United States Patent Office 3,246,655 Patented Apr. 19, 1966

3,246,655

SELECTIVE CIGARETTE FILTERS

Alexander W. Spears, William E. Routh, Chase W. Lassiter, and Jimmy H. Bell, Greensboro, N.C., assignors to P. Lorillard Company, New York, N.Y., a corporation of New Jersey

No Drawing. Filed Mar. 19, 1963, Ser. No. 266,212
9 Claims. (Cl. 131—10)

This is a continuation-in-part of application Serial No. 188,281, filed April 16, 1962.

This invention relates to a new cigarette filter which selectively removes certain products of combustion from tobacco smoke, and particularly to cigarette filters containing a chemical complexing agent to selectively remove phenol and phenolic compounds from tobacco smoke.

It has been thought by some to be desirable to provide a filter for cigarettes which removes phenol and phenolic compounds from the tobacco smoke. While smoke of a somewhat reduced content of phenolic compounds may be obtained by providing a more efficient filter, such filters have certain limitations. Highly efficient filters are not necessarily desirable from the standpoint of the average smoker because the increased efficiency of the filter tends to remove those compounds generally responsible for good body and flavor. In addition, an important limitation of filters of improved efficiency is that phenol and some of the phenolic compounds are in the vapor state, and hence not amenable to filtration no matter how efficient the filter may be.

According to the present invention, it has been found that cigarette filter fibers impregnated with certain polymeric compounds having the property of forming a complex with phenol and phenolic compounds in tobacco smoke are particularly advantageous. Such filters selectively adsorb phenol and phenolic compounds from tobacco smoke, while at the same time, if properly made, they do not remove excessive amounts of tar.

—O— which occurs between successive alkylene groups in the X chain.

The molecular weight of the complex-forming compounds (which determine y and z) is dependent on the specific material used. In selecting a suitable molecular weight, the vapor pressure of the complex-forming compounds should be sufficiently low that it will remain in the filter during storage and will not volatilize during the smoking of the cigarette. The vapor pressure of the complex-forming compounds should be from 0 mm. up to 40 mm. Hg at 25° C., however, the preferred range is between 0 mm. and 1 mm. Hg at 25° C.

A further limitation on the molecular weight of the polymeric compound is that the compound should be soluble in the plasticizer for the filter fiber base. Where compounds of Formulas 1 to 3 are used, the molecular weight may vary over a considerable range, to over several thousand. If di-isocyanates are used (e.g., Formula 4) it is preferred that the polymer contain no more than 1 or 2 urethane linkages. Where the urethane linkage is large with respect to the number of ether linkages, the polymer becomes insoluble in the plasticizer materials.

Particularly preferred compounds within the above class have the formula $R(XOH)_a$, a being 3, and especially glyceryl derivatives of propylene oxide, e.g., compounds of the formula $C_3H_5—(XOH)_3$, wherein X has the formula $(OC_3H_6)_z$.

Compounds of groups 1 to 4 are most conveniently prepared by reacting a lower alkyl mono-, di-, trihydric alcohol, a lower alkyl amine or diamine, or a lower alkyl isocyanate or di-isocyanate with a lower alkylene oxide or glycol. Useful compounds may also be prepared by reacting alkylene oxides or glycols containing alkylene groups of various chain lengths with alcohols, amines, or isocyanates containing alkyl groups of various chain lengths.

Several typical compounds are illustrated by, but not limited to the materials set forth in Table I.

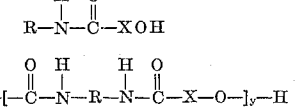
TABLE I

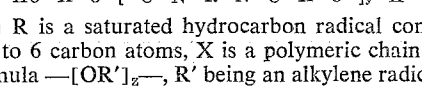
Formula of Polymeric Condensate	Approx. Mol. Wt.	Monomeric Units	Available From—	Trade Name	Manufacturer's Trade Bulletin
$C_3H_5[(OC_3H_6)_xOH]_3$	3,000	Glycerol, propylene oxide	Union Carbide Chemicals Co.	Niax LG 56	"Union Carbide Chemical Co. Technical Bulletin Niax Polyethers" (1961).
$C_3H_5[(OC_3H_6)_xOH]_3$	1,000	do	do	Niax LG 168	Do.
$C_6H_{11}[(OC_3H_6)_xOH]_3$	5,000	Hexanetriol, propylene oxide.	do	Niax LH T-34	Do.
$C_6H_{11}(OC_3H_6)_xOH$	4,100	Trimethanol propane, propylene oxide.	Wyandotte Chemicals Corp.	Pluracol 4040	"Wyandotte Chemicals Technical Bulletin Pluracol TP Triols," (1958).

Complex-forming compounds which are particularly useful in accordance with this invention are selected from the group consisting of

(1) $R(XOH)_a$

(2) $R—N_b(XOH)_{2b}$

(3)


(4)


wherein R is a saturated hydrocarbon radical containing from 2 to 6 carbon atoms, X is a polymeric chain having the formula $—[OR']_z—$, R' being an alkylene radical having from 2 to 6 carbon atoms, a is 3 and b is a small whole number from 1 to 2, said compounds being insoluble in water.

The foregoing compounds should contain from 10% to 37% ether oxygen, based on the weight of the compound, the preferred materials containing from 16% to 37% ether oxygen. By "ether oxygen" is meant the group

According to the present invention, the complex-forming compounds are incorporated into a conventional cigarette filter prepared from a fibrous material. Cellulose, regenerated cellulose, cellulose acetate or other cellulosic fibers are the most economical and commercially preferred materials. Such fibers are normally procured in the form of a rope or tow and the tow manufactured into a compact plug by methods known in the art to form a filter having a plurality of tortuous passages to trap solid and liquid particulate matter which may be present in the tobacco smoke.

In the manufacture of such cigarette filters it is known to add powdered materials such as water-soluble polyethylene oxide with the filter tow. In general, these additives provide increased surface area for the removal of condensed tars and other such matter. The polyethylene oxide when so added provides a filter having a high surface area particularly adapted to trap condensed tars from the tobacco smoke, thereby resulting in a filter of improved tar removal efficiency. While a good efficiency of tar removal is thought by some to

be desirable, the removal of large amounts of tar tends to make the taste of the tobacco smoke undesirably flat.

Accordingly to the present invention, the complex-forming compounds are dissolved in a plasticizer for the fiber base, and impregnated into the fibrous base by applying the plasticizer solution of the complexing compounds to the fibrous tow. By manufacturing methods well known in the art, the tow is thereafter formed into a compact filter plug. A sufficient amount of the plasticizer-complexing compound solution should be applied to the tow to provide a filter containing between 0.01% and 25% of the complexing compound, based on the weight of the fiber. The optimum range is between 0.3% and 2% by weight of the fiber.

Where a cellulosic based fiber is used, there are many well known plasticizers therefor. These include, but are not limited to, esters of glycerine with lower fatty acids, such as glyceryl triacetate, esters of citric acid with lower alkyl alcohols and lower fatty acids, such as triethyl citrate, acetyl triethyl citrate and esters of phthalic acid with lower alkyl alcohols. The preferred plasticizer is glyceryl triacetate. The plasticizer may be present in the finished filter in an amount up to 20% by weight.

Filters made in accordance with the method of this invention have a number of advantages. An important advantage lies in the improved simplicity of manufacture. According to the present invention, it has been discovered that materials containing high percentages of ether oxygen have the property of complexing with phenol and phenolic compounds in tobacco smoke. Typical compounds having this property are the polyalkylene oxides, as well as the condensates of alkylene oxides with alcohols or amines such as described above. In the complexing reaction it is believed that the ether oxygen acts as a Lewis base, and that accordingly the percentage of ether oxygen is a measure of the number of bonding sites per molecule.

In the manufacture of cigarette filters an additional consideration must be taken into account. As noted above, it is known to add powdered polyethylene oxides to tobacco smoke filters to improve the tar filtration efficiency. This improvement in filtration efficiency comes about merely by providing for tighter packing and greater internal surface area in the filter plug and results in a concomitant increase in the draw. Such filters can, if desired, be more loosely packed, thereby giving rise to a filter having a more acceptable draw but which then provides no improvement in total filtration efficiency. The preparation of cigarette filters in this manner, furthermore, requires additional, undesirable steps in the manufacturing process. It is preferred in the manufacture of cigarette filters that the additive be incorporated in the fiber tow in such a manner that additional steps are not required.

This is accomplished by using the materials of the present invention which, in addition to being polyethers capable of complexing with phenol and phenolic compounds, are characterized by being soluble in the plasticizers for the fiber base. Thus, a solution of the polyether complexing compounds may be prepared in a plasticizer, which, when added to the filter tow, is absorbed into the fiber. The fibrous tow is thereafter processed into filter plugs in the usual manner, yielding a plug having approximately the same draw without the need for additional steps in the filter manufacturing process. Such filters thereby have approximately the same total of filter efficiency, i.e., they remove the same amounts of tars, but selectively remove phenol and phenolic compounds from the tobacco smoke.

An additional advantage arises from the use of the compounds of the present invention. Besides being soluble in the usual plasticizers for cellulosic fibers, compounds of the present invention are insoluble in water. This is in contrast to the polyethylene oxides mentioned above, which are water-soluble. When prepared in the form of cigarette filters, the filter is potentially subjected to being wetted by saliva. If water-soluble materials are used in such a filter, the water-soluble resin would be reduced to a gummy mass, an obviously undesirable result. By contrast, the materials of the present invention being water-insoluble are not subject to this disability.

Cigarette filters according to the present invention are illustrated by the following examples. As a basis of comparison, a typical cellulose acetate filter is illustrated in Example 1. In Example 1 and subsequent illustrations, the cellulose acetate filter tow which was used to prepare the filter in the base case was impregnated with a solution of glyceryl triacetate and each of several complexing compounds in various proportions. The cigarettes resulting were machine smoked and the products passing through the filter were collected and analyzed, each test being run in quadruplicate.

The terms "filter efficiency" and "phenol filter efficiency" used in the following tables and examples refer to the percentages of particulate matter and of phenol and phenolic compounds, respectively, which are removed from the tobacco smoke.

Example 1

A solution of a complex-forming polymer and a suitable plasticizer were applied to the filter fiber. Each of several complexing materials was mixed with glyceryl triacetate in weight ratios varying from 1:3 to 1:15 parts of complexing compound per part of plasticizer and added to cellulose acetate fibers. The treated fibers were prepared into filter plugs in the usual manner. As a basis of comparison, a filter was prepared using glyceryl triacetate without a complex-forming additive. The results are set forth in Table II.

TABLE II.—FILTERS CONTAINING COMPLEXING COMPOUND MIXED WITH GLYCERYL TRIACETATE PLASTICIZER

Complex Forming Compound [1]	Weight Ratio of Complex-Forming Compound to Plasticizer	Weight Percent on Fiber	Denier of Fiber	Total Denier	Length, mm.	Plug Draw	Total Filter Efficiency, Percent	Phenol Filter Efficiency, Percent
None		[2] 6	3.2	55,000	17	0.48	47.0	71.5
Niax LHT-34	1:7	6	3.2	55,000	17	0.44	49	87
Niax LG-168	1:7	6	3.2	55,000	17	0.46	47.5	88
Niax LG-168	1:15	6	3.2	55,000	17	0.42	49	91
Niax LG-168	1:3	5	3.2	55,000	17	0.39	47	89.5
Niax LG-168	1:7	5	2	42,000	20	0.36	61	94
Niax LG-56	1:15	5	3.2	55,000	17	0.48	48	86
Pluracol 4040	1:7	5	3.2	55,000	17	0.45	47	86

[1] See Table I for identification of these compounds
[2] 6% plasticizer present in the fiber.

The data reported in Example 1 were all obtained within several weeks after sample preparation. However, in normal practice, the time lapse from production to consumer use may be approximately two months. Therefore, it is appropriate to determine the stability of these filters with respect to aging for a two month period.

Example 2

Filter-tipped cigarettes having a filter of cellulose acetate and glyceryl triacetate, as set forth in Table I, and filter-tipped cigarettes having a similar filter but containing Niax LG 168 in addition were stored for a period of two months and then tested for phenol filter efficiency. The Niax LG 168 as received contained an antioxidant (butylated hydroxy toluene, 250 p.p.m.) which was added by the manufacturer to improve the stability of the compound. The cigarettes thus prepared were separated into two groups, one of which was smoked immediately and the other group was smoked after storage for two months. The results are set forth in Table III.

TABLE III.—STABILITY OF CIGARETTE FILTERS

Sample	Phenol Filter Efficiency, Percent	Change in 2 Months, Percent
Cellulose acetate and glyceryl triacetate	67	−4.5
Niax LG 168 and glyceryl triacetate (1:15)	76	−15
Niax LG 168 and glyceryl triacetate (1:7)	89	0
Niax LG 168 and glyceryl triacetate (1:3)	88	0

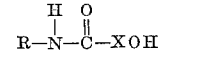
The various filters set forth in the preceding examples have been described for exemplary purposes only, and it will be understood that the invention is not limited thereto, but is to encompass all equivalents and variations within the scope of the following claims.

We claim:

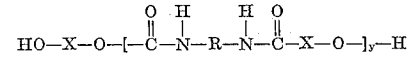
1. In a filter-tip cigarette having in combination an elongated cylindrical plug of shredded tobacco joined at one end thereof with a filter element having a fibrous base material, the improvement comprising the combination with said fibrous base material of (A) from 0.01% to 25% based on the weight of said fiber of a compound selected from the group consisting of

(1) $R(XOH)_a$

(2) $R{-}N_b(XOH)_{2b}$

(3)


and

(4)


wherein R is a saturated hydrocarbon radical containing from 2 to 6 carbon atoms, X is a polymeric chain having the formula $-[OR']_x-$, R' being an alkylene radical containing from 2 to 6 carbon atoms, *a* is 3 and *b* is a small whole number from 1 to 2, said compound containing from 10% to 37% ether oxygen, based on the weight of the compound and having a vapor pressure between 0 mm. and 40 mm. Hg at 25° C., and (B), a plasticizer for said fibrous base material, said compound (A) being insoluble in water and soluble in said plasticizer (B), and being impregnated in said fiber.

2. A filter-tip cigarette according to claim 1 wherein said compound (A) contains from 16% to 37% ether oxygen based on the weight of the compound.

3. A filter-tip cigarette according to claim 1 wherein said compound (A) has a vapor pressure of from 0 mm. to 1 mm. Hg at 25° C.

4. A filter-tip cigarette according to claim 1 wherein said compound (A) is present in an amount from 0.2% to 3% based on the weight of said fiber.

5. In a filter-tip cigarette having in combination an elongated cylindrical plug of shredded tobacco joined at one end thereof with a filter element having a fibrous base material, the improvement comprising the combination with said fibrous base material of (A) from 0.01% to 25%, based on the weight of said fiber, of a compound having the formula

$$R(XOH)_a$$

wherein R is a saturated hydrocarbon radical containing from 2 to 6 carbon atoms, X is a polymeric chain having the formula $(OR')_z$, R' being an alkylene radical containing from 2 to 6 carbon atoms, and *a* being 3, said compound containing from 10% to 37% ether oxygen based on the weight of the compound and having a vapor pressure between 0 mm. and 40 mm. Hg at 25° C., and (B) a plasticizer for said cellulosic material, said compound (A) being insoluble in water and being soluble in said plasticizer (B), and being impregnated in said fiber.

6. A filter-tip cigarette according to claim 5 wherein said compound (A) contains from 16% to 37% ether oxygen based on the weight of the compound.

7. A filter-tip cigarette according to claim 5 wherein said compound (A) has a vapor pressure of from 0 mm. to 1 mm. Hg at 25° C.

8. A filter-tip cigarette according to claim 5 wherein said compound (A) is present in an amount from about 0.2% to 3% based on the weight of the fiber.

9. In a filter-tip cigarette having in combination an elongated cylindrical plug of shredded tobacco joined at one end thereof with a filter element having a fibrous base material, the improvement comprising the combination with said fibrous base material of (A) from 0.01% to 25% based on the weight of said fiber and the compound having the formula $C_3H_5(XOH)_3$, wherein X is a polymeric chain having the formula $[OC_3H_6]_z$, said compound containing from 16% to 37% ether oxygen and having a vapor pressure between 0 mm. and 1 mm. Hg at 25° C., (B) a plasticizer for said cellulose, said compound (A) being insoluble in water and being soluble in said plasticizer (B), and being impregnated in said fiber.

References Cited by the Examiner

UNITED STATES PATENTS

Re. 24,486	6/1958	Mitchell	106—163
2,881,771	4/1959	Touey	131—208
2,904,050	9/1959	Kiefer et al.	131—208
2,943,737	7/1960	Norton.	
2,956,329	10/1960	Touey	131—208
3,026,226	3/1962	Touey et al.	131—208
3,101,723	8/1963	Seligman et al.	131—208
3,144,024	8/1964	Eichwald et al.	131—208

FOREIGN PATENTS

173,262	12/1952	Australia.

SAMUEL KOREN, *Primary Examiner.*

MELVIN D. REIN, ABRAHAM G. STONE, *Examiners.*