

1299 641

PATENT SPECIFICATION

(11) 1299 641

NO DRAWINGS

- (21) Application No. 13866/70 (22) Filed 23 March 1970
 (23) Complete Specification filed 19 April 1971
 (45) Complete Specification published 13 Dec. 1972
 (51) International Classification A24C 15/00
 (52) Index at acceptance
 A2C 18A3 18C1 20A 20D 20E1 20E2 20EX 20F 20H1
 20H2
 (72) Inventors JAMES BORTHWICK and JAMES FORRESTER
 MORMAN



(54) IMPROVED SMOKING MIXTURE

(71) We, IMPERIAL CHEMICAL INDUSTRIES LIMITED, of Imperial Chemical House, Millbank, London S.W.1, a British Company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to smoking mixtures and to methods of producing such mixtures.

Our co-pending U.K. Patent Application No. 13860/70 (Serial No. 1,299,296) relates to smoking mixtures which contain a minimum amount of organic combustible material as smoke-producing fuel and a maximum amount of inorganic filler as combustion modifier. In such a mixture the combustion modifying properties of the inorganic fillers are utilised to attain a reduction of any health hazard chemicals which arise from combustion of the organic fuel. The mixtures are tobacco-substitute based and the organic materials comprising the tobacco substitutes include carbohydrate, modified carbohydrate, certain aldol condensation products and carbon samples which produce acceptably low amounts of polycyclic aromatic hydrocarbons.

We have discovered that the underlying principle of the invention of our said co-pending Application can also be applied to tobacco based smoking mixtures.

Accordingly, the present invention is a smoking mixture comprising tobacco, a harmless inorganic filler and a binder, the proportion by weight of said filler in the mixture being from 40 to 65% and the combination of cations and anions in the filler being such as to impart to the mixture a commercially acceptable burning rate.

The harmless inorganic filler may comprise 15 to 65% by weight of the smoking mixture, preferably 40 to 60% by weight.

The smoking mixture may also contain a plasticiser/humectant in addition to the binder.

The present invention is also a method of manufacturing a smoking mixture comprising

ing mixing the aforesaid ingredients and thereafter forming a shaped article of the mixture.

The shaped article is preferably a film, extruded filament or tape.

For the purpose of this specification the term "harmless inorganic filler" is intended to include inorganic salts of some relatively simple organic acids. For example, formates, oxalates, tartrates and citrates can be used as "harmless inorganic fillers" but these are less satisfactory than the preferred completely inorganic materials on account of some undesirable pyrolytic decomposition. This decomposition becomes potentially more harmful as the size of the organic molecule increases.

The inorganic filler preferably comprises a mixture of two or more such inorganic salts admixed in quantities such as to obtain the balance of anions and cations most appropriate for imparting the desired rate of combustion to the preselected amount of tobacco in the mixture.

Although the balance of ions is advantageously obtained by varying the respective amounts of two or more inorganic salts, it is possible to attain the desired rate of combustion using an optimum amount of a single inorganic salt as the inert filler.

An acceptable rate of combustion is considered to be 10—12 puffs from a machine made cigarette containing 1.1 g. of the smoking mixture of the invention, which puff count corresponds to that of a standard class B British cigarette containing 1.1 g. tobacco where each puff is of 25 ml. volume in two seconds, followed by a 58 second rest or smouldering period and where the 70 mm. cigarette is smoked to a butt length of 20 mm.

Additives can advantageously be included in the smoking mixture of the invention, which additives include ingredients such as are normally used in smoking mixtures, for example materials to improve ash coherence and colour, flavourants, drugs and medicaments.

Such a smoking mixture containing a high content of inorganic material can advantageously be produced by a process similar to reconstituted tobacco processes which utilise film-forming agents or binders. This enables a relatively large proportion of inorganic material to be incorporated in the smoking mixture.

The film-forming agent or binder and the plasticiser/humectant incorporated in the mixture may be selected from those commonly used in the reconstituted tobacco art.

Preferred binders include carboxymethyl cellulose and its salts and other carboxylated carbohydrates. Where the amount of binder employed is very small, other less satisfactory binder materials such as natural gums, guar gums, locust bean gums and other synthetic binders such as methyl and other ethers of cellulose may be used.

Preferred plasticiser/humectants include glycerol, ethylene glycol and polyethylene glycol.

As indicated above, the choice of the harmless inorganic fillers to be incorporated in the smoking mixture determines the rate of combustion or how many normal puffs can be had from a given weight of the mixture. The art of making an acceptable burning composition involves choosing the correct proportions of fillers which relatively promote or retard the combustion. Hydrate and nonhydrated versions of the same salt can be used.

The art of retarding combustion without making a composition glowproof yet obtaining the correct puff count from a given weight of smoking mixture by the use of these inert fillers is another key to the success of the invention. Combustion accelerators are combined with combustion retardants in proportions which give a desirable rate of combustion for a particular tobacco and a particular amount thereof incorporated in the mixture. Whilst this principle is exemplified herein and also in our co-pending U.K. Patent Application No. 13860/70 with reference to commonly available inorganic fillers, it can be applied to any inorganic material which proves to be a safe filler for use in a smoking mixture. We have found that a number of commonly available acidic anions influence combustion rate and the order of the retarding ability of a section of them is formate < oxalate < silicate < carbonate = chloride < sulphate < phosphate < borate. Isolated exceptions to these orders indicated can, however, be found. Amongst the cations the order is sodium = potassium < calcium < magnesium. With the cations it would appear that their effect on the burning rate depends on their basicity, the most significant change being from calcium to magnesium. This implies, for ex-

ample, that in smoking mixtures which are identical except that A contains sodium sulphate and B contains the same proportion by weight of magnesium sulphate, A will burn away more quickly. Similarly, in two compositions identical except that A contains calcium carbonate and B contains calcium borate, composition A will burn away more quickly than composition B.

The fillers chosen should not have the power to retain excessive quantities of water, otherwise glowproofing will result.

It is preferable, but not essential, that the fillers should be insoluble in water. Calcium carbonate has been found to be a most useful agent for use as a combustion accelerator balanced against combustion retarding agents such as calcium orthophosphate ($\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$), magnesite (MgCO_3), light basic magnesium carbonate ($3\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$), and calcium sulphate ($\text{CaSO}_4 \cdot \frac{1}{2}\text{H}_2\text{O}$). Using these and other inorganic materials, film can be made which give shred with filling powers comparable to 100% tobacco shred.

The main advantage of the smoking mixtures of the invention is that the lower amounts of tobacco present mean that proportionately lower amounts of the harmful smoke products are formed whilst maintaining a desirable rate of combustion for the mixture. In this connection, tests carried out on smoking mixtures of the invention show a reduction in the benzpyrene and other harmful chemicals content of the smoke because of the reduced tobacco content.

The principle of ion balancing of the inert fillers is the same as described in our co-pending U.K. Patent Application No. 13860/70 (Serial No. 1,299,296) and as illustrated in Tables 1 to 9 of said Application. According to this principle the amount of tobacco to be used in the mixture is first selected and then ion balance tests are effected to determine the composition having the most desirable rate of combustion. As can be seen from said Tables 1 to 9, the ions are investigated separately in order to ascertain their relative suitabilities.

The technique of arriving at the composition of a suitable tobacco-based smoking mixture is illustrated in the Table below. Experiments to arrive at the correct puff number employed the technique of balancing the calcium carbonate against the light basic magnesium carbonate content of the mixture. The Table shows that in this instance light basic magnesium carbonate caused glowproofing. The composition listed in the extreme left hand column of this Table constitutes Example 1 of a smoking mixture according to the present invention.

Composition	Percentage by Weight					
	28	28	28	28	28	28
Tobacco	28	28	28	28	28	28
SCMC	10	10	10	10	10	10
Glycerol	14	14	14	14	14	14
CaCO ₃	35	31	29	27	25	25
3MgCO ₃ .Mg(OH) ₂ .3H ₂ O	—	4	6	8	10	10
K citrate	4	4	4	4	4	4
Citric acid	4	4	4	4	4	4
(NH ₄) ₂ SO ₄	5	5	5	5	5	5
Puff Number	11—12	Extinguished after 3—4 puffs	Would not remain alight	Would not remain alight	Would not light properly	Would not burn
Correct Puff Number						Glow- proofed
Cigarettes made with the paper used by United Kingdom cigarette manufacturers						

- 5 The composition of further Examples 2 and 3 of the invention were arrived at by a similar process but in these cases it was possible to utilize even higher proportions of inorganic material by balancing calcium carbonate against magnesite. In Example 3 locust bean gum binder is used. Examples 4 to 9 illustrate further variations in the parameters of the invention.
- 10 Example 1
4.7 parts ball-milled, fine-cured Virginia type tobacco, 5.8 parts calcium carbonate were suspended in 62.7 parts of water with stirring.
- 15 0.7 part potassium citrate, 0.8 part citric acid, 0.9 part ammonium sulphate and 2.3 parts glycerol were dissolved in 20.8 parts of water and added to the tobacco, chalk suspension. After 4 hour mixing 1.7 parts sodium carboxymethyl cellulose were added to the mixture and the whole mixed for a further hour.
- 20 The slurry was spread at 0.012 inches on a band drier and dried using overband heating at 150° C. The band speed was 25—30 ft./min. The film material was satisfactorily shredded and processed in tobacco machinery. 1.1 g. cigarettes gave 10 puffs.
- 25

The smoke was mild to taste.

The dry weight composition of Example 1 as indicated in the above Table was:—

		%
5	Ball-milled flue-cured Virginia type tobacco	28
	SCMC	10
	Glycerol	14
	CaCO ₃	35
10	Potassium citrate	4
	Citric acid	4
	(NH ₄) ₂ SO ₄	5

Example 2

- 3.9 parts tobacco rag were added to 75 parts of water and allowed to soak for 30 minutes. The mixture was then stirred and 5.3 parts of calcium carbonate, 1.0 part magnesite, 0.6 part potassium citrate, 0.6 part ammonium sulphate, 0.7 part citric acid and 1.0 part glycerol were added and the mixing continued for 10 minutes. The suspension was transferred to a Probst and Class mill and with a gap setting of 2 milled for 10 minutes. The gap setting was reduced to zero and the mixture milled for 10 minutes, at the end of which time 1.1 parts sodium carboxymethyl cellulose were added and the slurry discharged into a container. The slurry was stirred for 15 minutes and 11 parts of water added to reduce the viscosity of 80,000 cps.

- Film material was prepared on a 4 zone conventional band dried using overband heating at 150° C. and underband heating on last 3 zones with slurry spread at 0.030 in. and band speed of 12 ft./min.

- The film material was shredded and processed satisfactorily on cigarette making machinery.

- 1.1 g. cigarettes gave 10 puffs. The smoke was mild to taste.

- The dry weight composition of Example 2 was:—

		%
45	Tobacco	27.5
	CaCO ₃	37.2
	MgCO ₃	7.0
	Potassium citrate	4.3
	(NH ₄) ₂ SO ₄	4.3
	Citric acid	4.9
50	Glycerol	7.0
	SCMC	7.8

Example 3

- 9.6 parts tobacco rag were added to 45.6 parts of water and allowed to soak for 15 minutes, after which time the mixture was transferred to a Probst and Class mill and milled at setting 3 for 10 minutes. The suspension was returned to the stirrer and after the addition of 13.7 parts calcium carbonate, 1.4 parts potassium citrate, 1.4 parts ammonium sulphate, 2.4 parts glycerol, 2.4 parts

magnesite and 1.7 parts citric acid, it was stirred for 30 minutes. The batch was then processed in the mill, again at gap setting 2, for 5 minutes and at gap setting zero for 5 minutes. 1.7 parts of locust bean gum and 7.5 parts of water were added at this stage. The slurry was discharged immediately after dispersion of the locust bean gum into a container and stirred for a further 20 minutes. 12.7 parts of water were added to reduce the viscosity to 30,000 cps.

The slurry was spread at 0.02 in. gap on a band drier and film made using a combination of overhand hot air at 150° C. and steam underband heating. The band speed was 24 ft./min.

The film was satisfactorily shredded and processed in cigarette making machinery. 1.1 g. cigarettes gave 10 puffs. The smoke was mild to taste.

The dry weight composition of Example 3 was:—

		%	
	Tobacco	28	85
	CaCO ₃	40	
	MgCO ₃	7	
	(NH ₄) ₂ SO ₄	4	
	Potassium citrate	4	
	Citric acid	5	90
	Glycerol	7	
	Locust bean gum	5	

Example 4

- 2.04 parts glycerol and 1.17 parts potassium citrate were dissolved in 150 mls. of water. 2.88 parts SCMC were then added to the solution and stirred for 15 minutes. 6.0 parts ball milled, flue cured Virginia tobacco; 6.6 parts powdered chalk and 11.31 parts magnesium carbonate were mixed together in the dry state, then added to the previous mixture. The whole was stirred for 1 hour.

Film material was made by spreading the slurry at 0.02 inches on glass plates and left to dry. The film material was humidified and shredded.

Ten cigarettes were hand rolled with Imperial Verge paper. 1.1 g. cigarettes gave 12 standard puffs. The smoke was mild to taste.

Dry weight composition:—

		%	
	Virginia Tobacco	20.0	
	Glycerol	6.8	
	SCMC	9.6	
	CaCO ₃	22.0	115
	MgCO ₃	37.7	
	Potassium citrate	3.9	

Example 5

- 0.9 parts glycerol were dissolved in 150 mls. of water. 2.1 parts SCMC were then added to the solution and stirred for 15 minutes. 7.5 parts flue cured, ball milled Virginia

Tobacco; 4.8 parts sodium silicate and 14.7 parts calcium oxalate were mixed together in the dry state, then added to the previous mixture. The whole was stirred for 1 hour.

5 Film material was made by spreading the slurry at 0.02 inches on glass plates and left to dry. The film material was humidified and shredded.

10 Ten cigarettes were hand rolled with Imperial Verge paper. 1.1 g. cigarettes gave 10 standard puffs. The smoke was mild to taste.

Dry weight composition:—

	%
15 Virginia Tobacco	25.0
Glycerol	3.0
SCMC	7.0
Sodium silicate	16.0
Calcium oxalate	49.0

Example 6

20 2.1 parts glycerol, 1.2 parts ammonium sulphate; 1.2 parts potassium citrate and 1.5 parts citric acid were dissolved in 150 mls. of water. 1.5 parts sodium alginate were then added to the solution and stirred for 15

25 minutes. 2.1 parts magnesium carbonate; 12.0 parts powdered chalk and 8.4 ball-milled, fine cured virginia tobacco were mixed together in the dry state, then added to the previous mixture. The whole was stirred for 1 hour.

30 Film material was made by spreading the slurry at 0.02 inches on glass plates and left to dry. The film material was humidified and shredded. Ten cigarettes were hand rolled with Imperial Verge paper. 1.1 g. cigarettes gave

35 11 standard puffs. The smoke was mild to taste.

Dry weight composition:—

	%
40 Virginia Tobacco	28.0
Glycerol	7.0
Sodium alginate	5.0
CaCO ₃	40.0
MgCO ₃	7.0
Potassium citrate	4.0
45 (NH ₄) ₂ SO ₄	4.0
Citric Acid	5.0

Example 7

50 A smoking mixture having the dry weight composition listed below was prepared in a manner similar to that of Examples 1 to 6

	%
55 Tobacco	60.0
SCMC	10.0
Glycerol	10.0
Potassium Citrate	3.9
CaCO ₃	6.4
MgCO ₃	9.7

1.1 g. hand rolled cigarettes (Imperial Verge

Paper) gave 14 standard puffs. The smoke was mild to taste. 60

Example 8

A smoking mixture having the dry weight composition listed below was prepared in a manner similar to that of Examples 1 to 6 65

	%	
Tobacco	60.0	
SCMC	10.0	
Glycerol	10.0	
Potassium Citrate	3.9	70
CaCO ₃	12.4	
MgCO ₃	3.7	

1.1 g. hand rolled cigarettes (Imperial Verge Paper) gave 16 standard puffs. The smoke was mild to taste. 75

Example 9

A smoking mixture having the dry weight compositions listed below was prepared in a manner similar to that of Examples 1 to 6

	%	
Tobacco	60.0	80
SCMC	10.0	
Glycerol	10.0	
Potassium Citrate	3.9	
CaCO ₃	16.10	85

1.1 g. hand rolled cigarettes (Imperial Verge Paper) gave 15 standard puffs. The smoke was mild to taste.

We are aware of the Customs and Excise Act 1952 and we make no claim to the use of the invention in contravention of the law. 90

WHAT WE CLAIM IS:—

1. A smoking mixture comprising tobacco, a harmless inorganic filler and a binder, the proportion by weight of said filler in the mixture being from 40% to 65% and the combination of cations and anions in the filler being such as to impart to the mixture a commercially acceptable burning rate. 95

2. A smoking mixture as claimed in Claim 1 wherein the harmless inorganic filler comprises 40 to 60% by weight of the smoking mixture. 100

3. A smoking mixture as claimed in any one of the preceding claims wherein the inorganic filler comprises a mixture of two or more inorganic salts admixed in quantities such as to obtain the balance of anions and cations most appropriate for imparting an acceptable rate of combustion to the smoking mixture. 110

4. A smoking mixture as claimed in any one of Claims 1 to 3 wherein the inorganic filler comprises a single inorganic salt present in an amount such as to obtain an acceptable rate of combustion for the smoking mixture. 115

5. A smoking mixture as claimed in any



Tobacco; 4.8 parts sodium silicate and 14.7 parts calcium oxalate were mixed together in the dry state, then added to the previous mixture. The whole was stirred for 1 hour.

- 5 Film material was made by spreading the slurry at 0.02 inches on glass plates and left to dry. The film material was humidified and shredded.

- 10 Ten cigarettes were hand rolled with Imperial Verge paper. 1.1 g. cigarettes gave 10 standard puffs. The smoke was mild to taste.

Dry weight composition:—

	%
15 Virginia Tobacco	25.0
Glycerol	3.0
SCMC	7.0
Sodium silicate	16.0
Calcium oxalate	49.0

Example 6

- 20 2.1 parts glycerol, 1.2 parts ammonium sulphate; 1.2 parts potassium citrate and 1.5 parts citric acid were dissolved in 150 mls. of water. 1.5 parts sodium alginate were then added to the solution and stirred for 15 minutes. 2.1 parts magnesium carbonate; 12.0 parts powdered chalk and 8.4 ball-milled, flue cured virginia tobacco were mixed together in the dry state, then added to the previous mixture. The whole was stirred for 1 hour.

- 30 Film material was made by spreading the slurry at 0.02 inches on glass plates and left to dry. The film material was humidified and shredded. Ten cigarettes were hand rolled with Imperial Verge paper. 1.1 g. cigarettes gave 11 standard puffs. The smoke was mild to taste.

Dry weight composition:—

	%
40 Virginia Tobacco	28.0
Glycerol	7.0
Sodium alginate	5.0
CaCO ₃	40.0
MgCO ₃	7.0
Potassium citrate	4.0
45 (NH ₄) ₂ SO ₄	4.0
Citric Acid	5.0

Example 7

- 50 A smoking mixture having the dry weight composition listed below was prepared in a manner similar to that of Examples 1 to 6

	%
55 Tobacco	60.0
SCMC	10.0
Glycerol	10.0
Potassium Citrate	3.9
CaCO ₃	6.4
MgCO ₃	9.7

1.1 g. hand rolled cigarettes (Imperial Verge

Paper) gave 14 standard puffs. The smoke was mild to taste. 60

Example 8

A smoking mixture having the dry weight composition listed below was prepared in a manner similar to that of Examples 1 to 6 65

	%	
Tobacco	60.0	
SCMC	10.0	
Glycerol	10.0	
Potassium Citrate	3.9	70
CaCO ₃	12.4	
MgCO ₃	3.7	

1.1 g. hand rolled cigarettes (Imperial Verge Paper) gave 16 standard puffs. The smoke was mild to taste. 75

Example 9

A smoking mixture having the dry weight compositions listed below was prepared in a manner similar to that of Examples 1 to 6

	%	
Tobacco	60.0	80
SCMC	10.0	
Glycerol	10.0	
Potassium Citrate	3.9	
CaCO ₃	16.10	85

1.1 g. hand rolled cigarettes (Imperial Verge Paper) gave 15 standard puffs. The smoke was mild to taste.

We are aware of the Customs and Excise Act 1952 and we make no claim to the use of the invention in contravention of the law. 90

WHAT WE CLAIM IS:—

1. A smoking mixture comprising tobacco, a harmless inorganic filler and a binder, the proportion by weight of said filler in the mixture being from 40% to 65% and the combination of cations and anions in the filler being such as to impart to the mixture a commercially acceptable burning rate. 95

2. A smoking mixture as claimed in Claim 1 wherein the harmless inorganic filler comprises 40 to 60% by weight of the smoking mixture. 100

3. A smoking mixture as claimed in any one of the preceding claims wherein the inorganic filler comprises a mixture of two or more inorganic salts admixed in quantities such as to obtain the balance of anions and cations most appropriate for imparting an acceptable rate of combustion to the smoking mixture. 105

4. A smoking mixture as claimed in any one of Claims 1 to 3 wherein the inorganic filler comprises a single inorganic salt present in an amount such as to obtain an acceptable rate of combustion for the smoking mixture. 110

5. A smoking mixture as claimed in any

one of the preceding claims wherein the inorganic filler comprises an inorganic salt of an organic acid.

5 6. A smoking mixture as claimed in any one of the preceding claims wherein the anion of the inorganic filler comprises formate, oxalate, citrate, tartrate, silicate, carbonate, sulphate, phosphate or borate, and the cation of the inorganic filler comprises sodium, potassium, calcium or magnesium.

10 7. A smoking mixture as claimed in any one of the preceding claims wherein the inorganic filler comprises calcium carbonate, calcium orthophosphate, magnesium carbonate, calcium sulphate, calcium oxalate or mixtures thereof.

8. A smoking mixture as claimed in any one of the preceding claims comprising a plasticiser/humectant in addition to the binder.

20 9. A smoking mixture as claimed in claim 8 wherein the plasticiser/humectant comprises glycerol, ethylene glycol or polyethylene glycol.

25 10. A smoking mixture as claimed in any one of the preceding claims wherein the binder comprises carboxymethyl cellulose and its salts, other carboxylated carbohydrates, natural gum, guar gum, locust bean gum or cellulose ethers.

30 11. A smoking mixture as claimed in any one of the preceding claims comprising medicaments, flavourants, drugs and other known additives.

35 12. A smoking mixture substantially as hereinbefore described with reference to the Examples.

13. A method of manufacturing a smoking mixture comprising mixing the ingredients specified in claim 1 and thereafter forming a shaped article of the mixture.

40 14. A method as claimed in Claim 13 wherein the shaped article is a film, extruded filament or tape.

45 15. A method as claimed in Claim 13 or Claim 14 comprising combining an inorganic filler which is a combustion accelerator with an inorganic filler which is a combustion retarder in proportions which gives a desirable rate of combustion for a specific amount of tobacco in the smoking mixture.

50 16. A method as claimed in any one of Claims 13 to 15 wherein the anion of the inorganic filler comprises formate oxalate, citrate, tartrate, silicate, carbonate, sulphate, phosphate or borate and the cation of the inorganic filler comprises sodium, potassium, calcium or magnesium.

55 17. A method as claimed in any one of Claims 13 to 15 wherein the inorganic filler comprises calcium carbonate, calcium orthophosphate, magnesium carbonate, calcium sulphate, calcium oxalate, or mixtures thereof.

60 18. A method of manufacturing a smoking mixture substantially as hereinbefore described.

65 19. A smoking mixture prepared according to the method of any one of Claims 13 to 17.

A. McPHERSON,
Agent for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1972.
Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.

