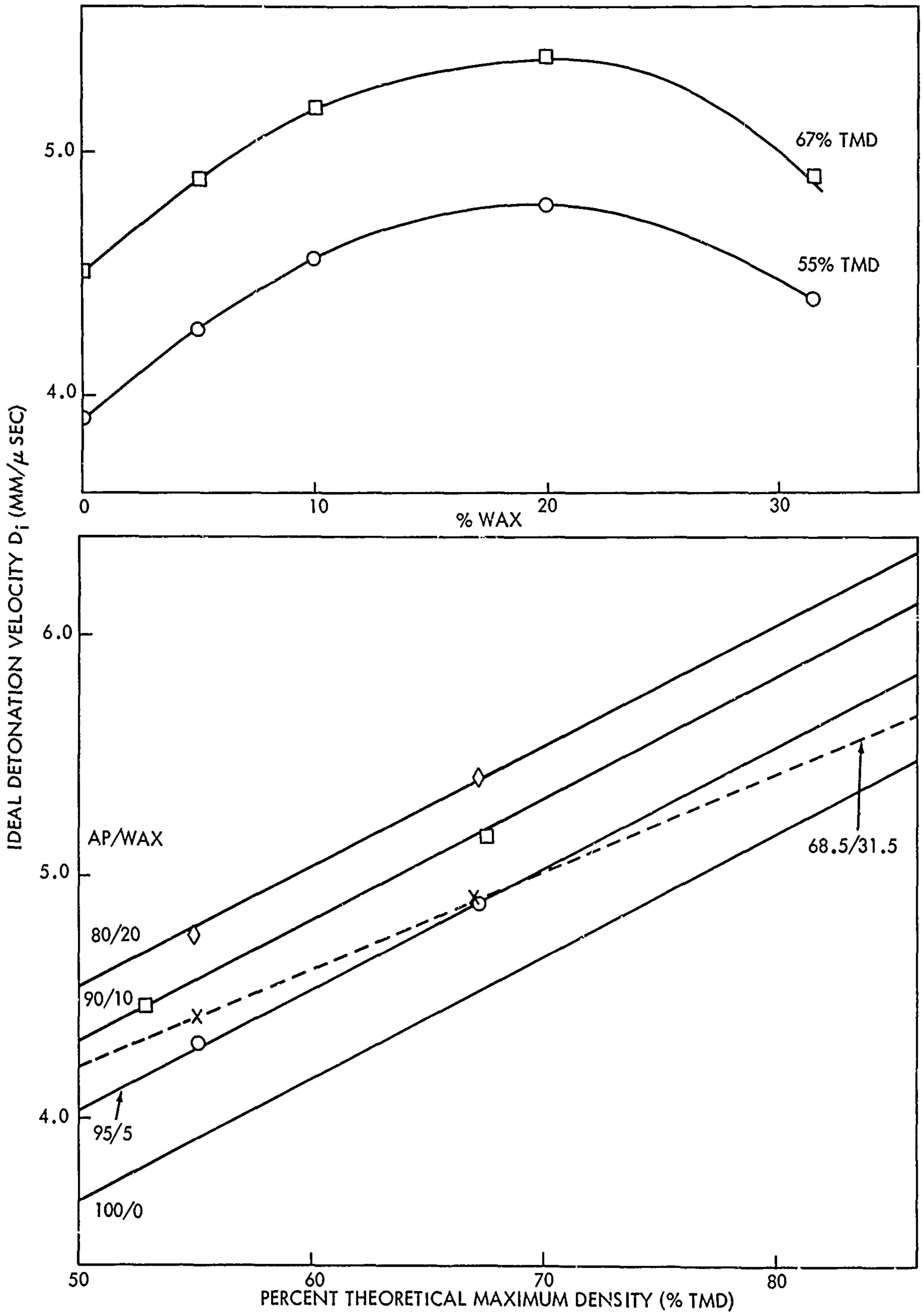

FIG. 9 EFFECT OF WAX ON D_i VALUE OF AP

with the Ruby code (TNT parameters). The results are compared in Table 10 with AP/Wax computations (NOL Propellant Code) for burning at one atmosphere.

The agreement between mechanism (a) and the Ruby results at 80% TMD is very good. They differ chiefly because of the products Cl and CCl_4 which were not considered in (a). Atomic Cl appears because of the very high temperature, but it is probable that CCl_4 is too complicated a molecule to form in the detonation and should not have been considered in the Ruby products. The net result is that mechanism (a) predicts the same amount of gas and a chemical energy about 4% lower than the Ruby values; most of this difference would be removed if CCl_4 were not considered a product. Mechanism (b) for 50% TMD produces results in good agreement with Ruby results at 50% TMD for the 80/20 mixture and in fair agreement for the 90/10 mixture. Here the difference (a net of +7% in chemical energy released) is caused chiefly by the shift in equilibrium products as a result of the high temperature (3174°K). The products from burning are not approximated well by either mechanism, but they are, of course, closer to the results of (b) which correspond to the lower % TMD and hence lower reaction pressure.

Figure 10 displays the results computed by Ruby for the effect of wax on AP. Note that the slope of the computed curve is the same as that of the experimental for pure AP although the absolute value of D_1 is too high by 0.7 mm/µsec. The slopes of the waxed mixtures are lower than those found experimentally, but this may result, in part, from the fact that the stoichiometric amount of wax is less than 10%. The slope of the experimental curve 68.5/31.5 (for which an excess of wax is undoubtedly present) is also lower than that of the AP curve (Figure 9). These Ruby computations give D_1 values for the waxed mixtures that exceed the experimental by at least as much as was the case for AP. In drawing the upper curve of Figure 10, D_1 vs % wax, straight lines were used to connect the few points; they were made to intersect at the appropriate maxima found with the arbitrary mechanisms. The computed values show convergence of the 55% and 67% TMD curves earlier than the experimental curves; this is comparable to the earlier occurrence of the maxima mentioned before.

TABLE 10 - Computations for AP/Wax

	AP/Wax, 90/10					AP/Wax, 80/20				
	Arbitrary b	50% TMD	80% TMD	Arbitrary a	Adiabatic* Burning Values	Arbitrary b	50% TMD	80% TMD	Arbitrary a	Adiabatic* Burning Values
D(mm/μsec)	--	5.526	6.638	--	--	--	5.305	6.397	--	--
P_j(kbar)	--	84.4	180.8	--	--	--	70.0	160.6	--	--
T_j(°K)	--	3174	2639	--	2686(T_p)	--	2816	2482	--	2054(T_p)
ρ_j(g/cc)	--	1.291	2.001	--	--	--	1.177	1.872	--	--
γ	--	2.221	2.470	--	--	--	2.295	2.343	--	--
Δe(cal/g)	1222	1140	1156	1120	913(Δh)	1085	1087	1088	1038	732(Δh)
n(moles/kg)	36.9	37.54	33.50	34.3	36.7	37.9	38.36	34.62	34.6	49.0
Products** (Moles/kg)										
N_2	3.83	3.82	3.82	3.83	3.77	3.40	3.31	3.39	3.40	3.40
Cl_2	0	0.37	0.09	3.83	0	0	0.03	0.11	3.40	0
Cl	0	1.36	0.62	0	1.17	0	2.64	0.97	0	0.02
HCl	7.66	5.53	0.69	0	6.49	6.81	6.49	1.83	0	6.79
H_2O	18.55	19.45	22.01	22.38	16.63	24.34	22.25	26.71	27.69	10.90
CO	1.36	2.25	0.55	0	2.96	3.35	3.22	0.18	0	12.18
CO_2	5.48	4.78	4.16	4.24	4.17	0	1.11	0.40	0	2.08
CCl_4	0	0.01	1.54	0	0	0	0	0.95	0	0
C(s)	0	0	0.59	2.6	0	10.89	8.87	12.13	13.67	0
H_2	0	0.14	0.01	0	1.80	0	1.03	0.03	0.05	13.56
NH_3	0	0.01	0.01	0	0	0	0.19	0.04	0	0
CH_4	0	0	0	0	0	0	0.47	0.02	0	0

** Other species considered were H, N_2O, NH_3, NO, NO_2, O, O_2, and OH

* Computed on NOL Propellant code at 1 atm. (other computations were on Ruby code with TNT parameters). For 90/10 mixture, also found 0.45H, 0.17 O, 0.66 O_2, 1.10 H and 0.12 NO. Also considered in flame equilibrium were C(s), ClO, NO, NO_2, and N_2O. In this computation, wax was approximated as $(CH_2)n$ with a heat of formation of 5.5n kcal/mole.

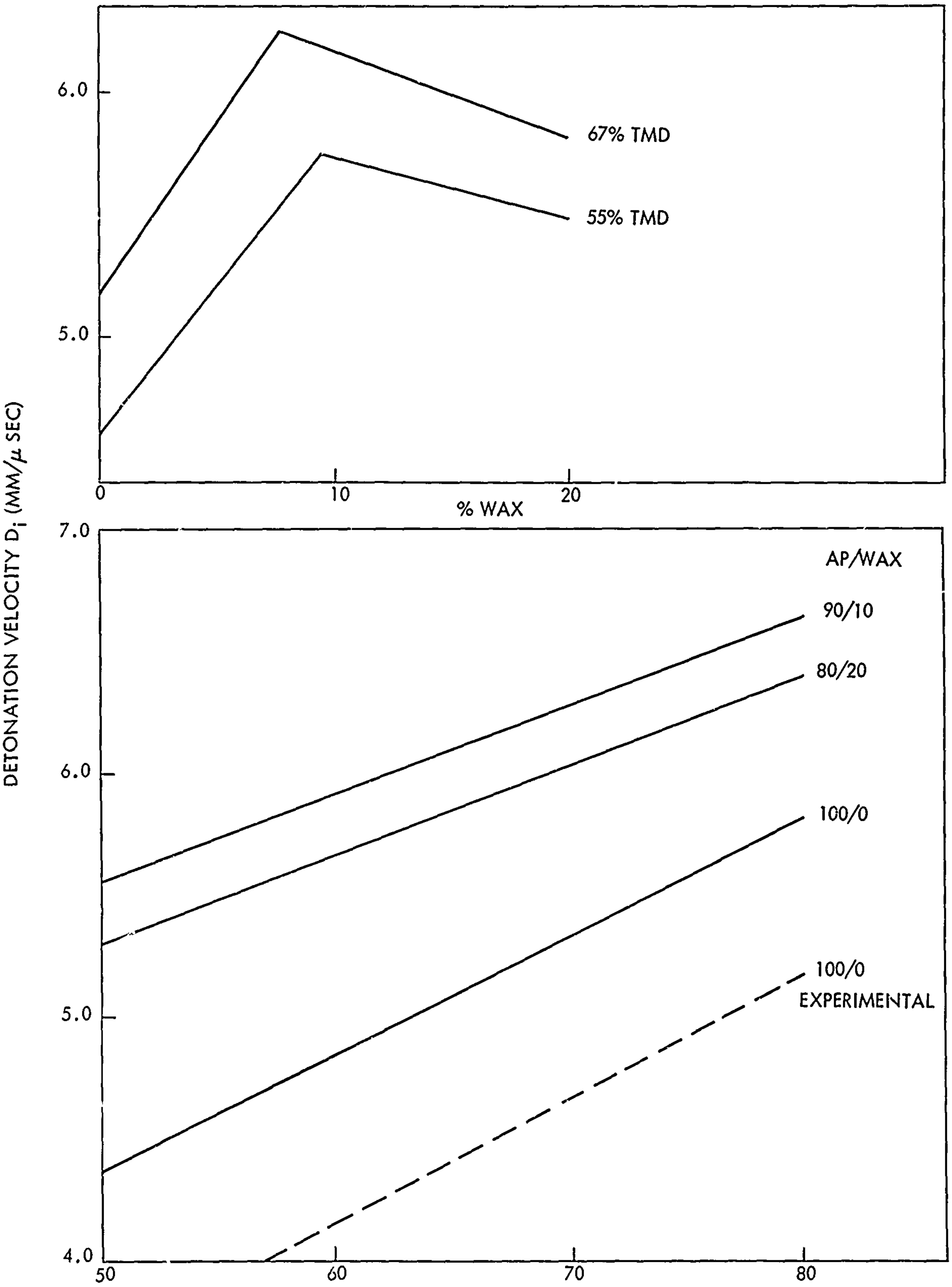

FIG. 10 COMPUTED EFFECT OF WAX ON D_i VALUE OF AP (RUBY CODE)

Figure 11 shows the computed detonation temperatures (T_j) for the AP/Wax mixtures together with their adiabatic flame temperatures which are considered a reasonable lower limit of T_j. Just as Ruby (with the TNT parameters) overestimates D_i, and consequently P_j, it appears to underestimate T_j. Even so, the relative positions of the curves in Figure 11 show, as would be expected, that the addition of a fuel to AP increases its temperature of reaction substantially.

The qualitative information offered by the computations indicates that wax and AP react. The resultant increase in chemical energy released causes about the expected increase in D_i and P_j. Experimentally, the maximum effect occurs not at the stoichiometric amount of wax, but at about twice this amount. This discrepancy is attributed to a number of effects, e.g., only the wax which has been vaporized and can be oxidized very rapidly by the AP decomposition products can contribute to the detonation phenomena. A higher than stoichiometric concentration of wax in the original mixture is evidently necessary to obtain the optimum concentration in the reaction zone.

The AP/Wax mixtures make very interesting explosives. In Figure 12, the arbitrary decomposition mechanism (a) has been used to show how addition of wax increases the reaction energy very rapidly to the stoichiometric; thereafter it decreases it, but very slowly. At the same time, the volume of gas products stays about constant from a wax content of zero to more than twice the stoichiometric amount. Beyond this, the gas products increase rapidly as a result of the simple decomposition of the excess wax (essentially CH_2). The same broad maximum and slow fall off found in reaction energy is reflected in the experimental D_i vs % wax curve of Figure 9, but the locations of the maxima differ, of course.

The most effective of the AP/Wax mixtures seems to be the 80/20. The organic explosive it approximates best is TNT. The comparison is shown in Table 11, and is made for D_i, P_j (approximated as $\rho_o D_i^2/4$), Δe and n. The last two values were computed on the Ruby Code, but for the AP/Wax the maximum of Table 10, i.e., that computed for 90/10, was used to approximate the value at the experimental point

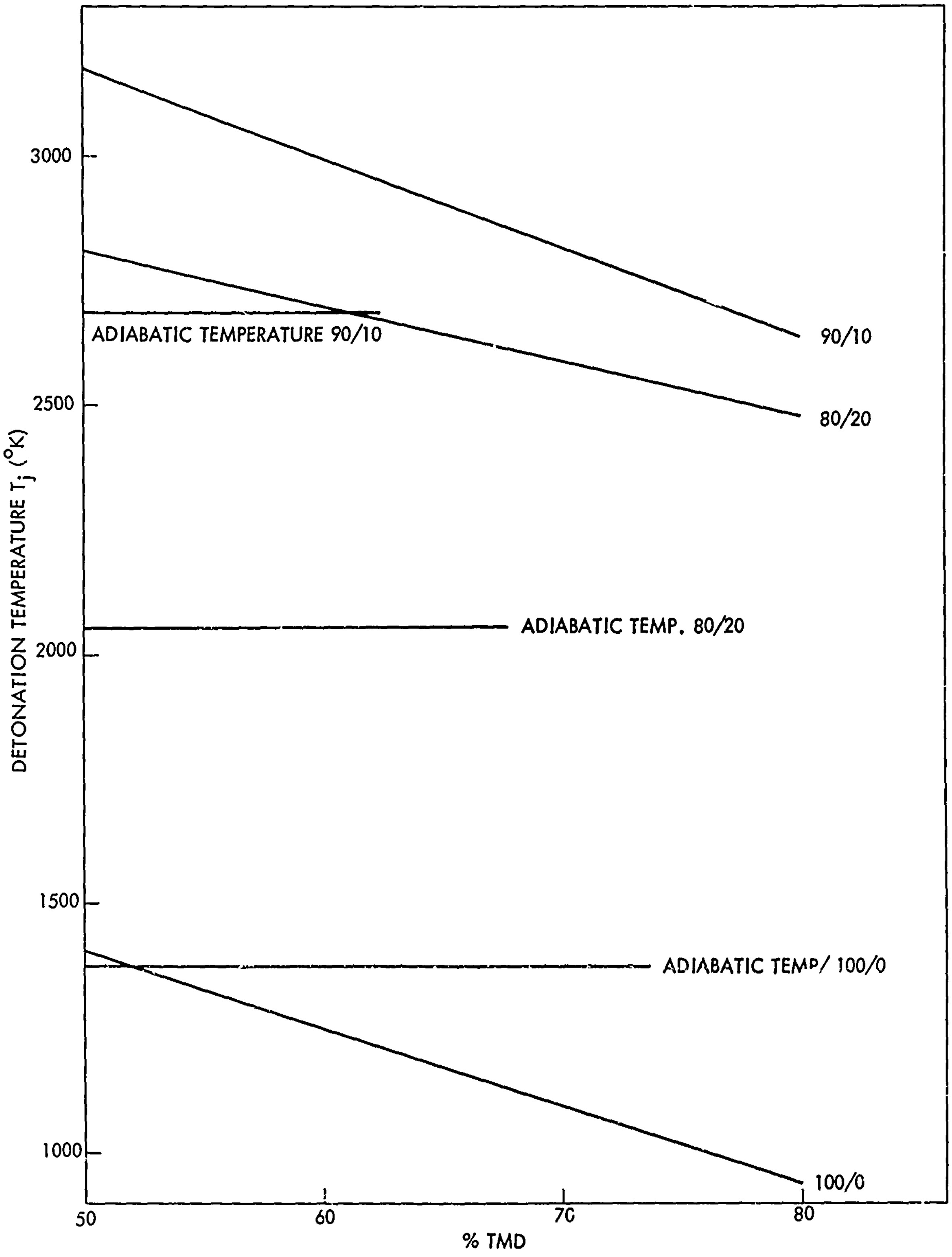

FIG. 11 COMPUTED DETONATION TEMPERATURES FOR AP/WAX MIXTURES

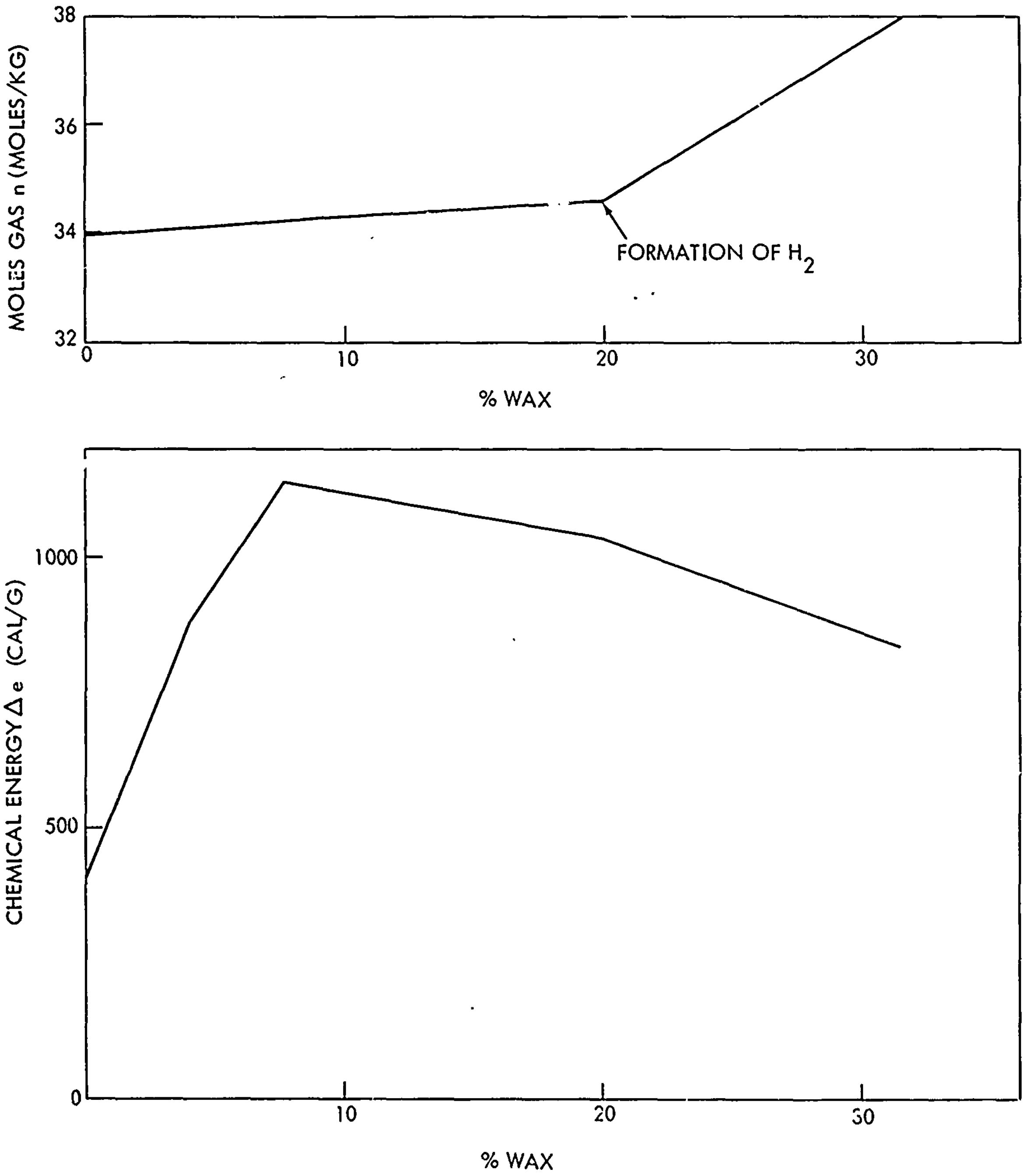

FIG. 12 VARIATION OF VOLUME OF GAS PRODUCTS AND CHEMICAL ENERGY RELEASE WITH AMOUNT OF WAX IN AP/WAX MIXTURE

TABLE 11 - Comparison of AP/Wax, 80/20, with TNT
25μ AP

Material	D_i mm/μsec	P_j [a] kbar	Δe [b] cal/g	n [c] moles/kg
55% TMD				
AP/Wax	4.79 [d]	51.7	~1140	~37.5
TNT	4.77	51.5	1092	30.8
67% TMD				
AP/Wax	5.39 [d]	79.8	~1156	~33.5
TNT	5.40	80.5	1130	28.4

a. Estimated as $\rho_o D^2/4$. TNT values are close to those of Reference (10), parallel the curve of Reference (11).

b. Ruby code computations. For AP/Wax, values at computed maximum, i.e., 90/10.

c. Ruby code computations. For AP/Wax, 90/10 mixture used; for TNT Reference (11).

d. Reference (12).

of maximum effect*. All comparisons are made at the same % TMD, but this is also nearly the same ρ_0 because the voidless density of AP/Wax, 80/20 is 1.64 g/cc; the crystal density of TNT is 1.654 g/cc. The two explosives have the same D_i and P_j at 55 and 67% TMD. The AP/Wax has an appreciably greater volume of gas products and probably a higher heat of reaction than the TNT. Its detonability region is more restricted than that of TNT (this will be described later), but within that region it is probably a slightly more effective H.E. [Ruby values of T_j and P_j have not been tabulated here because the T_j are probably wrong for all H.E., and the values for AP/Wax are either over-estimated (P_j) or underestimated (T_j)]. However, the subroutine of Ruby which computes equilibrium gives proper results for the P_j, T_j used, and the equilibrium shifts very little with T_j. Hence the Δe and n values may be useful for a relative rating.

Reaction of Wax with RDX

As we remarked initially, addition of wax has a very different effect on organic H.E. from that which it has on AP. This can be shown very simply with Ruby Code calculations on RDX/$(CH_2)_n$. The results are summarized in Table 12. (The computations were carried out for another project, but have not been previously reported.) It is evident that wax in RDX lowers D_i, P_j, T_j, and Δe although the volume of gas products stays about constant. Figure 13 displays the results for D_i vs % wax; the curve was extrapolated to 70% wax for comparison with an experimental value at that composition.

Also shown on Figure 13 are what we consider the best D_i measurements for RDX and PBX 9205 (approximating RDX/Wax, 92/8). The D_i for Comp A, also shown, is from data obtained on charges at a single diameter with the relatively slow streak camera available at that time[16]. Hence it is not considered as accurate as the D_i for RDX and PBX. Finally, Table 13 gives the Reference 17 data which were extrapolated to obtain D_i for the RDX/Wax, 31.5/68.5 mixture, also plotted in Figure 13. Contrary to the AP/Wax results, the agreement

---

* The Ruby values for 90/10 and 100/0 give D ratio values of 1.24 and 1.19 at 55 and 67% TMD respectively. Corresponding ratios from the experimental work (Figure 9) for 80/20 and 100/0 are 1.23 and 1.19.

TABLE 12 - Ruby Code Computations for RDX/$(CH_2)_n$*

Products \ %$(CH_2)_n$:	0[13]	15	25	35	45
ρ_0(g/cc)	1.80	1.55	1.43	1.33	1.24
$-\Delta H_f^\circ$(298°K) cal/g	-66.18	1.040	45.80	90.70	135.6
D_i(mm/μsec)	8.57	7.98	7.67	7.41	7.16
P_j(kbar)	341	249	209	180	154
T_j(°K)	2668	2403	2128	1847	1604
ρ_j(g/cc)	2.427	2.073	1.904	1.763	1.637
γ	2.882	2.962	3.016	3.068	3.121
Δe(cal/g)	1486	1325	1206	1079	952
n(moles/kg)	34.1	34.1	33.7	33.5	33.5
Products (moles/kg)					
CO	0.71	0.35	0.07	0.02	0
CO_2	6.66	1.23	0.16	0.04	0.01
H_2O	12.98	20.16	19.87	17.47	14.83
N_2	13.39	11.13	9.03	6.96	5.12
NH_3	0.23	0.71	2.19	3.65	4.62
CH_4	0.09	0.47	2.37	5.38	8.86
C(s)	6.05	20.14	25.34	28.30	30.64

* Original LRL parameters in code[13]. All mixtures are at approximately 100% TMD with density of polyethylene taken as 0.9 g/cc.

TABLE 13 - RDX/Wax Data Used for Extrapolation [17]

Diam. d in.	d^{-1} (in.)$^{-1}$	ρ_o g/cc	D mm/µsec
Batch 1 (31.5/68.5, regular casting)			
1.25	8.00	1.11	6.54
		1.11	6.67
1.50*	6.67	1.12	6.34
		1.12	6.19
1.75	5.71	1.13	6.73
		1.14	6.63
Batch 6 (30.75/69.25, vacuum casting)			
1.50*	6.67	1.08	6.13
1.63	6.14	1.11	6.52
		1.11	6.64
1.75	5.71	1.10	6.57
		1.10	6.50

* In both series the 1.5 in. values seem low.

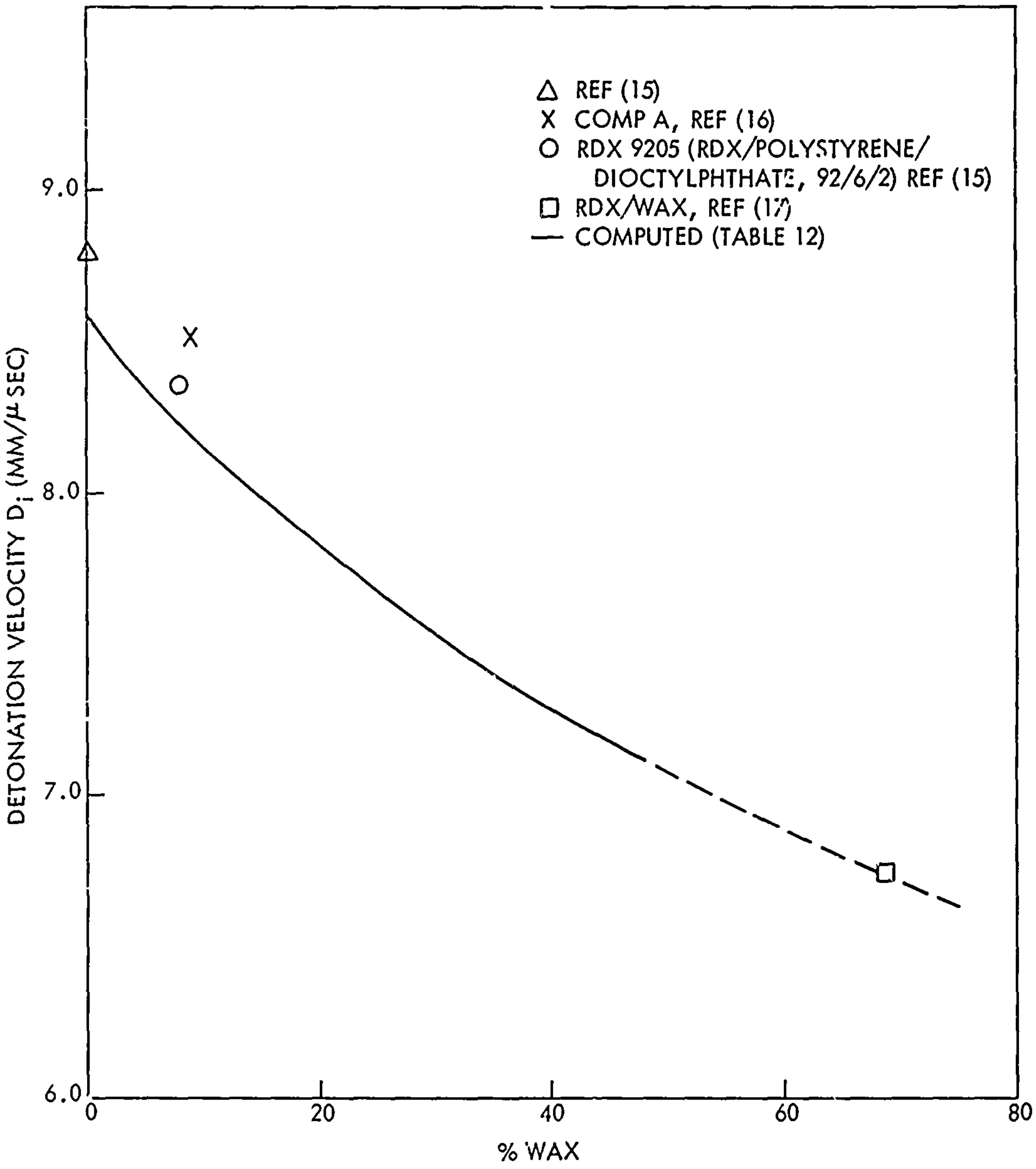

FIG. 13 EFFECT OF WAX ON DETONATION VELOCITY OF RDX

between absolute values for D_i computed and D_i measured is good here. It would probably have been better, had the RDX parameters been used in the BKW equation instead of the original Ruby values. Agreement with P_j, approximated as $\rho_o D_i^2/4$, is also good, of course.

A similar computation and excellent agreement with the experimental value of a HMX/polyethylene mixture will be found in Reference 18.

Reaction Zone Length and Reaction Time

The available diameter effect theories were summarized in Reference (3) where it was pointed out that the curved front or modified curved front theory is most consistent with our data for pure AP. If we treat the waxed AP in a similar manner, the Eyring reaction zone length is "a" in the expression

$$D/D_i = 1 - a/d \qquad (3)$$

and is related to the reaction time τ by

$$a = (D - \bar{u})\,\tau \qquad (4)$$

where $\bar{u}$ is the average particle velocity between the leading von Neumann shock and the C-J plane of the detonation front. (The ratio of two zone lengths should have the same value in either curved front theory although the absolute values of the computed zone lengths differ by almost an order of magnitude.) If Eq. (4) is applied to the infinite diameter conditions, and we assume that

$$D_i - \bar{u}_i = \beta D_i$$

where β is a constant, then

$$a_1/a_2 = (D_{1i}/D_{2i})\,(\tau_{1i}/\tau_{2i}) \qquad (5)$$

will give the desired ratios. The nominal reaction zone lengths (a) for the waxed AP mixtures are given in Table 9. To use Eq. (5), we need the corresponding "a" values for AP(N126) and AP(N127).

Values for the unwaxed APs were obtained by use of the D values for charges of d = 5.08 cm (Tables 1 and 4) and the D_i values (determined in earlier work[9]) in Eq. (3). The "a" values so calculated are listed in Table 14 and plotted in Figure 14. These are not very precise, of course, and the curves of Figure 14 could be drawn a number of ways. However, the curves, as drawn, show a

TABLE 14 - Nominal Reaction Zone Lengths
for AP (N-126) and AP (N-127)

ρ_o g/cc		a (mm)
	N-126[a], 25μ	
0.90		17.5
0.95		17.6
1.015*		15.5
1.10		16.5
1.20*		17.0
1.28		17.9
1.29		17.7
1.33		20.9
1.35		20.8
	N-127[b], 28μ	
0.90		21.7
1.00		19.2
1.10		18.9
1.22		19.2
1.27		20.6

a. Data from Table 1 used in Eq. (3).
b. Data from Table 4 used in Eq. (3).

* Average of two values.

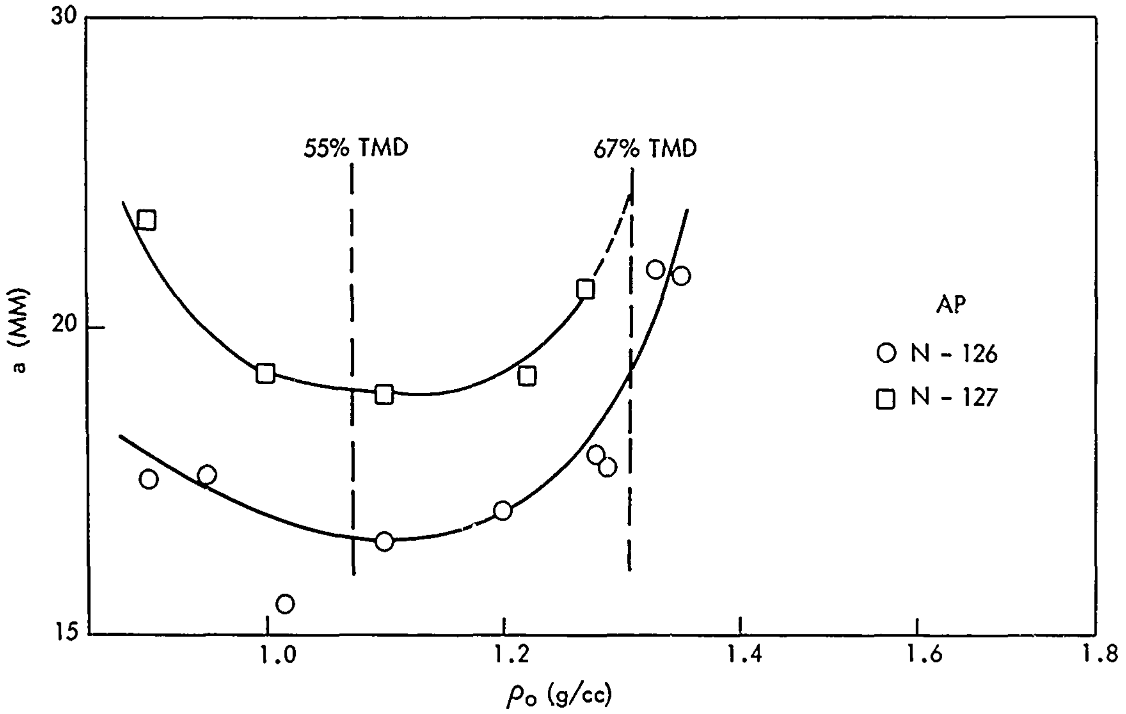

FIG. 14 VARIATION OF NOMINAL REACTION ZONE LENGTH WITH DENSITY FOR TWO AP'S

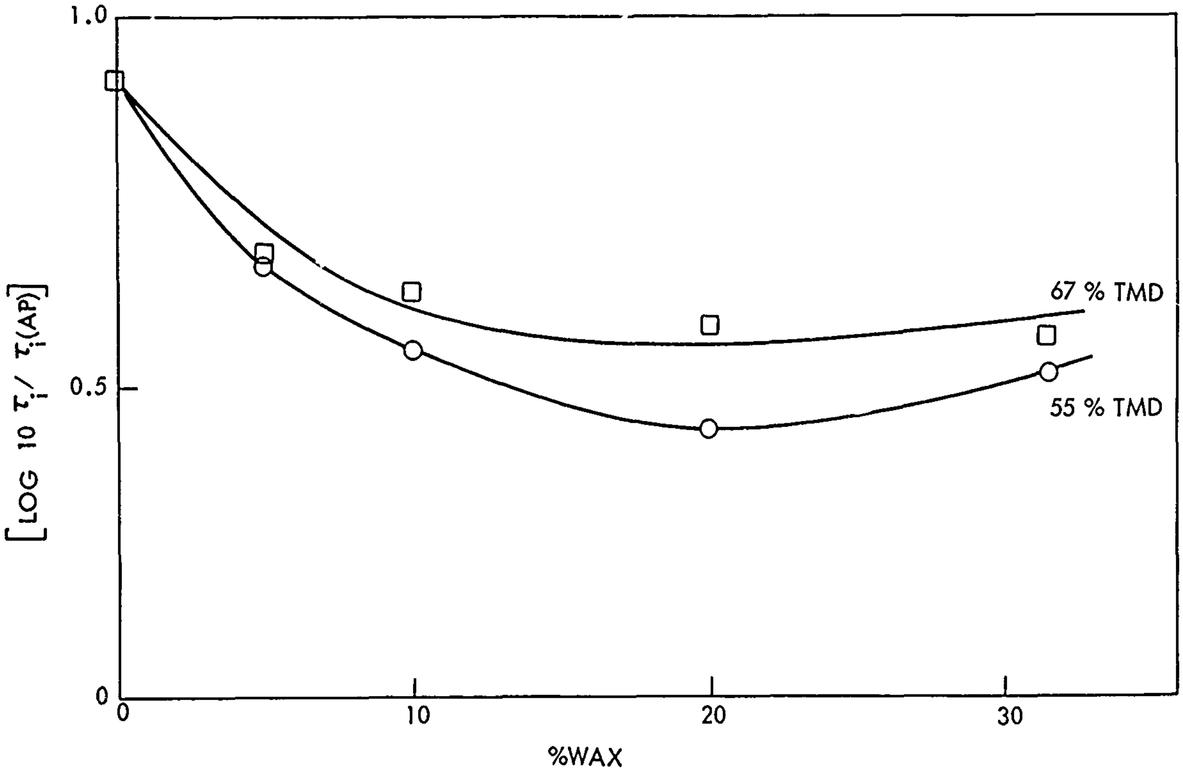

FIG. 15 EFFECT OF WAX ON REACTION TIME OF WAXED AP RELATIVE TO UNWAXED

reasonable trend of "a" with ρ_o, confirm that found earlier (and with different D_i values),[2] and provide approximate numbers to use in Eq. (5). The relative reaction rates thus computed are listed in Table 15 and plotted on a semi-log scale in Figure 15. The addition of wax to AP reduces the reaction time to about 0.3 and 0.4 its initial value at 55 and 67% TMD, respectively. These are maximum effects and occur at about 20% wax.

In a previous report[2], we compared reaction times for the <u>same</u> reaction at the <u>same</u> temperature, but at different grain sizes. Here we are comparing reaction times at nearly the same grain size (N126 and N127 do not differ greatly) but at <u>different</u> temperatures and quite possibly for different reactions, e.g., a diffusion controlled reaction between wax vapor and AP decomposition products compared to simple AP decomposition. The latter reaction follows an Arrhenius law and decomposes with the rate $Ze^{-A/RT}$. Hence its relative rate at two <u>different</u> temperatures is

$$\frac{\tau(T_1)}{\tau(T_2)} = e^{A/R(1/T_1-1/T_2)}$$

A frequently used activation energy A for AP is 20 to 22 kcal/mole. With this value, $T_1 = 2000^oK$ (maximum estimate for T_j of AP) and $T_2 = 2686^oK$ (maximum T_j computed by Ruby for waxed AP)

$$\frac{\tau(2000^oK)}{\tau(2686^oK)} = 0.28 \text{ to } 0.25$$

In other words a 686°K rise in temperature from 2000°K will decrease the decomposition time of AP to just about the extent addition of 20% wax decreases the resultant reaction time.

As Figure 15 shows, wax is less effective in reducing the reaction time τ at higher than at lower % TMD. The 67% TMD curve also shows more scatter and a less marked minimum than does the 55% TMD curve for the higher porosity charges. This may be fictitious in that it could be caused by the same difficulty in extrapolation of data from the waxed charges that occurred for the unwaxed AP. Certainly the results at 55% TMD seem better.

TABLE 15 - Reaction Times of Waxed AP
Relative to Unwaxed AP

% Wax	D_i [a] mm/µsec	$\tau_i/\tau_i(AP)$ [b]
	55% TMD	
0	3.90	1.0
5	4.27	0.495
10	4.57	0.362
20	4.79	0.271
31.5	4.40	0.331
	67% TMD	
0	4.51	1.0
5	4.88	0.521
10	5.17	0.452
20	5.38	0.396
31.5	4.89	0.382

a. From smoothed data of Figure 9.

b. Computed from Eq. (5) with a values for 55 and 67% TMD, respectively: 16.5 and 19 mm for AP (N126) and 19 and 22 mm for AP (N127). See Figure 14.

Data for AP(N119)/Wax, 90/10, have been included in Tables 5 and 9. N119 is a coarse material of about 200μ average particle size. Unwaxed, it will not detonate at its pour-density in a 7.62 cm diameter. The data of Table 5 show that the addition of 10% wax makes this material detonable at 1.073 g/cc (the pour density of the mixture) or 60.3% TMD. Extrapolation of the two points at d of 6.35 and 7.62 cm gives D_i(60.3% TMD) = 4.88 mm/μsec and a = 7.2 mm. The D_i value compares well to the interpolated one of 4.83 mm/μsec for AP(N126)/Wax, 90/10 (see Figure 9), but unfortunately we cannot interpolate the "a" values for this mixture. At 55 and 67% TMD, they were, respectively, 7.0 and 9.2 mm. It seems likely that the "a" values of the two waxed AP's are much the same, i.e., that addition of the wax has largely eliminated the difference in "a" values resulting from the large difference in particle size of the two AP's. However, the calculation of "a" for the N119 mix involves the small difference of two large numbers and could easily be in error by a factor of 2 to 3.

Before leaving the subject of reaction times it is of interest to compare values for the AP/Wax, 80/20, with corresponding ones for TNT. For this purpose, we can use "a" values for TNT (determined as in the present work) midway between those of the confined charges of Reference (12) and the unconfined charges of Reference (19); the particle size range of the TNT seemed much the same (70-200 μ) in these two investigations, but the particle size distribution curves were probably different. Thus the values of 3.5 and 2.3 mm were chosen for 55 and 67% TMD (0.91 and 1.11 g/cc). The corresponding D_i are given in Table 11, but since TNT and 80/20, AP/Wax have the same D_i values, Eq. (5) reduces to

$$\tau_i(80/20)/\tau_i(\mathrm{TNT}) = a(80/20)/a(\mathrm{TNT}).$$

Hence the waxed AP has reaction times 1.6 and 4 times greater than those of TNT at 55 and 67% TMD. This is one basic difference between the two explosives and could result from the slowness of the diffusion processes necessary for the AP/Wax reaction. This means that the low porosity range (higher % TMD) of the AP/Wax may, in some cases, not provide a practical explosive.

The "a" values of 80/20, AP(N126)/Wax, are very near those of AP(XP-17). In other words, addition of 20% wax to a 25μ AP results in an "a" value approximately equal that of a 10μ AP. If wax added to a 10μ AP has the same relative effect as its addition to a 25μ AP, the resultant "a" value of the 80/20 mix would be lower than that of TNT at 55% TMD but still above that of TNT at 67% TMD. Hence a very fine grained AP/Wax might exhibit a reaction time equal to or less than that of TNT at the lower % TMD. If so, the AP must have an average particle size of 10μ whereas that of the TNT might be about 125μ.

Detonability

The failure limit data in the d-ρ_0 plane appear throughout the tables. They are summarized in Table 16, References (2) and (3) for AP(N126), and Reference (20) for TNT. They are plotted in Figure 16 which gives a number of comparisons. Addition of 10 or 20% wax to 25μ AP lowers its d_c by a factor of 5 or more. The limit curves of the 90/10 and 80/20 mixtures are indistinguishable up to 70% TMD above which the 80/20 shows a smaller d_c. Addition of 10% wax to 200μ AP lowers its d_c from > 76 mm to about 40, or that of the 25μ AP at 60% TMD, but does not comparably lower the d_c value at 67% TMD. The AP particle size effect on the 90/10 mixtures is qualitatively that found for pure AP[2,3], i.e., it shifts the failure curve toward lower % TMD for increasing particle size. Thus for the 200μ AP, we find the steep portion of the 90/10 detonability curve at 60-67% TMD whereas for the 25μ AP it occurs at 76-82% TMD. Both the 90/10 and the 80/20 mixtures with 25μ AP have $d_c \leq$ that of TNT (70-200μ) at a % TMD $\leq$ 64. At lower porosities their d_c is larger than that of the TNT.

Gor'kov and Kurbangalina[21] have published an interesting study of the detonability of AP. They showed the effect of particle size, water content, temperature, and small amounts of fuel on the limit curve d_c vs ρ_0. In particular, working at about 66% TMD, a particle size of about 50μ, and glass confinement, they found that raising the initial temperature from 25° to 200°C lowered d_c from 23 to 12 mm. Moreover, adding either 0.9% carbon black or 2.4% RDX lowered the d_c at 25°C from 23 to 14-15 mm. At 200°C these mixtures had a d_c of

TABLE 16 - Summary of Detonability Data*

Material	AP	ρ_o g/cc	%TMD	d(cm)		d (cm)	ρ_o		% TMD	
				+	-		+	-	+	-
AP/Wax	N126	0.901	50.6		0.64					
90/10	25µ	0.907	51.0	0.95						
		1.212	68.1		0.64					
		1.202	67.5	0.95						
						5.08	1.447	1.516	81.3	85.2
	N119	1.071	60.3		3.49					
	200µ	1.075	60.4	5.08						
		1.201	67.5		7.62					
80/20	N126	0.916	55.8		0.64					
	25µ	0.934	56.9	0.95						
		1.105	67.4		0.95					
		1.103	67.3	1.27						
						5.08	1.424	--	86.8	--

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* From Reference (2) and Tables 5 and 6

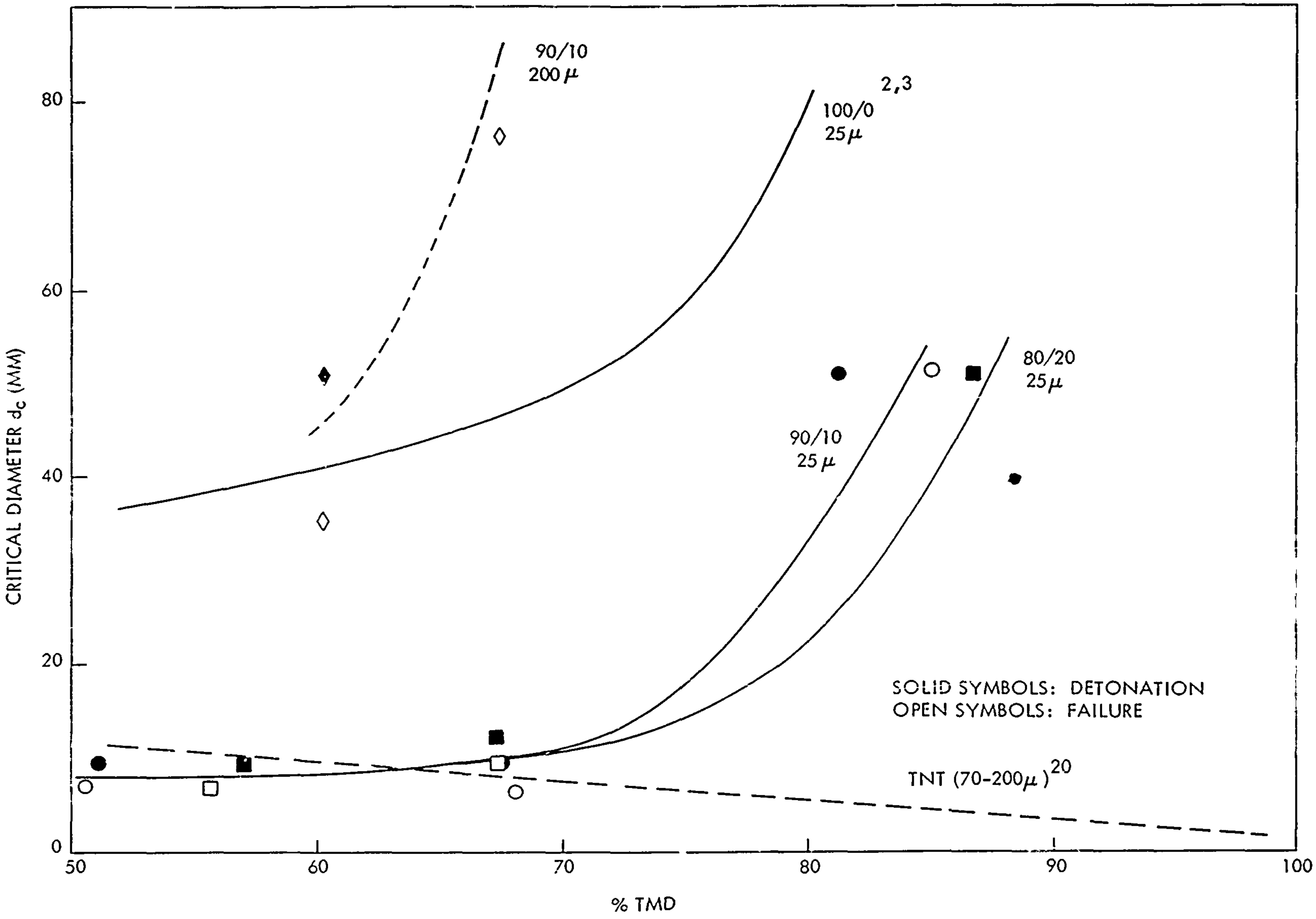

FIG. 16 DETONABILITY CURVES FOR AP/WAX AND TNT

11-12 mm. They concluded that the addition of fuel increased the reaction temperature and thereby decreased reaction time and d_c. This seems a reasonable conclusion, as far as it goes, and is equally applicable to our results.

Shock Sensitivity

Shock sensitivity data obtained on AP/Wax mixtures in the NOL Large Scale Gap Test are listed in Table 17. They, together with the analogous data for AP(N126)[2] and TNT[22], are plotted P_g vs % TMD in Figure 17. It is evident that addition of wax sensitizes AP to shock; it lowers the sensitivity curve and extends it to the right. The extension results from the extension of the detonability range to higher % TMD (Figure 16). As in the case of detonability, 90/10 and 80/20 mixtures of the 25μ AP are indistinguishable at lower % TMD; at higher % TMD the 80/20 mix remains shock sensitive up to a higher % TMD than does the 90/10. These shock sensitivity curves have been made vertical at the % TMD which fits the data and is also in accord with observed reaction limits. These reaction limits are indicated by vertical lines at the top of the figure: a thin line indicates that a shock-induced reaction less vigorous than detonation was observed at that density; a thick line, that no shock-induced reaction could be observed. Dead pressing for the conditions of the gap test occurs at a density slightly lower than that shown by the first limit (thin line).

The 200μ AP/Wax, 90/10, is only slightly (possibly insignificantly) less sensitive than the 25μ AP mixes at 79-86% TMD despite its vast difference in detonability. This indicates that both 90/10 mixes show about the same ease of ignition by shock, although the unconfined 200μ mix is much less able to propagate detonation than is the 25μ one. This implies about the same activation energy for ignition of the two 90/10 mixes, and little effect of available AP surface area on hot spot ignitions.

Finally, the waxed mixtures show a sensitivity curve very close to that of TNT; the 80/20 mix is comparably sensitive up to 90% TMD where it diverges because of its approach to the dead pressed condition. Under proper confinement, it is possible that this mix could replace TNT as a practical explosive. (Pressed TNT exhibits a

TABLE 17 - Shock Sensitivity of AP/Wax Mixtures

AP/Wax	AP	ρ	%TMD	50% Values No. Cards	 P_g kbar	Comment
90/10	N126	1.67	93.8	0*	Neg.	No reaction
	(25µ)	1.63	91.8	59*	70	Reaction but probably no detonation
		1.60	89.3	110-125*	45.6-40.0	
		1.52	85.4	186	20.2	
		1.32	74.3	225	13.7	
		1.08	60.7	240	12.1	
80/20	N126	1.57	95.6	0*	Neg.	No reaction
	(25µ)	1.54	93.9	0*	Pos.	Reaction
		1.52	92.7	0*	Pos.	Reaction
		1.50	91.5	178*	22.1	
		1.46	89.0	194*	18.5	
		1.39	84.6	213	15.2	
		1.23	75.0	236	12.5	
		1.00	61.0	236	12.5	
90/10	N119	1.53	86.3	175	22.9	
	(200µ)	1.42	79.4	199	17.5	

* Extended test, cast Comp B witness

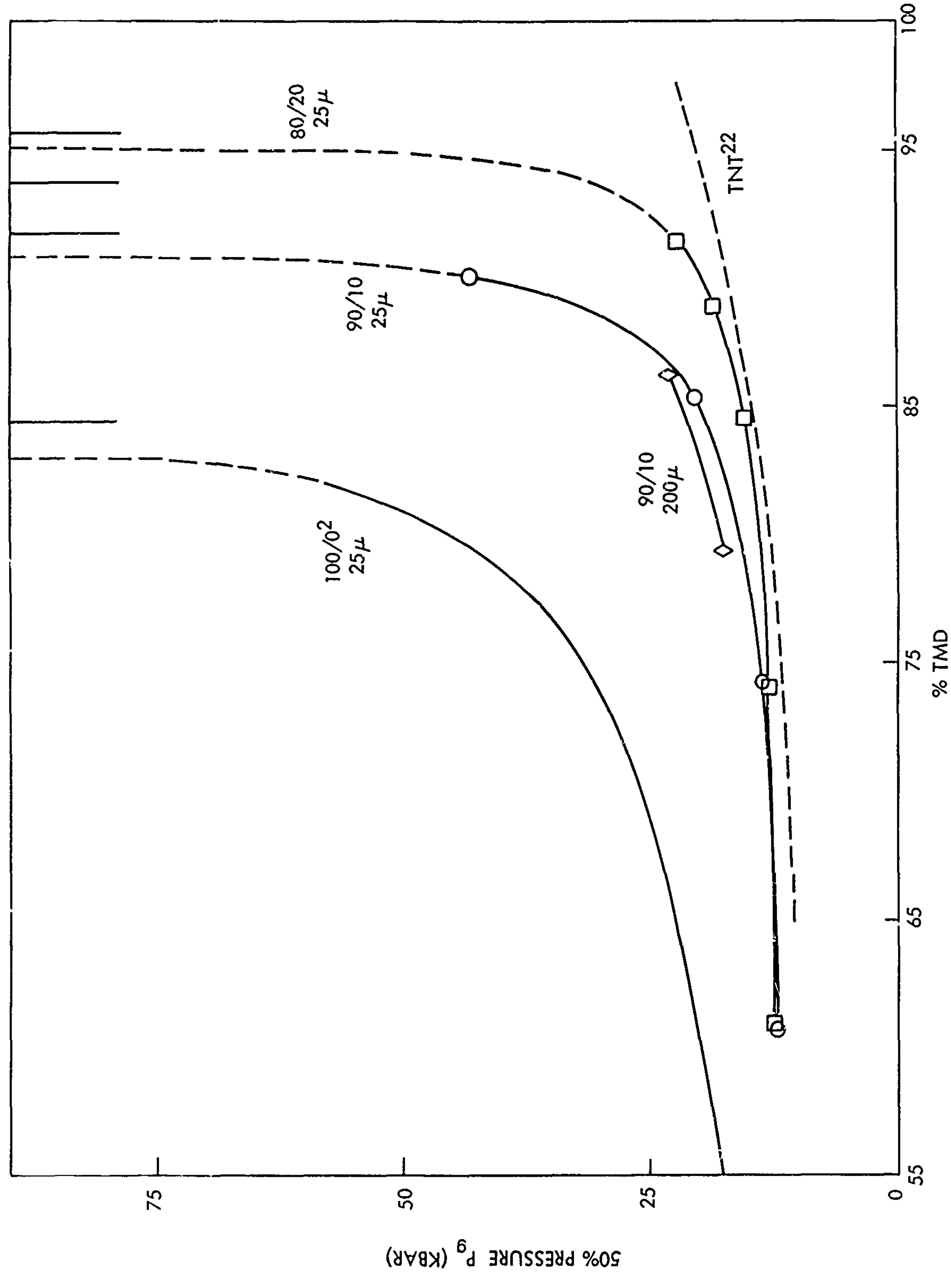

FIG. 17 SHOCK SENSITIVITY CURVES FOR AP/WAX

dead-pressing phenomenon in this geometry only if it is precompressed. However non-detonable TNT charges of these dimensions can be prepared by slow cooling of a cast charge.) The curves of Figure 17 in conjunction with the other explosive characteristics of the 80/20 mix also demonstrate the potential detonability of simple composite explosives under appropriate conditions. Although the wax is probably a more volatile fuel than the common propellant matrices, a small amount of carbon black, which is less volatile, has a similar effect in increasing detonability[21] and in increasing sensitivity[23].

Information from Burning Rate Studies

The use of arbitrary decomposition equations or Ruby Code computations and the assumption of a single-stage equilibrium reaction is a gross simplification of very complex behavior. It is justified only insofar as it offers guidance in planning experimental elucidation of the processes and predicts good relative ratings. Thus the maximum effect of wax was correctly indicated in the present work (though not the concentration required to produce that effect).

AP and its mixtures have been studied for many years as propellants. Recently two exhaustive reviews[25,26] of such work have appeared. They exhibit striking similarities between the results of deflagration studies and those of detonation studies, such as ours on AP/Wax. Successful propagation (and its failure) in burning are governed chiefly by transport processes, whereas detonation and its failure limits are determined chiefly by hydrodynamic phenomena; we can therefore interpret the failure diameter in deflagration as a heat loss by conduction, convection, or both, and the failure diameter in detonation as a heat loss by reaction quenching, caused by lateral rarefaction waves. In both cases, the dominant factor is the relative energy loss rather than the mechanism whereby it occurs. It seems very likely that the chemical reactions are the same in both burning and detonation although the products and rates will reflect the different pressure and temperature ranges. Consequently, a brief review of the available information from the burning rate studies seems indicated.

Factors affecting the burning rate of AP are charge diameter, density, particle size, confinement, initial temperature, and initial pressure. In general, these variables affect the burning rate and the detonation rate in the same way. There are a few situations for which there is no parallel in the two fields, e.g., initial pressure effect. However, many trends are the same, and deflagration, like detonation, is a multidimensional effect. Relatively few failure limit studies are available in either field.

AP is a somewhat exceptional propellant as well as an unusual H.E. Its linear burning rate decreases with decreasing % TMD whereas that of the common organic H.E. and of mixtures (AP/fuel) increases.[26] It is now generally accepted[25] that the initial step in its thermal decomposition is the sublimation-dissociation reaction*

$$NH_4ClO_4 \rightleftarrows NH_3 + HClO_4 \qquad \Delta H = 58 \text{ Kcal/mole} \qquad (6)$$

with an activation energy of about 32 kcal/mole. The reaction sustaining steady burning (and producing the flame) is then the gas phase oxidation of NH_3 by $HClO_4$. DTA studies reveal an endotherm at 240°C (crystal transition from orthorhombic to cubic) and two exotherms at about 300°C and 440°C corresponding to "low" and "high" temperature decomposition.

AP will not exhibit steady burning at 20-25°C and 1 atm. However, if it is preheated or if a small amount of volatile fuel is present, it will burn, and then, under comparable conditions, its burning rate is higher than that for pure AP. However, the amount of fuel is critical; very small amounts can decrease the rate or increase the critical diameter for burning[26].

There are numerous models for AP/fuel mixtures. Many suggest an oxidizer flame supplying heat to vaporize the fuel and thus provide a diffusion flame, e.g., $HClO_4$/fuel. Since addition of fuel increases the burning rate, some energy from the diffusion flame must be fed back to the solid mix although it may serve only to increase the rate of the reaction of Eq. (6). The reactions controlling the burning rate are completed near the solid surface, e.g., within 200μ, but chemical reaction may go on several mm beyond the surface.

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* This is a multi-step reaction and the rate determining step is not clear.

This leads to a concept of "zone of influence" which includes only the portion of the total reaction which can affect the burning rate. Thus the final flame temperature (measured by thermocouples) may be higher than and some distance downstream from the temperature at the beginning of the reaction zone.

The necessary preheating for steady state burning at 1 atm. has been reported as initial temperatures of 200°C[26] and 280°C[25]. The latter, for a specific heat[25] of 0.309 cal gm^{-1} deg C^{-1} amounts to 87.5 cal/g energy supplied to the AP. (The minimum preheating by radiation gives the value 95 cal/g[27].) The minimum fuel to effect the same result seems to be 3.85% paraformaldehyde or 2.6% metaldehyde, equivalent to 107 cal/g extra heat[27]. In this case, the extra energy must not only initiate the reaction of Eq. (6) but also vaporize the fuel.*

It seems clear in the case of our AP/Wax mixtures that the AP can decompose exothermally at about 300°C, which is below the fire point of the wax. For a given thermal imput for initiation, the vapor phase will probably be richer in oxidizer than in fuel. This difference in concentration of fuel in solid and vapor phases could account for the maximum effect at a fuel concentration in the solid mixture approximately twice the stoichiometric. The requirement of excess fuel (presumed necessary to keep composition of the gas phase in the "zone of influence" constant and corresponding to maximum rate of reaction) is commonly observed in burning rate studies. For volatile organic fuels, the maximum effect on burning rate generally lies between 20 and 30% excess fuel over the stoichiometric. But greater excesses have been reported, and, in particular, a homogeneous premixed flame of $HClO_4/CH_4$ exhibits two distinct flame fronts at atmospheric pressure and at concentrations on the fuel rich side of stoichiometric[28]; the second flame front is 100-150°C hotter than the first and seems to be essentially a CO flame. The maximum burning velocity for this gas mixture occurs at a concentration considerably on the fuel-rich side of the stoichiometric. This is apparently

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* The minimum flame temperatures observed in steady-state burning at 1 atm were 970°C (preheated AP) and 1000°C (AP/fuel)

associated with the lag of the oxidation of CO to CO_2 behind the other reactions of burning; a similar lag in detonation reactions might not be entirely eliminated by the higher pressures.

Variables affecting the burning rate of AP/fuel mixtures are, of course, the same as those determining the burning rate of pure AP with the addition of the concentration as a variable for the mixture. Adams and his coworkers studied burning rates as a function of pressure, mixture composition, and particle size. They concluded that the dependence of the rate on any one of these variables was affected by the value of the other two; that there was a complex dependency and not a simple, separable effect. These conclusions seem to be generally accepted[25,26].

In conclusion, it seems well established that burning of AP/fuel is a multidimensional, multistep, multistage process, dependent on both kinetic and diffusion factors. We know that detonation of AP/fuel is a multidimensional process, as all detonations are, and must depend on diffusion processes as well as kinetic since adding a volatile fuel increases the D_i value above that of pure AP. There is every probability that detonation is also a multistage process and quite as complicated as deflagration.

SUMMARY AND CONCLUSIONS

Addition of wax to AP results in a large increase of D_i, and a D_i vs % wax curve with a broad maximum at about 20% wax. The maximum increase $\left[D_i(\text{AP/Wax})-D_i(\text{AP})\right]$ is about that computed on the Ruby code although the absolute values of the code are about 0.7-0.9 mm/µsec too large. The computations also show a maximum at a stoichiometric composition of less than 10% wax. On the other hand, Ruby code computations correctly predict both absolute D_i values and the trend for RDX/Wax, i.e., monotonic decrease in D_i with increase in % wax. The effect of wax on the D_i of AP is attributed to the reaction between the volatile fuel and the detonation products of the AP. The observation of maximum effect at a concentration of wax approximately twice the stoichiometric is believed caused by the kinetics of the diffusion processes necessary in the oxidation-reduction reaction.

Determination of the effect of wax on D_i and reaction time was at 55 and 67% TMD, but detonability curves and shock sensitivity measurements were carried to as high a % TMD as our facilities permit. The results will be illustrated by data for the AP/Wax, 80/20 mixture which showed the largest effect of wax on each explosive characteristic studied. At 55% TMD the wax reduced reaction time (curved front theory) by a factor of 3.7. It increased detonability at d = 5.08 cm from a critical value of 71% TMD to 88% TMD; these determinations were on unconfined charges. In the confinement of the gap test, wax shifted the point of dead-pressing from about 83% TMD to about 96% TMD; it also increased sensitivity (lowered P_g) over the range of detonability. The AP/Wax, 80/20, mix is most similar to pressed TNT. It has about the same voidless density, the same D_i at the same % TMD, approximately the same d_c at % TMD $\leq$ 70, and approximately the same shock sensitivity curve at % TMD $\leq$ 90. It differs from TNT chiefly in exhibiting greater reaction time and lower detonability at high % TMD.

These results show very clearly that a simple composite (AP/organic matrix) is potentially detonable. This was expected since a composite (AP/matrix/Al in which the matrix contained no oxidizing groups or explosive substances) has been detonated at essentially voidless density and d = 72 in. However, the present data offer detailed information on how the point of detonability is approached and what explosive behavior to expect of granular mixes prior to their compaction to voidless density.

APPENDIX

USE OF AN EXPONENTIAL FUNCTION FOR EXTRAPOLATION

Because we had some initial success in fitting the data for the 90/10 and 80/20 AP/Wax mixtures to an exponential function[24], all data were fitted to the curve

$$D = D_i \left[1-(a/d)^K \right] \qquad (1)$$

as well as to its linear form (K = 1). It is evident that the linear form cannot fit the data over the whole range of d, e.g., see Figure 4.

As indicated in the text, this project was unsuccessful. To be sure, it fit all the data about as well as a straight line fit the data for larger diameters. The results are given in Table A1 and plotted in Figure A1. (Compare with Figure 9 of text.) But these results cannot be reasonably interpreted and they are, in many cases, much higher than the maximum computed D_i values from Ruby. We know that Ruby values are much too high for AP, and assume that this might also be the case for AP/Wax.

Finally, the extrapolated values are supersentitive to the number of data points considered. For example, the 90/10 mix at 52.8% TMD, has an extrapolated value of 5.58 mm/µsec when all 8 points are used and of 4.27 mm/µsec when only 4 are treated (in the same handling, the exponent K changes from 0.38 to 1.35!). Despite the fact that a straight line cannot be expected to fit all points well (see Figure 4), the linear fit of 8 points gives 4.21 mm/µsec compared to 4.46 for 4 points.

For these reasons, the use of Eq. (1) ($K \neq 1$) to extrapolate to D_i cannot be justified even by the empirical arguments used for the linear treatment of D vs d^{-1}. The latter, on the other hand, has been most successful in the present work in producing consistent data which can be interpreted, at least qualitatively, in terms of chemical reaction.

TABLE A1 - Data Fitted to $D = D_1 \left[1 - (a/d)^K \right]$

AP/Wax	AP	ρ_o	%TMD	Range Used in d, cm	D_1 mm/μsec	K	q.m.e.	
95/5	N127	1.025	55.1	1.9 - 7.6	3.84 (2.6%)	1.650 (13.5%)	0.0542	All points (6)
				2.5 - 7.6	4.26 (2.6%)	0.891 (11.0%)	0.0113	5 points
90/10	N126	0.940	52.8	0.95 - 7.6	5.58 (16.8%)	0.382 (41.3%)	0.0737	All points (8)
				2.5 - 7.6	4.27 (11.2%)	1.347 (86.8%)	0.0875	4 points
80/20	N126	0.901	54.9	0.95 - 7.6	5.05 (1.9%)	0.703 (7.3%)	0.0326	All points (13)
				1.9 - 7.6	5.16 (6.4%)	0.631 (29.4%)	0.0359	11 points
				3.5 - 7.6	6.38 (57.3%)	0.303 (169%)	0.0291	8 points
68.5/31.5	N127	0.826	55.1	2.5 - 7.6	4.53 (5.8%)	0.867 (30.7%)	0.0256	All points (5)
95/5	N127	1.250	67.2	2.5 - 7.6	7.58 (21.2%)	0.306 (39.0%)	0.0200	All points (5)
				3.5 - 7.6	5.69 (8.9%)	0.597 (24.7%)	0.0115	4 points
90/10	N126	1.200	67.5	0.95 - 7.6	6.70 (3.4%)	0.345 (7.0%)	0.0144	All points (8)
				5.1 - 7.6	6.96 (227%)	0.322 (594%)	0.0283	4 points
80/20	N126	1.100	67.1	1.3 - 7.6	5.93 (1.7%)	0.585 (5.0%)	0.0146	All points (8)
				1.9 - 7.6	5.16 (6.4%)	0.631 (29.4%)	0.0359	7 points
				5.1 - 7.6	Cannot fit data			4 points
68.5/31.5	N127	1.006	67.1	2.5 - 7.6	4.87 (0.5%)	1.026 (2.3%)	0.0875	All points (4)

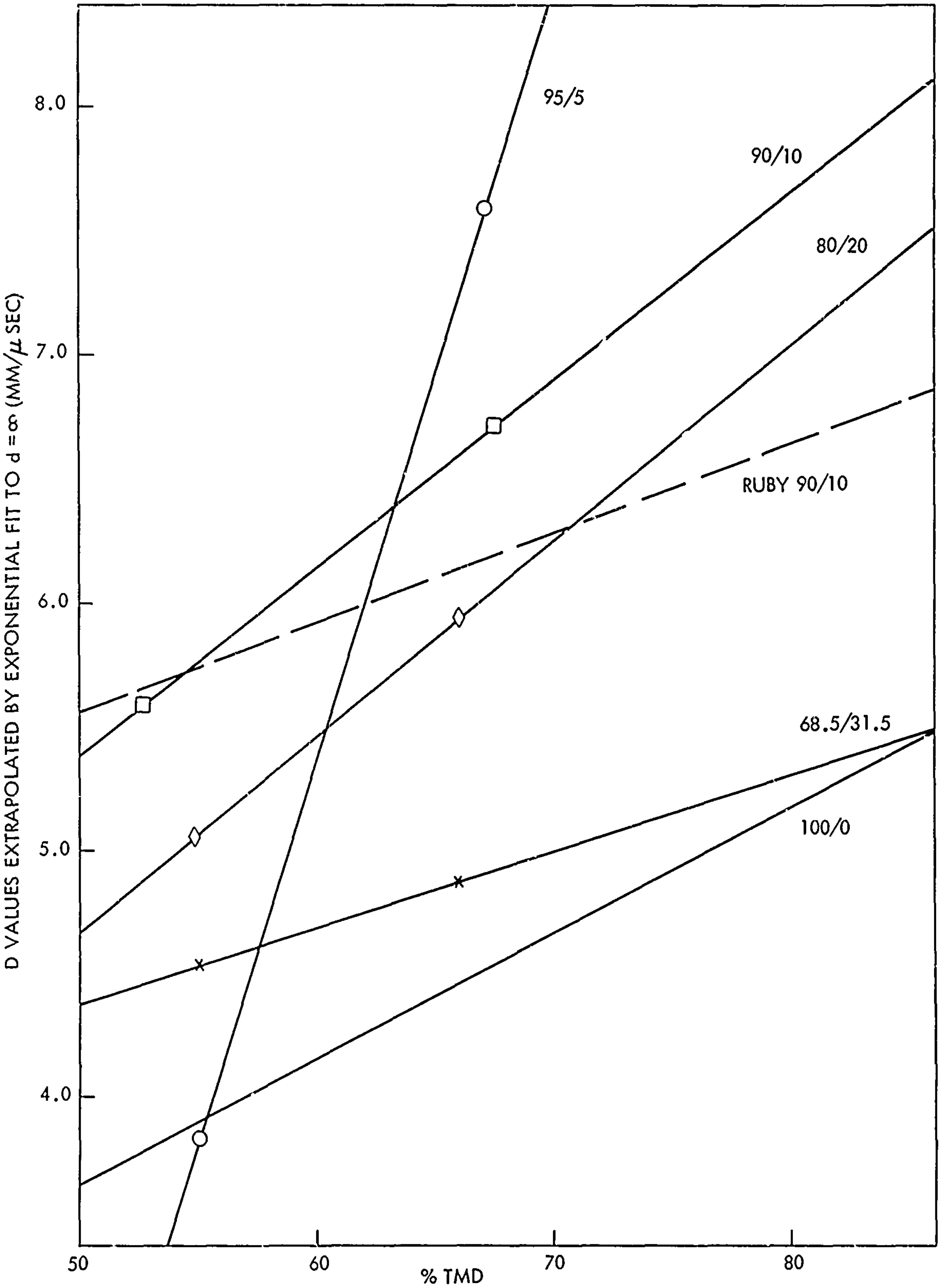

FIG. A1 EFFECT OF EXPONENTIAL EXTRAPOLATION OF VALUES FOR AP/WAX MIXTURES

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AP/Wax mixtures were used to model a simple composite propellant. Addition of wax to the AP increased its infinite diameter detonation velocity, increased its shock sensitivity, decreased its critical diameter and its reaction zone length. The maximum of these effects occurred at about 20% wax. At this composition, the model is an explosive comparable to TNT.

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Detonation velocity						
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