

DEVELOPMENT OF A SYNTHETIC SMOKING MATERIALObjective

The development of a smoking material with smoke characteristics which are acceptable in terms of taste and flavour and biologically inactive.

Present knowledge(a) Biological

The specific biological activity of smoke condensate depends, in part, on the type of tobacco used; thus Flue-cured tobacco > Burley tobacco > Air cured fermented tobacco (1). There are also indications that smoke condensate from stem has a lower specific biological activity than that from lamina and that smoke condensate from reconstituted tobacco has a lower specific biological activity than that from tobacco (2).

Amcel have developed a synthetic smoking material, based on α -cellulose, which has little or no biological activity (3). Whilst a number of additives are incorporated to modify combustion, indications are that the pyrolysis of cellulose alone yields a smoke condensate of low biological activity.

(b) Chemical

Examination of smoke condensates from cigarettes made from lamina, C.R.S. and PCL shows that the benzpyrene product efficiencies are lamina > PCL > CRS (4). That PCL is intermediate between lamina and C.R.S. is as would be expected since PCL is made from three parts of lamina plus one part of stem binder which has been obtained by the extraction of two parts of stem with water under pressure.

If, instead of a ligno-cellulose stem binder, pure cellulose is substituted, there is a reduction in tar delivery which could indicate a lower biological activity (5). If this is so, then it may be reasonable to assume that biological activity can be ranked as follows :-

lamina > stem > extracted stem > cellulose.

A number of additives have been found effective in reducing the biological activity of tobacco smoke condensate. These are indicated below, when considering means of controlling the biological activity of smoke from a synthetic smoking material.

(a) Physical

Physical methods, such as changes in cuts per inch and cigarette paper porosity may be used to reduce the delivery of benzpyrene (6,7). Specific activity, however, is only marginally altered and thus these methods will not be considered further here. Likewise, the effect of filters has been excluded from the present proposals.

100050133

An examination of reconstituted tobacco (Folie B) indicates that increasing the base weight from 40 g./m² to 90 g./m² reduces the specific biological activity of the smoke condensate (8), thus the physical nature of a synthetic smoking material may have a considerable bearing on its smoke characteristics.

Benzpyrene delivery may be affected by smoulder temperature (9). This would be in line with pyrolysis experiments which indicate a 20-fold increase in benzpyrene when the temperature is increased from 700° C. to 900° C. (10). As the smoulder temperature of Burley tobacco is lower than that for flue cured tobacco (11), this may account for the lower specific biological activity of Burley compared with flue cured tobacco.

Another possibility for reducing the specific biological activity of smoke condensate lies in attempts to obtain more complete combustion by increasing the combustion temperature (12). A similar effect may be achieved by the addition of carbonates, bicarbonates and oxidising agents (3).

From the above it is clear that means are available for reducing the specific biological activity of tobacco smoke. These are being examined in R. & D.E. and it is not proposed to duplicate this work. The aim of the work outlined here is, as previously stated, to develop a synthetic smoking material.

Choice of approach and justifications

(a) Choice of base material

The range of materials, from which a suitable base material may be selected, is wide and includes the following :-

- (1) Polysaccharides such as cellulose, either as natural fibres or as synthetic fibres e.g. viscose, pectates, alginates, starch and guar gum.
- (2) Modified celluloses such as methyl cellulose, carboxymethyl cellulose, hydroxyethylcellulose, cellulose acetate and carbonized and oxidised celluloses.
- (3) Polyesters including Terylene.

In the first instance it is proposed to examine the following :-

(1) Cellulose

Besides being a pure material, thus making it ideal for pyrolysis studies, the work of Ancel and also our own use of cellulose as a binder in PCL, indicate that cellulose may produce a smoke condensate of low biological activity. This, coupled with economic attractiveness and the "ease" of producing cigarettes, makes cellulose an ideal choice, at least for initial examination. In common with all cellulosic materials, the acrid character of the smoke is likely to be a major problem in the development of a tobacco substitute.

100056134

(2) Oxidized cellulose

Amcel noted (3) that when cellulose is replaced by oxidized cellulose - a polymer of anhydroglucuronic acid - the smoke is sweeter and much less irritating than that obtained from pure cellulose. If further work confirms this, oxidized cellulose, which may be obtained by the oxidation of cellulose by nitrogen dioxide, could be a very attractive base material.

(3) Carbonized cellulose

If one considers the base material simply as a heat sump, which on combustion volatilizes nicotine and flavours, it may be possible to carbonize cellulose partially and still obtain a material which will smoulder in a satisfactory manner. Whilst this may give a useful insight into the requirements of a synthetic smoking material, the practicality of carbonizing large quantities of cellulose may be questioned.

(4) Methyl Cellulose and Carboxy Methyl Cellulose

These additives have been used in the manufacture of cigar wrapper and binder and considerable work undertaken to ameliorate their peculiar smoke taste (13-16). Since, like other cellulose derivatives, methyl and carboxy-methyl cellulose may be formed into films which could readily be cut for cigarette manufacture, their examination as a base material is considered worthwhile.

(5) Natural Polysaccharides

Materials such as pectins, alginates and guar gum have frequently been used in the manufacture of reconstituted tobaccos (17-19). As with the cellulose derivatives mentioned above, work has been carried out on their pyrolysis and again their examination is considered worthwhile.

(6) Dialdehyde Starch

Starch itself produces, on combustion, a very acrid smoke. It is believed that dialdehyde starch has been considered as a binding agent in reconstituted tobacco and thus it is proposed to examine its possible use as a base material.

(7) Polyesters

Polyesters have been proposed for a number of reconstituted tobacco processes (20-22). During the initial work on the development of PGL in 1956-1957, laboratory-made polyesters based on acids such as citric, maleic and fumaric and alcohols such as glycerol were examined. They had an attractive smoke taste and thus it may be worthwhile considering these polyesters anew as possible base materials. Of the various commercial polyesters available, Terylene could be examined.

100050135

(b) Choice of additives

Additives will be required for the following :-

- (1) Burn control.
- (2) Flavour control.
- (3) Control of biological activity.
- (4) Provision of nicotine.
- (5) Possibly colour.

(1) Burn control

By burn control, in this context, is meant not only control of the rate of combustion but, also, if necessary, the amelioration of undesirable characteristics of the smoke such as the acrid components associated with the burning of cellulose.

Ameliorants used in the tobacco industry include citric, lactic, tartaric and hydrochloric acid, magnesium chloride, sodium bisulphate and cocoa butter. Likewise combustion agents include the chloride, acetate and nitrate of sodium and potassium carbonate and citrate.

In the development of SM.65, Amcel have found that hydrated salts, such as magnesium sulphate, sodium sulphate, alumina and calcium tartrate, are effective both as ameliorants and as reducers of the rate of combustion (3). In addition they found that a number of agents, including the chloride, sulphate and nitrate of potassium, the chloride and nitrate of magnesium, the oxide and hydroxide of ferric iron, bentonite and infusorial earths, effective in maintaining combustion during the smoulder period. Certain salts also assist in favouring a more complete combustion by dilating the combustion zone. These include the carbonates and bicarbonates of sodium, potassium, magnesium and ammonia.

(2) Flavour control

In developing a synthetic smoking material it will be necessary to build in a flavour character acceptable to the smoker. "Flavour character" rather than "tobacco-like character" has been used intentionally to describe the smoke taste since it may be necessary, by a process of gradual education, to convert the smoker to a new flavour sensation. When one considers the success that has been achieved with mentholated cigarettes, this may not be as difficult as at first imagined, particularly if it could be demonstrated that the cigarette was safer. This is not to say that it may not be possible to make a synthetic cigarette of tobacco-like character, but the possibility should be borne in mind.

Besides the many additives, well known in the tobacco industry, for imparting particular flavours, the considerable work carried out by the Reynolds Tobacco Company and divulged in numerous patent applications (23-44), will require examination.

100050136

(3) Control of biological activity

Investigations indicate that a number of additives, such as magnesium oxide, cupric nitrate, aluminium silicate, nickelous acetate and sodium nitrate may be effective in reducing the specific biological activity of cigarette smoke condensate (1, 45-47). Ammonium sulphamate has been found effective in reducing the benzpyrene delivery of cigarettes (48). It is believed that ammonium salts, such as the sulphate, carbonate, persulphate and perchlorate, are also effective in reducing benzpyrene delivery (3).

The production of benzpyrene is temperature dependent (10), thus any additives which, in particular, reduce the smoulder temperature may be effective in reducing the delivery of benzpyrene.

(4) Provision of nicotine

Although nicotine may be a contributing factor in certain cardiovascular conditions, it is a basic assumption of the present work that the synthetic smoking material must be a vehicle for the administration of the alkaloid i.e. the smoker requires nicotine "dressed up" in an attractive guise.

It will be necessary to obtain nicotine either by synthesis or by the extraction of tobacco. The form and level of addition of nicotine will require examination.

(5) Colouring

If a product can be developed which has the physical appearance of tobacco, it may be desirable, for marketing purposes, to impart a yellow colour. Care must be taken in choosing suitable dyestuffs but at least one - Tartrazine - would appear acceptable.

(c)

Choice of process

A number of processes might be considered :-

(1) Conventional - cutting a sheet or film which would enable cigarettes to be made in a normal manner.

(2) "Bonded" cigarette rod - similar to bonded filters.

(3) Extrusion of a continuous rod either in filament or blown form.

Whilst it is preferable to develop a suitable smoking material and then devise a method of manufacture, it should be remembered that the conventional and bonded rod processes have a distinct advantage over extrusion, in that they would allow blending with natural tobacco. This is desirable as it would permit the gradual introduction of the synthetic material.

100056137

Development of processes, methods of test and evaluation criteria

(a) Work to be carried out by PCL Group

The PCL Group will undertake an examination of the pyrolysis of the various base materials proposed.

For example, with cellulose, various types of absorbent papers will be examined primarily to see if the physical form of the sheet has a material effect on smoke delivery. From this the most suitable paper will be chosen for further work. This will involve a study of additives to achieve burn control and also to ameliorate any undesirable smoke taste. Following this additives for flavour control and the minimisation of biological activity should be examined. Finally, the incorporation of nicotine and any required colouring in the product must be investigated.

Other base materials will be examined in a similar fashion.

In addition to the above, consideration will also have to be given to the following :-

(1) Means of incorporating additives. This may require a pilot plant coating or spray unit.

(2) Means for cutting small batches of the synthetic smoking material so that cigarettes may be made for evaluation.

Whilst the initial aim will be to develop a synthetic smoking material which may be utilized either in a conventional manner or in a bonded cigarette form, the practicality of extruding a continuous rod either in a filament or in a blown form will be examined. In this context some work carried out by A.M. & F. is of interest (49).

(b) Ancillary requirements

(1) Smoke Group

Since present evidence indicates that a part, at least, of the activity of smoke condensate may be due to the presence of aromatic polycyclic hydrocarbons, it is suggested that changes in the delivery of A.P.H. should be used as a guide to the examination of new smoking materials. Two procedures being developed by Smoke Group could be useful :-

(i) A short method for determining gross changes.

(ii) For materials which successfully pass the first test, a complete analysis.

100050138

(1) A short method for determining gross changes

A short method for benzpyrene does not, at present, appear feasible. However, a quick method for the determination of a number of closely related C20 A.P.H. does appear possible. Using either the Nickel 63 electron capture detector or the Aminco-Bowman spectrofluorimeter it is hoped to develop a rapid method, based on smoking 10-20 cigarettes.

(ii) Complete analysis

The existing method, based on the flame ionization detector of the gas chromatograph, is being developed further with the aim of quantitatively determining a number of A.P.H. It is hoped to get one result per assistant per week or better.

In addition to the above, methods of analysis using total integrated fluorescence will be examined in the hope of developing a quick routine method for the determination of A.P.H.

Smoke analyses will also be required to relate the smoke to observed ciliastatic and necrotic effects and to minimize the delivery of undesirable components such as phenols, aldehydes, hydrogen cyanide and oxides of nitrogen.

Smoke Group or the PCL Group may also make use of the R. & D.E. furnace as a means of evaluating the effect of additives. It must be emphasised, however, that the furnace can only be used to study the effect of additives which operate by altering the gas phase reactions and not those that alter the puff and smoulder temperatures of cigarettes. Previous mention has been made of the various types of additives required. It is suggested that Smoke Group, with their background of knowledge, will be able to assist particularly with the choice of additives for the control of flavour and biological activity and also for the provision of nicotine.

(2) Bio-assay section

Where possible short term tests should be used to guide the development of the synthetic smoking material. Such tests which it may be possible to carry out in R. & D.E. include :-

- (1) The Hanging-drop Paramecium test.
- (ii) The Second Paramecium test.
- (iii) The Necrotic test.
- (iv) Any other tests which may be developed.

(3) JANUS

If and when a synthetic smoking material is developed, which has an acceptable smoke character and which, by chemical and bio-assay tests, meets the objective, it will be necessary to carry out a long term mouse painting experiment before giving the new material final clearance.

100050139

Staff, time scale and costs

Besides the considerable assistance that will be required for the ancillary studies of the smoke, two graduates will be required for the initial development work. One, yet to be recruited, will be needed for the preparation and examination of materials which may be processed either in conventional or "bonded" cigarette form, whilst a second will examine the feasibility of extrusion of a continuous rod either in filament or blown form.

Due to the ad hoc nature of the work no date can be given for the completion of the project. Within 12 months of inception however, a preliminary examination of the selected base materials and the feasibility of extrusion should be completed. This would cost approximately £50,000, after which it is hoped that lines of development may be more clearly defined and an overall time scale predicted. As a guide line it has been estimated (50), that the development of the PCL process cost £400,000 over a four year period and it is unlikely that a synthetic smoking material would be developed for less.

100050140

References

- (1) Wynder E.L. and Hoffmann D. Deut. Med. Wochschr. 1963. 88,623.
- (2) T.P.C. Document D.917.
- (3) Belgian Patent No. 639,243, Ancel Product Bulletin PBS-1.
- (4) R. & D.E. Report No. 210-R.
- (5) R. & D.E. Report No. 121-R.
- (6) Wynder E.L. and Hoffmann D. Unpublished work 1963.
- (7) R. & D.E. Report No. 246-R.
- (8) Prof. Kracht, Eppendorf Klinik, Hamburg-Eppendorf. Unpublished work.
- (9) R. & D.E. Report No. 319-R.
- (10) R. & D.E. Report No. 314-R.
- (11) R. & D.E. Report No. 331-R.
- (12) Muth F. - Unpublished work.
- (13) Frankenburg W.G. and Garbo P.W. - U.S. Patent 2,592,553.
- (14) Frankenburg W.G. - " 2,598,680.
- (15) Frankenburg W.G. - " 2,706,695.
- (16) Frankenburg W.G. - " 2,797,689.
- (17) Frankenburg W.G. - " 2,592,554.
- (18) Samfield M.M. et al - " 2,708,175.
- (19) Bandel D. - U.K. Patent 824,324.
- (20) Carmellini A.E. and Hotelling E.B. - U.S. " 2,949,117.
- (21) Carmellini A.E. and Hotelling E.B. - U.S. " 2,976,873.
- (22) Hotelling E.B. and Kelly T.E. - U.S. Patent 2,957,478.
- (23) Jones S.O. - U.S. Patent 2,766,145.
- (24) Ashburn J.G. - " 2,766,146.
- (25) Rowland R.L. - " 2,766,147.
- (26) Rowland R.L. - " 2,766,148.
- (27) Rowland R.L. - " 2,766,149.
- (28) Teague C.E. - " 2,766,150.
- (29) Teague C.E. - " 2,869,557.
- (30) Teague C.E. - " 2,872,360.
- (31) R.J. Reynolds Tobacco Co. - U.K. Patent 838,856.
- (32) Hoover K.H. - U.S. Patent 2,886,042.
- (33) Giles J.A. - U.S. Patent 2,905,575.
- (34) Schumacher J.N. - U.S. Patent 2,905,576.
- (35) Schumacher J.N. - U.S. Patent 2,978,365.
- (36) Keaton J.L. - U.S. Patent 3,006,347.
- (37) Henley W.M. - U.S. Patent 3,041,211.
- (38) Mins S.S. - French " 1,346,170.
- (39) Fredrickson J.D. - U.S. Patent 3,124,140.
- (40) Rowland R.L. - U.S. Patent 3,211,157.
- (41) Roberts D.L. & Schumacher J.N. - U.S. Patent 3,217,716.
- (42) Roberts D.L. - " 3,217,717.
- (43) Roberts D.L. - " 3,217,718.
- (44) R.J. Reynolds Tobacco Co. - U.K. Patent 1,061,090.
- (45) Wynder E.L. and Hoffmann D. - Acta Pathol. Microbiol. Scand. 1961,52,119.
- (46) Kiefer J.E. and Mumpower R.G.) - Tennessee Eastman Co.,
- (47) Leonard R.E. and Miller E.G.) - Private communication.
- (48) Cuzin J.L. et al - Z. Praventivmedizin 1963, 8, 125.
- (49) American Machine & Foundry Co. - U.K. Patent 1,013,303.
- (50) R. & D.E. Report No. RD.375-R.

100050141