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Development of Sustainable, Epoxy-Bound Mg/$NaNO_3$ Compositions for the U.S. Army's 40 mm Yellow Illuminant Flares

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ABSTRACT: We report here on the development of novel yellow-emitting pyrotechnic flares based on ground magnesium, sodium nitrate ($NaNO_3$), and a two-part epoxy thermoset. This work was aimed to eliminate the chemical exposure risk posed by the toxic and solvent-based polyester thermoset specified for two of the U.S. Army's 40 mm yellow illuminant flares, the M585 star cluster and M583A1 star parachute signals. In particular, we describe efforts to develop a common replacement composition for both flares based on a two-part epoxy thermoset free of volatile organic chemicals (VOCs) or phthalate plasticizer. In addition, for the M583A1, we compare static burn performance of flares encased in Viton-laminated phenolic tubes to those in unlaminated tubes. A role for the Viton laminate is also proposed.

KEYWORDS: *Formulation, Thermoset, Illuminant, Epoxy, Lifecycle risks, Laminate*

INTRODUCTION

To ensure proper training and readiness of soldiers, it is critical to mitigate lifecycle risks posed by the chemicals that comprise modern weapon systems and ammunition.[1−11] One particularly common problem suffered by most legacy U.S. military pyrotechnic flares is the Laminac 4116/Lupersol DDM binder system.[12] In this system, Laminac 4116 is an acetone solution of a polyester resin and styrene monomer, whereas Lupersol DDM is a solution of methyl ethyl ketone peroxide (MEKP) and dimethyl phthalate. After mixing at ambient temperature, this two-part system cures by the chemical cross-linking of polyester strands with styrene to form a hardened thermosetting polymer. The resulting polymer matrix functions to promote homogeneity between the fuel and oxidizer and to impart rigidity to a flare after consolidation.[13,14]

Despite the advantages of using Laminac/Lupersol in flare assembly, there are numerous lifecycle risks posed by both parts of this binder system. First, the volatile acetone fumes emitted by the Laminac solution prove objectionable during signal production at the typical batch size (~54.48 kg of flare composition). In addition, the styrene[15] present in Laminac 4116 and the dimethyl phthalate[16] in Lupersol DDM have been characterized as unacceptably toxic. Also importantly, the Laminac 4116 solution is a supply chain bottleneck as its commercial availability is unreliably cyclical and limited to usually only one supplier.

In light of these problems, our group at the Armament Research, Development and Engineering Center (ARDEC) has made significant advances recently to replace the Laminac/Lupersol platform with a commercially available epoxy/polyamide binder system (Epon 813/Versamid 140)[17] for several illuminant flares. This new reagent combination comprises an alternate two-part thermoset that does not pose any of the above-mentioned lifecycle risks suffered by the Laminac/Lupersol system. Moreover, it has been implemented for many other legacy Army/Navy pyrotechnics with favorable long-term aging results.[18−21]

With a suitable binder replacement in mind, we turned our attention to two of the U.S. Army's yellow illuminating signals currently configured in the polyester platform: the M585 star cluster and the M583A1 star parachute signals (Figure 1). Both of these signals are part of the 40 mm product line but have some key geometric differences. First, each M585 unit has the same diameter as the M583A1 but is much shorter. This is because five packed M585 cluster tubes are typically stacked into the signal hardware without a parachute, whereas the M583A1 has only one full-sized pellet but is fixed with a parachute. In addition, the M585 has a hollow core to allow for radial propagation at all exposed surfaces (core-burning), whereas the M583A1 has a solid core useful for linear propagation (end-burning). Lastly, the M585 cluster is encased in an unfinished Kraft cardboard tube, whereas the M583A1 is encased in a phenolic resin tube having a Viton-laminated inner diameter. The Viton laminate for the M583A1 introduces significant labor costs since it must be applied manually to each tube. Thus, developing a prototype with an unlaminated

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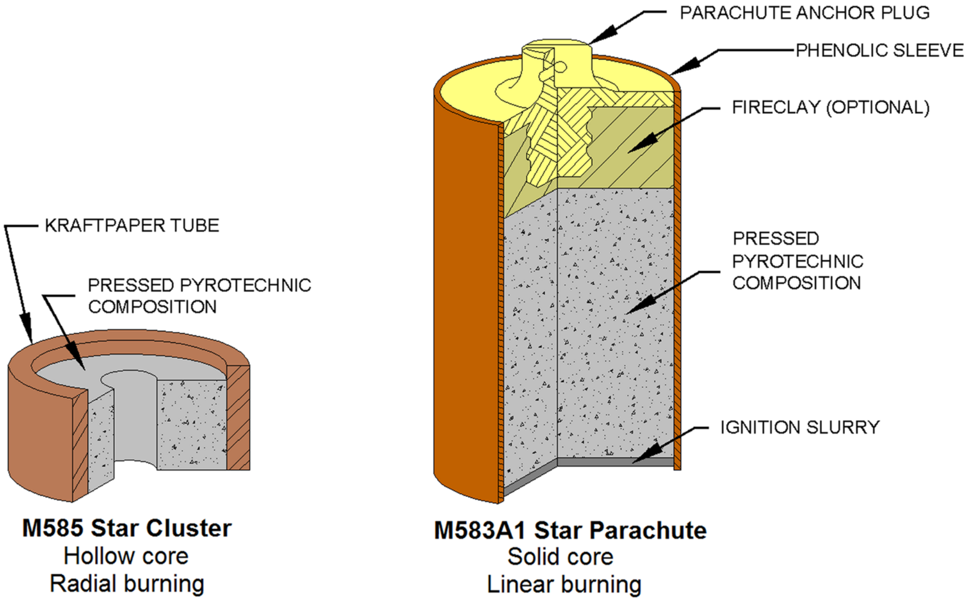

M585 Star Cluster
Hollow core
Radial burning

M583A1 Star Parachute
Solid core
Linear burning

Figure 1. Cut-away drawings of the 40 mm white M585 and M583A1 illuminant signals (not to scale).

phenolic tube would present additional lifecycle cost savings (roughly $4 US per signal!).

Despite the obvious geometric differences between the M585 and M583A1 signals, both employ very similar chemical compositions to display a yellow visible signature upon burning (Table 1). Here both of the compositions are unique from other illuminants recently studied,[18−22] owing mainly to the absence of a chlorine donor. In fact, the dominant emission exhibited by the sodium atom renders any chlorine donor unnecessary.[23] Also different from our previous work, the fuel content in both compositions is split between two different cuts of magnesium powder, a coarse cut corresponding to the 30/50 sieve size range, along with a fine cut corresponding to the 50/100 range. This is probably done to balance the intensity and duration of the resulting flame.[24] Interestingly, the two compositions differ in the overall fuel-oxidizer ratio (~7/6 for the M583A1 and ~2/3 for the M585) and in the total binder content (11 wt % for the M583A1 and 7.5 wt % for the M585). In light of the differences between the M585 and M583A1 compositions, combined with the dreaded binder problems, ARDEC was compelled to develop a common epoxy-based composition that meets the requirements of both signals. An added objective of this program was to use unlaminated M583A1 phenolic tubes, instead of the expensive Viton-laminated tubes, to economize further our epoxy-based prototype for this signal.

Table 1. Chemical Composition of the Visible Illuminant Charges for the M583A1 Parachute Signal Flare and M585 Star Cluster

	percent composition by weight				
composition	$NaNO_3$	Mg 30/50	Mg 50/100	Laminac 4116	Lupersol DDM
M583A1 control	41	28	20	10.45	0.55
M585 control	56.5	18	18	7.125	0.375

RESULTS AND DISCUSSION

The initial task was to prepare a range of compositions based on ground magnesium, granular $NaNO_3$, and 4:1 Epon/Versamid, followed by consolidation of each composition into both form factors (cluster and tube) and then static burn testing in parallel with both of the baseline compositions. Table 2 shows three families of compositions (**A**−**C**, **D**−**F**, and **G**−**H**), each set at a different oxidizer level: 40, 50, and 60% by weight. Here, the binder content for all three families was set at 10 wt %, the oxidizer content within a family was set at one of the three above-mentioned values, and the remaining balance was partitioned between two different cuts of magnesium powder, 30/50 ellipsoidal and 50/100 atomized, as they appear to be most commonly used in military yellow illuminants. This design was implemented to probe the tolerance of performance (burn time and intensity) to metallic particle size; it was hypothesized that as the content of the coarser magnesium cut was ramped up across a family, the burn time would be extended while the luminous intensity would correspondingly decrease. This hypothesis was grounded in the well-known correlation that exists between particle size/morphology and performance.[23]

Table 2. Chemical Makeup of Epoxy Replacement Compositions A−H

	percent composition by weight[a]		
mix ID	$NaNO_3$	Mg 30/50	Mg 50/100
A	40	10	40
B	40	30	20
C	40	50	0
D	50	10	30
E	50	25	15
F	50	40	0
G	60	15	15
H	60	30	0

[a] Weight balance for compositions **A**−**H** was 10% Epon 813/Versamid 140 (4:1 mix ratio).

Table 3 below shows the results of static performance testing of compositions **A**−**H** relative to those of the control compositions for the cluster and tube configurations. Note especially that there are two rows of data for the tube format, one for flares encased in Viton-laminated tubes and the other for flares in unlaminated tubes. Interestingly, the baseline composition for the M583A1 flare showed a very drastic performance difference between these two configurations. In a Viton-laminated tube, the flare burned for the requisite 40 s approximately, but in the unlaminated tube the entire flare was consumed in less than 18 s! Also surprising was the performance of our epoxy-based candidates, none of which was able to meet the performance metrics of both control compositions. Although close inspection confirms the trend of increased burn times with increased coarse magnesium content in both configurations, the right balance between burn time and intensity was not obtainable by any composition within **A**−**H**. Furthermore, it is noteworthy that the specifications of the M585 cluster (average 30 000 cd over 6.0 s) were met by compositions **D**−**F** (all at 50 wt % $NaNO_3$), but no composition was capable of meeting the requirements of the M583A1 (average 70 00 cd over 35 s).

Table 3. Performance of Baseline Compositions and Replacements A−H in Native M583A1 and M585 Formats

composition	M583A1 performance[a] burn time (s)	M583A1 performance[a] luminous intensity (cd)	M585 performance[b] burn time (s)	M585 performance[b] luminous intensity (cd)
M585 control			7.17	54 414
M583A1 control[c]	39.91	97 108		
M583A1 control	17.62	120 716		
A	15.01	138 244	4.23	73 703
B	17.69	131 967	4.91	70 976
C	21.32	89 664	5.23	61 396
D	28.76	72 024	6.38	49 212
E	38.63	55 594	8.05	42 486
F	47.65	39 496	8.84	30 500
G	59.21	16 141	11.39	21 470
H	83.10	11 441	14.61	13 066

[a] M583A1 static test requirements: 30 000 candlepower minimum, 6.0 s burn time minimum. [b] M585 static test requirements: 70 000 candlepower minimum, 35.0 s burn time minimum. [c] Encased in Viton-laminated phenolic tubes; all other M583A1 samples encased in unlaminated tubes.

The performance differences between flares in laminated and unlaminated tubes may be explained by deducing a role for the Viton laminate specified for the M583A1 control. We propose that the laminate serves as a thermal barrier to propagation ahead of the propagation front; flares encased in unlaminated tubes suffer heat transfer down the column with a corresponding change in the burning mode from linear to radial. The Viton fluoropolymer lining the inner diameter of the laminated tubes, however, may block such heat transfer and prevent the change in burning mode. Nevertheless, this observation of performance dependence on inner diameter coating was held in consideration for future development, but the next iteration of development would reattempt to match the baseline performance in an unlined tube to economize our prototype.

Despite the insufficient burn times exhibited by the three families described above in the unlaminated parachute tube configuration, a second attempt at developing a common composition for both configurations was made (Table 4). This time the Epon/Versamid binder content was reduced to 5 wt % and the fuel was again partitioned between two different magnesium cuts, but the oxidizer was fixed at 45 wt % (intermediate between the first two families). In addition, the coarser magnesium content was increased at the expense of the fine going down the series. This approach was envisioned to improve on the performance exhibited by compositions **A**−**H**.

Table 4. Chemical Makeup of Compositions I−K

mix ID	percent composition by weight[a] $NaNO_3$	Mg 30/50	Mg 50/100
I	45	15	35
J	45	25	25
K	45	35	15

[a] Weight balance was 5% Epon 813/Versamid 140 (4:1 mix ratio) for compositions **I**−**K**.

With compositions **I**−**K** prepared, the time came again to compare the static burn test performance to the M585 and M583A1 (laminated and unlaminated) baseline configurations (Table 5). Notice again how the epoxy candidates smother out in less than 30 s in the unlaminated phenolic tube, but this time the composition change also hurt the performance of the cluster format. The 6 s burn time requirement for the M585 could not be met by any member of this series either.

Table 5. Performance of Baseline Compositions and Replacements I−K in Native M583A1 and M585 Formats

composition	M583A1 performance[a] burn time (s)	M583A1 performance[a] intensity (cd)	M585 performance[b] burn time (s)	M585 performance[b] intensity (cd)
M585 control			7.17	54 414
M583A1 control[c]	39.91	97 108		
M583A1 control	17.62	120 716		
I	18.12	160 137	4.55	119 851
J	17.88	153 298	4.80	113 915
K	21.80	141 281	5.63	98 509

[a] M583A1 static test requirements: 30 000 candlepower minimum, 6.0 s burn time minimum. [b] M585 static test requirements: 70 000 candlepower minimum, 35.0 s burn time minimum. [c] Encased in Viton-laminated phenolic tubes; all other M583A1 samples encased in unlaminated tubes.

As a result of the performance shortfall of compositions **I**−**K**, an additional series of compositions was to be developed and tested. This time, the combined approach to develop a common composition was abandoned and a different series of compositions was planned for each illuminant configuration. Moreover, the compositions planned for the M583A1 would be consolidated into Viton-laminated phenolic tubes only, since no range of compositions seemed to work for the unlaminated tubes. Accordingly, Table 6 below shows both the ingredient percentages and performance metrics of compositions **L**−**N**. Here, the Epon/Versamid binder content was increased to 7.5 wt %, the $NaNO_3$ content was fixed at 42.5 wt %, and the remaining balance was again partitioned between the two cuts of magnesium powder, with the coarse particle content increasing down the series. To our delight, composition **N** closely matched the performance of the baseline flare, burning

Table 6. Composition and Performance of Epoxy Replacements for the M583A1 White Parachute Signal

	percent composition by weight[a]			performance[b]	
mix ID[c]	$NaNO_3$	Mg 30/50	Mg 50/100	burn time (s)	luminous intensity (cd)
M583A1 control	41	28	20	39.63	78 270
L	42.5	15	35	32.83	119 124
M	42.5	25	25	35.31	101 346
N	42.5	35	15	39.71	89 825

[a] M583A1 baseline weight balance was 11% Laminac/Lupersol binder system; all others had 7.5% Epon/Versamid. [b] M583A1 static test requirements: 70 000 candlepower minimum, 35.0 s burn time minimum. [c] Encased in Viton-laminated phenolic tubes.

at >89 000 cd for almost 40 s! Lastly, the expected positive correlation between coarse fuel content and burn time was confirmed by this set of experiments.

With a valid prototype for the M583A1 flare in hand, we focused our attention on finalizing a separate composition for the M585 cluster. Our next attempt at meeting the performance requirements for this signal is described in Table 7, showing the ingredient percentages and burn test data for new compositions **O**−**Q**. This time, all three of the epoxy candidates contained only 7.5 wt % Epon/Versamid, 52.5 wt % $NaNO_3$, and the remaining balance was again partitioned as above for the series comprised by compositions **L**−**N**. Interestingly, all three of these compositions met the minimum performance requirements for the cluster. However, we felt it best practice to down-select to the candidate that most closely matched the performance data as the control sample, in this case composition **O**.

Table 7. Composition and Performance of Epoxy Replacements for the M585 White Star Cluster Signal

	percent composition by weight			performance[a]	
composition[b]	$NaNO_3$	Mg 30/50	Mg 50/100	burn time (s)	luminous intensity (cd)
M585 baseline	56.5	18	18	6.79	68 157
O	52.5	10	30	6.40	77 026
P	52.5	20	20	7.57	61 656
Q	52.5	30	10	8.76	50 171

[a] M585 static test requirements: 30,000 candlepower minimum, 6.0 s burn time minimum. [b] M585 baseline weight balance was 7.5% Laminac/Lupersol binder system; all other compositions had 7.5% Epon/Versamid.

CONCLUSION

In summary, we have developed two separate epoxy-based pyrotechnic compositions to replace the chemical fillers currently specified for the U.S. Army's M585 and M583A1 yellow illuminant flares. The Viton laminate for the inner tube diameter of the M583A1 is essential to proper function of this signal, as evidenced by the substantial loss of performance exhibited by flares encased in unlaminated tubes. This nicely complements our previous work as all four colored signals for the 40 mm family of illuminants each now have an epoxy variant of the current specification.[25,26]

EXPERIMENTAL SECTION

Materials. Ellipsoidal Mg 30/50 (MIL-P-14067, Type IV), ellipsoidal Mg 50/100 (MIL-P-14067, Type IV), and atomized Mg 50/100 (MIL-P-14067, Type I) were purchased from Magnesium Elektron (Manchester, NJ). $NaNO_3$ (prilled) was purchased from Hummel Croton (South Plainfield, NJ) and $NaNO_3$ (MIL-S-322, grade B, Class 2) was purchased from Atlantic Equipment Engineers (Upper Saddle River, NJ). Laminac 4116 was purchased from Ashland Chemical Company (Budd Lake, NJ). Lupersol was purchased from Norac (Azusa, CA). Epon 813 was purchased from Momentive (Columbus, OH). Versamid 140 was purchased from BASF (Cincinnati, OH). Kraft cardboard tubes for the M585 prototypes were purchased from New England Paper Tube Company (Pawtucket, RI). Lined and unlined phenolic parachute tubes for the M583A1 prototypes were purchased from Chemring Ordnance (Perry, FL).

Blending of Pyrotechnic Mixtures. Oxidizers were dried in the oven for 18 h at 60 °C prior to blending and all chemicals were weighed out according to the weight percentages given in the previous tables. A binder system (95:5 Laminac 4116/Lupersol DDM for the M585 and M583A1 controls, 80:20 Epon 813/Versamid 140 for compositions **A**−**Q**) was weighed in a Hobart air-mixing bowl and vigorously mixed by hand with a wooden tongue depressor for 1 min. To the bowl was added ellipsoidal magnesium 30/50 (MIL-P-14067, Type IV) and magnesium 50/100 (ellipsoidal MIL-P-14067, Type IV for the M583A1 control; all others used atomized 50/100 Mg per MIL-P-14067, Type I). The mixture was blended with the aid of a B-blade at 207 kPa for 10 min, after which time the air was turned off. To the mixture was then added $NaNO_3$ (prilled for the M585 control; all other compositions used powdered MIL-S-322, grade B, Class 2) and the mixture was blended for another 10 min at 207 kPa. The air was turned off and the resulting pyrotechnic mixture was poured from the air-mixer bowl to a ceramic evaporating dish. All prepared compositions were dried in air overnight at ambient temperature and pressure before consolidation.

Prototyping of M583A1 Flares. Flares were prepared by consolidating the cured compositions into a phenolic tube (height of 5.54 cm; inner diameter of 5.15 cm) with the aid of a tooling die (inner diameter of 3.37 cm) and a manual hand press. Each flare was consolidated in one increment (4435 kg with 10 s dwell) after adding three individual powders to the tooling die in the following order: (i) 1 g Class 7 black powder, (ii) 3 g thermate-based igniter slurry,[27] and (iii) 80 g of each cured composition. Between 83.5 and 83.9 g of energetic material was loaded into each tube, and five candles were prepared and tested for each composition. After consolidation, all candles were cured for 16 h in the oven at 60 °C.

Prototyping of M585 Clusters. Clusters were prepared by consolidating the cured compositions into a cylindrical kraft cardboard tube (height of 1.397 cm; inner diameter of 2.870 cm) with the aid of a tooling die (inner diameter of 3.37 cm) and a manual hand press. Each cluster was consolidated in one 15.3 g increment at a consolidation dead load of 3629 kg. The resulting cluster pellets were coated with the above thermate-based igniter[27] (as acetone slurry) and fitted with two one-inch strips of black match. Between 14.6 and 15.1 g of illuminant composition was loaded into each cluster tube, and 5 clusters were prepared and tested for each composition. After coating, all candles were cured for 16 h in the oven at 60 °C.

Characterization. Static ignition testing of all M583A1 and M585 prototypes was performed in a 100 ft indoor flare tunnel at ambient temperature and pressure using an electric match pulsed with an energy source of 20 V. Optical emissive properties were characterized by a single element silicon detector (33 mm^2) and quartz window (SED 033 from International Light), coupled to a photopic filter (Y-filter) and a field of view limited H-hood. The current output of the detector was converted to voltage using a DL Instruments 1211 transimpedance amplifier. Voltage output was collected and analyzed from the amplifier using a NI-9215 National Instruments data card and in-house developed Labview based data acquisition and analysis software.

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Author Contributions

The paper was written through contributions of all authors. All authors have given approval to the final version of the paper.

Notes

The authors declare no competing financial interest.

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