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3,310,060

CELLULOSIC FILTER FOR TOBACCO SMOKE
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This invention relates to improved cellulosic filters for tobacco smoke, and particularly to improved paper filters characterized by unusual capacity for the removal of certain ingredients from tobacco smoke.

It has been known that cigarette filter tips of absorbent paper can readily be made, which remove a high proportion of the particulate matter, generally referred to as tars, from the cigarette smoke passing therethrough. High filtration efficiencies can readily be attained, without undue increase in the resistance to gas flow, through the proper mechanical treatment of a paper web and conformation of the treated web to final filter form. However, it has also become known that the smoke after passage through a cellulosic filter of the above character contains certain undesired ingredients, such as phenolic compounds. Further, such filtered smoke has been recognized as bearing an off-taste to many smokers. A practical solution for the alleviation of such deficiencies has not been available even after extensive investigations.

For example, the incorporation of a polyhydric alcohol in the filter, as disclosed in Australian Patent 201,558, while effecting some purifying action, is found to cause undesirable drying of the smoke because of the hygroscopicity of such compounds. Such agents also cause an undue softening of the filter tip and display a tendency to migrate into the neighboring cigarette elements such as the tobacco column and the paper wrapper.

At least some of these deficiencies likewise characterize other additives for cigarette filters which have been proposed heretofore, such as the solid ethylene oxide and similar polymers of U.S. 3,032,445 and the finely divided particles of wax, such as the high-molecular weight polyethylene glycols ("Carbowax") of U.S. 2,904,050.

Further, while some limited increase is found in the purifying action of freshly made filters composed of filamentary cellulose acetate, containing a plasticizer such as triacetin and a sorbitan ester or other finishing agents as disclosed in U.S. 2,953,837 and 2,953,838, such effect is found to decrease during storage of the filters, apparently due to decreased concentration near the filament surfaces as the agents diffuse into the filament interiors.

In accordance with the present invention, it has been found that cellulosic filters for tobacco smoke are substantially improved by the addition thereto of certain liquid absorbents having a chemical structure as specified below. Cellulosic filters which have been thus treated are characterized by high affinity for phenolic compounds, and thus by excellent capability for removing these and other undesired ingredients of tobacco smoke, thereby enhancing the flavor and increasing the acceptability.

The highly effective agents in accordance with this invention are compounds which are liquid and have very low volatility (low vapor pressure) at ordinary atmospheric temperatures (0° C. to about 40° C.) and are characterized by a molecular structure corresponding to



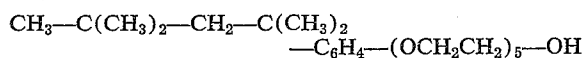
wherein R is a monovalent hydrocarbon radical, chosen from the group consisting of alkyl and alkaryl containing 10 to 20 carbon atoms, and n is a number between about 5 and 15, representing the average number of moles of alkylene oxide combined with a mole of the starting monohydric alcohol or alkylated phenol R-OH.

These poly (oxyethylene) compounds result from the

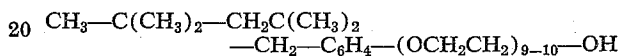
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condensation of desired proportions of ethylene oxide with the monohydric alcohols, preferably saturated, or alkylated phenols R-OH containing 10 to 20 carbon atoms, or of mixtures thereof. In the formation of these adducts, ethylene oxide may in part be replaced by propylene oxide. The filter treating agent may likewise consist of a mixture of poly (oxyalkylene) compounds as defined above.

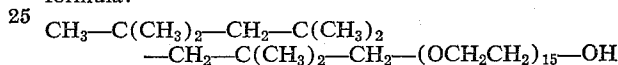
Preferred agents are adducts based on n-butylphenol, p-tertiarybutylphenol, isoamylphenol, dibutylphenol, diisoamylphenol, n-hexylphenol, p-cyclohexylphenol, decylphenol, dodecylphenol, iso-tridecylphenol butyl-cyclohexylphenol, p-tertiaryoctylphenol and p-tertiarynonylphenol. The latter two are most highly preferred, corresponding in structure to the following formulae:



and



The preferred monohydric alcohol adduct is one based on iso-tridecyl alcohol corresponding to the following formula:



Other adducts useful in accordance with this invention are those resulting from combining alkylene oxide, as above defined, with monohydric alcohols, preferably saturated, containing 10 to 20 carbon atoms, including decyl, dodecyl, tetradecyl, hexadecyl, cyclohexyldecyl, butylcyclohexyl and octadecyl alcohols.

The agents as above defined are liquids of low volatility at ordinary temperatures which avidly and selectively remove phenolic compounds and readily absorb other acidic compounds from gaseous mixtures, particularly tobacco smoke. While soluble or readily dispersible in water, they are not so absorptive with respect to water vapor as to cause undue drying of tobacco smoke. Some of these adducts particularly alkylphenols with higher proportions of alkylene oxide may tend to solidify partly at room temperature as oily solids but are readily liquefied by adding thereto a different adduct containing less oxide or based on a different starting material within the above definition.

The additive is suitably incorporated in a cellulosic filter which may consist of crepe or other absorbent paper, or of regenerated cellulose filaments, sponge or film, assembled to provide tortuous passages so that the filtration of tobacco smoke passing therethrough is accomplished efficiently but without undue resistance to gas flow. For best results, the filter is formed of an absorbent paper web of enhanced filtration properties imparted by mechanical treatment, preferably in accordance with the procedure disclosed in U.S. 2,954,036. In accordance therewith, the starting paper web is moistened, then provided with parallel longitudinal grooves by treatment between suitable rolls, being simultaneously provided with randomly spaced longitudinal rents, then dried to evaporate excess moisture, and is next compressed between smooth-surfaced rolls to flatten the grooves and cause the adjacent ones partly to overlap each other. The treated web is then gathered to rod form and cut to desired lengths.

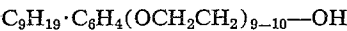
Preferably, the agent is added in desired proportion by uniformly dispersing the same in the moistening liquid, which is best applied to the web by spraying, although other conventional immersion or roll-coating methods may be used. The treatment is carried out so that the paper web just prior to the grooving operation contains about 10% to 55% by weight of the moistening liquid,

preferably about 25% by weight, the concentration of the agent therein being maintained at a desired value such that the resulting filter rod has an effective content of the agent. Following the grooving step, the grooved web is subjected to drying, as by circulating heated gas thereover or by other suitable means as by infra-red lamps, in order to remove excess moisture. After drying, the grooved web should have a moisture content of a few percent, for example about 3% to 6%.

The grooved and dried web may then be assembled to filter rod displaying good filtration effectiveness. However, for best results, an additional operation is preferred wherein the grooved and dried web is compressed by passage through the nip of smooth-surfaced pinch rolls, whereby the groove ridges are flattened so that the adjacent ones partly overlap each other, resulting in a filter sheet of desirably increased density wherein the exposed sheet surfaces are covered by projecting fibers and fiber ends. In cross-section, such treated sheet displays a "mushroom" or open "hour glass" appearance in contrast to the starting appearance of oppositely faced connected loops having side walls parallel to each other.

Finally, the treated web is passed through a forming cone, such as is used conventionally in the assembly of cigarette rods, together with a wrapper strip, and is therein laterally gathered and radially compressed to result in a wrapped consolidated filter rod.

In preferred embodiments in accordance with this invention, filter rods were made containing 10% and 20% by weight respectively of a liquid agent consisting of the nonylphenol adduct containing an average of 9 to 10 moles of ethyleneoxide per mole of the phenol, corresponding to the formula:



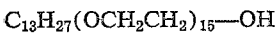
Filter tips 15 mm. in length were attached to individual cigarettes selected from a uniform commercial lot and were tested in a standard analytical smoking apparatus, yielding the following data in direct comparison with some of the same lot of cigarettes (a) having no filter, and (b) having identical paper filters containing no added agent. These 15 mm. filter tips displayed low resistance to gas flow corresponding to an average loss in head of 1.9 inches of water at an air flow rate of 17.5 milliliters per second. The smoking apparatus was operated to take a 35 milliliter puff of two seconds duration at a rate of one per minute. For each sample, the values represent the average of five closely agreeing determinations. The phenol determinations were obtained chromatographically and are considered to be parallel to the results which would be obtained for all of the phenolic constituents of the smoke if these were determined individually.

TABLE I

Sample	Filter Tip	Percent Reduction of Particulates (Tars)	Percent Reduction of Phenol
A	None	0	0
B	With no agent	48.5	53.2
C	With 10% agent	55.5	84.7
D	With 20% agent	54.8	89.3

It is evident from the above comparative results that the filter tips containing agent in accordance with this invention selectively remove most of the phenolic constituents from the smoke passing therethrough.

Another preferred selective agent is polyethoxylated tridecyl alcohol containing an average of about fifteen moles of ethylene oxide combined with a mole of alcohol, represented by the formula



The following table lists the comparative data obtained with paper filter tips provided with a series of proportions of this agent as described above.

TABLE II

Sample	Filter Tip	Percent Reduction of Particulates	Percent Reduction of Phenol
1	None	0	0
2	With no agent	48.5	53.2
3	With 5% agent	59.5	81.2
4	With 10% agent	53.4	82.4
5	With 15% agent	55.8	86.8
6	With 20% agent	53.7	88.0
7	With 25% agent	54.0	94.6
8	With 30% agent	55.5	95.2
9	With 50% agent	54.8	96.7

Highly selective removal of phenol is likewise established by the above results, the extent of removing phenol from the smoke by the 15 mm. filter tips of paper containing the agent being far greater than the extent of removal of the particulate matter.

It is seen that the selective action increases significantly with increased percentage of the agent in the filter between about 5% and 25% by weight, while further increase in agent concentration is accompanied by only slight increase in efficiency.

Accordingly, the liquid agents of this invention are best used in the range of 5% to 25% of the cellulosic filter, with a range of about 10% to 15% being most highly preferred.

The following table lists comparative data obtained in smoking tests, as described above, wherein nonylphenol adducts having different proportions of combined ethylene oxide were applied to paper filters.

TABLE III

Sample	Agent	Percent Reduction of Particulates	Percent Reduction of Phenol
1	n=5, 10% by wt	49.8	73.3
2	n=5, 15% by wt	47.3	80.2
3	n=10, 10% by wt	47.5	84.1
4	n=15, 10% by wt	49.2	81.0
5	n=15, 13% by wt	49.2	84.0
6	n=15, 17% by wt	46.5	85.8

In smoking tests by taste panels, filter-tip cigarettes, having cellulosic filters impregnated with a selective liquid agent as above specified, have been found to provide smoke of improved taste and mildness as compared with cigarettes differing only in that no agent was used in the filter.

Being substantially non-volatile, the above agents add no taste or flavor of their own to the cigarette smoke, while efficiently removing phenolic compounds as well as other acidic smoke constituents. Also, these agents are non-hygroscopic and thus do not cause any undesirable drying of the smoke. Further, the selective absorption properties are retained without substantial decrease even after extended storage of the treated filter rods or of cigarettes provided with tips thereof.

What is claimed is:
1. A filter for tobacco smoke comprising a mechanically worked absorbent paper sheet consolidated in rod form, said sheet having incorporated therein about 5% to 25% by weight of a normally liquid polyethoxylated alkylphenol, containing an average of about five to fifteen moles of ethylene oxide per mole of said alkylphenol, said alkylphenol containing 10 to 20 carbon atoms.

2. A filter in accordance with claim 1, wherein said paper contains about 10% to 15% by weight of nonylphenol combined with an average of 9 to 10 moles of ethylene oxide per mole of said nonylphenol.

3. A filter for tobacco smoke comprising a mechanically worked absorbent paper sheet consolidated in rod form, said sheet having incorporated therein about 5% to 25%

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by weight of a normally liquid polyethoxylated saturated alcohol, containing about five to fifteen moles of ethylene oxide per mole of said alcohol, said alcohol containing 10 to 20 carbon atoms.

4. A filter in accordance with claim 3, wherein said paper contains about 10% to 15% by weight of tridecyl alcohol combined with an average of 15 moles of ethylene oxide per mole of said tridecyl alcohol.

5. A filter for tobacco smoke comprising a carrier element of cellulose consolidated in rod form, said carrier element having uniformly incorporated therein about 5% to 25% by weight of a normally liquid polyethoxylated monohydric derivative of a hydrocarbon, containing about five to fifteen moles of ethylene oxide per mole of said derivative, said derivative being selected from the group

consisting of alkylphenols and saturated alcohols and containing 10 to 20 carbon atoms.

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