

Flame Resistant Cotton Fabrics Prepared by Radiation-Initiated Polymerization with Vinyl Phosphonate Oligomer and N-Methylolacrylamide

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Synopsis

Radiation-initiated polymerization of vinyl phosphonate oligomer (molecular weight 500–1000) and N-methylolacrylamide from aqueous solutions was investigated with cotton printcloth, flannelette, and sateen fabrics and with cotton (50%)–polyester (50%) flannelette fabrics. Determinations were made of the effects of radiation dosage, mole ratio of vinyl phosphonate in the oligomer to N-methylolacrylamide in aqueous solution, concentration of reactants, wet pickup of solutions on fabrics, and irradiation of both dry and wet fabrics on efficiency of conversion of oligomer and monomer in solution to polymer add-on. The effects of vinyl phosphonate oligomer and N-methylolacrylamide radiation-initiated polymerization on some of the textile properties of cotton printcloth and on flame resistances of cotton and cotton–polyester fabrics were evaluated. The breaking strength of modified cotton printcloth was about the same as that of unmodified fabric; however, the tearing strength and flex abrasion resistance of modified fabric were reduced. The textile hand of the modified fabric was comparable with that of unmodified fabric. Microscopic examinations of the modified printcloth fabrics that had flame resistance indicated: interaction between cellulose and vinyl phosphonate oligomer–poly(N-methylolacrylamide) and uniform deposition in the fibrous cross section (transmission electron microscopy); surface areas of heavy deposits of oligomer–polymer (scanning electron microscopy); and phosphorus located throughout the fibrous cross section (energy dispersive x-ray analysis). Polymerization of vinyl phosphonate oligomer and N-methylolacrylamide was radiation initiated with cotton–polyester fabric; however, this modified fabric did not have flame-resistant properties.

INTRODUCTION

The development of flame retardant compounds and of processes for applying them to textile fabrics continues to be of interest. For about three centuries it has been known that the treatment of fabrics with ammonium salts of sulfuric, hydrochloric, and phosphoric acids reduced their flammability.¹ In 1953 the Flammable Fabrics Act was passed in the United States. Since that time extensive research has been conducted on developing nitrogen-, halogen-, and phosphorus-containing compounds and applying them to fabrics to reduce their flammability.² One objective has been to increase the durability of these fabric treatments to laundering, dry cleaning, and normal usage.² More recently there has been interest in increasing the durability of flame retardant treatments of fabrics in normal usage, because there is evidence that some flame retardants have long-term toxicological and biological effects.³

Free-radical-initiated grafting, primarily by irradiation and redox systems of vinyl and allyl monomers that contain phosphorus, nitrogen, and halogens

has been reported with cotton, regenerated cellulosic fibers, cellophane, wood, and other cellulosic materials.⁴⁻¹⁴ The free-radical-initiated interactions between unsaturated polymer and cellulose increased the durability of the treatments. Monomers that contained phosphorus included vinyl phosphonate,¹⁵⁻¹⁷ bis (2-chloroethyl)vinyl phosphonate,^{16,18} diethyl vinyl phosphonate,¹⁶ vinyl phosphoric acid,¹⁹ phenylvinyl phosphinic acid,²⁰ allyl phosphate,²¹ dihydroperfluorobutyl vinyl phosphonate,²² methacrylate phosphorodiamidate,²³⁻²⁷ diethylphosphatoethyl methacrylate,²⁸ cyclotriphosphazene,²⁹ cyclotetraphosphazene,²⁹ and other phosphorus compounds.³⁰⁻³⁷ Monomers that contained chlorine included vinyl chloride,³⁸ vinylidene chloride,^{16,21,39-41} allyl chloride,^{42,43} and copolymers of methacrylate and chlorinated⁴⁴ and brominated⁴⁵ monomers. Monomers that contained nitrogen included vinyl pyridine,^{46,48} methyl vinyl pyridine,⁴⁹ acrylamide,^{19,32,33,50} N-methylolacrylamide,¹⁵ and acrylonitrile.^{16,18} Eisenberg and Weil recently reviewed the use of phosphorus-containing vinyl and allyl monomers in flame retardancy.^{7,51} They reported the preparation of a polyunsaturated vinyl phosphonate oligomer for use with N-methylolacrylamide as a coreactant to impart flame resistance to textiles.⁵² Ammonium and potassium persulfates were used as catalysts to initiate free-radical curing of the oligomer and monomer on textile fabrics.

This report will deal with radiation-initiated, free-radical curing of vinyl phosphonate oligomer and N-methylolacrylamide to form a copolymer with cotton fabrics of different constructions and with cotton-polyester fabric. The initiating free radicals are formed on the cellulose molecule when the fabrics are exposed to high-energy radiation,⁵³ so that there is a maximum of interaction between fiber and copolymer and a minimum of homopolymer formation.⁵⁴ The efficiency of conversion of vinyl phosphonate oligomer and N-methylolacrylamide to copolymer on the fabrics and the flame resistance of the modified fabrics are reported.

EXPERIMENTAL

Materials

Cotton fabrics used were a commercial grey printcloth (about 3.4 oz/yd², 84 × 77), flannelette (about 4 oz/yd², 48 × 47), and sateen (about 7.5 oz/yd², 84 × 52). The fabrics were desized, alkali scoured, bleached, washed with water, and air dried. Cotton (50%)–polyester (50%) flannelette (about 4 oz/yd², 50 × 42) was used. Samples of the fabrics were equilibrated overnight in air at about 25°C and 45% R.H. to constant weight.

A polyunsaturated vinyl phosphonate oligomer (molecular weight 500–1000; 22–23 wt-% phosphorus) that was made by condensation polymerization of vinyl phosphonate monomer^{7,51,52} and was designated FYROL 76 was obtained from the Stauffer Chemical Company. The oligomer was soluble in water. N-methylolacrylamide, a commercial product, was used as received. Other chemicals used were reagent grade.

TABLE I
Effects of Mole Ratios of Vinyl Phosphonate Oligomer to N-Methylolacrylamide on Radiation-Initiated Polymerization with Cotton Printcloth

Sam- ple no.	Radia- tion dosage, Mrad	VP:NMA, ^a mole ratio	Solids in solu- tion, %	Wet pick- up, %	Polymer add-on, %	Effi- ciency of monomer conversion ^b
Effect of Radiation Dosage						
1	0.00	1::1.08	31	150	0.5	0.01
2	0.27	1::1.08	31	150	6.7	0.15
3	0.41	1::1.08	31	120	36	0.80
11	0.55	1::1.08	31	150	37	0.80
14	0.82	1::1.08	31	150	41	0.89
19	1.2	1::1.08	31	150	42	0.93
29	1.6	1::1.08	31	150	43	0.95
30	1.8	1::1.08	31	140	38	0.93
33	2.3	1::1.08	31	150	46	1.0
Effect of VP:NMA ^a						
4	0.41	1::1.08	31	150	33	0.72
5	0.41	1::1.36	35	130	40	0.80
6	0.55	1::0.83	28	130	30	0.75
11	0.55	1::1.08	31	150	37	0.80
12	0.55	1::1.66	38	130	43	0.84
13	0.55	1::1.91	42	130	48	0.86
15	1.3	0::1.00	30	114	31	0.90
16	1.3	1::0.00	30	115	23	0.67
17	1.3	1::0.66	30	114	32	0.92
18	1.3	1::0.88	30	115	33	0.94
20	1.3	1::1.11	30	116	34	0.97
21 ^c	1.3	0::1.00	30	116	34	0.98
22 ^c	1.3	1::0.00	30	121	0.8	0.02
23 ^c	1.3	1::0.66	30	119	25	0.69
24 ^c	1.3	1::0.88	30	118	25	0.71
25 ^c	1.3	1::1.11	30	118	26	0.73
26	1.8	1::0.00	17	130	12	0.54
27	1.7	1::0.54	24	130	27	0.86
28	1.9	1::0.83	28	130	31	0.83
30	1.8	1::1.08	31	140	38	0.93
31	1.9	1::1.36	35	140	43	0.92
32	2.3	1::0.28	21	130	24	0.89
33	2.3	1::1.08	31	150	46	1.0
Effect of Solids in Solution						
7	0.55	1::1.08	10	130	5.7	0.44
8	0.55	1::1.08	15	130	12	0.60
9	0.55	1::1.08	20	130	18	0.69
10	0.55	1::1.08	25	130	23	0.69
11	0.55	1::1.08	31	150	37	0.80

^a VP = Vinyl phosphonate oligomer; NMA = N-methylolacrylamide; mole ratio calculated on basis of monomer composition of oligomer.

^b Efficiency of monomer conversion = polymer add-on/solids in wet pickup.

^c Fabric samples irradiated wet.

TABLE II
Effects of Vinyl Phosphonate Oligomer and N-Methylolacrylamide Radiation-Initiated Polymerization on Some Textile Properties of Cotton Printcloth

Sample no. ^a	Breaking strength, lb	Tearing strength, g	Flex abrasion, cycles	Wrinkle recovery angle, °(W + F)	
				Conditioned	Wet
1	36	753	808	223	151
2	34	633	706	223	155
12	38	307	190	193	216
13	37	333	178	191	247
26	34	660	534	217	157
27	36	433	206	200	203
28	31	367	177	231	213
30	31	333	138	211	247
31	34	300	124	211	262
Control	34	740	747	214	137

^a Sample no. refers to polymerization reaction given in Table I.

TABLE III
Effects of Wash and Dry Cycles on Flame Retardance of Modified Cotton Printcloth

Sample no. ^a	Char length, in.				
	0 ^b	5	10	40	50
3	2.1	2.0	2.4	5.1	3.8
5	2.4	1.5	1.6	2.1	2.8
6	2.4	2.1	1.8	2.5	3.6
14	2.1	1.6	2.0	1.7	—
19	1.8	2.0	2.8	2.4	—
29	2.4	1.8	2.0	2.0	—
33	2.8	2.3	1.9	2.0	—

^a Sample no. refers to polymerization reaction given in Table I.

^b Number of machine wash and dry cycles.

TABLE IV
Effects of Wash and Dry Cycles on Phosphorus and Nitrogen Contents of Modified Cotton Printcloth

Sample no. ^a	Phosphorus, %		Nitrogen, %	
	0 ^b	50 ^b	0 ^b	50 ^b
28	2.51	1.74	1.28	1.44
30	3.27	1.95	1.64	1.91
31	2.98	1.66	2.21	2.26

^a Sample no. refers to polymerization reaction given in Table I.

^b Number of machine wash and dry cycles.

Methods

The SRRC ⁶⁰Co radiation source previously described⁵⁵ was used. During the period of this investigation, the dose rate of the source ranged from 0.5 to 0.6×10^6 rad/hr as determined by ferrous-ferri dosimetry.⁵⁶

TABLE V
Effect of Vinyl Phosphonate Oligomer and N-Methylolacrylamide Radiation-Initiated Polymerization on Flame Retardance

Sam- ple no.	Radia- tion dosage, Mrad	VP:NMA ^a mole ratio	Solids pickup, %	Polymer add-on, %	Number of machine wash and dry cycles	Char length, in.
			Cotton Flannelette			
34	0.88	1:0.97	55	51	25	1.4
					50	2.0
35	1.0	1:0.80	38	32	0	1.6
					1	1.8
					10	1.7
					25	2.8
					40	2.1
					50	2.2
36	1.0	1:1.08	36	34	0	1.3
					1	1.4
					10	1.1
					25	2.3
					40	2.6
					50	2.8
			Cotton Sateen			
37	0.55	1:0.89	31	21	0	2.0
					5	5.0
					10	4.4
					40	2.8
			Cotton (50%)–Polyester (50%) Flannelette			
38	1.0	1:1.08	38	35	0	BEL ^b
39	1.8	1:0.97	55	52	25	BEL
					50	BEL

^a VP, vinyl phosphonate oligomer; NMA, N-methylolacrylamide; mole ratio calculated on basis of monomer composition of oligomer.

^b BEL, burned entire length.

Samples of fabrics (maximum size about 6 × 36 in., limited by dimensions of radiation source) were immersed in aqueous solutions of vinyl phosphonate oligomer and N-methylolacrylamide at 25°C. To saturate the fabrics thoroughly, vacuum (about 25 torr) was applied for 2–3 min and then released with nitrogen. The samples were padded to the desired wet pickup. Some of the samples were irradiated wet. Other samples were air dried overnight at room temperature and then irradiated to the desired dosage. The irradiated samples were washed in hot water, allowed to line dry, and then pressed with a cool iron on a wool setting. After equilibration to constant weight at about 25°C and 45% R.H., the increase in weight of the irradiated, modified fabric, as compared with the weight of an equivalent sample of unmodified fabric, was recorded as polymer add-on. The efficiency of conversion of monomer and oligomer to polymer add-on was calculated as polymer add-on/solids in wet pickup.

The irradiated, modified fabrics were laundered and dried in accordance with AATCC Test Method 124-1969, as specified in the DOC standards.⁵⁷ Flammability measurements were made in accordance with DOC FF 3-71.⁵⁸ The amounts of phosphorus present in the samples were determined by the reduced

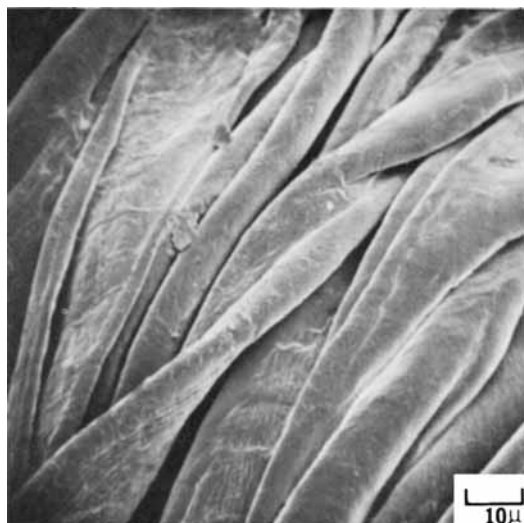


Fig. 1. SEM surfaces of cotton fibers from printcloth treated with vinyl phosphonate oligomer and N-methylolacrylamide (Table I, sample 27).

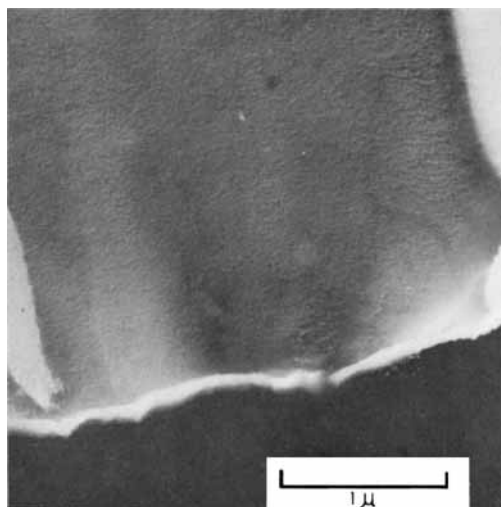


Fig. 2. TEM cotton fiber cross section from printcloth treated with vinyl phosphonate oligomer and N-methylolacrylamide (Table I, sample 27).

molybdate colorimetric method⁵⁹; the amounts of nitrogen present were determined by the Kjeldahl method.⁶⁰ Physical properties of the fabrics were determined by ASTM methods for breaking strength,^{61(a)} tearing strength,^{61(b)} flex abrasion resistance,^{61(c)} and conditioned and wet wrinkle recovery, Monsanto.^{61d}

Microscopic analyses⁶² were used to determine the interaction between cellulose and vinyl phosphonate oligomer-poly(N-methylolacrylamide) and the location of the oligomer-polymer within the fibrous cross section (transmission electron microscopy); surface areas of deposits of oligomer-polymer (scanning

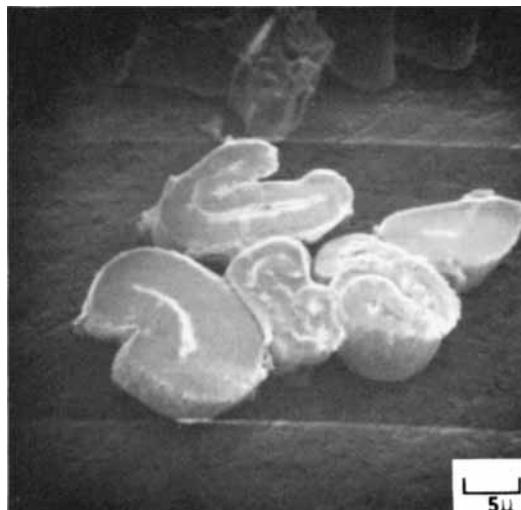


Fig. 3. Cotton fiber cross sections from printcloth treated with vinyl phosphonate oligomer and N-methylolacrylamide (Table I, sample 27).

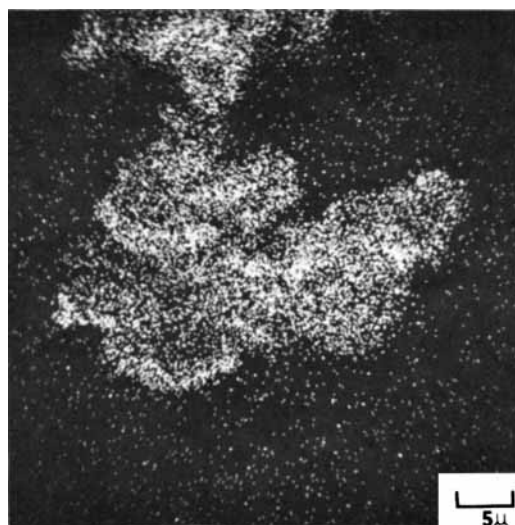


Fig. 4. EDX map showing location of phosphorus in cotton fiber cross sections shown in Fig. 3.

electron microscopy); and phosphorus location within the fibrous cross section (energy dispersive x-ray analysis).

RESULTS AND DISCUSSION

The effects of mole ratios of vinyl phosphonate oligomer (VP) to N-methylolacrylamide (NMA) on radiation-initiated polymerization with cotton printcloth are shown in Table I. The effects of several dependent variables (radiation dosage 0 to 2.3 Mrad; VP:NMA mole ratios 1:00 to 1.91; 0::1; solids in solution, 10% to 42%; and irradiation of samples both wet and dry) on polymer add-on on

TABLE VI
Microscopic Analyses of Cotton Printcloth Modified by Radiation-Initiated Polymerization of Vinyl Phosphonate Oligomer and N-Methylolacrylamide

Sam- ple no. ^a	Phos- phorus, %	Nitro- gen, %	Char length, in.	Cuene solubility of TEM sections ^c	SEM surface ^d	EDX analysis P map ^e
1	0.00	0.03	BEL ^b	dissolved	small deposits	no pattern
13	2.31	2.95	2.1	slightly swelled, thinned	heavy areas of deposit, many areas clean	P throughout section, heavy deposits between fibers
30	3.27	1.64	1.9	slightly swelled, thinned	heavy areas of deposit, many areas clean	P throughout section, heavy deposits between fibers
26	2.45	—	BEL ^b	dissolved, polymer residues	heavy areas of deposit, many areas clean	P throughout section
27	2.96	0.92	2.3	thinned	heavy areas of deposit, many areas clean	P throughout section, heavier on outer surface

^a Sample no. refers to polymerization reaction given in Table I.

^b BEL, burned entire length.

^c Solubility of TEM (transmission electron microscopic) ultrathin cross section in cuene (0.5M cupriethylenediamine dihydroxide).

^d SEM (scanning electron microscopic) surface.

^e EDX (energy dispersive x-ray) analysis, P (phosphorus) map.

cotton printcloth were determined. For example, at a mole ratio of VP:NMA 1:1.08 and at about 30% solids in the padding solution (see samples 2 and 33), efficiency of conversion of VP and NMA to polymer on irradiated, dried fabrics increased from 0.15 to 1.0 as the radiation dosage increased from 0.27 to 2.3 Mrad. Solutions that contained only NMA were more efficiently converted to polymer add-on than solutions that contained only VP (compare sample 15 and 16). In general, as the relative amount of NMA was increased in VP-NMA solutions, efficiency of conversion was increased; as the percentages of solids in the padding solutions were increased, efficiency also increased (compare samples 7-11). When fabric samples were irradiated wet, VP and NMA conversion to polymer add-on was less than when comparable samples were irradiated dry (compare samples 15-20 with 21-25).

The effects of VP and NMA radiation-initiated polymerization on some textile properties of cotton printcloth are shown in Table II. Samples of cotton printcloth were selected so that radiation dosages ranged from 0 to 1.9 Mrad, VP to NMA mole ratios ranged from 1:0.00 to 1:1.91, and polymer add-on ranged from 0.5% to 48%. The breaking strengths of modified fabrics were about the same as that of unmodified fabric; however, both tearing strengths and flex abrasions of modified fabrics were less than those of unmodified fabrics. The textile hands of modified fabrics (0.5% to more than 40% polymer add-on) after

one to five machine wash and dry cycles were about the same as that of unmodified fabric. Note: Textile hands are subjective assessments of textile fabrics obtained from the sense of touch, particularly as related to softness, roughness, and pliability. For example, modified fabric sample 27 (about 27% polymer add-on) compared with unmodified fabric, had flame resistant properties, retained 100% of the breaking strength, about 60% of the tearing strength, and about 30% of the flex abrasion. The textile hand of this modified fabric was comparable with that of unmodified fabric.

The effects of wash and dry cycles on flame resistance and on phosphorus and nitrogen contents of VP-NMA-modified cotton printcloth fabrics are shown in Tables III and IV. Samples of cotton printcloth were selected so that radiation dosages ranged from 0.41 to 2.3 Mrad, VP:NMA mole ratios ranged from 1:0.83 to 1:1.36, and polymer add-on ranged from 30% to 46%. Generally, the flame resistance of modified fabrics did not change markedly, as measured by char length, as the number of wash and dry cycles increased. There was a decrease in phosphorus content with no loss in nitrogen content as the number of wash and dry cycles increased.

Also, the effects of wash and dry cycles on flame resistance of VP-NMA-modified cotton flannelette and sateen and cotton (50%)–polyester (50%) fabrics are shown in Table V. Modified cotton fabrics that had been impregnated with aqueous solutions of VP and NMA in about 1:1 mole ratios, dried, and irradiated at dosages ranging from 0.4 to 1.0 Mrad to give products with about 30% polymer add-on, passed the DOC FF-3-71 flame resistant test.⁵⁸ Similar modification of cotton (50%)–polyester (50%) fabrics with VP and NMA did not impart flame resistant properties to these fabrics as measured by the vertical flame and DOC FF-3-71 tests.⁵⁸

Microscopic analyses of VP-NMA-modified cotton printcloth fabric are shown in Figures 1–4 and are summarized in Table VI. Samples of fabrics that were flame resistant (no laundry cycles involved) had (1) heavy surface areas of deposit of VP and NMA (copolymer), as shown by scanning electron microscopy (Fig. 1); (2) deposits of VP and NMA (copolymer) throughout the fibrous cross section which were resistant to dissolution in cupriethylenediamine dihydroxide, as shown by transmission electron microscopy (Fig. 2); and (3) phosphorus throughout the fibrous cross section, as shown by energy dispersive x-ray analysis (Figs. 3 and 4).

Free-radical-initiated polymerization of binary solutions of monomers with macroradicals on cotton^{63,64} and regenerated cellulosic fibers⁶⁵ has been shown to increase the monomer reactivity of one or both of the monomers. We can conclude, therefore, that the inclusion of NMA in the solutions increased the efficiency of conversion of the unsaturated VP oligomer and NMA to polymer on fabrics when irradiated either in the wet (samples 21–25) or dry (samples 15–20) conditions. The efficiency of conversion of VP oligomer and NMA to polymer on fabrics was also shown to be dependent on radiation dosage, that is, the number of macroradicals formed on the cellulose molecules.⁵³ The use of other nitrogen-containing monomers with VP oligomer could possibly increase the efficiency of conversion of oligomer and monomer to polymer on fabric at lower radiation dosages. However, the major energy consumption in the radiation-initiated, free-radical process for making flame resistant cotton fabrics, outlined in this report, would probably be in the drying of the modified fabrics.

The use of VP oligomer and NMA, which are soluble in water, has the obvious advantage in the drying operation that the solvent (water) being removed is compatible with the biological and ecological environment.

Trade names are given as part of the exact experimental conditions. Mention of a trademark on proprietary product does not constitute a guarantee or warranty of the product by the U.S. Department of Agriculture and does not imply its approval to the exclusion of other products that may also be suitable.

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