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### Temperature Variation of Electron Diffraction Patterns of Some Organic Films

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#### Abstract

Temperature variation of electron diffraction patterns reflected from films of grease, vaseline and paraffins was examined. It was observed that at some elevated temperatures the parallel arrangement of the molecules of the substances is gradually destroyed and with further rise of temperature it becomes haphazard. This transition temperature of the arrangement of the molecules differs with different substances and with different thickness of the films.

It is known<sup>1</sup> that when certain organic substances of long-chain molecules are smeared on a polished metal surface in a thin film, the molecules arrange themselves parallel to each other with their long axes perpendicular to the metal surface. Electron diffraction patterns reflected from such films generally consist of layer lines with some spots. Such pattern is usually called "grease-pattern." This pattern is explained as follows<sup>2</sup>: The layer lines are the directions of maximum intensity of the radiation scattered by the carbon atoms in a chain molecule when they scatter the radiation independently of each other. The spots on these lines correspond to the side-by-side spacings of the chains when the molecules are packed closely together in some regular manner. When the molecules are arranged so that the carbon atoms of different chains are in a set of planes parallel to the metal surface, the central spots are obtained on each even order layer line.

It is expected that when such films are heated the parallel arrangement of the long-chain molecules may be destroyed owing to the heat motion of the molecules, and the diffraction pattern may vary with the rise of the temperature of films. To see this effect four substances—grease, vaseline and two kinds of paraffin—were selected. Each of these substances gives rise to the grease-pattern at room temperature.

Two kinds of films, thick and thin, were examined for each substance. Thick film was prepared by putting a drop of the substance

<sup>1.</sup> C. A. Murison: Phil. Mag., 17, 201 (1934)

L. T. Andrew: Trans. Faraday Soc., 32, 607 (1936)

<sup>2.</sup> C. A. Murison: Ibid.

upon a small piece of polished copper, which was then heated to melt the drop and allow it to drain off. Thin film was prepared by melting the thick one and then wiping it with clean cloth so as to get a uniform coating of the substance so thin that in most cases it showed interference colours. The specimen thus prepared was set in a small electric-furnace, and these together were mounted in the diffraction apparatus. The varying diffraction patterns were continuously observed on a fluorescent screen and their photographs were taken at suitable intervals.

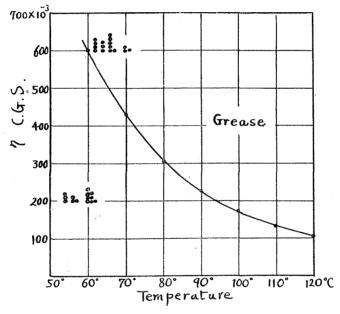


Fig. 1

One set of such photographs taken with a thick film of Cenco stop-cock grease (15520 B) by using the cathode rays of about 70 KV. ( $\lambda$  = 0.045 A. U.) is shown in Plate I. In this case the regular arrangement of the molecules is destroyed gradually at the temperatures above 65°C and their arrangement becomes almost haphazard at about 71°C.

Such temperature as 71°C, mentioned above, at which the arrangement of the molecules become haphazard will be called the "transition-temperature" of the film. The transition-temperatures observed with different specimens of the same grease as above are plotted as dots in Fig. 1. The dots above the curve refer to thick films and those below the curve to thin films. The transition-temperatures of vaseline

and two kinds of paraffins (M. P. 40°-42° C and M. P. 68°-72° C) are also shown in Figs. 2, 3 and 4 in the same manner as in Fig. 1. The tran-

sition-temperature is not definite for a substance. In general, thick films have higher transition-temperatures than thin films.

It is noteworthy that the transitiontemperature is different from the melting point of the substance; and no appreciable change in diffraction pattern was observed at the temperatures above and below the melting point of any substance. When the

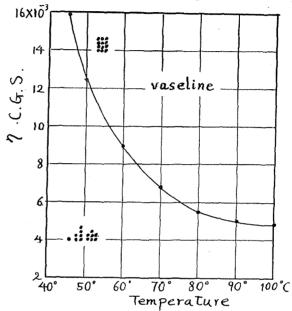


Fig. 2

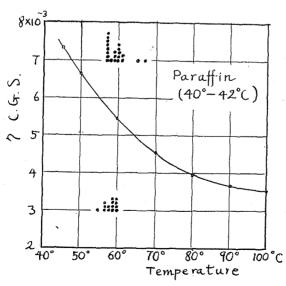


Fig. 3

is repeatedly heated and cooled, the transition-temperature was almost, but not exactly, the same for the same part of the same specimen. The transition from parallel to random arrangement heating takes place at a temperature higher by a few degrees than transition from random to parallel arrangement by cooling.

It is conceivable that the viscosity of the substance changes abruptly by passing through the transition-temperature. To study this possibility the coefficient of viscosity  $\eta$  of each substance at different temperatures was measured by Ostwald's viscosimeter. The results are shown by the curves in Figs. 1, 2, 3 and 4. Contrary to our expectation, the coefficient of each substance decreases regularly with the rise

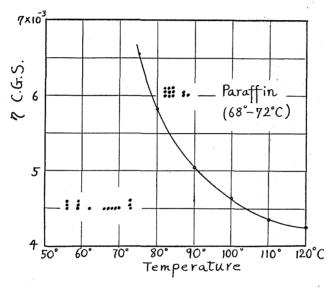


Fig. 4

of temperature, and the anticipated discontinuity at the neighbourhood of the transition-temperature was not observed. This fact, considered in connection with the discovery that the transition is unrelated to the melting temperature of the substance, seems to indicate that the regular arrangement of the chain molecules as described before, is confined to the outer surface of the substance or to its very thin film coating the flat surface of the solid substance.

The writer wishes to express his sincere thanks to Prof. U. Yoshida for his invaluable suggestions.

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## Plate I

