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## *Small-angle X-ray scattering of polyethylenesulphide fibres*

In a recent paper, Hendra and Majid [1] have used the Raman method to show that polyethylene sulphide, being a highly crystalline material, forms lamellar crystallites and that these lamellae thicken on annealing as function of temperature. Apparently attempts to prove the lamellar structure by small-angle X-ray diffraction observations so far have been unsuccessful [1].

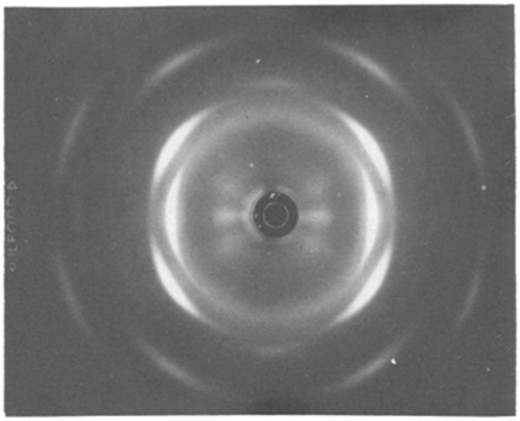

*Figure 1* Wide-angle diffraction pattern of spin-oriented polyethylenesulphide fibre. Ni-filtered Cu$K\alpha$ radiation. Sample to film spacing, 5 cm.

In the course of melt spinning studies leading to oriented crystalline fibres (spin-orientation) we have produced a polyethylene sulphide morphology completely analogous to that of a spin-oriented polypropylene [2], i.e. we observe a fan-like meridional SAXR diffraction pattern in the range from 60 to 140 Å, the centre being at about 75 Å (Fig. 2a). The orientation of the crystallites is quite good, as shown in the wide-angle X-ray pattern of Fig. 1. (Similar to the sample prepared by Y. Takahashi *et al.* [3].)

On annealing these samples between 140 and 180° C for 1 h, the small-angle pattern changed considerably, the long fan-like spacing breaking into two meridional spots, the outer one increasing from 82 (140° C) to about 100 Å (180° C), the inner one being much sharper and having small lateral dimensions (Fig. 2b and c, 160, 180° C).

From comparison with polypropylene [2], there is no doubt that the meridional scattering is based on a lamellar structure, the lamellae showing a range of thicknesses and being arranged normal to the extrusion direction. However, in polypropylene, the lamellar fan gradually sharpens on annealing as the lamellae become more uniform; at 135° C and higher a weak overtone becomes visible.

On annealing polyethylenesulphide, the second

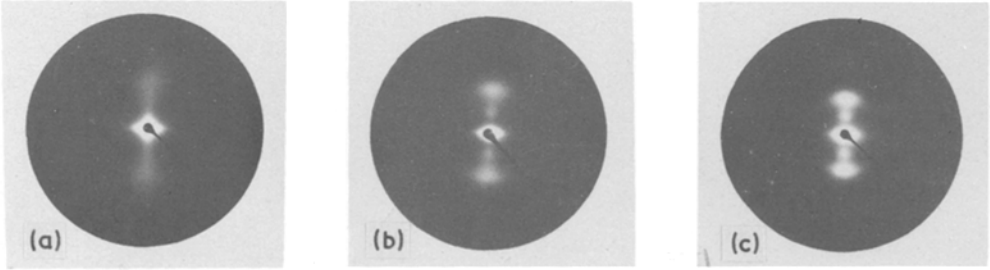

*Figure 2* SAXR diffraction pattern of polyethylenesulphide fibres: (a) as-spun, (b) annealed 160° C for 60 min (c) annealed 180° C for 60 min. Ni filtered Cu$K\alpha$ radiation, Rigaku Denki Rotating Anode Generator 100 mA, 50 kV, sample to film 300 mm, exposure 18 to 20 h.

strong diffraction spot appears with a spacing of 170 Å (150 to 160° C) to 200 Å (180° C). This spacing strongly suggests a doubling of the original long period (74 to 100 Å). A similar observation was reported by Keller *et al.* [4–6] for a series of nylons. There, an abrupt doubling of the thickness of the lamellae rather than a gradual increase as observed in polyethylene, polypropylene, etc. was noted. Independently, the same observation was made with spin-oriented nylon 6 and 6, 6 fibres on annealing at high temperatures and in liquid crystallizing systems [7]. The difference in the degree of orientation of the two SAXR diffraction spots in polyethylenesulphide allows the suggestion that the doubling of the lamellar size takes place preferentially in areas where the chain orientation is parallel to the extrusion direction.

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## *A novel route for the production of β′ sialon powders*

It has been demonstrated [1, 2] that certain ceramic nitride powders (e.g. $Si_3N_4$, AlN) may be produced by reaction of ammonia with the appropriate oxides. The inital oxide must, however, be in a finely divided, highly surface active state. There is at present a great deal of interest in ceramics based on silicon, aluminium, oxygen and nitrogen with a view to their potential application as components in gas turbines. A number of workers [3–7] have shown that β-$Si_3N_4$ can take into solid solution considerable amounts of aluminium and oxygen, the properties of such materials being equivalent if not superior to those of the pure nitride. Lumby [6] has given the composition of these "β′ sialons" as $Si_{(6-z)}Al_zN_{(8-z)}O_z$. β′-sialons are normally produced by hot-pressing appropriate mixtures of AlN, $Al_2O_3$, $SiO_2$ and $Si_3N_4$. This is an expensive process in which substantial difficulties are encountered in producing a reproducible, fully homogeneous product.

In view of these investigations, the present author considered that a possible and economical method of producing β′ material is by reaction of ammonia with kaolin. Kaolin $2(Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O)$ occurs naturally as a clay which, when dried, produces a very fine particle size powder. Heating at about 500° C removes the structural water to produce finely divided, highly surface active metakaolin.

$$\underset{\text{kaolin}}{2(Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O)} \xrightarrow{500^\circ\,C} \underset{\text{metakaolin}}{(2Al_2O_3 \cdot 4SiO_2)} + 4H_2O$$

The kaolin used in the present work was obtained from English China Clays Sales Co Ltd, St. Austell, Cornwall,. Chemical analysis and particle size distribution are given in Tables I and II.

Weighed amounts of kaolin powder contained in alumina boats were reacted for various times and at various temperatures in an atmosphere of pure dried ammonia at flow rates of about 200 $cm^3\,min^{-1}$. Reactions were also studied for a fixed time (22 h) and a fixed temperature (1400° C) but with varying ammonia–hydrogen ratios. A platinum wound, alumina tube furnace was employed for these experiments.

From infra-red analysis and observed weight changes it was concluded that reaction occurs at temperatures as low as 900° C although the product is mainly amorphous and contains a substantial amount of mullite ($3Al_2O_3 \cdot 2SiO_2$). At higher temperatures (~ 1400° C), mullite is