Influence of Moisture on the Flame Retardance of Textile Fabrics

S. H. ZERONIAN, M. S. ELLISON, and K. ALGER, *Division of Textiles and Clothing, University of California, Davis, California 95616*

Synopsis

For cotton fabric not treated with a flame retardant (FR), the oxygen index (OI) increases slowly but at a constant rate with moisture content (MC). Above a MC of roughly 33% which roughly equals the water fiber saturation point of the cotton fiber, the increase becomes more rapid but the relation appears to remain linear. The relation between OI and MC is also linear for all the FR cotton fabrics examined up to an OI of at least 40. It is suggested that the moisture in the FR samples is not reducing flammability by entering into solid and/or gaseous phase reactions but by absorbing thermal energy owing to the endothermic process of heating and vaporization. Empirical relations appear to be present between the inherent hydrophilicity of the FR cotton fabrics and the OI of the dry fabric $(OI)_0$ and also between $(OI)_0$ and the temperature of the zenith of the first exothermic peak for the FR cotton fabrics as determined by differential thermal analysis. OI–MC relations for wool fabric and wool treated with an FR are similar to those found for the cotton fabrics except that the slopes of the lines found for the wool samples are lower than those of the equivalent cotton samples. The relation reported by Van Krevelen between OI and char residue for polymeric materials is tested for both the cotton and wool samples. Agreement with Van Krevelen's relation is not good when $(OI)_0$ values are used. Better agreement can be obtained if OIs are determined on samples with MCs between 2 and 10%.

INTRODUCTION

Since the consideration and introduction of the U.S. standard for the flammability of children's sleepwear FF3-71,[1] knowledge related to the influence of moisture on the flame retardance of textile fabrics has become increasingly important. The standard requires the samples to be tested in the dry state. It has been suggested that this requirement is unrealistic since it discounts the inherent moisture present in air dried fibers.[2] On the other hand, it has been found that the amount of moisture present in apparel can be low, and testing dry fabric allows a margin of safety.[3] A number of studies have been made on this topic because of its interest.[2–6] Moisture in a fabric can impede burning by absorbing thermal energy because of the endothermic process of heating and vaporization of the water. However, some researchers have claimed that the relation between oxygen index (OI) and moisture content (MC) is neither linear nor curvilinear for flame retardant cotton fabrics. Thus, they suggest that the effect of moisture might be more than simple heat utilization and that water is possibly entering into solid and/or gaseous phase reactions during burning.[2,4,5] Our study was conducted to examine this suggestion for cotton fabrics and to determine the relation between OI and MC for wool fabrics. In addition, empirical relations were sought between the OI of dry FR cotton fabrics and characteristics of thermal analysis curves. The suggestion[7] that there is a linear relation between char residue (CR) and OI for flame retardant cotton fabrics was tested. This relation was also considered for wool fabrics.

Journal of Applied Polymer Science, Vol. 25, 1311–1322 (1980)

0021-8995/80/0025-1311$01.20

EXPERIMENTAL

Materials

The following fabrics of 100% cotton fiber content were used: printcloth (89.2 g/m^2), twill (247 g/m^2), and sateen (271 g/m^2). The printcloth fabric (PC) was treated with the following flame retardants (FR) by the Southern Regional Research Center, U.S.D.A., New Orleans: THPOH–NH_3, Pyrovatex CP, THPOH–amide, and THPCl–urea. The twill (TL) and sateen (SN) fabrics were treated with THPOH–NH_3 by the Southern Regional Research Center and by Reeves Bros., Columbus, Georgia, respectively. A 100% wool serge fabric (353 g/m^2) was also used. This fabric (SE) was treated with potassium hexafluorozirconate (HFZ) by the Technical Service Center, The Wool Bureau Inc., Long Island, NY. All fabrics were treated with sufficient FR to meet the FF3-71 sleepwear standard.[1] Finish add-ons are given in Table I.

Methods

Fabric Weight. ASTM procedures were used to determine fabric weights.[8].

Water Retention. The procedure for determining water retention at 21°C has been described previously.[9] The tests were conducted on yarn unraveled from the fabrics and cut to approximately 1-cm lengths. For this measurement, water swollen samples are centrifuged at 900 g for 30 min, and the water retention value [(WRV), %] is calculated from the formula

$$[(W_1 - W_2)/W_2] \times 100$$

in which W_1 is the weight of sample immediately after centrifuging and W_2 is the weight of the dry sample.

TABLE I
Fabric Weights and Flame Retardant (FR) Add-Ons

Fabric type	Fabric weight[a] (g/m^2)	FR	FR add-on[b] %
Cotton fabric			
Printcloth (PC)	89.2	None	0
Twill (TL)	247.2	None	0
Sateen (SN)	270.9	None	0
Printcloth (PC)	109.5	Pyrovatex CP	22.8
Printcloth (PC)	109.9	THPCl–urea	23.2
Printcloth (PC)	110.2	THPOH–amide	23.5
Printcloth (PC)	106.5	THPOH–NH_3	19.4
Twill (TL)	280.4	THPOH–NH_3	13.4
Sateen (SN)	319.1	THPOH–NH_3	17.8
Wool fabric			
Serge (SE)	352.5	None	0
Serge (SE)	402.8	HFZ[c]	14.3

[a] Measurements made on samples conditioned at 65% relative humidity and 21°C.
[b] Calculated from fabric weights.
[c] HFZ denotes potassium hexafluorozirconate.

OI Determinations. Fabric samples (6.35 × 8.90 cm) were soaked in distilled water, passed through a pad to remove excess water, and allowed to dry in the ambient atmosphere to approximately the desired moisture content. For wool fabrics it was necessary to add 0.5 ml surfactant per liter of water to wet out the fabric. The samples were rinsed thoroughly before passing the samples through the pad. A set of 14 samples (including five controls for moisture content measurement) were sealed in a plastic bag for two days to allow samples to equilibrate to the same moisture content. OI was then determined in the manner described previously[10] using an MKM model JD-14 Oxygen Index Tester. To determine moisture contents, controls were placed in the tester for 1 min, the length of time required for an OI test, removed, placed immediately into a tared container, and weighed. The placing of controls in the tester was done periodically throughout the testing of a set of samples of a specific moisture content. The dry weight of the control samples was determined by drying samples for 4 hr at 105°C. Moisture contents are the average of the five determinations.

Thermal Analyses. Samples were ground in a Wiley Mill to pass a 20-mesh screen. Differential thermal analysis (DTA) was run on a Mettler TA 2000 Modular Thermal Analysis System under air at a heating rate of 10°C/min. Thermogravimetric analysis (TGA) was run on a Perkin-Elmer TGS-1 thermogravimetric analyzer under nitrogen at a heating rate of 40°C/min. The percent char residue was obtained from TGA curves by dividing the weight of the sample at 850°C by the initial weight of the sample after the loss of physically bound water. The latter reading was taken off the curve at roughly 150°C before the gradient of the curve began to change noticeably.

RESULTS AND DISCUSSION

Oxygen Index–Moisture Content Relation for Cellulosic Fabrics

For non-FR (i.e., starting) cotton fabric, OI initially increased slowly but at a constant rate with MC (Fig. 1). Above an MC of roughly 33% the increase

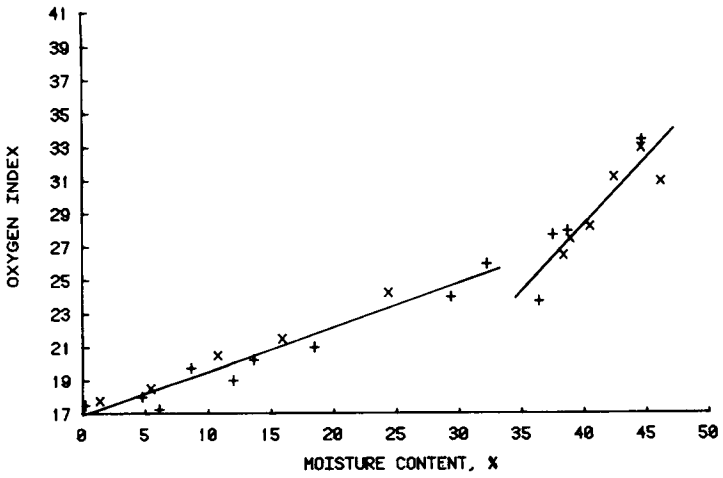

Fig. 1. Relation between oxygen index and moisture content for untreated cotton fabrics: (+) printcloth; (x) twill.

became more rapid but the relation appears to remain linear. Regression lines were calculated from the data. It should be noted that the weight of the fabric did not appear to affect the relation since the data for Figure 1 was collected from both PC (89.2 g/m^2) and TL (247 g/m^2) fabric. For each section of the curve, the relation between OI and MC can be represented by

$$OI = ZM + (OI)_0$$

where OI is the oxygen index of the FR sample at a moisture content of M, $(OI)_0$ is the oxygen index of the dry FR sample, and Z is the gradient of the line. Values for Z and $(OI)_0$ are given in Table II.

The relation between OI and MC is also essentially linear for flame retardant PC fabrics up to an OI of 40 (Fig. 2) and regression lines were calculated again from the data (Table II). It will be noted that the gradients of the lines are similar to each other and to that of non-FR cotton fabrics of high MC (i.e., MC > 33%). The data for the Pyrovatex and THPOH–amide samples overlapped and thus a single line was put through the points in Figure 2.

The moisture regain at which the slope of the OI–MC relation changes for non-FR cotton fabrics is close to the fiber saturation point (FSP) of the cotton. The MC at the inflection point is about 33%, which is equivalent to a moisture regain of 49%. From previous work[11,12] (Table III), the water retention value (WRV) for the non-FR cotton is between 43 and 47%. WRVs measured by centrifuging water-swollen cellulosic samples at 900 g for 30 min have been found empirically to give a quantitative measure of FSP.[13] FSP is defined as the total amount of water held in the cell walls of water-swollen fibers. It appears then that the change in slope of the OI–MC relation occurs for non-FR cotton when liquid water occurs in increasing quantities in interfiber and interyarn spaces. This water appears to contribute more strongly to reducing the flammability of the fabric than water present in the fabric below the FSP moisture content. Below the FSP moisture content the water is present primarily in the fiber cell wall and is either strongly hydrogen bonded to the hydroxyl groups of the cellulose or is in the amorphous regions or pores of the fibers. It is speculated that the reason, at least in part, that a fabric becomes more difficult to burn for a given increase in MC when the absolute MC of the fabric is greater than the FSP of the fiber is as follows: It can be deduced from the work of Preston and Chen[14] that the liquid water present in the interfiber and interyarn capillaries of a fabric can diffuse relatively easily through the fabric since the capillaries form a continuous channel throughout the fabric. In contrast, when the water in a fabric is contained only inside the fiber cell walls, diffusion of water from one part of the fabric to another is difficult since the capillary channels between fibers and between yarns will not be filled with water. When a flame is held to a fabric with an MC higher than the FSP of the fiber, the heat will initially cause any water in the fabric in the vicinity of the flame to evaporate as it absorbs thermal energy. As the fabric dries in the vicinity of the flame, liquid water present in the fabric further away from the flame will diffuse through the continuous interfiber and interyarn capillary channels towards the flame. This water will absorb thermal energy as it evaporates. Thus, a fabric containing liquid water remains difficult to ignite until the liquid water remaining in the material ceases to move towards the flame. This would occur when there is insufficient water to fill the interfiber and interyarn capillaries. Consequently, high oxygen concentrations are required in the OI tester so that ignition of fabrics of high MC can take place.

TABLE II

Value of Constants [Z and $(OI)_0$] in Equation[a] Relating Oxygen Index (OI) and Moisture Content (M) for Various Flame Retardant Cotton and Wool Fabrics

Fabric type[b]	Flame retardant	DF[c]	$(OI)_0$[d]	Z[d]	R[e]	DF[c,f]	$(OI)_0$[d,f]	Z[d,f]	R[e,f]
Cotton fabric									
PC and TL	None	14	16.9 ± 0.33	0.26 ± 0.02	0.96	10	−4.10 ± 5.5	0.98 ± 0.10	0.91
PC	Pyrovatex CP	4	28.9 ± 0.51	0.97 ± 0.08	0.99	—	—	—	—
PC	THPCl–Urea	4	28.1 ± 0.89	0.92 ± 0.11	0.99	—	—	—	—
PC	THPOH–Amide	4	29.0 ± 0.69	0.98 ± 0.10	0.99	—	—	—	—
PC	THPOH-NH_3	4	25.4 ± 0.39	1.01 ± 0.05	1.00	—	—	—	—
TL	THPOH–NH_3	4	26.3 ± 1.01	0.73 ± 0.09	0.99	—	—	—	—
SN	THPOH–NH_3	4	26.0 ± 1.87	0.82 ± 0.15	0.97	—	—	—	—
Wool fabric									
SE	None	3	24.0 ± 0.14	0.20 ± 0.01	1.00	5	24.9 ± 9.7	0.29 ± 0.25	0.56
SE	HFZ[b]	5	29.9 ± 1.19	0.43 ± 0.08	0.95	—	—	—	—

[a] $OI = ZM + (OI)_0$.
[b] Code given in Table I.
[c] Degrees of freedom.
[d] Standard error of regression coefficient given also.
[e] Correlation coefficient.
[f] Linear portion at high MC.

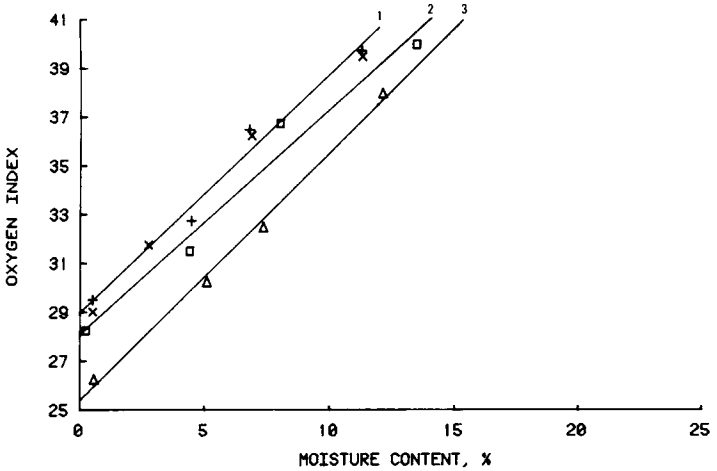

Fig. 2. Relation between oxygen index and moisture content for cotton printcloth treated with THPOH–amide (+), Pyrovatex CP (x), THPCl–urea (□), or THPOH—NH_3 (△): (1) Pyrovatex and THPOH–amide; (2) THPCl–urea; (3) THPOH—NH_3.

Again, when a flame is held to a fabric with an MC less than the FSP of the fiber, the heat will initially cause any water in the fibers of the fabric in the vicinity of the flame to evaporate as thermal energy is absorbed. However, in this case, when the fabric is dry in the vicinity of the flame, water trapped inside fibers further away from the flame will not be able to diffuse as easily toward the flame as there are no continuous capillary channels containing liquid water between the fibers or yarns. Thus, the concentration of oxygen in the OI tester does not need to be as high to cause fabric of low MC to burn.

The OI value of the non-FR cotton samples is approximately 26 (Fig. 1), which is close to $(OI)_0$ of the FR printcloth fabrics (Table II) when the rate of increase

TABLE III
Water Retention Values (WRV) of Samples Prepared from Various Fabrics

Fabric type[a]	FR	WRV[b,c]
Cotton sample		
PC	None	47.1
TL	None	42.9[d]
PC	Pyrovatex CP	41.2
PC	THPCl–urea	34.7
PC	THPOH–amide	34.8
PC	THPOH–NH_3	42.2
TL	THPOH–NH_3	39.1[d]
SN	THPOH–NH_3	38.3
Wool sample		
SE	None	45.0
SE	HFZ[a]	41.3

[a] Code given in Table I.
[b] g water/100 g dry sample.
[c] WRV of PC fabrics taken from ref. 11.
[d] Ref. 12.

of OI with MC becomes more rapid. Also, the rate of increase of OI with MC for the cotton fabrics above an MC of 33% becomes similar to that of the FR printcloth fabrics (Table II). It can be deduced from these observations that moisture is having a similar retardation effect on burning on both the non-FR (MC > 33%) and FR samples, indicating that water is reducing flammability not by entering into solid and/or gaseous phase reactions in the FR samples but by absorbing thermal energy because of the endothermic process of heating and vaporization. It should be noted that the water is held differently in the FR samples. The moisture uptakes of the FR samples, expressed as regains, were less than 18% when OIs of 40 were approached (Fig. 2). These regains are far lower than the WRV of the samples (Table III). However, the finishes are hygroscopic and are partly located on fiber and yarn surfaces.[11] Thus, water could be transferred towards the heated end of the fabric through the FR finish present on fiber surfaces.

To determine whether FR add-on and fabric weight affected the relation between OI and MC, a further set of experiments was made on heavier weight cotton fabric (i.e., TL and SN) treated with sufficient THPOH–NH_3 to pass the FF3-71 sleepwear standard. Linear relations were found for both these fabrics, and since the slopes and intercepts were not significantly different (Table II), both sets of data were combined to compare with that of the THPOH–NH_3-treated PC fabric in Figure 3. The slope of the regression curve for the PC fabric differs from that of the SN fabric (Table II), and these fabrics have similar THPOH–NH_3 add-ons (Table I). Thus, fabric weight can affect this parameter. Since the slope of non-FR treated PC cotton at MC < 15% differs markedly from that of the THPOH–NH_3-treated PC fabric of similar MC (Table II), it is assumed that the slope at low MCs can be affected by the FR add-on also. Changes in fabric weight and FR add-on probably affect the rate function of the MC–OI relation by such parameters as diffusibility of moisture in the system, and distribution of FR in the fabric.

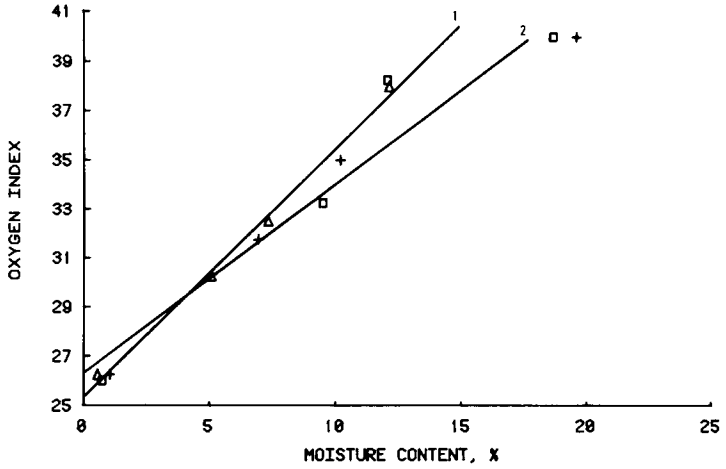

Fig. 3. Relation between oxygen index and moisture content for various cotton fabrics [(△) printcloth, (□) sateen, (+) twill)] treated with THPOH—NH_3: (1) printcloth; (2) sateen and twill.

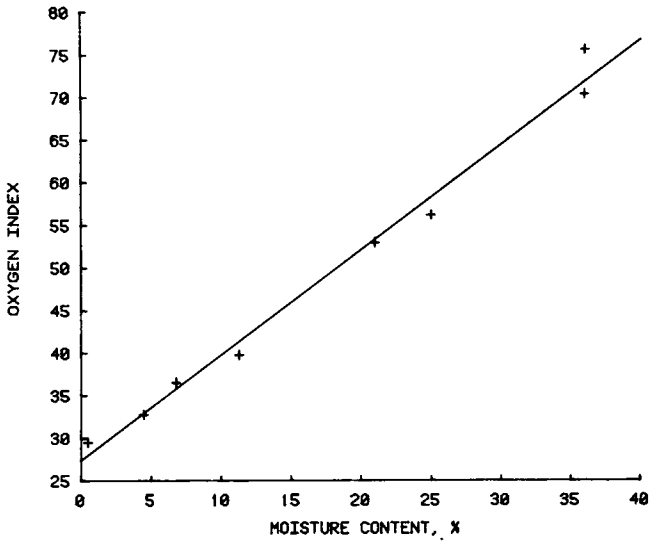

Fig. 4. Relation between oxygen index and moisture content for THPOH–amide-treated printcloth fabric.

To establish the effect of moisture uptakes greater than their WRVs on OI of FR samples, additional experiments were conducted on THPOH–amide-treated PC fabric and THPOH–NH_3-treated TL fabric. The WRV of these samples expressed as moisture contents is 25.8 and 28.1%, respectively. The OI–MC relation for the THPOH–amide-treated PC fabric appears to be linear up to the MC limit tested (Fig. 4). In contrast, the OI–MC relation for the THPOH–NH_3-treated TL fabric contains an inflection at an MC of roughly 20% and then continues as a straight line with a slope approximately similar to that of the initial straight line portion (Fig. 5). Thus, it appears that WRV for cotton

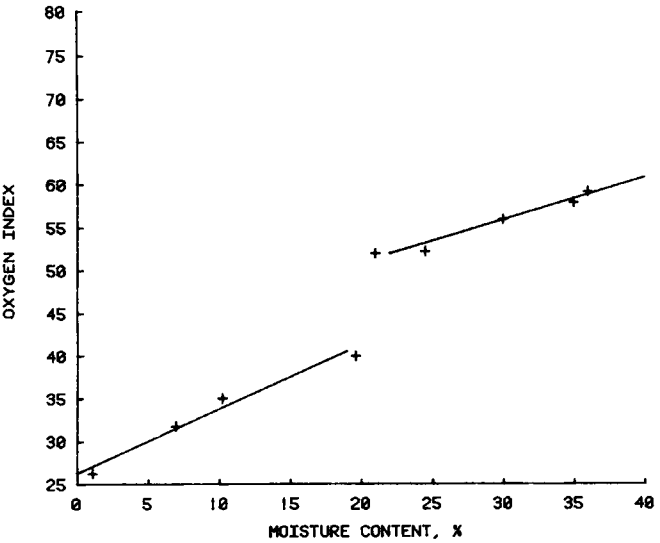

Fig. 5. Relation between oxygen index and moisture content for THPOH–NH_3-treated twill fabric.

fabrics treated with FR finishes does not define a change in slope in the OI–MC relation as clearly as it does for nontreated cotton fabrics. The diffusibility of water in an FR-treated cotton fabric at MC greater than the WRV appears to be affected by the type of finish applied as well as fabric weight, FR add-on, and FR distribution in the fabric.

We have shown in an earlier study[11] that at a 59% relative humidity and 21°C the desorption moisture regain of the Pyrovatex CP-, THPOH–amide-, THPCl–Urea-, and THPOH–NH_3-treated PC cotton samples are 7.35, 8.37, 8.48, and 9.77%, respectively. These fabrics are equivalent in that all pass the FF3-71 standard and have roughly similar FR add-ons. $(OI)_0$ for these materials decreases approximately in this order (Table II), supporting the suggestion of Drake et al.[2] that OI is related to the inherent hydrophilicity of the fabric. Drake et al. have stated that for FR cotton fabrics, noncrosslinking flame retardants pick up more moisture at relative humidities up to 65% than those treated with crosslinking types.[2] These workers have stated that up to about 10% moisture content the OI values for fabric treated with a crosslinking finish will be higher than that of the noncrosslinked finished fabric, and they imply that it may be related to the inherent hydrophilicity of the fabric. Our findings do not support the suggestion that crosslinking may be involved in this relation. Our data[11] indicates that THPCl–urea- and THPOH–amide-treated PC fabrics are heavily crosslinked in comparison to THPOH–NH_3- and Pyrovatex CP-treated PC fabrics which may be crosslinked but to a lesser extent. We quoted the moisture regains for these materials above and their $(OI)_0$ values are given in Table II. It can be seen then that there does not appear to be a correlation either between moisture regain and degree of crosslinking or between $(OI)_0$ and degree of crosslinking. Although an empirical relation between the inherent hydrophilicity, as determined by desorption regain at 59% relative humidity and 21°C of an FR fabric, and $(OI)_0$, independent of degree of crosslinking, is indicated by our data, we are unable to explain the relation in terms of a molecular degradation mechanism.

Relation Between OI and MC for Wool Fabrics

Measurements were also made on untreated wool and the same fabric treated with HFZ. The OI–MC relation for these fabrics (Fig. 6) followed the same pattern as that for the cotton fabrics. For untreated wool, OI increases slowly, but at a constant rate until an MC of 25–35% is reached. The OI then increases more rapidly but still appears to remain linear. The regain equivalent to an MC of 25–35% is 33–54%, which brackets the WRV for wool (45%). The slope of the line for nontreated wool at high MCs is intermediate between that of nontreated wool at low MCs and that of HFZ wool (Table II). The change in slope of the OI–MC relation of untreated wool at high MCs can be explained in terms similar to that given above for untreated cotton fabric.

It is interesting to note that a comparison of the gradient of the OI–MC relation for untreated cotton fabric and untreated wool fabric indicates that moisture has less effect on the OI of wool. A similar observation can be made with FR-treated samples. Namely, moisture has less effect on the OI of FR-treated wool fabric than on the OI of FR-treated cotton fabric.

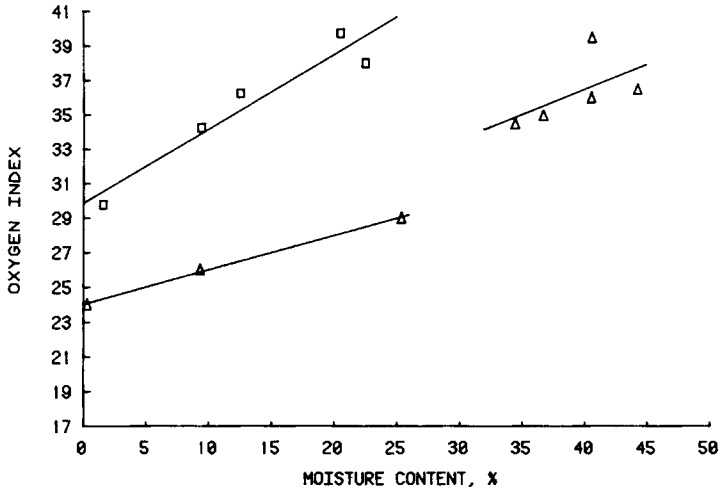

Fig. 6. Relation between oxygen index and moisture content for wool fabrics: (△) untreated wool fabric; (□) wool fabric treated with potassium hexafluorozirconate.

Relation Between $(OI)_0$ and Thermal Analysis Data

Since it requires a relatively large amount of time and material to determine $(OI)_0$, correlations were sought between $(OI)_0$ and parameters that could be determined by thermal analysis. Differential thermal analysis (DTA) curves for the FR cotton samples in an atmosphere of air are given in Figure 7. It will be seen that each curve consists of multiple exothermic peaks. A comparison of the curves for the THPOH–NH_3-treated samples indicates that the shape of the DTA curve can vary for fabrics treated with a sufficient amount of the same FR to pass the FF3-71 sleepwear standard depending on the weight of the fabric,

TABLE IV
Thermal Analysis Data

Fabric type[a]	Flame retardant	CR[b]	Calculated[c] OI	Empirical $(OI)_0$	Zenith of first exothermic peak, °C
Cotton fabric					
PC	None	0	17.5	16.9[d]	—
PC	THPOH–NH_3	34.4	31.3	25.4	284[e]
SN	THPOH–NH_3	33.0	30.7	26.0	311
TL	THPOH–NH_3	30.1	29.5	26.3	300[e]
PC	THPCl–urea	36.4	32.1	28.1	314
PC	Pyrovatex CP	32.8	30.6	28.9	319
PC	THPOH–amide	33.3	30.8	29.0	319
Wool fabric					
SE	None	19.8	25.4	24.0[d]	—
SE	HFZ[a]	38.9	33.1	29.9	—

[a] Code given in Table I.
[b] Char residue.
[c] Calculated from: OI = 17.5 + 0.4 CR.
[d] Initial portion of curve.
[e] Shoulder.

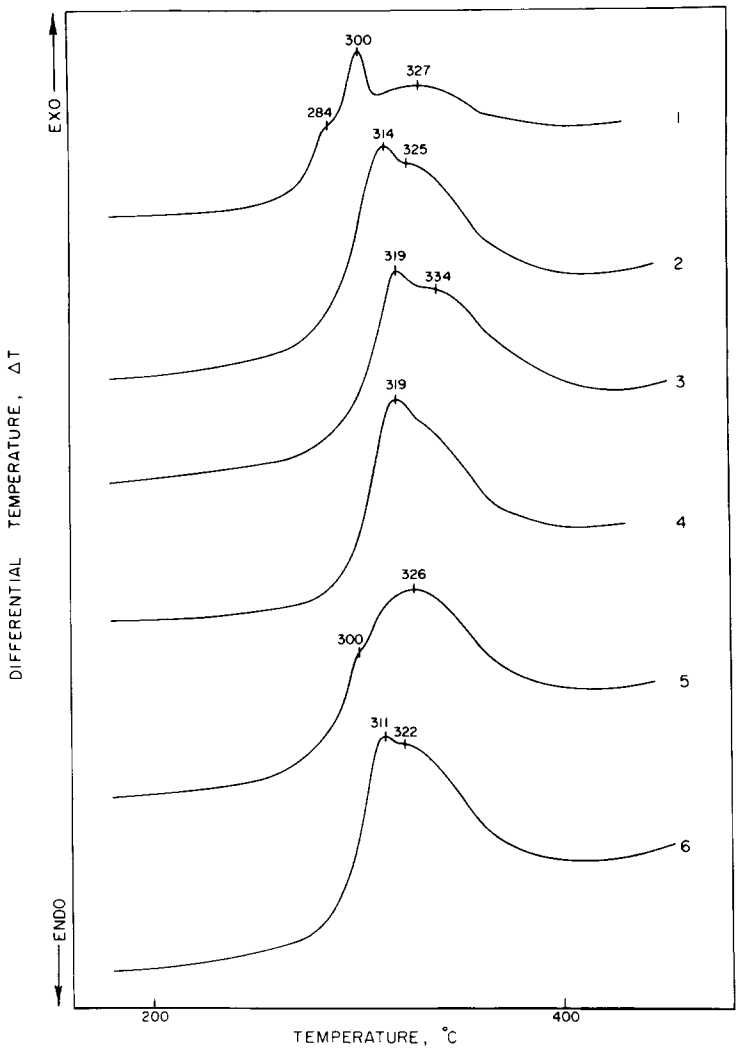

Fig. 7. DTA curves of (1) THPOH—NH_3-, (2) THPCl–urea-, (3) Pyrovatex CP-, and (4) THPOH–amide-treated printcloth fabrics, (5) THPOH—NH_3-treated twill fabric, and (6) THPOH—NH_3-treated sateen fabric.

the amount of FR add-on and possibly the method of application of the FR. There appears to be a correlation between $(OI)_0$ and the zenith of the first exothermic peak for the samples we tested. As $(OI)_0$ increased, the zenith shifted to higher temperatures (Table IV), indicating that the initial decomposition reactions occur at higher temperatures with the samples of higher flame resistance.

Van Krevelen[7] has reported a significant correlation between char residue (CR) and OI for polymers. The equation expressing the relation is OI = 17.5 + 0.4 CR. He has suggested that the same relation can be applied to cellulose when the effect of the FR additive is to have a dehydrating effect. We checked the relation for both our cotton and wool samples. With the exception of the Pyrovatex CP- and THPOH–amide-treated PC fabrics and of the non-FR wool

fabric, our data for CR and $(OI)_0$ (Table IV) did not fall close to the linear relation obtained by Van Krevelen between OI and CR. We used $(OI)_0$ since this is the intrinsic flammability of the fabric not influenced by water. We calculated OI from our CR data using the Van Krevelen equation, and the data is presented in Table IV. It will be noted that in each case the calculated OI is higher than the empirical $(OI)_0$. It appears then that better correlation can be obtained between our data and the data presented by Van Krevelen when our OIs are determined on samples which have MCs between 2 and 10% since the presence of moisture raises OI. From a fundamental viewpoint, however, it would be preferable to develop the OI–CR relation for dry materials. It should be noted that it appears a universal relation between OI and CR can include certain wool samples as well as other polymeric materials.

References

1. "Standard for the Flammability of Children's Sleepwear," *Text. Chem. Color.*, **4**(9), 71 (1972).
2. G. L. Drake, Jr. et al., *Text. Chem. Color.*, **4**, 287 (1972).
3. Natl. Bur. Stand. Tech. Note, U.S. Department of Commerce, **749**, 6 (1972).
4. J. F. Hendrix, G. L. Drake, Jr., and W. A. Reeves, *Text. Chem. Color.*, **5**, 144 (1973).
5. J. E. Hendrix, J. V. Beninate, G. L. Drake, Jr., and W. A. Reeves, *Text. Res. J.*, **41**, 854 (1971).
6. B. Miller, J. R. Martin, B. C. Goswami, and C. H. Meiser, Jr., *Text. Res. J.*, **45**, 328 (1975).
7. D. W. Van Krevelen, *J. Appl. Polym. Sci. Appl. Polym. Symp.*, **31**, 269 (1979).
8. ASTM D 1910–64, *Annual Book of ASTM Standards*, Part 32, Textiles, ASTM, Philadelphia, 1977.
9. S. H. Zeronian and K. E. Cabradilla, *J. Appl. Polym. Sci.*, **16**, 113 (1972).
10. I. Hardin, A. J. Treece, S. H. Zeronian, R. L. Galbraith, and B. R. Farthing, *J. Consum. Prod. Flammabil.*, **4**, 121 (1977).
11. J. Chandler and S. H. Zeronian, *Text. Chem. Color.*, **11**, 56 (1979).
12. J. Chandler, M. S. thesis, University of California, Davis, 1977.
13. A. M. Scallan and J. E. Carles, *Sven. Papperstidn.*, **75**, 699 (1972).
14. J. M. Preston and J. C. Chen, *J. Soc. Dyers Colour.*, **64**, 60 (1948).

Received October 3, 1979
Revised December 4, 1979