STUDY OF THE ENVIRONMENTAL DEGRADATION OF CIGARETTE FILTERS: A Simulation of the Roadside or Parking Lot Environment.

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In recent years, there has been increasing public awareness of items which may be discarded as litter with particular attention being given to cigarette filters (1). Some studies have concluded that when cigarettes filters are introduced into the environment, they either do not degrade or degrade very slowly (2). Previous work in our laboratories has demonstrated that cigarette filters biodegrade readily in environments where mixed microbial populations can thrive (3). The purpose of this study was to examine the changes occurring in cigarette filters over time in an environment similar to that of 2 parking lot or roadside area. In this environment, exposure to sunlight, moisture and wind occur, but there is limited exposure to microbial attack. Two items of special interest to us were the effect of the triacetin plasticizer that is used to provide firmness to the filter, and the influence of the tipping paper and plug wrap. The triacetin works as a solvent bonding plasticizer which softens the fibers and welds them together into a cross-linked structure. One concern had been that this cross-linked structure might prove difficult to disperse and could slow the degradation of the filter. The tipping paper and the plug wrap are paper elements which provide some measure of protection from environmental exposure to the cellulose acetate filter. Previously, we had been unable to measure the extent of this protection and its effect on the degradation of the filter material.

We chose to simulate the roadside or parking lot environment by utilizing the roof top of our Research Laboratory in Kingsport, Tennessee. This asphalt covered, flat roof provided the necessary exposure to sunlight, rain, and extremes of temperature while providing a secure, restricted area over the course of many months. During weathering, the filters were placed in baskets constructed of stainless steel mesh with 3 X 3 wires per inch. This container allowed exposure to the environment while keeping the samples separate and preventing them from scattering over (or off the roof these baskets did allow the wind to agitate the samples and this provided the only mechanical stress. The baskets were placed on top of supports to keep them above the water that occasionally collects on the roof. This environment approximates the conditions experienced by the filter in the roadside of parking lot environment. These include sunlight, rain, snow, heat and cold, but do not include an abundance of soil, humus, or other materials which one would expect to enhance microbial degradation.

The Effect of Triacetin Plasticizer on Degradation

The simplest way to determine if the presence of plasticizer actually slows the dispersal and degradation of die filter into the environment is to compare the performance of filter tips with plasticizer to filter tips without plasticizer. Both types of filters were prepared, attached to tobacco columns and smoked under standard conditions. After smoking, the tipping paper and plug wrap were removed to eliminate this variable from the process. The samples were placed in the cages and exposed to the roof top environment. Periodically, the samples were brought back into the laboratory, allowed to equilibrate in a controlled humidity environment and weighed before being returned to the roof Figure I shows the relative weight changes of the filters with and without plasticizer over a period of 10 months. If the crosslinking caused by the plasticizer were slowing dispersal and degradation, the weight loss of the unplasticized sample should be more rapid and the final weight should be less. As the results show, this is not the case. Both samples show the same relative weight loss which indicates that the presence or absence of triacetin has no significant effect on the rate or extent of degradation under these conditions. There is, however, a visible difference in appearance between the two samples. The sample without plasticizer has a less defined shape, but is still held together, even without a bonding agent. Although the individuals fibers are not fused or welded together, the high degree of crimping of the fibers creates entanglements which do not readily disperse. While some definition of shape is lost after a period of time, the fibers remain entangled enough to form a mat of fibers (Figure 2). At the end of 10 months of exposure, these filters are still distinguishable in this mat form and have actually degraded no more or less than the plasticized samples.

The Effect of Plug Wrap and Tipping Paper on Degradation Rate

To investigate the influence of the paper wrapping around the cellulose acetate filter tip, three commercial cigarette brands were chosen to be representative of the market. The three brands covered a wide range of ventilation and included a mentholated, full flavor brand. Forty cigarettes of each type were smoked and the filter tips collected. Twenty tips from each brand were stripped of the tipping paper and plug wrap before exposure, while the remainder was left intact. Removal of the paper elements allowed us to distinguish the intrinsic dispersion and degradation of the filter tips from the influences due to a particular cigarette's construction.

These samples were placed on the roof in the cages as described above. Periodically, the filter tips were brought into the laboratory and Bowed to equilibrate in a controlled humidity environment, weighed, and placed back on the roof. During this period, a few filters were deformed enough to permit them to escape from the holes in the cages. Average filter tip weights were then based on the remaining filter tips. After 12 months of exposure, the experiment was terminated and the filters were analyzed to determine what changes had occurred in the polymeric structure of the cellulose acetate during the year of weathering. Liquid chromatographic determination of acetyl level and gel permeation chromatographic determination of molecular weight were used to measure chemical changes related to degradation while scanning electron microscopy was used to examine the surface of the filters for a visual estimation of the degradation. Since the core of the filter tip is protected by the outer cellulose acetate fibers on the surface, we checked for gross differences in degradation by analyzing the acetyl level and molecular weights of portions taken from the surface (outside) and from the center of the filter tips (inside).

Figure 3 shows the average filter tip weight for samples of each brand over the course of a year's exposure. The samples which had their paper removed initially show a downward trend in weight due to loss of material during exposure. The samples exposed with paper intact, initially show a more rapid weight reduction due to paper loss, but after all the paper is lost, the remaining cellulose acetate material has the same weight and weight loss rate, as material which has not been protected by a paper covering. Table I presents results after 12 months of exposure. The first column distinguishes the brand of cigarette tested and indicates which samples were originally exposed with the paper wrap intact. It should be noted that by the tenth month, A paper had been lost on the samples which were originally paper covered. The relative weight of the filter is the weight of remaining material after 12 months of exposure, based on the starting weight of the cellulose acetate filter tip (without tipping paper or plug wrap). Since the relative weights with and without paper are equal at 12 months, we see that under these conditions the early loss of paper does not change the extent of degradation.

The percent acetyl measurement tracks the degree of esterification of the cellulose acetate. As the acetate groups are removed, this number decreases and the material eventually would be equivalent to ordinary cellulose wood pulp. The fact that all the samples have a lower average percent acetyl level after 12 months of exposure indicates that acetate groups are being removed. In Table 2, changes to the cellulose acetate are tracked by measuring the number average molecular weight and the polydispersity. In this study, samples were taken from the surface of the filter tip and the interior core of the filter to track the differences in exposure and degradation in these regions.

Cellulose acetate is a polymeric material composed of long chains of molecules. The number average molecular weight is an indicator of the average chain length since the chain length is directly proportional to molecular weight. The data indicate that the average polymer chain is shorter on the surface than in the core, showing degradation probably resulting from photolysis. The average chain length in the core is also shorter than in the starting material, which indicates some degradation is occurring even in this protected area. This data set also shows that the early removal of paper does enhance the breakdown of the polymeric chains in all cases although the extent of this effect is insufficient to increase dispersion and weight loss in these samples.

Polydispersity is a measure of the variance in chain lengths of a polymer. If all of the molecules in a polymer had chains of exactly the same length and molecular weight, the polydispersity would equal 1.0. Most polymeric materials tend to have chains of varying lengths. The initial polydispersity values for cellulose acetate reflect this. As the variation in the lengths of the chains increases, the polydispersity measure gets larger. When a polymeric sample is degraded, the chains are broken at random points to produce shorter chains with a variety of lower molecular weights. This mixture of short and long chain polymers produces a higher number for polydispersity. A further indication of the degradation of the filter tips during weathering is the increased values for polydispersity shown in Table 2.

Scanning electron micrographs, Figures 4 through 6, show the highly magnified outer surface of the filters after exposure. It is interesting that the samples exposed with paper attached seem to show more obvious surface pitting, even though the weight loss was equivalent for the samples regardless of the presence of paper. The presence of the paper may cause this effect by holding moisture which speeds hydrolysis or by providing a starting point for biodegradation, but these effects are insufficient to result in significant loss of the filter mass.

CONCLUSIONS

Previous work has demonstrated that cigarette filters biodegrade readily in environments where mixed microbial populations can thrive (3). This work demonstrates that current commercial cigarette filters also degrade when exposed to an environment which is not optimal for microbial biodegradation.
The process is somewhat slow but can be measured by both chemical and physical techniques. The process is probably composed of a mixture of mechanisms involving photolysis, hydrolysis, and microbiotic attack.

The evidence indicates that the triacetin plasticizer does not inhibit either the dispersal or the degradation of the filter. Therefore an alternative, non-bonding plasticizer would be of little aid in increasing filter dispersion. The effect of early removal of tipping paper and plug wrap is somewhat mixed. Although the early removal of the plug wrap does facilitate the breaking of the polymeric chains on the exposed surface of the filter, the extent of this process is not sufficient to result in greater weight loss or increased degradation under the conditions of this experiment.

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3. The Fate of Cellulose Acetate in the Environment: Aerobic Biodegradation of Filter Tow Fibers. C.M. Buchanan, R.M. Gardner, R.J. Komarek and D.V. Strickler, Eastman Chemical Co. Paper presented at 45th Tobacco Chemist's Research Conference, Asheville, NC. 1991.

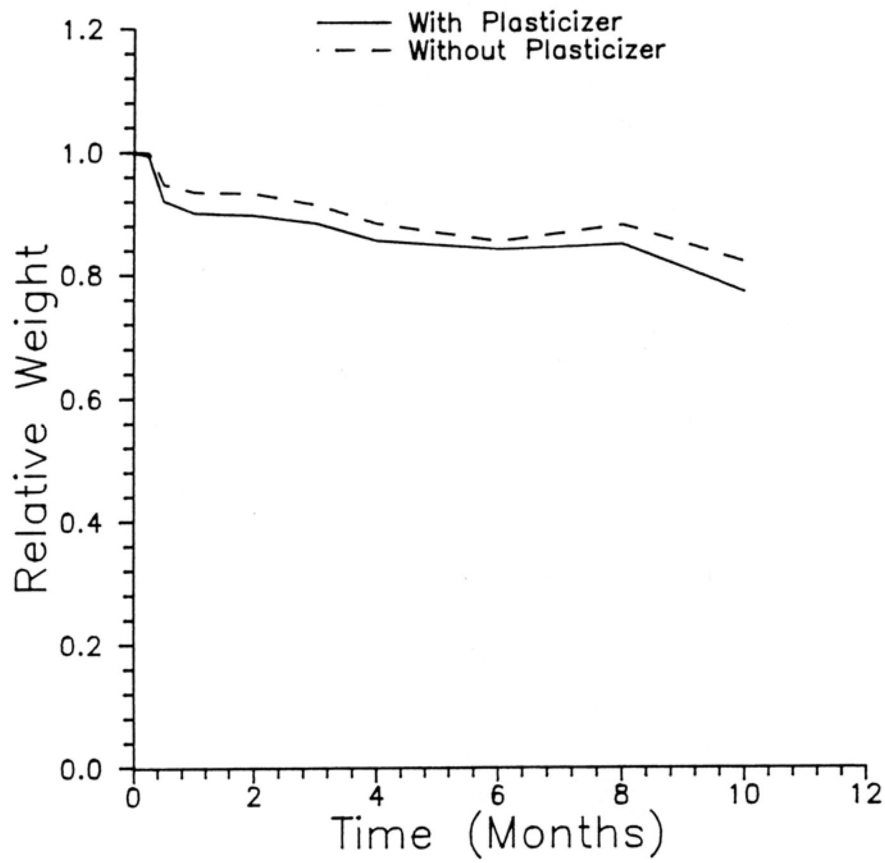

Figure 1. Weight Loss of Filter Tips with and without plasticizer.

Filters with Plasticizer **Filters without Plasticizers**

WEATHERED 10 MONTHS

Figure 2. Effect of Plasticizer on Appearance of Filter Tip.

Effect of Paper Wrap on Degradation

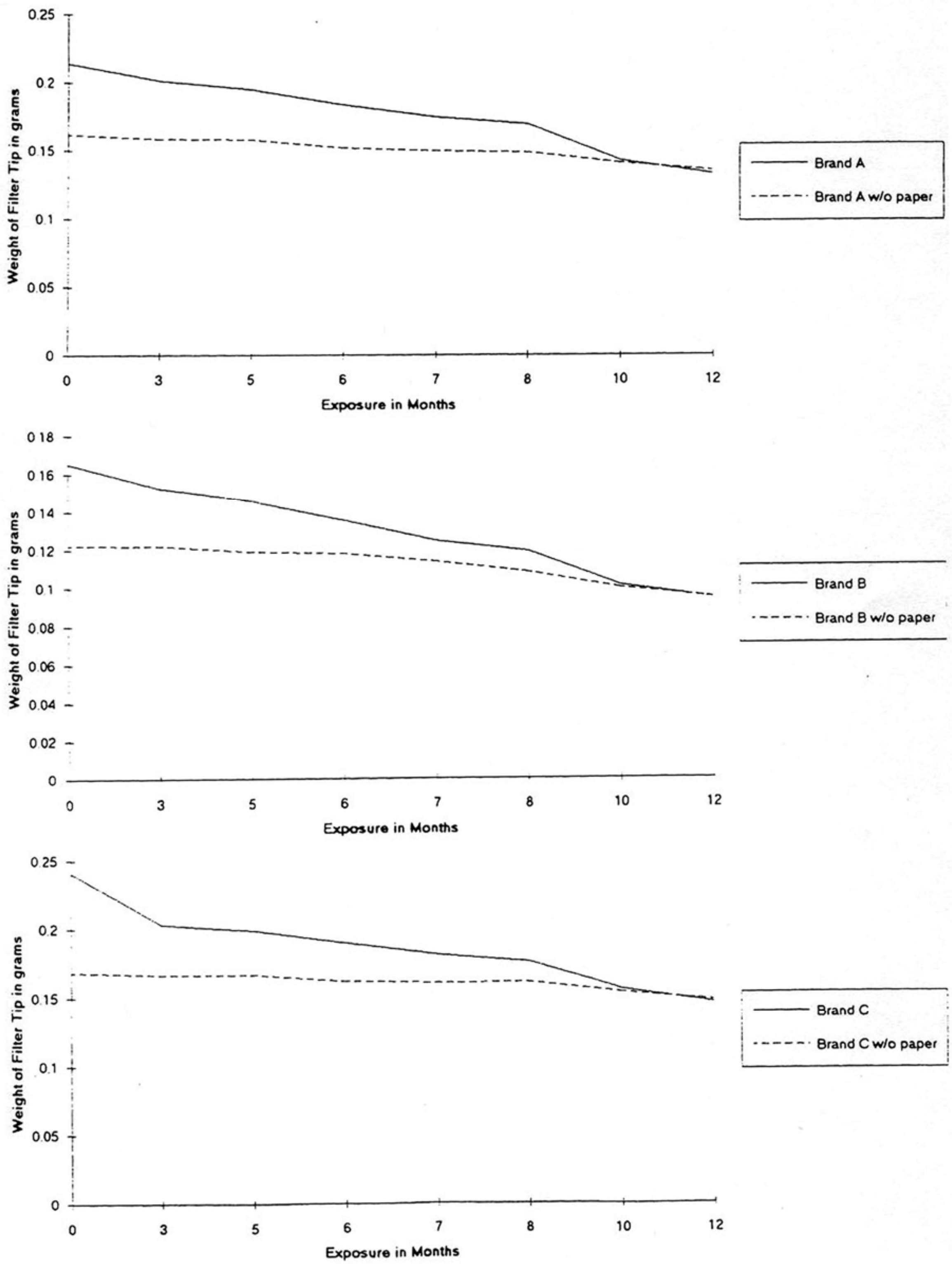

Figure 3. The effect of paper wrapping on filter tip weight loss during weathering.

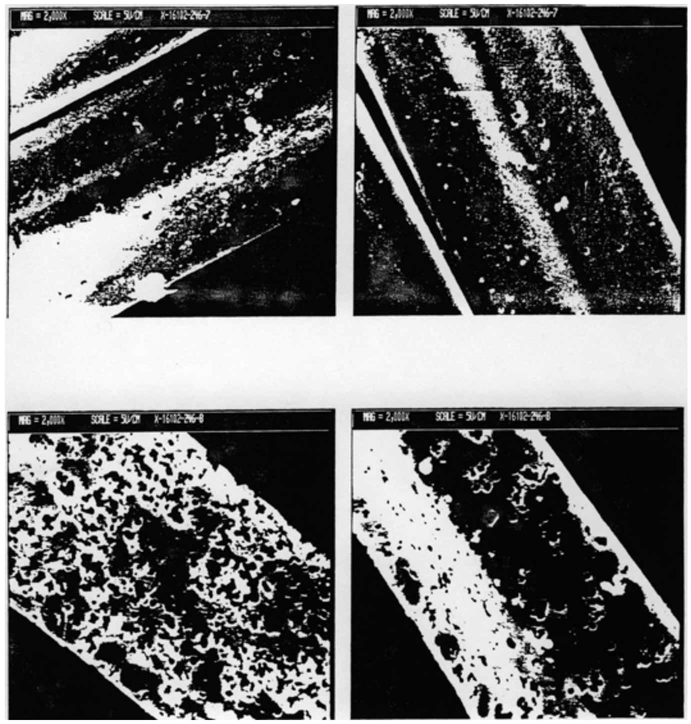

Figure 4. Brand A showing the appearance of the filter tip surface which had the tipping paper and plug wrap removed before exposure to the environment (top) and the appearance when the paper tipping and plug wrap are allowed to weather away naturally (bottom).

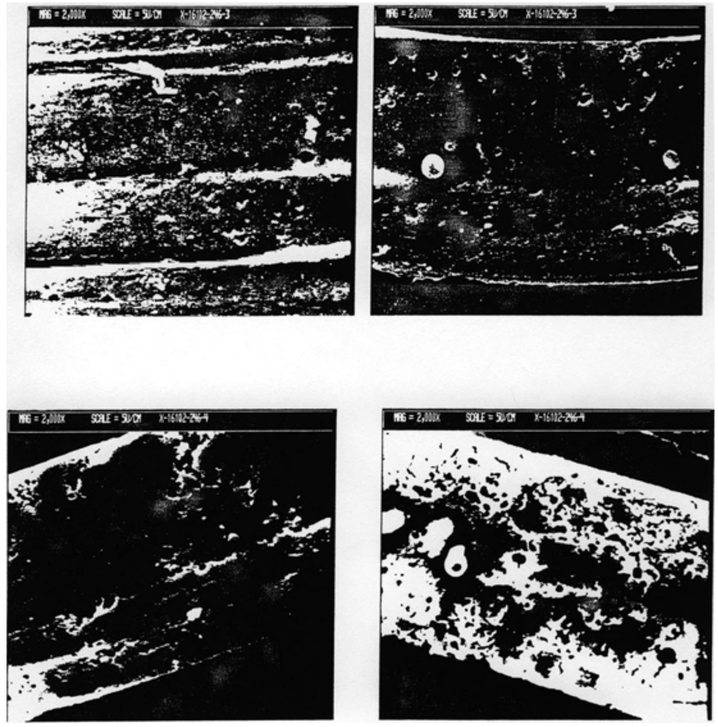

Figure 5. Brand B showing the appearance of the filter tip surface which had the tipping paper and plug wrap removed before exposure to the environment (top) and the appearance when the paper tipping and plug wrap are allowed to weather away naturally (bottom).

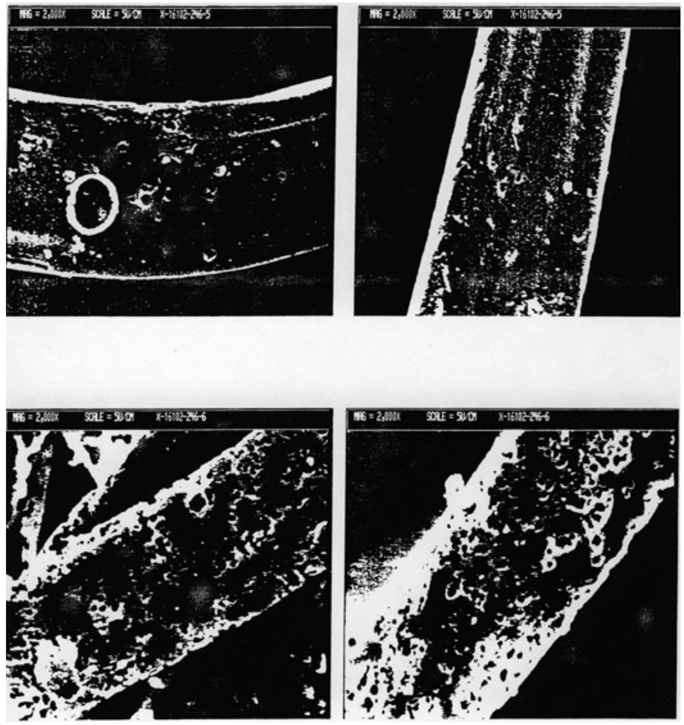

Figure 6. Brand C showing the appearance of the filter tip surface which had the tipping paper and plug wrap removed before exposure to the environment (top) and the appearance when the paper tipping and plug wrap are allowed to weather away naturally (bottom).

Table 1. Changes in filter weight and acetyl content during weathering.

Cigarette	Relative Wt. of Filter Tip** without Paper after 12 Months Exposure (Initial Weight = 1)	Initial % Acetyl Content	Average % Acetyl Content after 12 Months Exposure
Brand A		39.2	
w/paper *	0.82		38.6
paper removed before exposure	0.83		38.2
Brand B		39.2	
w/paper *	0.77		38.5
paper removed before exposure	0.77		37.7
Brand C		39.1	
w/paper *	0.87		38.8
paper removed before exposure	0.87		38.7

* These filter tips started with the paper attached, but all paper had weathered away before 10 months had passed.

**Results are based in most cases on an average of 20 filter tips. In a few instances, the filter degraded to the point where it was lost through the holes in the cage. Results are based on remaining filters in these cases.

Table 2. Molecular weight and polydispersity changes to the exterior and interior of filter tips after weathering.

	Position of Cellulose Acetate in Filter Tip	Initial Number Average Molecular Weight	Number Average Molecular Weight after 12 Months Exposure	Initial Poly-dispersity	Polydispersity after 12 Months Exposure
Brand A		66,100		2.77	
w/paper					
	inside		60,300		2.46
	outside		40,000		3.19
w/o paper					
	inside		55,800		2.68
	outside		24,300		2.98
Brand B		67,400		2.53	
w/paper	inside		55,400		2.72
	outside		34,500		3.04
w/o paper	inside		43,800		3.03
	outside		30,100		3.19
Brand C		71,500		2.47	
w/paper	inside		63,400		2.51
	outside		44,300		2.60
w/o paper	inside		61,100		2.61
	outside		37,000		3.42

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