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The effect of batch and source variation on the crystallinity of microcrystalline cellulose

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Summary

Crystallinities of samples of microcrystalline cellulose of different particle sizes from different batches obtained from a variety of sources have been measured using both an X-ray diffraction and an infrared method. Batchwise variation was found to be of a magnitude that neutralises any effect of particle size and/or source of material.

Microcrystalline cellulose is used extensively in the production of granules, extrudates, spheroids and tablets. It is now obtainable from sources/manufacturers in many countries often in several grades with different particle sizes/moisture content. Recent work on granulation (Rowe and Sadeghnejad, 1987; Parker and Rowe, 1991; Parker et al., 1992) extrusion and spheronisation (Raines et al., 1989; Newton et al., 1992) and tabletting (Landin et al., 1993c) has indicated that microcrystalline powder from various sources is not interchangeable even allowing for changes in processing variables such as binder/water concentration.

An important property of the material and one which has recently been correlated with its interaction with water as measured by immersion calorimetry (Landin et al., 1993a, b) is its crystallinity denoting the proportion of crystalline (as opposed to amorphous) cellulose in the sample. Data on the crystallinity of microcrystalline cellulose are extensive but often difficult to compare between workers because of the differences not only in the techniques used, e.g., X-ray powder diffraction (Schurz and Klapp, 1976; Nakai et al., 1977; Nurnberg and Gerst 1979; Nagavi et al., 1983; Soltys et al., 1984), infrared (Nakai, 1977; Nagavi et al., 1983), density measurements (Nurnberg and Gerst, 1979) and NMR (Sterk et al., 1987), but also in the method of data manipulation and analysis. Consequently, definitive trends in, for instance, the effect of particle size and source are inconsistent. Soltys et al. (1984) have, for example, claimed that the crystallinity of microcrystalline cellulose (Avicel) decreases with decreasing particle size and yet have shown a similar decrease between Avicel PH101 and PH103 of similar particle size. Doelker et al.

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(1987) using an X-ray diffraction method have claimed that Avicel PH101 has a significantly higher crystallinity than material obtained from other sources, a finding disputed by Sterk et al. (1987) using an NMR method.

A criticism of all the work undertaken is that no account has ever been taken on the possible contribution of interbatch variation. In this work this problem has been addressed by performing measurements on samples of microcrystalline cellulose of different particle sizes from different batches obtained from a variety of manufacturers using two well defined methods of assessing crystallinity.

Samples of microcrystalline cellulose were obtained from FMC International (Avicel PH101, 102, 103, 105, 200), Edward Mendell Co. Inc. (Emcocel and Emcocel 90M), Unitika Rayon Ltd (Unimac MG100, MG101) and Rettenmaier and Sohne GmbH (Vivacel 101, 102). All were used as received.

Crystallinity was assessed using both X-ray diffraction (XRD) and infrared (IR) methods. In the XRD method diffraction patterns of samples compressed into a holder were measured on a Siemens X-ray diffractomer Model D5000 using monochromatic CuKα radiation at a scanning rate of 0.25° 20 min^{-1} over the range 5–32°2θ. Samples were spun horizontally at 30 rpm in an attempt to minimise any possibility of preferential alignment of the material under test. The crystallinity index was calculated as suggested by Doelker et al. (1987) using a computer program to draw the demarcation line between the surface of the crystalline reflections and the diffuse halo of the amorphous regions (Fig. 1). Repeat experiments on 10 samples of one batch of material gave a coefficient of variation of 1.33% of the mean.

In the IR method samples (0.5% w/w in compressed discs of potassium bromide) were measured using a Nicolet FTIR Spectrometer Model 20SXC in transmission mode over the range 400–4000 cm^{-1}. The crystallinity index was calculated as suggested by Nelson and O'Connor (1964) using a computer program to determine the ratio of the transmission at wave numbers 1372 and 2900 cm^{-1} (Fig. 2). Repeat experiments on four samples of one batch of material gave a coefficient of variation of 1.82% of the mean.

Results on all the materials tested are shown in Table 1. Three features require emphasising.

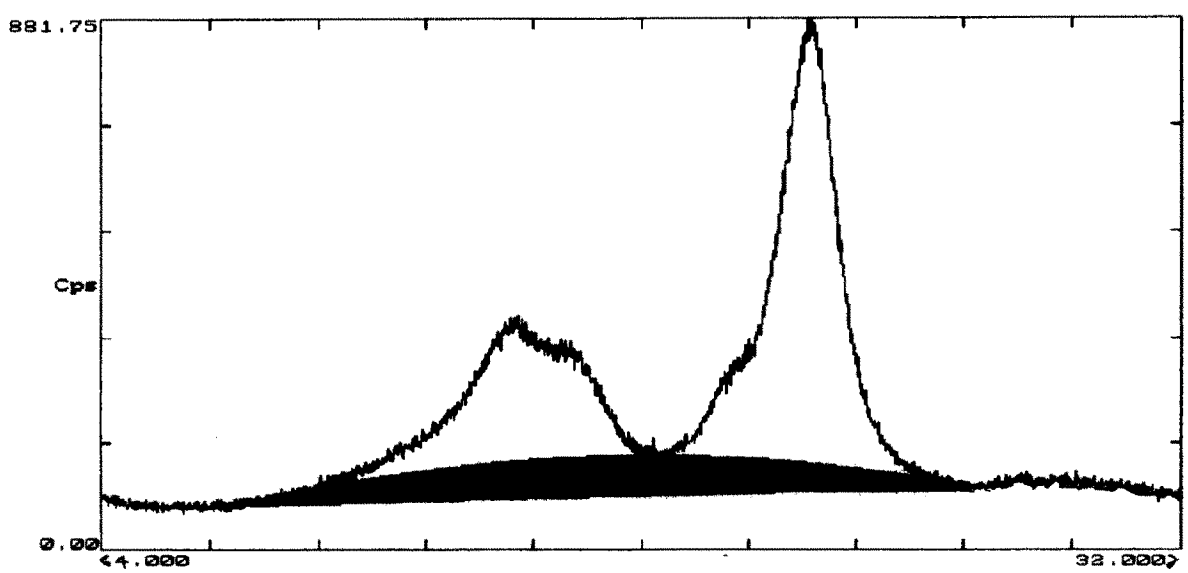

Fig. 1. X-ray diffraction pattern for microcrystalline cellulose showing computed halo of amorphous region. The crystallinity index was calculated by subtracting this area from the total area and expressing the difference as a percentage of the total area.

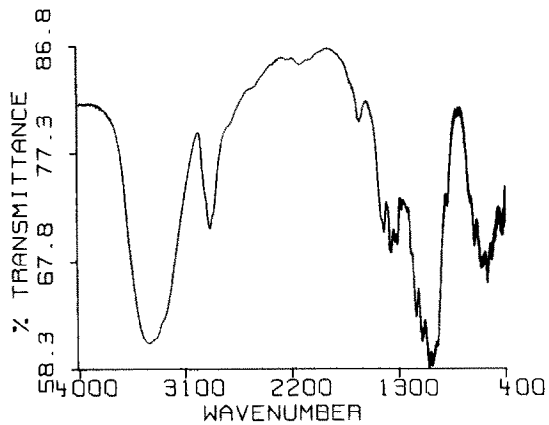

Fig. 2. Infrared spectra of micrystrystalline cellulose in the transmission mode.

(a) Values of the crystallinity from this work are similar to those reported using similar methodology.

(b) The IR method gives slightly different results from the XRD method with no apparent trend confirming data from other sources.

TABLE 1

Crystallinity data on microcrystalline cellulose

Grade	Batch	Particle size (μM)	Crystallinity index (%)	
			XRD	IR
Avicel PH101	1	50	61.8	62.3
	2	50	65.3	62.0
Avicel PH102	1	100	62.5	57.9
	2	100	62.4	58.0
	3	100	62.6	58.3
Avicel PH103	1	50	63.5	60.8
Avicel PH105	1	20	64.4	68.7
Avicel PH200	1	200	63.3	65.5
Emcocel	1	51	65.0	68.5
	2	51	65.0	69.4
Emcocel 90M	1	91	64.4	61.3
Unimac MG100	1	38	63.0	62.2
	2	38	61.8	68.3
Unimac MG200	1	105	60.6	61.6
	2	105	63.1	61.6
Vivacel PH101	1	-	62.7	57.7
Vivacel PH102	1	-	62.4	58.0

[a] Manufacturer's literature.

(c) Batchwise variation obviously exists and is of a magnitude that neutralises any effect of particle size and/or source of material.

It must be concluded, therefore, that differences seen in the processing (especially granulation and extrusion) of these materials must be due to some other property other than crystallinity. Whether the effect is due solely to the macroscopical features such as particle size and shape as suggested by Parker and Rowe (1991) or differences in the lignin and hemicellulose sugar content as suggested by Landin et al. (1993b) or even to a complex interaction of both remains to be confirmed. However, it is becoming increasingly apparent that much more work will need to be done to identify the reason and define analytical tests that will differentiate between materials.

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