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RADIATION-INDUCED POLYMERIZATION I

A PYROTECHNIC BINDER

DEPARTMENT OF THE NAVY
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RADIATION-INDUCED POLYMERIZATION I
A PYROTECHNIC BINDER

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RADIATION-INDUCED POLYMERIZATION I
A PYROTECHNIC BINDER

I. INTRODUCTION

One of the major problems in pyrotechnic research is the development of an effective binder material which is safe to use, provides good bonding, gives desirable burning characteristics and lends itself to rapid mass production techniques. Presently, there are two basic methods in use: (1) casting, which suffers from high binder concentrations, and (2) pressing, which does not lend itself to continuous processing methods. Both of these methods suffer from problems, such as, hydrogen production from moisture attack on the magnesium, poor bonding of the composition to the casing and chunking of the candle during burnings. These problems and many others have long been recognized and in most cases are related to the binder.

A study of new binders and production methods has long been under consideration. One method presently under investigation is the extrusion of mixes similar in composition to those used in cast systems. Typically, they involve precipitation of polymers such as, Viton A or Estane 5702 with a nonsolvent like 2-propanol to coat the fuel and oxidizer.[1,2]

A technique which has been previously described illustrates a method of microencapsulating magnesium and sodium chloride with poly(methyl methacrylate). These compounds were dry mixed in an atmosphere of the monomer, and the encapsulated particles so treated were pressed, and the monomer polymerized by irradiation. Since the particles were in intimate contact after pressing, radiation-induced polymerization bonded the particles together. Irradiation to two megarads appeared to produce the maximum strength for the system. This procedure has the potential advantage that the coated particles would be moisture resistant, easy to handle, could be continuously mixed, pressed, and polymerized at any time.[3] Although the idea of radiation-induced bonding of microencapsulated flare components is new, microencapsulation of fuels with liquid oxidizers has been tried previously.[4] The method of microencapsulating flare components was recently investigated at this facility. It was found that this method has inherent difficulties in microencapsulating the sodium nitrate and particularly the magnesium.[5]

Presently under consideration are several other applications of radiation-induced polymerization which have potential commercial applications. For example, the development of concrete-polymer materials[2] is an interesting application where the concrete specimen is loaded with approximately 6 percent monomer and polymerized with either gamma irradiation or catalytically. Monomers which have been used to date are methyl methacrylate, styrene, acrylonitrile and copolymers with trimethylolpropane trimethacrylate. These materials have increased the physical properties by 300 to 500% over standard concrete.

Another example of the use of gamma radiation is to polymerize monomers impregnated into materials in the production of wood-plastic products. Commercial production of wood-plastic began in the U.S. in 1969. Three companies (American Novawood Company, Atlantic Richfield Company, and Lockheed Georgia Company) are currently using radiation to cure monomer impregnated wood. Application of these materials include the production of parque flooring, which has been used in the Kansas City International Airport where there is approximately ten acres of flooring. These references are given only to illustrate that the commercial application of ionizing radiation to polymerize vinyl monomers at least in the area of wood plastic material has been considered economically sound. Production considerations have not only been in the U.S., but as evidenced by the participation in the International Conference sponsored by the International AEC Agency held in Bangkok, November 1967, indicates that this industrial application has far reaching international implications.[7] Studies of the economics of the production of wood-plastic materials indicate that the cost of curing (radiation cost) are no more than ten percent of the processing cost.[8]

It is the intent of the work described in this report to illustrate yet another process which readily lends itself to an irradiation curing process, viz., the production of pyrotechnic materials. As mentioned previously, production methods used to date suffer heavily from the lack of automation and present many inherent safety hazards, many of which are based on the requirement that many of the production operations must be handled by individuals, in particular, the batch mixing processes. It has been the intent of the work described here to illustrate that: (1) such a method of producing pyrotechnic materials is feasible, and (2) this method produces items that can be applied to future uses.

II. EXPERIMENTAL

A. Materials

The monomers used in this study were of standard commercial purity. No attempt was made to remove the inhibitors added by the manufacturer since it was the intent of this investigation to simulate actual production conditions. Removal of inhibitors by distillation would add to the cost of manufacturing and to the inconvenience of the method.

The magnesium and sodium nitrate used was the standard Navy production material. The magnesium was 30/50 mesh, and the sodium nitrate was 32 ± 10 micron. These materials were not pretreated before preparation of either the test candles or the ampoule polymerizations.

B. Irradiation Facility

All irradiation in this investigation was carried out in a nominally 10,000 curie cobalt-60 source at the Radiation Laboratory of Indiana State University.[8] This facility is a "cave-type" design with water well source storage. An automatic source lowering system also is installed which would remove the cobalt-60 from the irradiation chamber in the event of a fire, thus preventing a radiation hazard.

The radiation dosimetery was based on the Fricke ferrous sulfate method with a $G(Fe^{+3}) = 15.5$.[10]

C. Conversion Studies

Although several methods of preparing samples to determine the degree of conversion of the monomers in the pyrotechnic mixture were tried, the most effective method is the one ultimately used in this study. Since in production irradiation in vacuum would not be practical, all samples reported here were prepared under ambient conditions in a sealed vial. A mixture, consisting of 60% $NaNO_3$ and 40% Mg, was added to the monomer system. In most cases, a polymer was added to the monomer to increase its viscosity. A sample for polymerization usually consisted of 15% of the monomer system and 85% of the pyrotechnic mix. This higher monomer concentration was used to aid in polymer analysis and probably had some effect on the polymerization reactions. Analysis of polymers varied but generally consisted of gravimetric methods.

1. Styrene

The starting binder contained a 15% mixture of polystyrene and styrene. A simulate flare was prepared with 10.9% binder, 54.4% sodium nitrate and 35.7% magnesium. Approximately 4 gms of this mixture was placed in a glass vial and sealed at ambient pressures. The ampoules were irradiated at 3×10^5 rad/hr. The polymer was analyzed by dissolving the monomer polymer system in toluene which contained 0.1% hydroquinone. The hydroquinone was added to inhibit the polymerization of the residual monomer during the polymer work-up. Approximately 100 ml of toluene was added to the sample and the resulting mixture stirred for one hour. This mixture was then filtered to remove the magnesium and sodium nitrate. The toluene solution was concentrated to a volume of 25 ml; then 300 ml of methanol was added. This precipitated the polymer as a fine suspension. This polymer suspension was heated then set aside for 24 to 36 hours so that the polymer would settle and leave a clear liquid above the sample. The polymer was removed by filtering into a tared filter crucible and weighed. The percent conversion of monomer to polymer was recorded.

2. Styrene-Benzoyl Peroxide

The binder contained 2.1% benzoyl peroxide, 12.1% polystyrene, and 85.8% styrene. The simulated flare mixture contained 17.7% binder, 46.5% sodium nitrate, and 34.1% magnesium. Approximately 5 gms of this mixture was added to the sample vials and irradiated. The polymer work-up was similar to the one used for styrene.

3. Styrene Oxide-Benzoyl Peroxide

The binder contained 0.5% Benzoyl peroxide, 15% polystyrene, and 80% styrene oxide. The simulated flare mixture contained 19.7% of the monomer mixture along with 48.2% sodium nitrate and 32.1%

magnesium. The polymerization and polymer work-up of this system was similar to the styrene system.

4. 2-Nitro-2-Methyl Propyl Methacrylate (2N2MPMA)

The polymerization and polymer work-up for this system was somewhat different from the styrene and styrene oxide systems. A mixture of 17.5% monomer, 50.0% sodium nitrate, and 33.4% magnesium was prepared. Approximately 5 gms of this mixture was added to glass vials for irradiation. Analysis of the polymer was done in two ways. The low converted material was soluble in acetone, the higher converted material was not. For conversions less than 10%, approximately 200 ml of acetone which contained hydroquinone was added to each sample. This system was stirred for 1 hour and then the magnesium and sodium nitrate removed by filtration. The filtrate was concentrated to 25 ml, and then 300 ml of methanol was added to this solution. The polymer precipitated as a white solid. This solution was set aside for 2 days. The polymer present was isolated by filtration in a tared filter crucible. The yield was determined by standard gravimetric techniques. The higher converted samples were analyzed by dissolving the magnesium and sodium nitrate in 300 ml of a 10% solution of hydrochloric acid. After stirring for two hours, the polymer-monomer mixture was isolated by filtration. This precipitation was washed with methanol; a solvent for the monomer was added to remove any unreacted monomer. The remaining white solid was isolated and the percent conversion determined by standard gravimetric techniques.

5. DER 321

A mixture of 11.4% DER 321 (a Dow Chemical Co. epoxy resin), 53.5% sodium nitrate and 35.1% magnesium was prepared. Approximately 5 gms of this mixture was added to glass sample vials for irradiation. Analysis of the polymer was unsatisfactory so no quantitative data could be obtained. There was some indication that the sample had polymerized but the extent could not be quantitatively measured.

6. Formrez F-17-80

This monomer is a carbonate terminated unsaturated polyester supplied by the Witco Chemical Corp.. The binder is a prepolymer with a molecular weight of approximately 1500. A mixture of 15.3% Formrez, 51.2% sodium nitrate, and 35.5% magnesium was prepared. Approximately 5 gms of this mixture was added to sample vials for irradiation. In the dose range used, no significant conversions were observed, however, this could be due to the lack of a satisfactory polymer isolation method. The only real change was a loss of tackiness above 2.5 megarads.

D. Experimental Candles and Controls

1. Preparation of the Monomer

Polystyrene (15% by weight) was mixed with styrene and styrene oxide monomers to obtain a more viscous material which could easily be mixed. This technique was tried with vinyl acetate, but the mixture with magnesium and sodium nitrate was always too dry to press. The other monomers and prepolymers, 2N2MPMA, DER 321, and Formrez F-17-80, were viscous enough to mix as purchased. The binder for the control candles was DER 321 resin and DEH 14 catalyst purchased from the Dow Chemical Co.

2. Preparation of the Candles

The composition used for the control and experimental candles was magnesium 58%, sodium nitrate 38%, and binder 4%. The composition for the candles was mixed in a Hobart blender which had been converted to an air drive.[11] The composition was pressed into fishpaper tubes at 8,500 psi, and the candles were approximately 2" diameter and 3" long, and contained approximately 230 gms.

III. RESULTS

A. Conversion Studies

Results of the conversion studies for styrene, styrene plus 2% benzoyl peroxide, styrene oxide plus 5% benzoyl peroxide and 2-nitro-2-methyl propyl methacrylate are illustrated in Figures 1 and 2. Conversion data for DER 321 and Formrez have not been included due to the difficulty in obtaining quantitative data.

B. Candle Data

The results of the candle data obtained are shown in Tables I-III. It should be noted that each individual candle was measured for length, thus, some variances in burning time and rate should be expected.

One or two sets of controls were burned with each set of experimental candles to allow for normal tunnel discrepancies.

IV. DISCUSSION

A. Radiation Yields

Examination of the results of the polymerization of styrene indicate that 100% conversion to polymer is obtained after approximately 7.5 megarads while upon the addition of 2% of benzoyl peroxide, this is lowered to a value of approximately 5 megarads. In addition, the benzoyl peroxide appears to have two effects: one, it tends to shorten the induction period and increase the rate of polymerization.

The polymerization of styrene oxide plus 0.5% benzoyl peroxide is illustrated in Figure 2. Here it can be seen that there is no significant conversion of the styrene oxide for doses as high as 9 megarads. The polymerization of 2N2MPMA,also illustrated in Figure 2, shows that 100% conversion is obtained only for doses in excess of 11 megarads. There appears to be a significant induction period which may last to doses as high as five megarads. The polymerization of Formrez and analysis of polymer did not lend itself to standard analytical methods, however, there was indication of an insoluble material that was not soluble in acetone, a known solvent for Formrez Since this material is an unsaturated polyester with a molecular weight of approximately 1500, there is the ever present possibility that conversion to a higher molecular weight polymer proceeds via a cross-linking type mechanism as opposed to the conventional additional polymerization methods. This would probably account for the change in the viscosity observed in the range of one to two megarads.

The polymerization of DER 321 also suffered from the difficulty of analyzing the resultant polymer since no satisfactory solvent-nonsolvent system was developed. Attempts to remove the magnesium and sodium nitrate only lead to hydrolysis of the polymer which was known to be present based on solvent extractions with toluene. However, since this solvent is not completely effective in dissolving the polymer, it could not be used in an analytical method.

Results from the polymerization of styrene in wood plastic combinations have indicated that 100% conversion would be obtained after a dose of approximately five megarads based on a copolymer of 60% styrene and 40% acrylonitrile, which is somewhat lower than the dose required for this study. However, one very interesting feature of the wood plastic combination studies is the fact that the dose required for 100% conversion is a function of the irradiation intensity. For example, in the polymerization of methyl methacrylate the dose for 100% conversion at an intensity of 1×10^5 rads/hr is approximately 0.6 megarads, while at an intensity of 8×10^5 rads/hr the dose required is 1.8 megarads. Thus, a decrease in intensity by a factor of eight decreases the dose required to reach 100% conversion by a factor of three. This would be an important design factor to be considered in any future commercial application of this method. Also, the gamma radiolysis of sodium nitrate produces several radical species[10] and the presence of these radicals could hinder polymerization.

The use of peroxide to lower the dose required for 100% conversion is illustrated in Figure 1. Here a factor of approximately two was gained on the dose required for 100% conversion.

B. Comparison of Controls and Experimental Candles

From the Data Summary on the following page, it can be seen that the experimental candles exhibited higher candlepower and faster burning rates than the control candles without loss in the efficiency. Particularly the styrene candles showed extremely high candlepower and efficiency.

The fast burning rates of the experimental candles can be explained in part by the incomplete conversion of the monomer or prepolymer since the candles were irradiated between 2.5 to 3.5 megarads. If the monomer was not completely converted to polymer, there is the possibility that vaporization of the unreacted monomer could be immediately proceeding the advancing flame front. The fate of the unreacted monomer is a matter of conjuncture and should be investigated.

DATA SUMMARY

		Candlepower	B.T. (sec)	Eff. $\frac{cp\text{-}sec}{gm}$	B.R. Sec/in
Table I	Control	171,400	62	46,000	18.63
	Styrene	332,900	35	50,100	9.97
	Styrene Oxide	240,400	37	47,800	11.78
Table II	Control #1	159,700	63	43,400	17.29
	Control #2	175,200	56	42,400	15.38
	DER 321	217,100	47	44,100	12.78
	Styrene + B.P.	296,800	35	44,600	9.68
	Styrene Oxide + B.P.	248,900	40	43,700	11.16
	2N2MPMA	321,800	32	45,000	8.87
Table III	Control	184,300	55	43,900	15.48
	Styrene	327,600	34	48,000	9.34
	Styrene Oxide	251,200	41	44,700	11.08
	Formrez F-17-80	226,100	47	46,600	13.54

V. CONCLUSIONS

A. The systems studied to date do not lend themselves to existing flare configurations (such as the Mk 45), but may be applied to future uses. However, the systems studied have not been prepared to 100% conversion of the monomer and it is possible that this binding technique can be applied to existing flare configurations.

B. In the past, binder systems were limited to systems where the catalytically initiated polymerization exotherm was low. With radiation-initiated polymerization, this is not a problem and the number of monomers which can be investigated is now unlimited.

C. Based upon the styrene-Benzoyl Peroxide system, addition of peroxide sensitizers significantly decrease the radiation dose required for 100% conversion.

VI. FUTURE PLANS

Based upon the work reported here, several obvious experiments are suggested. Study of these parameters would be necessary before application of this technique could be considered for production.

A. Effect of 100% conversion of the monomer on the burning characteristics of the flares, with some consideration given to the fate of the unreacted monomer.

B. Although styrene appears to be the most satisfactory binder studied to date, a program for investigation of other candidate monomers and comonomers should be established.

C. A full scale investigation of all polymerization parameters for the styrene should be initiated. This appears practical for the following reasons:

1. Styrene gives the best burning characteristics studied to date.
2. Styrene is inexpensive and readily available.
3. Good handling characteristics.

This investigation should include the following studies:

1. Degree of conversion on burning characteristics.
2. Optimization of composition including percent binder, peroxide, sodium nitrate, magnesium, and particle sizes.
3. Radiation intensity study.
4. Copolymerization of styrene with other monomers.

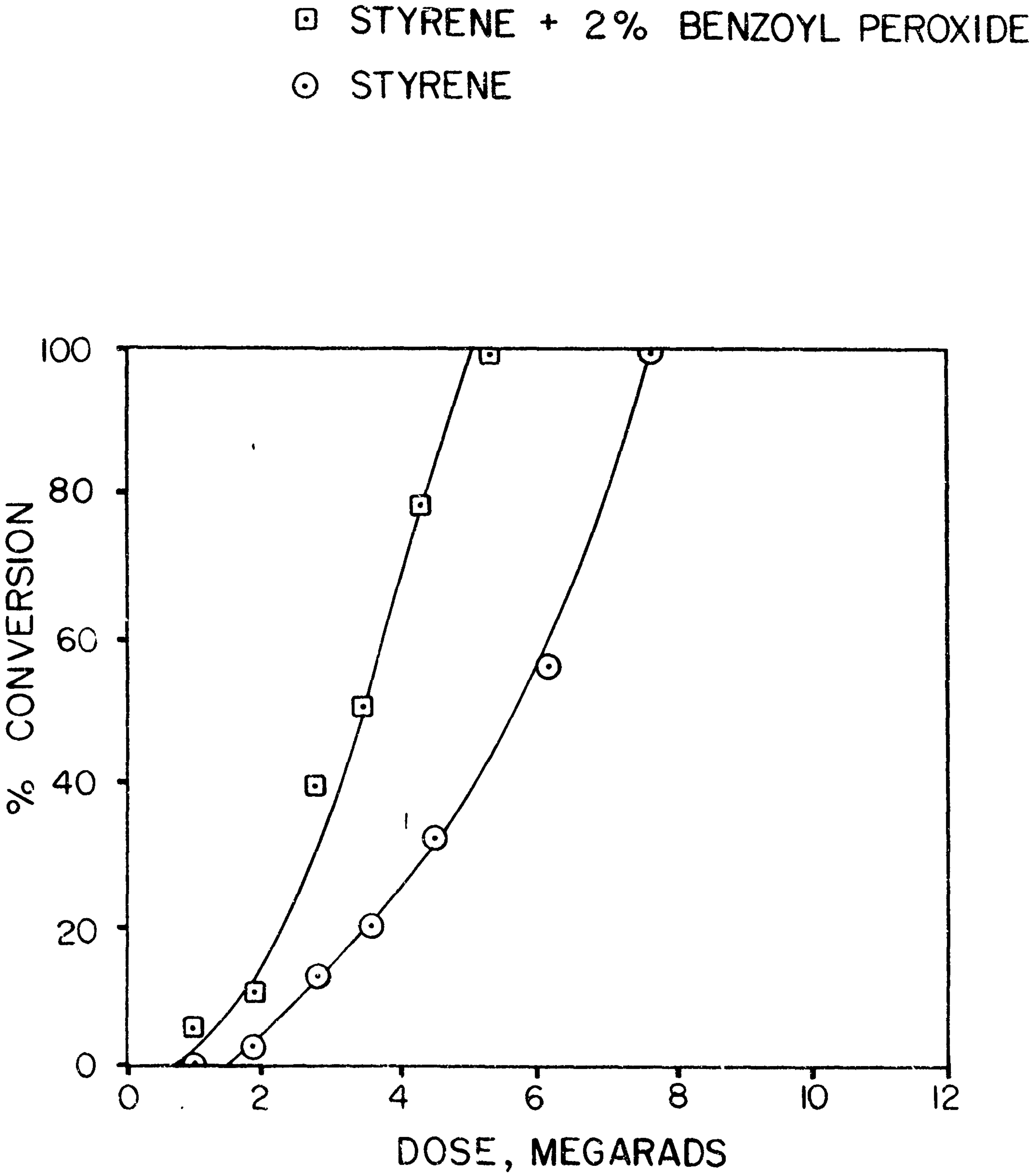
⊡ STYRENE + 2% BENZOYL PEROXIDE
⊙ STYRENE
100
80
60
40
20
0
% CONVERSION
0
2
4
6
8
10
12
DOSE, MEGARADS

FIGURE 1

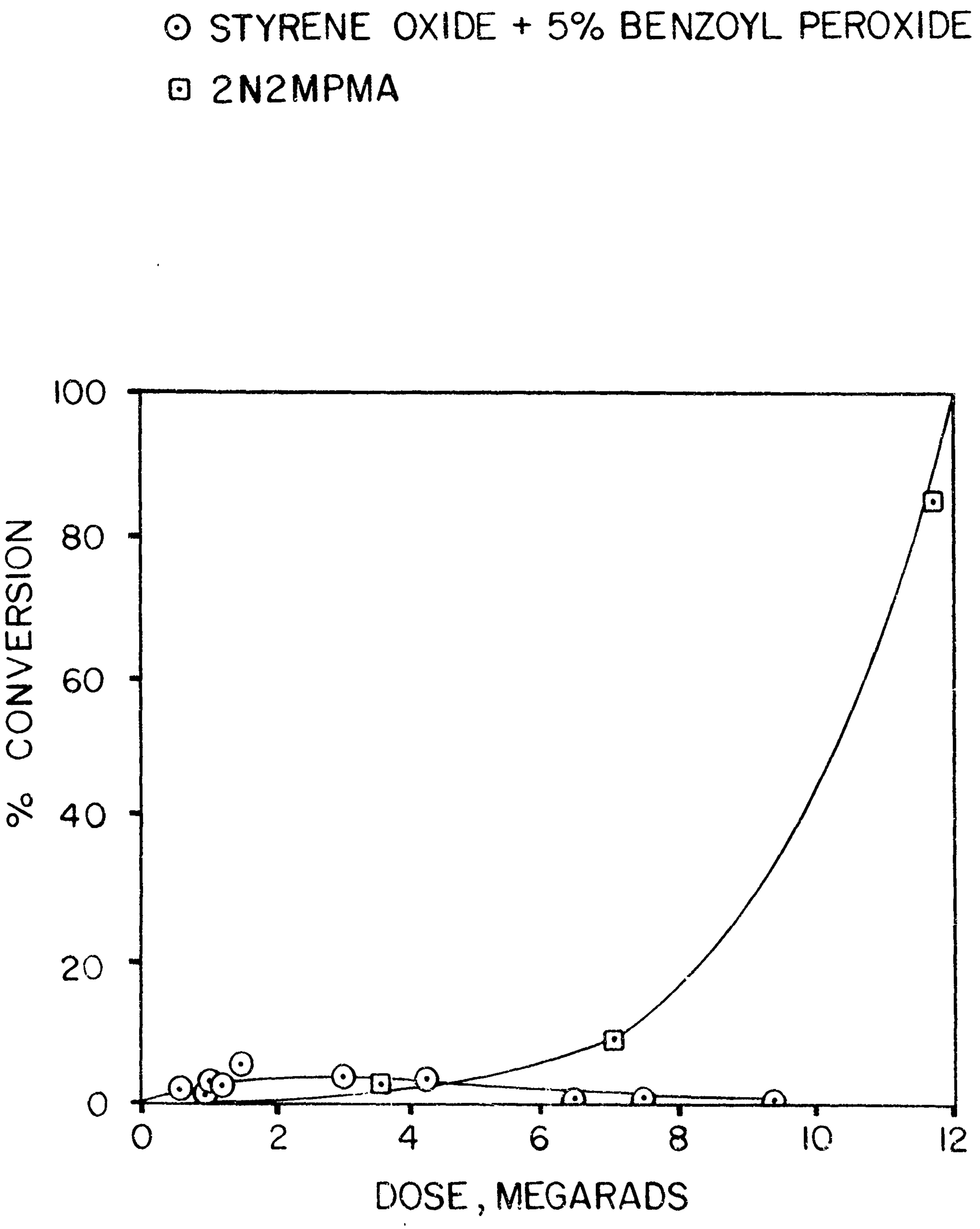
⊙ STYRENE OXIDE + 5% BENZOYL PEROXIDE
⊡ 2N2MPMA
100
80
60
40
20
0
% CONVERSION
0
2
4
6
8
10
12
DOSE, MEGARADS

FIGURE 2

TABLE I

CONTROLS

Candle No.	Candlepower (C.P.)	Burning Time (B.T.) sec	Efficiency (Eff.) C.P.·sec/gm.	Burning Rate (B.R.) sec/in.	Burning Rate (B.R.) gm./sec
1	153,000	63	41,900	18.92	3.65
2	171,000	63	46,800	18.92	3.65
3	165,000	64	45,900	19.22	3.59
4	176,000	63	48,200	18.92	3.65
5	192,000	57	47,600	17.18	4.04
Avg.	171,400	62	46,000	18.63	3.72
STYRENE					
1	226,000	40	39,300	11.43	5.75
2	301,000	36	47,100	10.29	6.39
3	306,000	34	45,200	9.71	6.76
4	379,000	34	56,000	9.71	6.76
5	359,000	34	53,100	9.71	6.76
6	360,000	34	53,200	9.71	6.76
7	321,000	35	48,800	10.00	6.57
8	368,000	33	52,800	9.43	6.97
9	376,000	34	55,600	9.71	6.76
Avg.	332,900	34.9	50,100	9.97	6.61
STYRENE OXIDE					
1	247,000	43	46,200	12.29	5.35
2	250,000	34	37,000	9.71	6.76
3	256,000	43	48,000	12.29	5.35
4	277,000	44	53,000	12.57	5.23
5	296,000	43	55,000	12.29	5.35
6	262,000	43	49,000	12.29	5.35
7	237,000	42	43,000	12.00	5.48
8	288,000	38	47,600	10.86	6.05
9	291,000	41	51,900	11.71	5.61
Avg.	240,400	37.1	47,800	11.78	5.62

TABLE II

CONTROL #1

Candle No.	Candlepower (C.P.)	Burning Time (B.T.) sec	Efficiency (Eff.) C.P.·sec/gm	Burning Rate (B.R.) sec/in	Burning Rate (B.R.) gm./sec
1	159,200	60	41,530	16.58	3.83
2	169,000	62	45,556	17.13	3.71
3	158,000	64	43,965	17.68	3.59
4	154,800	63	42,401	17.40	3.65
5	154,000	67	44,860	18.50	3.43
6	152,200	64	42,351	17.68	3.59
7	166,000	61	44,026	16,85	3.77
8	153,300	63	41,990	17.40	3.65
9	155,300	62	41,863	17.13	3.71
10	175,000	60	45,652	16.58	3.83
Avg.	159,700	62.6	43,400	17.29	3.68
CONTROL #2					
1	167,900	57	41,610	15.75	4.04
2	179,300	51	39,757	14.11	4.51
3	181,000	57	44,856	15.75	4.04
4	168,900	55	40,389	15.19	4.18
5	175,000	58	44,130	16.00	3.97
6	182,700	53	42,100	14.63	4.34
7	176,000	57	43,617	15.75	4.04
8	172,700	56	42,048	15.47	4.12
9	173,700	57	43,047	15.75	4.04
Avg.	175,200	55.7	42,400	15.38	4.14
DER 321					
1	211,700	48	44,180	13.01	4.79
2	224,200	46	44,840	12.47	5.00
3	218,700	48	45,641	13.01	4.79
4	226,600	46	45,320	12.47	5.00
5	220,500	47	45,058	12,74	4.89
6	213,200	50	46,347	13.55	4.60
7	218,600	48	45,620	13.01	4.79
8	207,000	49	44,100	13.27	4.69
9	211,300	46	42,260	12.47	5.00
10	224,200	45	43,865	12.20	5.11
11	207,900	46	41,580	12.70	5.00
12	212,100	47	43,342	12.98	4.89
13	221,900	45	40,520	12.42	5.12
14	222,000	46	44,400	12.70	5.00
Avg.	217,100	47	44,100	12.78	4.91

TABLE II (continued)

STYRENE + 0.5% Benzoyl Peroxide

Candle No.	Candlepower (C.P)	Burning Time (B.T.) sec	Efficiency (Eff.) C.P.·sec/gm.	Burning Rate (B.R.) sec/in	Burning Rate (B.R.) gm/sec
1	305,000	34	45,086	9.40	6.76
2	312,000	35	47,478	9.67	6.57
3	305,000	34	45,086	9.40	6.76
4	324,000	34	42,730	9.40	6.76
5	322,000	35	49,000	9.67	6.57
6	289,000	36	45,234	9.96	6.39
7	294,000	34	43,460	9.40	6.76
8	269,200	36	42,135	9.96	6.39
9	282,000	36	44,139	9.96	6.39
10	266,000	36	41,666	9.96	6.39
Avg.	296,800	35	44,600	9.68	6.57
STYRENE OXIDE + 0.5% Benzoyl Peroxide					
1	264,000	40	115,391	11.05	5.75
2	249,000	41	44,386	11.32	5.61
3	260,000	41	46,347	11.32	5.61
4	245,700	40	42,730	11.05	5.75
5	243,200	40	42,295	11.05	5.75
6	259,000	40	45,043	11.05	5.75
7	241,500	41	43,050	11.32	5.61
8	248,500	39	42,136	10.75	5.90
9	229,000	41	40,821	11.32	5.61
10	249,000	41	44,386	11.32	5.61
Avg.	248,900	40.4	43,700	11.16	5.70
2 NITRO - 2 METHYL PROPYL METHACRYLATE					
1	324,000	32	45,078	8.85	7.19
2	307,000	33	45,482	9.12	6.96
3	336,000	31	45,286	8.56	7.42
4	337,000	32	46,886	8.85	7.19
5	339,000	32	47,165	8.85	7.19
6	334,000	32	46,469	8.85	7.19
7	313,000	33	44,908	9.12	6.96
8	324,000	31	43,669	8.56	7.42
9	302,100	33	43,334	9.12	6.96
10	302,000	32	42,017	8.85	7.19
Avg.	321,800	32.1	45,000	8.87	7.17

TABLE III

Candle No.	Candlepower (C.P.)	Burning Time (B.T.) sec	Efficiency (Eff) C.P.·sec/gm.	Burning Rate (B.R.) sec/in.	Burning Rate (B.R.) gm/sec
			CONTROLS		
1	193,300	58	48,745	16.38	3.97
2	197,600	50	42,956	14.12	4.60
3	188,000	59	48,226	16.66	3.90
4	191,100	58	48,190	16.38	3.97
5	186,100	54	43,693	15.25	4.26
6	182,600	58	46,046	16.38	3.97
7	174,800	55	41,800	15.54	4.18
8	170,600	52	38,570	14.69	4.42
9	183,000	50	39,782	14.12	4.60
10	175,700	54	41,251	15.25	4.26
Avg.	184,300	54.8	43,900	15.48	4.21
			STYRENE		
1	331,600	35	50,460	9.64	6.57
2	324,500	34	47,969	9.67	6.76
3	324,900	35	49,441	9.64	6.57
4	339,500	33	48,710	9.09	6.97
5	351,200	34	51,916	9.67	6.76
6	318,060	33	45,634	9.09	6.97
7	311,050	35	47,333	9.64	6.57
8	315,600	33	45,281	9.09	6.97
9	323,400	33	46,400	9.09	6.97
10	335,900	32	46,733	8.82	7.19
Avg.	327,600	33.7	48,000	9.34	6.83
			STYRENE OXIDE		
1	262,700	41	46,829	11.11	5.61
2	256,000	42	46,747	11.38	5.48
3	251,000	39	42,679	10.57	5.90
4	245,800	43	45,953	11.65	5.35
5	260,300	40	45,269	10.84	5.75
6	222,900	42	40,703	11.38	5.48
7	261,500	41	46,661	11.11	5.61
8	242,400	42	44,264	11.38	5.48
9	248,100	40	43,147	10.84	5.75
10	261,600	39	44,358	10.57	5.90
Avg.	251,200	40.9	44,700	11.08	5.63

TABLE III (continued)

FORMREZ F-17-80

Candle No.	Candlepower (C.P.)	Burning Time (B.T.) sec	Efficiency (Eff.) C.P.·sec/gm	Burning Rate (B.R.) sec/in.	Burning Rate (B.R.) gm/sec
1	249,200	46	49,840	13.14	5.00
2	226,000	49	48,147	14.00	4.69
3	234,600	45	45,900	12.86	5.11
4	225,500	50	49,021	14.29	4.60
5	230,800	48	48,166	13.71	4.79
6	230,400	48	48,083	13.71	4.79
7	245,000	47	50,065	13.43	4.89
8	230,000	48	48,000	13.71	4.79
9	203,000	50	44,130	14.29	4.60
10	237,400	48	49,544	13.71	4.79
11	229,000	47	46,795	13.43	4.89
12	218,300	48	45,558	13.71	4.79
13	224,400	45	43,904	12.86	5.11
14	214,100	49	45,612	14.00	4.69
15	219,400	47	44,833	13.43	4.89
16	211,800	48	44,201	13.71	4.79
17	216,200	47	44,180	13.43	4.89
18	226,130	46	45,226	13.14	5.00
19	222,700	46	44,540	13.14	5.00
20	228,900	46	45,780	13.14	5.00
Avg.	226,100	47.4	46,600	13.54	4.86

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