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The refractive indices of polymer film formers, pigments and additives used in tablet film coating: their significance and practical application

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The refractive indices of both the polymer film formers and additives are a fundamental factor in defining the appearance of the subsequent film coatings. The refractive indices of the polymer film formers are all in the region of 1·5 but those of pigments and additives vary between 1·5–3·2. Most pigments and additives are also anisotropic, i.e. they possess different refractive indices depending on their orientation. These observations have implications in the use of pigments and additives for the production of both opaque films with good hiding power and film-coated tablets with highlighted intagliations.

Additives in the form of pigments (e.g. aluminium lakes of water soluble dyes), opacifiers (e.g. titanium dioxide) and various inorganic materials (e.g. iron oxides, calcium carbonate, talc) are often included in film coating formulations (Pickard & Rees 1974; Porter 1980; Rowe 1982). The refractive indices of these materials and the polymer film formers used are a fundamental factor defining the appearance of the subsequent film coatings. We have collected literature values for the refractive indices of these materials and discuss their significance and practical application in tablet film-coating.

Theory

All materials, including pigments, additives and polymer film formers, can be divided optically into two groups: those that are isotropic and so possess one refractive index, and those that are anisotropic and therefore possess different refractive indices along different axes. Anisotropic materials are usually referred to as uniaxial, i.e. they possess two refractive indices designated by ε and ω, or biaxial, i.e. they possess three refractive indices designated by α, β and γ.

The capacity of a film formulation containing dispersed additives or pigments to reflect light, its hiding power or opacity and depth of colour all depend on the respective refractive indices of the dispersed solid and the polymer film former. These properties all derive from the fact that at each polymer/additive interface a certain fraction of the incident light is reflected. The amount of light reflected at this interface (R), assuming normal incidence and no absorption, can be estimated by the formula given by Cooper (1948):

$$R = \left[\frac{n_1 - n_2}{n_1 + n_2}\right]^2 \quad (1)$$

where n_1 is the refractive index of the additive and n_2 is the refractive index of the polymer film former. Fig. 1 shows the change in R for a polymer film former, $n_2 = 1{\cdot}5$, containing additives of increasing refractive index n_1. It can be seen that if $n_1 = n_2$ then R will be zero and the film will appear transparent. If, however, $n_1 \neq n_2$, then the larger the difference between n_1 and n_2, the larger the value of R. If there is a sufficient number of interfaces i.e. in the case of the additive being finely dispersed throughout the film, then the combined effect of all the reflected light and subsequent scattering will make the film more and more opaque. It can also be seen that for anisotropic materials, the amount of reflected light will change depending on their orientation or alignment within the film. If the materials orientate equivalent to the state of their lowest refractive indices, then R will be smaller than if they orientate equivalent to the state of their highest refractive indices. The degree of opacity of a film will thus vary with the orientation of these materials.

Practice

These observations are important in the film coating of tablets especially in the formulation of opaque coatings with good hiding power to overcome the problem of interbatch colour variation in tablet cores, and in the production of tablets with highlighted intagliations to improve product identification.

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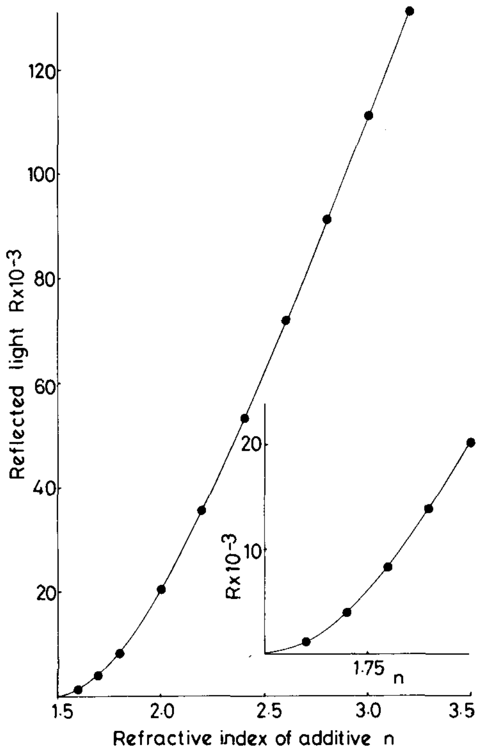

FIG. 1. The increase in the reflected light at the polymer/additive interface with change in refractive index of the additive for a polymer film former of refractive index 1·5.

Formulation of opaque film coatings

Many drug substances like the tetracyclines are coloured and exhibit interbatch variation in colour leading to interbatch colour variation in tablet cores. When this occurs a film formulation with good opacity and hiding power is required to ensure good batch reproducibility in the final film coated tablets. For this purpose the incorporation of an additive with a high refractive index is needed to increase reflection at the polymer/additive interface and subsequent scattering. It can be seen (Table 1) that titanium dioxide fulfils this criterion, hence its widespread use alone or as mixtures with the aluminium lakes. The use of aluminium lakes alone in this situation would be contraindicated since the films prepared would be essentially transparent with little hiding power. Red iron oxide is also a useful pigment in producing opaque films with good hiding power. For both titanium dioxide and red iron oxide the orientation of the materials within the film is irrelevant since the lower refractive indices of both materials are very high. This is not the case with yellow iron oxide with its acicular particles and a

Table 1. The refractive indices of some additives and pigments.

Additive	Refractive indices	Reference
Alumina hydrate	1.50–1.56	Gettens & Stout (1966)
Aluminium lakes	1.50–1.54*	Manufacturer's data, Warner-Jenkinson Co.
Calcium carbonate	ε1.510, β1.645	Gettens & Stout (1966)
Calcium sulphate	α1.57, β1.575, γ1.614	Gettens & Stout (1966)
Magnesium carbonate	ε1.516, β1.710	Partington (1953)
α-Lactose	α1.517, β1.553, γ1.555	Partington (1953)
Talc	α1.539, β1.589, γ1.589	Gettens & Stout (1966)
Kaolin	α1.558, β1.564, γ1.565	Gettens & Stout (1966)
Titanium dioxide (anatase)	ε2.493, ω2.554	Partington (1953)
Red iron oxide	ε2.94, ω3.22	Partington (1953)
Yellow iron oxide	α1.94, β2.20, γ2.51	International Critical Tables (1928)

* These values will be dependent on the dye content of the lake (Cooper 1948).

relatively wide spread in its refractive indices. This oxide is known to cause problems in paint films since it tends to orientate in the direction of the brush strokes resulting in stripes or streaks appearing either darker or lighter than the surrounding surface depending on the direction from which they are viewed (Kresse 1966).

Production of tablets with highlighted intagliations

Recent work with calcium carbonate and talc in tablet film coatings prepared from hydroxypropyl methylcellulose has shown that both these materials show orientation (Rowe 1983). Over the main surface of an intagliated tablet these materials were found to orientate equivalent to their state of lowest refractive index, i.e. the film was essentially transparent, but within the intagliation they were randomly orientated or orientated equivalent to their state of highest refractive index, resulting in a white opaque film. When such a formulation was applied to coloured tablet substrates the intagliations were found to be highlighted in white. This observation has recently been shown to have wide applicability (Forse & Rowe 1981, 1982).

This highlighting effect will be most distinct, if, firstly, the anisotropic material used has its lowest refractive index equivalent to that of the polymer film former but, secondly, has a spread of refractive indices as large as possible. When this occurs the film will be totally transparent over the main surface of the tablet and very opaque within the intagliations. It can be seen from Tables 1 and 2 that, for most of the polymer film formers, calcium carbonate and magne-

FIG. 2. Coloured intagliated tablets coated with hydroxypropyl methylcellulose films containing magnesium carbonate.

FIG. 3. Examples of the colour combinations obtained when the tablets shown in Fig. 2 are further coated with hydroxypropyl methylcellulose films containing water-soluble dyes.

Table 2. The refractive indices of some film coating polymers.

Polymer	Refractive Index	Reference
Methylcellulose	1·50	Manufacturer's data, Dow Chemical Co.
Ethyl cellulose	1·47	Savage (1965a)
Hydroxyethylcellulose	1·51	Savage (1965b)
Hydroxypropyl cellulose	1·56	Manufacturer's data, Hercules Inc.
Hydroxypropyl methylcellulose	1·49	Manufacturer's data, Shin-Etsu Chemical Co.
Sodium carboxy methylcellulose	1·52	Klug (1965)
Cellulose acetate	1·48	Manufacturer's data, Hercules Inc.
Acrylic resins	1·48	Manufacturer's data
Shellac	1·52	Manufacturer's data

sium carbonate fulfil both criteria. Various coloured intagliated tablets coated with hydroxypropyl methylcellulose containing magnesium carbonate are shown in Fig. 2. In every case the intagliations are highlighted in white against a coloured background.

Aqueous tablet film coatings based on the water soluble cellulose ethers have led to water soluble dyes gaining in popularity as colouring agents. A dyed film in this context is transparent with no hiding power. If such a coloured film is applied to an already coloured tablet with highlighted intagliations, or if an anisotropic material such as calcium carbonate or magnesium carbonate is included in such a formulation and applied to a coloured substrate, then numerous colour combinations are possible, the intagliations normally being seen as a pale version of the colour of the dye used. If the colour of the tablet substrate and the dyed film are so-called complementary colours, then the main body of the tablet will appear black. Examples of such colour combinations are shown in Fig. 3.

In both of the above applications the degree of opacity of the applied film will be dependent on the number of polymer/additive interfaces implying that the particle size of the additive is a very important factor. This has indeed been shown to be the case in optimizing the hiding power of paint films (Gerstner 1966) but no data as yet exist for tablet film coatings.

In conclusion, it can be seen that the refractive index is a fundamental property of both the polymer film formers and additives playing an essential role in defining the appearance of the subsequent film, and is thus of practical importance in the film coating of tablets and other oral dosage forms.

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