Wool Cigarette Filters

Part II: Modifications to Promote the Selective Removal of Biologically Active Vapour Phase Components*

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In the past two decades the carcinogenic activity of cigarette-smoke condensate has been indicated by numerous experiments with animals and animal tissues. These findings have led to the wide acceptance of fibrous cigarette filters that reduce the delivery of particulate matter (PM). More recently, attention has been focused on the biological activity of the vapour-phase (VP) components of cigarette smoke. VP components are associated with increased pulmonary carcinogenesis (1), abnormal growth of mammalian cells in culture (2), ciliatoxicity (3, 4, 5), immuno-suppression (6), and the inhibition of sulphydryl-containing enzymes (7–9). In this respect, filters that reduce the delivery of VP components as well as PM are desirable.

Fibrous filters are specifically designed to retain some PM by mechanical filtration and, generally, are incapable of retaining VP constituents. However, by the use of certain additives, fibrous filters can be modified to remove some of the VP components. Numerous examples of such modified filters have been described in the patent literature (10), but very few have been used commercially.

Additives that have been employed in fibrous filters to enhance the retention of specific VP components include solid adsorbents, liquids of low volatility and compounds that chemically interact with smoke constituents. Solid adsorbents, such as charcoal, are the best known and most effective additives but have the

disadvantage of poor selectivity. Liquid additives, such as triacetin, that are employed to harden cellulose acetate filters, remove volatile and semivolatile components by solvent adsorption. However, filters containing liquid additives retain only a limited range of components and deteriorate in performance over a short period (11). Another disadvantage of solid and liquid absorbents is the tendency for some of the absorbed VP components to be released as the filter temperature increases during smoking. The third type of additive incorporated in filters promotes chemisorption of specific VP components. Such filters chemically bind volatile components to the filter material and are usually highly selective.

Wool in the form of an adhesive-bonded random web was shown previously to provide filters with acceptable firmness and pressure drop without the addition of plasticizers or hardeners (12). For this reason, wool is a particularly suitable substrate for the application of filter additives. In the present work, filters prepared from wool modified chemically or by the application of additives were evaluated for their effectiveness in selectively retaining specific biologically active VP components by chemisorption. Additives were selected on the basis of their chemical reactivity with the weak acids or aldehydes shown in Table 1, the biological effects of which have been well documented (2–5, 8, 9, 13, 14). Basic additives were employed to promote the selective removal of the weakly acidic components and additives containing amino, sulphydryl or hydroxyl groups were utilized to retain the volatile aldehydes.

Table 1. Biologically active volatile and semivolatile components of tobacco smoke that may be selectively removed by chemisorption.

Smoke components	Phase*	Chemical classification	Biological activity
Formaldehyde Acetaldehyde Acrolein	VP	Reactive carbonyl compounds	Ciliatoxic (4,5), cytotoxic (2,13), and enzyme inhibiting (8,9)
Hydrogen cyanide Hydrogen sulphide	VP	Weak acids	Ciliatoxic (3,5) and enzyme inhibiting
C_1-C_2 carboxylic acids Fatty acids Phenols	VP/PM	Weak acids	Tumour promoting (14) and ciliatoxic (5)

* VP = vapour phase.   PM = particulate matter.

Hygroscopic additives were also investigated as a means of facilitating the adsorption of these water-soluble smoke components.

MATERIALS AND METHODS

Materials

Either combed top (11 g/m) prepared from scoured Merino wool (60–64's/22.0 μ) or an adhesive-bonded random web (60 g/m²) prepared from carbonized peroxide-bleached Merino wool (60–64's/22.0 μ) was employed as the filter material. The polyethylenimines used as filter additives were commercial grades as follows: PEI 6 (MW 600), PEI 18 (MW 1800), PEI 600 (MW 40 to 60,000), PEI 1000 (MW 50—100,000), PEI 1090 [Dow Chemical]; Polymin P, and Polymin SN [BASF]. Poly(ethylene glycols) were commercial grades: PEG 200 (MW 200) and PEG 1000 (MW 1000) [ICI]. Poly(styrene potassium sulphonate) (PSPS) and poly-(ethylene sodium sulphonate) (PESS) were synthesized. The ionene polymer, tetramethyl-2,3-ionene bromide, was prepared from N,N,N',N'-tetramethylethylenediamine and 1,3-dibromopropane. Other filter additives were laboratory-reagent grade. Standard machine-made tobacco columns were employed to evaluate filter performance and were conditioned at 60% relative humidity for 48 h prior to smoking.

Chemically Modified Wools

Wool was esterified under anhydrous conditions with methanol containing hydrogen chloride (0.1 M) (15). Reduced wool was prepared by treating top (75 g) with thioglycollic acid (0.5 M, pH 4.8) at 50° C for 3 h. Tributylphosphine-reduced wool was obtained by the method of *Sweetman* and *MacLaren* (16). Wool top was chlorinated by treatment with sodium hypochlorite (1.6% Cl_2) at pH 4, washed with sodium bisulphite and rinsed with water. Oxidation of chlorinated top was accomplished by treatment with peracetic acid (3.1%) for 5 min.

Wool with Internally Deposited Polymers

Wool top was impregnated with ferrous ammonium sulphate (0.1%) and air-dried (17). A portion of the top was heated (3 h) at 50° C in a solution containing water (1.8 l), methacrylic acid (100 g), sulphuric acid (0.5 M, 100 ml) and 20 vol. hydrogen peroxide (0.2 ml) to give an increase in weight of 25.3% after washing and air-drying. Another portion of the top was heated (19 h) at 50° C in water (110 ml) containing ethylenimine (10 ml) and 2 vol. hydrogen peroxide (80 ml) to give a weight increase of 8%. A third portion of top was similarly treated with water (160 ml), 2 vol. hydrogen peroxide (4 ml), oleic acid (2 ml) and dimethylaminoethyl methacrylate (16 ml) at 50° C for 5 h to give a weight increase of 5.2%.

Application of Additives to the Filter Material

Aqueous solutions (200 ml) of the additives containing the surfactant Antarox CO 630 [GAF] (10%, 0.2 ml) were poured into the nip of a Peter pad-mangle. Slivers of top or bonded web were passed through the pad and squeezed to about 100% expression. The treated filter material was dried (10 min) at 90° C in a forced-draught oven and equilibrated at 65% relative humidity for 24 h before fabrication into filters. The quantity of additive applied to the slivers was determined by accurately weighing them before and after the expression of pad-liquors and was varied by adjusting the concentration of the initial solutions. The pH of the polyethylenimine (PEI) solutions was adjusted by stirred additions of concentrated hydrochloric acid or sodium hydroxide (1 M).

Preparation of Filters

Each sliver was carefully drawn through hollow paper tubes (100 mm) prepared from commercial cigarette paper and sealed with adhesive tape to give filter rods of diameter 7.5 mm. The rods were squarely cut to give 20 mm filter plugs. The moisture content of the filters was adjusted prior to testing by equilibrating in atmospheres of 65%, 80% or 93% relative humidity for 48 h. Filter pressure drops were determined with a water manometer at a rate of flow of air of 17 ml/s. Filters required for assays were chosen in the pressure-drop range 5.6—7.0 cm of water. The pH of treated filters was determined by shaking the filter material (1.0 g) with distilled water (20 ml) for 1 h, decanting the liquors and measuring with a pH meter.

The Determination of Moisture Content (Regain)

The paper wrappers from three filters were removed and the plugs accurately weighed in a weighing bottle. The weighing bottles were placed in a tube and heated at 100° C in vacuo (0.05 mm of Hg) for 1 h. On cooling, the weighing bottle and contents were reweighed. Percentage regain was calculated from the wet (A) and dry (B) weights of the wool by the following expression:

$$\text{Regain (\%)} = \frac{A-B}{B} \times 100 \; .$$

Smoking Procedure

Smoking was accomplished with a CSM 100 automatic machine using the standard conditions specified by *Coresta* (18). A CM-113 Cambridge filter pad (44 mm) and assembly were employed to separate the PM and VP. Filters were attached to the tobacco columns with adhesive tape prior to smoking. PM was collected on the Cambridge filter pads whilst specific VP components were trapped in two scrubbing bottles, containing the appropriate reagent, that were placed between the cigarette and smoking machine. The smoking machine was adjusted to the correct smoking conditions with the collection apparatus attached. Cigarettes were smoked to a constant butt length (27 mm) and a clearing puff was taken in between cigarettes.

Paramecia Bioassay

The method employed was similar to that described by *Weiss* (19), the VP of cigarette smoke being passed through a chamber in which was suspended a drop of buffered culture containing *Paramecium aurelia*. The protozoa were viewed microscopically, the number of puffs required to immobilize the entire colony being recorded. The exposure chamber was connected to a CSM 10 smoking unit programmed to take 17.5 ml puffs of 1 s duration at 1 min intervals. A three-way solenoid connected in front of the chamber enabled a 17.5 ml clearing-air puff to be taken 15 s after the cigarette puff.

Chemical Assays

Hydrogen cyanide was collected by passing whole smoke through scrubbing bottles containing sodium hydroxide (1 M) and determined spectrophotometrically as described by *Aldridge* (20). The chromotropic acid procedure (21) was employed to determine formaldehyde collected by passing the VP through scrubbing bottles containing sodium metabisulphite (1 %). Acetaldehyde in the VP was collected in gas-scrubbing bottles containing cold water ($1°$–$2°$ C) and determined by gas-solid chromatography (22). Acrolein in the VP was trapped in gas-scrubbing bottles containing cold ($1°$–$2°$ C) 95 % ethanol and determined spectrophotometrically using the n-hexylresorcinol method (23). For the determination of total steam-volatile phenols, the Cambridge filters were extracted with sodium hydroxide (0.1 M, 10 ml) and acidified with 0.2 M hydrochloric acid (25 ml) and the mixture was steam-distilled until 250 ml of distillate was collected. 4-Aminophenazone was used to determine phenols in an aliquot of the distillate (24). Total steam-volatile acids were determined by titration with standard alkali (25) after collection by steam distillation as described for total phenols.

RESULTS AND DISCUSSION

Bioassays utilizing ciliated protozoa, such as *Paramecia*, provide a rapid means of assessing the delivery of cytotoxic smoke components, in particular formaldehyde, acetaldehyde and acrolein (13). Thus, the effectiveness of a filter in retaining these compounds is reflected in the bioassay result. In this work, the bioassay was employed for the initial screening of filter treatments. Filters that gave a significant reduction in the VP cytotoxicity were subjected to broader assessment using chemical assays.

Chemically Modified Wools

As indicated by the *Paramecia* survival times in Table 2, filters fabricated from chemically modified wool or from wool containing internally deposited polymers, failed to retain significant quantities of the VP cytotoxic components. By comparison, filters containing activated charcoal substantially reduced the delivery of cytotoxic

Table 2. The effect of filters containing chemically modified wools on the level of cytotoxic components in the vapour phase of cigarette smoke.

Filter type	Paramecia survival time* (number of puffs)
Untreated wool	21
Cellulose acetate**	21
Cellulose acetate/charcoal**	26
Chemically treated wool	
Esterified	17
Thioglycollic-acid reduced	20
Tributylphosphine reduced	18
Chlorinated	17
Chlorinated/oxidised	19
Polymer-modified wool	
Polymethacrylic acid	21
Polyethylenimine	20
Poly(2-dimethylaminomethacrylate)	20

* Mean of 4 determinations. ** Commercial 20 mm filters.

smoke components. Although wool modified by esterification, reduction or the graft polymerization of ethylenimine contains nucleophilic groups capable of interacting with aldehydes, their distribution throughout the fibre would be dependent on its complex morphology and largely unknown protein structure. Obviously permeation of the fibre surface by these volatile components with subsequent chemical binding by reactive groups within the fibre is not a significant factor in their removal. Apparently, the velocity of the smoke stream is too high for this relatively slow process to be effective. To facilitate the chemisorption of VP components it appears necessary to provide adequate contact between the smoke stream and a high concentration of reactive groups. This can be achieved only by applying appropriate additives to the surface of the wool fibres.

Filters Containing Additives

At moisture regains below 15 %, bioassay results with the low-molecular-weight additives given in Table 3 were essentially the same as those obtained for untreated control filters. In contrast, filters containing the polymeric additives in Table 4 in most cases gave longer *Paramecia* survival times, thus indicating selective removal of some cytotoxic VP components. Examination of the treated fibres by electron microscopy revealed extensive surface deposits on fibres treated with the polymers but only scattered deposits on fibres containing the low-molecular-weight additives. Not unexpectedly, the latter type of additive penetrates the fibre surface and so does not come into contact with the smoke stream. This undoubtedly explains the poor retention of VP components by wool filters containing low-molecular-weight additives. Clearly, additives for wool filters need to be of relatively high molecular weight

Table 3. The effect of wool filters containing low-molecular-weight additives on the level of cytotoxic components in the vapour phase of cigarette smoke.

Treatment	Quantity of additive (% w/w)	*Paramecia* survival time** (number of puffs)		
		65% r.h.*	80% r.h.*	93% r.h.*
Untreated	—	21	25	23
Na₂CO₃	2.0	23	20	29
Na₂CO₃	5.3	24	29	36
PEG 200	9.0	19	20	25
PEG 200 } Na₂CO₃	9.4 } 4.7	22	34	43
Diethylene glycol	24	17	20	22
Diethylene glycol } Na₂CO₃	26 } 2.6	19	28	40
Sorbitol	20	18	22	25
Calcium chloride	8.7	20	22	30
Diethanolamine	9.9	19	26	40
Triethylenetetramine	5.9	20	24	32

* Filters were equilibrated at 65, 80 and 93% relative humidity for 48 h prior to testing.
** Mean of 4 determinations.

Table 4. The effect of wool filters containing polymeric additives on the level of cytotoxic components in the vapour phase of cigarette smoke.

Treatment	Quantity of additive (% w/w)	*Paramecia* survival time** (number of puffs)		
		65% r.h.*	80% r.h.*	93% r.h.*
Untreated	—	21	25	23
PSPS	6.2	20	21	25
PSPS } Na₂CO₃	6.4 } 2.6	24	22	35
PESS } Na₂CO₃	6.8 } 2.7	21	23	30
Ionene polymer } Na₂CO₃	4.6 } 4.6	25	26	35
PEG 1000	5.0	20	23	28
PEG 1000 } Na₂CO₃	5.0 } 5.2	23	25	32
PEI 1000	5.2	33	38	52
PEI 1000 } Na₂CO₃	5.2 } 2.4	27	32	36

* Filters were equilibrated at 65, 80 and 93% relative humidity for 48 h prior to testing.
** Mean of 4 determinations.

to prevent permeation of the fibre surface. A further advantage of such additives is their low volatility.

Tables 3 and 4 show that in most cases higher moisture levels enhance the efficiency of the treated filters. Because of the hygroscopicity of the additives they occur essentially as aqueous solutions on the fibre surface,

Table 5. The retention of hydrogen cyanide (HCN) and acetaldehyde (CH₃CHO) by filters treated with commercial polyethylenimines.

Polyethyl-enimine	Weight on wool (% w/w)	Removal (%)*		*Para-mecia*** (number) of puffs)	Filter pH
		HCN	CH₃CHO		
PEI 6	3.6	47	18	27	8.7
PEI 18	3.8	46	19	27	8.6
PEI 600	3.8	46	19	27	8.1
PEI 1000	3.2	45	18	26	8.1
PEI 1090	5.2	32	28	33	7.0
Polymin P	3.4	23	19	25	8.4
Polymin SN	3.4	32	16	25	7.3

* With respect to an untreated wool filter.
** Mean of 4 determinations.

or within the fibre, depending on their molecular weight. The marked improvement in performance at higher moisture levels indicates that adsorption by water plays a primary role in the retention of VP components by the treated filters. The undesirable components of smoke listed in Table 1 are all water-soluble and would therefore be preferentially absorbed by treated filters with high moisture levels. Very probably the additives supplement the action of the liquid phase by increasing the rate of adsorption and chemically bind the absorbed components, thus preventing their revolatilization into the smoke stream.

The polyethylenimines (PEI) appeared most effective in retaining cytotoxic components at lower moisture levels and, in consequence, several commercial polyethylenimines were examined for their selectivity by bioassay and chemical assays (Table 5). Differences in the effectiveness of the polyamines are apparently unrelated to the differences in molecular weight. With one exception

Table 6. The effectiveness of PEI-1000-treated wool top filters+ in removing specific undesirable smoke components.

Smoke component	Average concentration in each puff (μg/puff)			Removal* (%)
	Unfiltered	Untreated wool filter	PEI 1000/ wool filter	
Hydrogen cyanide	27.7	24.0	13.2	45
Formaldehyde	4.9	4.1	2.0	51
Acetaldehyde	90	92	75	18
Acrolein	7.2	7.4	6.3	15
Total phenols (s. v.)++	18.3	7.4	5.7	23
Total acids (s. v.)++	100	62	49	21

* Of the PEI-1000-treated filter with respect to the untreated filter.
+ Equilibrated at 65% relative humidity.
++ Steam-volatile.

Figure 1a. The effect of moisture content on the retention of hydrogen cyanide and acetaldehyde by PEI-treated wool filters (hydrogen cyanide X, acetaldehyde ●, filter material: combed wool top —, adhesive-bonded web - - -).
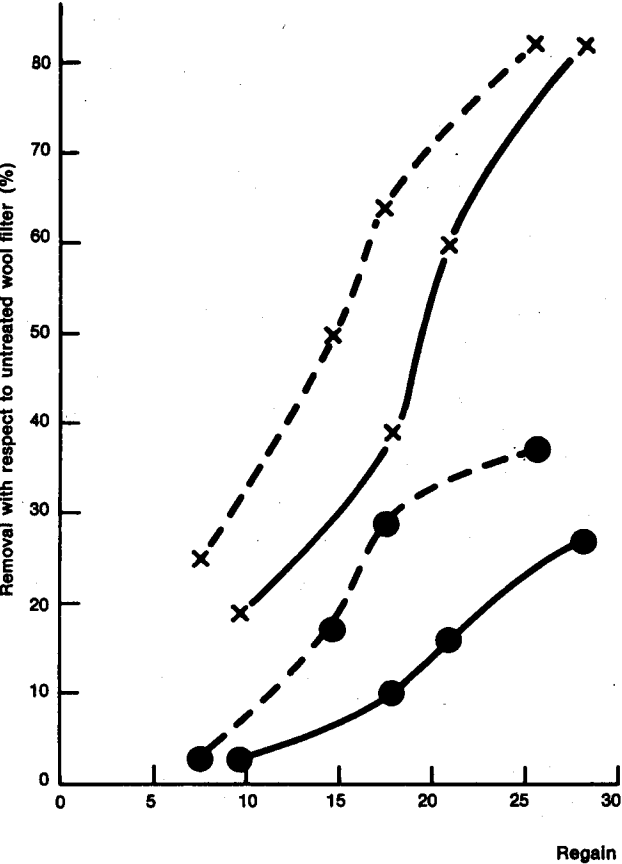
Figure 1b. The effect on Paramecia survival time (adhesive-bonded web X, combed wool top ●, untreated adhesive-bonded web △).
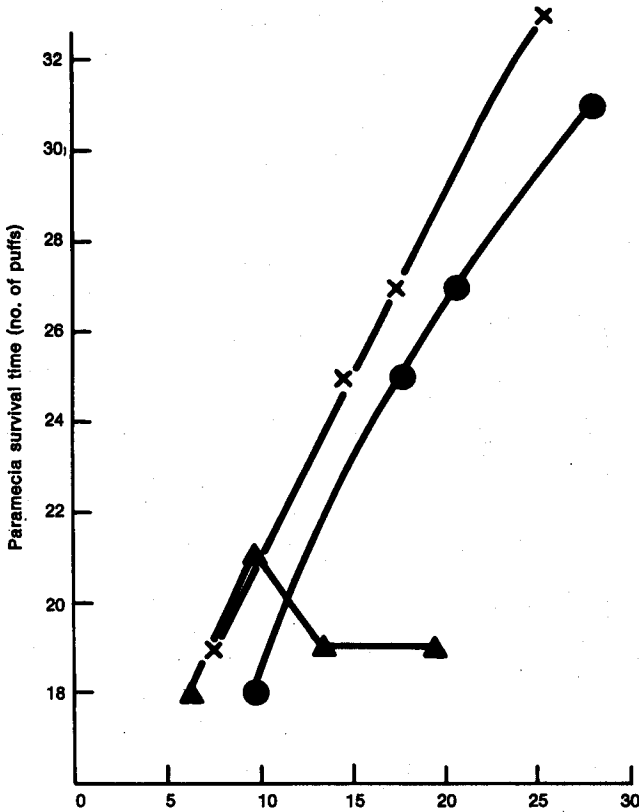

Regain of filter material (%)

Figure 2a. The influence of the pH of PEI-treated wool filters on the retention of hydrogen cyanide and acetaldehyde at two different moisture levels (filters equilibrated at 65 % [X] and 93 % [●] relative humidity).
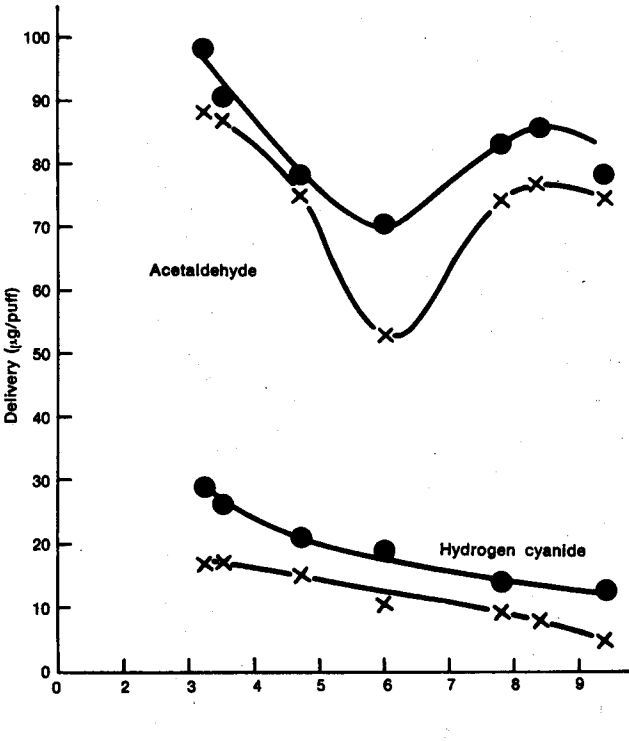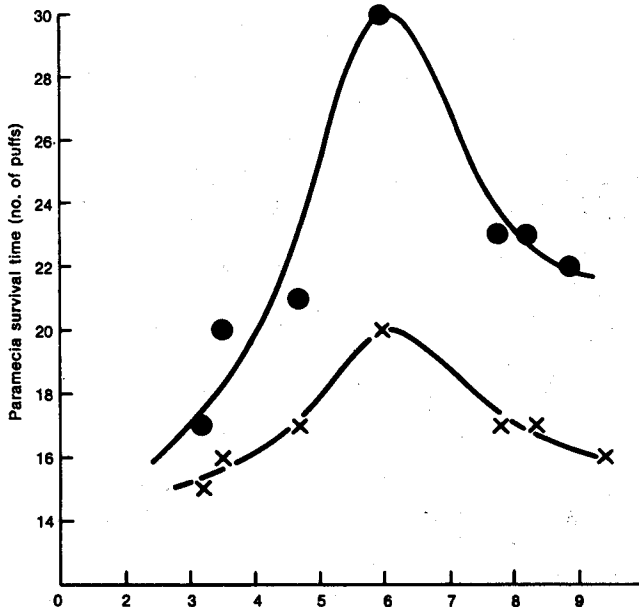
Figure 2b. The effect on Paramecia survival time (filters equilibrated at 65 % [X] and 93% [●] relative humidity).

Filter pH

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the more basic polyethylenimines retained larger quantities of hydrogen cyanide and presumably other weak acids. PEI 1090 provided the best retention of the cytotoxic volatile aldehydes, but is precluded from use as a filter additive by its dark colour and unpleasant odour.

PEI-1000-treated filters removed various proportions of volatile aldehydes and weak acids but showed the highest selectivity towards formaldehyde and hydrogen cyanide (Table 6). The selectivity for the volatile aldehydes decreases markedly as their molecular weight increases. The semivolatile phenols and other weak acids occur predominantly in the PM and are therefore removed to a large extent by mechanical filtration. However, some transfer of these constituents must occur to the VP as has been previously suggested (26) because the treated filters remove a larger proportion of these compounds.

Applications in the range of 3—7 % by weight of PEI on wool top or bonded fabric gave optimum filter performance. Apparently, quantities in this range cover the fibre surface in such a way that a maximum surface area of polymer is exposed to the smoke stream. Smaller amounts inadequately cover the fibre surface and reduce the filter performance, whereas larger quantities fail to significantly increase the surface area exposed to the smoke stream and have little effect on the filter performance.

The Effect of Moisture Content and Filter Material

Moisture regains higher than 15 % cause PEI-treated wool to become tacky and therefore unsuitable for processing on filter-making machines. However, lower moisture contents adversely affect the removal of VP components. Hence, a study was undertaken to define more closely the relationship between moisture content and filter performance. Figures 1a and 1b show the marked effect of moisture content on the retention of hydrogen cyanide, acetaldehyde and cytotoxic VP components by PEI-treated wool top and random web.

Clearly, at regains below 5 %, insignificant quantities of these components would be removed from the smoke stream. Thus, the aqueous phase on the fibre surface is essential for the initial adsorption process to occur. The PEI then completes the process by irreversibly binding the absorbed components in the aqueous phase.

Figures 1a and 1b also demonstrate that random-web filters remove the VP components more effectively than wool-top filters with an equivalent moisture content. This occurs despite the fact that the wool-top filters contain 20 % by weight more wool than the random-web filters and therefore expose a larger surface area of additive to the smoke stream. The random arrangement of fibres evidently provides a more effective medium for chemisorption than the essentially co-linear mass of fibres in the wool-top filters. Random-web wool filters are also superior in their mechanical filtration properties (12).

The Effect of pH on the Performance of PEI Filters

The extent of adsorption and reaction of certain VP constituents, particularly weakly acidic compounds, is likely to depend on the pH of the PEI-treated filters. Figure 2a shows the influence of the filter pH on the efficiency of removal of acetaldehyde and hydrogen cyanide at two different filter moisture-levels. As expected, the retention of hydrogen cyanide, and presumably other weak acids, improves as the filter pH increases. The number of free amino groups available for reaction with the weak acids decreases appreciably below pH 8.0 due to protonation, and ion exchange becomes the only mechanism by which these components can be bound to the surface film of PEI. It is apparent that the effectiveness of acetaldehyde retention is extremely sensitive to the filter pH, optimum removal occurring at pH 6.0—6.2. Clearly, a certain proportion of protonated and free amino groups facilitates the reaction of PEI with aldehydes. Upsetting this balance by altering the pH in either direction lowers the retention of acetaldehyde and, presumably, the other volatile aldehydes. The optimum pH for removal of aldehydes obviously differs by several units from that for the removal of weak acids.

The *Paramecia* bioassay (Figure 2b) exhibited a pH dependence similar to that observed for the removal of acetaldehyde. A maximum survival time was recorded for filters at pH 6.0; altering the pH to higher or lower values resulted in a marked reduction in the survival time. This result supports the existing evidence that exposures of ciliated protozoa to cigarette smoke provide an assessment of the delivery of the cytotoxic C_1–C_3 aldehydes (13).

Figures 2a and 2b further demonstrate the marked effect of the moisture content of the filter on the efficiency of removal of VP components. At the optimum pH for the removal of aldehydes, it is evident that the beneficial effect of a higher moisture level is enhanced considerably.

SUMMARY

Modified-wool cigarette filters have been evaluated for their efficiency in selectively retaining specific biologically active volatile and semivolatile smoke components. Filters containing chemically modified wools or wool treated with low-molecular-weight additives were ineffective. Polymeric additives reduced the cytotoxic level of cigarette smoke by varying degrees. Polyethylenimines were particularly effective additives and selectively removed portions of formaldehyde, acetaldehyde, acrolein, hydrogen cyanide, phenols and other weakly acidic compounds. Increasing the moisture content of the treated filters markedly improved their performance. The effectiveness of polyethylenimine-treated filters in removing volatile aldehydes was strongly dependent on the pH of the filter, most efficient removal occurring at pH 6.0–6.2.

ZUSAMMENFASSUNG

Aus modifizierter Wolle bestehende Cigarettenfilter wurden auf ihre Wirksamkeit untersucht, spezifische, biologisch aktive flüchtige und halbflüchtige Rauchinhaltsstoffe selektiv zurückzuhalten. Filter, die chemisch modifizierte Wollarten oder Wolle enthielten, die mit Zusätzen niedrigen Molekulargewichts behandelt wurden, waren unwirksam. Polymere Additive verminderten die Zelltoxizität des Cigarettenrauches in unterschiedlichem Ausmaß. Polyäthylenimine erwiesen sich als besonders wirksame Zusätze, die Teile von Formaldehyd, Acetaldehyd, Acrolein, Zyanwasserstoff, Phenolen und anderen schwach sauren Verbindungen selektiv entfernten. Die Erhöhung des Feuchtigkeitsgehaltes der behandelten Filter verbesserte deutlich deren Leistung. Die selektive Retention flüchtiger Aldehyde durch Filter, die mit Polyäthyleniminen behandelt wurden, war in hohem Maße abhängig von dem pH-Wert des Filters; die stärkste Wirksamkeit zeigte sich bei einem pH-Wert von 6,0—6,2.

RESUME

On a comparé l'efficacité de différentes modifications de filtres de cigarettes en laine, quant à leur propriété de retenir sélectivement des composés volatiles ou semivolatiles biologiquement actifs contenus dans la fumée. Les filtres s'avérant inefficaces sont ceux qui contenaient des laines modifiées chimiquement ainsi que des laines traitées au moyen d'additifs au poids moléculaire bas. Les additifs polymères s'avéraient réduire de façon variable le niveau cytotoxique de la fumée de cigarettes. Les polyéthylènimines (PEI) se sont avérés des additifs particulièrement efficaces et supprimaient sélectivement des portions de formaldéhyde, d'acétaldéhyde, d'acroléine, d'acide cyanhydrique, de phénols et d'autres composés légèrement acides. En augmentant la teneur en humidité des filtres traités, on augmentait sensiblement leur influence. Le pH des filtres traités au PEI semblait jouer un rôle important dans l'efficacité à supprimer les aldéhydes volatiles; la suppression la plus efficiente semblait se manifester à un pH de 6,0—6,2.

REFERENCES

1. Leuchtenberger, C., and R. Leuchtenberger: Morphology of experimental respiratory carcinogenesis; United States Atomic Energy Commission 1970, 329.
2. Thayer, P. S., and C. J. Kensler: Science 146 (1964) 642.
3. Battista, S. P., and C. J. Kensler: Arch. Environ. Health 20 (1970) 318.
4. Dalhamn, T., and A. Rosengren: Arch. Otolaryng. 93 (1971) 496.
5. Wynder, E. L., and D. Hoffmann: Tobacco and tobacco smoke, Studies in experimental carcinogenesis; Academic Press, N. Y., 1967, 253.
6. Holt, P. G., W. N. Bartholomaeus, and D. Keast: Aust. J. exp. Biol. med. Sci. 52 (1974) 211.
7. Powell, G. M., and G. M. Green: Biochem. Pharmacology 21 (1972) 1885.
8. Benedict, R. C., and R. L. Stedman: Experientia 24 (1968) 1205 / Tobacco Sci. 13 (1969) 166.
9. Schabort, J. C.: J. South African Chem. Inst. 20 (1967) 103.
10. Bednarcyk, N. E.: Tobacco smoke filters; Noyes Data Corporation, New Jersey, 1972, 140.
11. George, T. W.: Selective removal of components of tobacco smoke by filtration; Nat. Cancer Inst. Monogr. 28, 237, 1968.
12. Evans, D. J., M. Lipson, and R. J. Mayfield: J. Text. Inst. 66 (1975) 325.
13. Comber, R.: Chem. and Ind. 1972, 64.
14. Hoffmann, D., and E. L. Wynder: J. Nat. Cancer Inst. 48 (1972) 1855.
15. Alexander, P., D. Carter, C. Earland, and O. E. Ford: Biochem. J. 48 (1951) 629.
16. Sweetman, B. J., and J. A. MacLaren: Aust. J. Chem. 19 (1966) 2347.
17. Lipson, M., and J. B. Speakman: J. Soc. Dyers and Colourists 65 (1949) 390.
18. Coresta standard method No. 10, Information Bulletin 1969—1, 24.
19. Weiss, W.: Arch. Environ. Health 17 (1968) 62.
20. Aldridge, W. N.: Analyst 69 (1944) 262.
21. Altshuller, A. P., D. L. Miller, and S. F. Slera: Anal. Chem. 33 (1961) 621.
22. Evans, D. J., and R. J. Mayfield: Analyst 100 (1975) 540.
23. Cohen, I. R., and A. P. Altshuller: Anal. Chem. 33 (1961) 726.
24. Lorentzen, G., and G. Neurath: Beitr. Tabakforsch. 2 (1963) 73.
25. Rothwell, K., and C. A. Grant: Standard methods for the analysis of tobacco smoke; Tobacco Research Council Research Paper 11, London 1972.
26. Williamson, J. T.: Bull. Inf. Coresta Spécial 1974, 79.

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