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**CARBON FIBRE
REINFORCED POLYMERS
AS SELF-LUBRICATING
MATERIALS**

by

J. P. Giltrow

J. K. Lancaster

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SUMMARY

This Report describes an investigation of the friction, wear and mechanical properties of polymers reinforced with different proportions of two types of carbon fibres. During dry sliding against steel, coefficients of friction of the composites lie within a common range of 0.25-0.35 at high fibre concentrations, and rates of wear can be reduced by factors of more than 10^3 . Fibre reinforcement can also greatly improve the strength, stiffness and resistance to deformation under load. The most important factor influencing the wear rate is the amount of fibre present: the strength is more dependent on adhesion at the fibre-polymer interface.

The practical applications of carbon-fibre reinforced polymers are discussed.

Departmental Reference MAT 15

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1 INTRODUCTION

Thermosetting and thermoplastic polymers are frequently reinforced with fibres, such as glass and asbestos, or lamellar solids, such as graphite and MoS_2 , to improve their performance in applications as dry bearings, dynamic seals and gears. The recent development of high strength, high modulus carbon fibres at R.A.E.^{1,2} is of considerable potential interest for the reinforcement of polymeric bearing materials, because carbons themselves generally exhibit good friction and wear properties. Preliminary experiments, reported elsewhere^{3,4}, have shown that, during dry sliding against steel, carbon fibre reinforced polymers in general yield lower coefficients of friction and wear rates than the parent matrices. The fibre orientation can play a part in the wear process, and minimum friction and wear were obtained with the fibres oriented normally to the sliding surface. In a limited number of materials incorporating a constant amount of randomly oriented carbon fibres, the fibres reduced the coefficients of friction to a level which was almost independent of the matrix material. The range of wear rates with reinforced composites was also appreciably narrower than for the original polymers.

The present work extends the earlier investigations to include a wider variety of polymer matrices, and examines in detail the effect of fibre concentration upon coefficient of friction, rate of wear, flexural strength, elastic modulus in flexure and deformation of composites under static compressive load. The practical implications of the results are discussed

2 MATERIALS AND APPARATUS

2.1 The carbon fibres

High temperature treatment of synthetic organic fibres has produced carbon fibres of two general types². Heat-treatment at 2500°C yields the type I fibre, a partly graphitic material with an ultimate tensile strength and elastic modulus of $300 \times 10^3 \text{ lb/in}^2$ and $60 \times 10^6 \text{ lb/in}^2$ respectively. Treatment at 1500°C produces essentially non-graphitic fibres - type II - with a higher tensile strength, $400 \times 10^3 \text{ lb/in}^2$, but lower elastic modulus of $30 \times 10^6 \text{ lb/in}^2$. Both types of fibre are of the order of $3 \times 10^{-4} \text{ in}$ (7 microns) in diameter.

Carbon fibres have a fluted external structure⁵ and an internal structure of fibrils lying parallel to the fibre axis⁶. The fibrils are

formed by chains of turbostratic graphite crystallites with their c-axes normal to the axis of the fibre. It is thought that the fibre modulus is a function of the orientation of the graphite crystallites and that the fibre strength is dependent on the interfibrillar bonding.

2.2 Polymeric materials

The polymers used in this work are listed in the legend to Fig.1. Those materials which were unobtainable in the required powder form were ground at a low temperature (77°K) to pass 60 mesh (250 microns).

2.3 Preparation of composites

All the materials examined contained randomly oriented, short fibres. Composites with thermoplastic matrices were prepared by blending chopped carbon fibres (about $\frac{1}{8}$ in long) with a water dispersion of the powdered polymer in a liquidiser. The mixture was vacuum dried at 80°C and compression moulded into a bar at a temperature appropriate to the particular polymer matrix. Some composites were also made in a similar manner using carbon fibres which had been precoated with nickel or silver by an electroless deposition technique.

Composites of carbon fibres with thermosetting resins were prepared by pouring liquid resin over a random mat of fibres in a mould and applying light pressure during the cure period.

2.4 Measurement of friction, wear and mechanical properties

Rods of diameter $\frac{1}{4}$ in and length about 2 in were machined from the composite bars prepared as above. After finishing by abrasion on 600 grade silicon carbide paper flooded with water, the rods were loaded against the cylindrical surface of a 4 in diameter rotating cylinder of mild steel (B.S.15), with the two axes at right angles. The surface of the steel cylinder was prepared by random abrasion on 600 grade silicon carbide paper to a roughness of about $6\text{ }\mu$ in c.l.a., and cleaned by scrubbing with tissue soaked in petroleum ether. The speed of rotation of the steel cylinder was 100 rpm (54 cm/sec) and the applied load was 1.2 kg. The rate of wear of the specimens was calculated from periodic measurements of the major axis of the elliptical wear scar, up to a total time of sliding equivalent to approximately 20000 revolutions. Coefficients of friction were measured by a torque transducer interposed between the steel cylinder and the driving motor.

The ultimate flexural strength and elastic modulus in flexure of the composites were measured using a calibrated Hounsfield 'Tensometer'.

Determinations of the deformation under load were made by a modification of an ASTM test⁷. Composite cylinders, $\frac{1}{4}$ in diameter and $\frac{1}{4}$ in long were subjected to a compressive stress of 1500 lb/in² across their plane faces and the decrease in length of the cylinders measured as a function of time.

3 RESULTS

Fig.1 illustrates the reduction in coefficient of friction and rate of wear which occurs when 25%-30% by weight of carbon fibres are incorporated into a wide range of polymers. The coefficients of friction of the polymers are reduced from values ranging from 0.25-0.75 to an approximately common level of 0.25-0.35 for all the composites. Wear rates of the polymers are reduced by factors of up to 7000 and, apart from three examples, lie within a single decade; in contrast, the wear rates of the unreinforced materials range over more than three decades. The results indicate that the friction and wear properties of the composites are predominantly controlled by the carbon fibres rather than by the matrix, and this suggests that the carbon fibres support the major part of the applied load. Direct evidence in support of this suggestion is given by the electron micrographs, Figs.2a-e. The photographs show parts of the wear scars on four materials which have been shadowed with gold at a glancing angle to provide relief. Some carbon fibres clearly project above the mean surface level of the matrix, this being particularly marked for the relatively soft polypropylene and PTFE. With polypropylene, Fig.2a, the discontinuity visible at the fibre-polymer interface in the top right-hand corner suggests that there has been appreciable visco-elastic recovery of the surrounding polymer after removal of the stress, tending to raise the fibre above the surface of the matrix. Such an effect will be most pronounced for fibres oriented in the plane of the surface. For short fibres oriented normally to the worn surface, visco-elastic recovery may raise both the fibre and the surrounding polymer, as shown in the top left-hand corner of Fig.2b for PTFE. An extreme situation arises with relatively long fibres oriented normally to the sliding surface. Because such fibres are attached to the matrix in regions remote from those affected by the stress, fibre movement is no longer possible and only the

surface layers of the matrix can recover visco-elastically. Fig.2c shows an example for polyimide containing unidirectionally oriented long fibres, where it can be seen that the fibre ends are well beneath the matrix surface. The pattern of ridges on the sides of the depressions in the polyimide is characteristic of the surface structure of the carbon fibres. The particular photograph in Fig.2c was taken some weeks after the production of the wear scar in order to emphasize the effects of visco-elastic recovery; similar photographs of freshly prepared wear scars showed much less recovery of the matrix.

Although considerations of visco-elastic recovery complicate the detailed interpretation of the photographs in Fig.2, some carbon fibres must nevertheless support the load preferentially by projecting above the matrix surface. Fig.2b for PTFE shows that appreciable amounts of wear debris have collected along one side of a number of fibres and there is a groove in the centre of the photograph which terminates at a fibre. In extreme cases, the collection and aggregation of wear debris may partially mask the carbon fibres, as shown in Fig.2d.

In relatively hard matrices, a commonly observed feature is the presence of fibres which must have been fractured during processing of the composite. Fig.2e shows an example for a Friedel-Crafts resin⁸. Fractured fibres are rarely, if ever, seen with the softer, more ductile matrices.

3.1 Fibre concentration

The influence of carbon fibre concentration on the wear rate, coefficient of friction, flexural strength and flexural modulus for six polymer matrices is shown in Fig.3. Coefficients of friction are continuously reduced by increasing amounts of carbon fibre. Flexural strengths often reach a maximum value at 15%-25% reinforcement, although for some polymers, notably ABS and polyphenylene oxide (not shown), no increase in strength was observed. Flexural moduli, in general, increase with increasing reinforcement, although 'Delrin' exhibits a maximum.

The effect of fibre concentration on the rate of wear appears to be specific to each polymer. Thus, whilst increasing amounts of fibre continuously reduce the wear rates of ABS, polysulphone and PTFE, 'Delrin' and nylon exhibit a minimum wear rate at 15%-20% reinforcement, and high

density polyethylene wears more rapidly when reinforced. There appears to be no general correlation between the mechanical properties of the composites and their wear resistance.

Similar relationships are shown in Fig.4 for acetal copolymer reinforced with the two different kinds of carbon fibre. The type II fibres (non-graphitic) are more effective than type I (graphitic) in improving the wear resistance, strength and stiffness of the composites, but at the expense of higher coefficients of friction. Very similar results were obtained for 'Delrin'.

Both types of fibre were also incorporated into PTFE and Fig.5 shows that composites with type II fibres are again superior to those containing type I in all respects except friction. The coefficient of friction of about 0.6 obtained at high fibre concentration is extraordinarily high for a material based on PTFE.

Included in Fig.5 are the effects of incorporating third components, such as bronze flake, tungsten carbide/cobalt and MoS_2 , into composites containing both type I and type II fibres, and also of coating type I fibres with nickel or silver before incorporation into the composite. Nickel and bronze additions (Nos. 3, 4 and 8) yield wear rates lower than any obtainable with binary type I fibre - PTFE mixtures, although at some sacrifice to the strength and stiffness of the composite. Bronze (No. 12) is also an effective addition with type II fibres. MoS_2 (Nos. 9, 10 and 11) has no significant influence on the wear rate of PTFE composites containing either type of fibre.

3.2 Deformation under load

Fig.6a shows the deformation under compressive load (1500 lb/in^2) as a function of time for a variety of PTFE-based composites, some of which are available commercially. It is evident that fibrous materials, such as glass, asbestos and type I carbon fibres are not as effective in reducing deformation as graphite, coke, bronze and mica. Type II carbon fibres, however, are extremely effective, the composite showing a total deformation of only about 1% after 24 hours under stress. The permanent deformation, that remaining after a further 24 hours without load, was only 0.2%. It may be noted that the wear rate of this particular composite is about 14 times lower than that of the PTFE-mica material, the only other binary composite to show comparable resistance to deformation.

The marked difference between the two types of carbon fibre in reducing deformation under load is particularly evident in Fig.6b, where it may be seen that the addition of 45% of type I fibre to PTFE is no more effective than only 5% of the type II fibre. Third components in PTFE composites containing the type I fibres, however, can have a pronounced effect, and composites containing nickel (Nos. 1 and 3), tungsten carbide/cobalt (No. 7) and bronze (No. 8) are comparable in performance to the binary mixtures of PTFE and type II fibres.

3.3 Effect of fluid lubricants

A limited number of experiments has been carried out to examine the wear of carbon fibre reinforced polymers sliding against mild steel lubricated by n-hexadecane. Results for two materials, typical of the general behaviour of most of the other polymers, are shown in Fig.7. The lubricant is most effective in reducing wear at low concentrations of fibre, and becomes relatively ineffective at high fibre concentrations. It is postulated that hexadecane lubricates contacts between the polymer matrix and the steel only: thus, the effect of the lubricant at various fibre concentrations depends on the extent to which the fibres are able to support the load, and hence on the nature of the matrix material as well as on the fibre concentration.

3.4 Reproducibility

Some of the results given in this work are derived from single experiments, but where previously established trends appeared to alter in any way, multiple experiments were made to check their validity. In general, the reproducibility of wear rates, flexural strengths and elastic moduli in flexure were of the order of $\pm 25\%$, $\pm 10\%$ and $\pm 20\%$ respectively. One particular series of tests on nineteen separate PTFE structures of the same composition showed a standard deviation of 6% on the ultimate flexural strength.

4 DISCUSSION

It has been demonstrated, both here and in the earlier work^{3,4}, that carbon fibre reinforcement is very effective in reducing both the coefficient of friction and the rate of wear of polymers during continuous sliding against steel. The carbon fibres are exposed at the surface of the composite and support a significant part of the applied load. Since the fibres are somewhat abrasive⁴, they smooth the counterface, reducing localised stresses to

within the limits of elastic deformation⁹. Evidence in support of the conclusion that carbon fibres support the major part of the applied load may be summarised as follows:-

- (a) Direct observation from electron micrographs, e.g. in Fig.2.
- (b) The coefficients of friction of all polymer matrices with relatively high fibre contents are approximately of the same value ($\mu \sim 0.25-0.35$, Fig.3).
- (c) The high coefficient of friction of PTFE containing type II fibre ($\mu \sim 0.6$) can only be explained by postulating that the PTFE matrix carries little or no part of the applied load.
- (d) Lubrication of carbon fibre reinforced composites is relatively ineffective when the fibre content is high.
- (e) The wear rates and coefficients of friction of carbon fibre composites are almost independent of the speed of sliding and of temperature, in contrast to those of the unreinforced polymers⁴.
- (f) Electrical contact resistance measurements with unidirectionally oriented composites are of the order of magnitude to be expected, if the fibres are supporting the greater part of the applied load⁴.
- (g) The range of wear rates obtained for carbon fibre reinforced composites with different matrices is much narrower than that for the unreinforced polymers - Fig.1.

If the carbon fibres preferentially support the load, rates of wear should, ideally, be independent of the matrix polymer and characteristic of those for bulk carbon sliding on steel. Comparison shows that the values obtained are, in fact, within the range commonly observed for many carbons¹⁰. However, Figs.1 and 2 suggest that the matrix polymer is not wholly irrelevant to the wear process for the following reasons:-

- (a) Polymer debris may obscure the carbon fibres.
- (b) For matrices which are relatively soft and of high ductility, the incorporation of carbon fibres greatly reduces the elongation to break. The wear rate of the composite may then become greater than that of the matrix alone, as observed for high density polyethylene. This aspect is particularly important in conditions of abrasive wear⁴.

(c) Fibres may become detached from the surface layer if the matrix has a high intrinsic wear rate, or if the interfacial bonding between fibre and matrix is relatively weak.

The bonding between carbon fibres and the polymers is one of the most uncertain aspects impeding the detailed interpretation of the results in this Report. The simplest theory tenable is that bonding is primarily mechanical, resulting from interlocking of the polymer and the surface irregularities of the fibres. In support of this is the fact that treatments known to increase the surface roughness of the fibres increase the strengths of composites with thermosetting resin matrices¹¹. Such treatments, however, appear to be less effective with thermoplastics. It must be admitted that at the present stage of the technology of carbon fibre reinforced thermoplastics, interfacial bond strengths appear to be very much lower than their potential maxima. The increases in strength and modulus of thermoplastic composites, e.g. in Fig.3, are much less than simple theory predicts¹² and appreciably lower than those attainable with thermosetting resin matrices¹³. One reason could be the high melt viscosity of many thermoplastics, which will affect not only the degree of mechanical interlocking at the interface, but may also prevent complete encapsulation of each fibre, leading to voids or fibre-fibre contacts. The latter may explain why the flexural strengths (and sometimes the elastic moduli) tend to decrease at high fibre concentrations in many of the composites.

A further factor which may affect the fibre-polymer bonding is the physical structure of the fibres. There is a marked difference in the strength and stiffness of acetal copolymer and PTFE composites containing the two types of carbon fibre. This cannot be attributed to differences in topography of the fibre surfaces, since they are similar¹⁴. The graphite crystallites in type I fibres are oriented with their c-axes normal to the fibre axis. It is tempting to suppose that the weaker composites produced with type I fibres are a consequence of a relatively low shear strength in the surface layers of the fibre. Type II fibres, which are essentially non-graphitic, will be superior in this respect.

The higher coefficients of friction obtained with type II fibres in PTFE and acetal copolymer strongly suggest that the friction of carbon fibres is

directly related to their degree of graphitisation. This conclusion is in contrast to the observations of Midgley and his co-workers¹⁵, who have shown that the friction of carbons (in bulk) is largely independent of their graphite content. The reason for the discrepancy is not yet known but may well be connected with the fibrillar internal structure of the carbon fibres; this structure is currently being studied in detail. The high coefficients of friction obtained with PTFE containing type II fibres also suggests that additions of lamellar solid lubricants to the composites as third components are unlikely to cause further appreciable reductions in friction and wear. Additions of MoS_2 (up to 10% by weight) have, in fact, been shown to be relatively ineffective. There are two possible reasons. Firstly, if the carbon fibres support the load preferentially, it will be difficult for any lamellar solid lubricant incorporated in the matrix to reach the regions of true contact. Secondly, because carbon fibres are mildly abrasive, any lamellar solid transferred to the counterface will persist there for a short time only.

Additions of metals to the composites as third components have been shown to be more effective in reducing wear than MoS_2 . Metal particles may act as additional load bearing areas and also by facilitating removal of frictional heat from the composite. In addition, bronze, which appears to be particularly good, could function by transferring to the steel counterface, thus providing a surface which is more readily smoothed, by the abrasive action of the carbon fibres, than the original steel. It has also been suggested that the presence of bronze or other copper alloys in PTFE facilitates chemical reaction and bonding of transferred films of PTFE on steel¹⁶.

The improvements in wear resistance obtained by additions of metals to the fibre-reinforced composites are coupled with some loss in strength and stiffness. This result is merely one facet of the general conclusion that there is no significant correlation between the bulk strength of carbon fibre reinforced polymers and their wear rates. The absence of such a correlation emphasizes the paucity of our understanding of some of the factors affecting the friction, wear and deformation of composites, and points to the considerable amount of work which remains to be done.

5 PRACTICAL APPLICATIONS

The results described in this paper were obtained from experiments with a pin and ring apparatus under constant, arbitrary conditions of sliding. Whilst this type of test (non-conforming geometry) is unrivalled for the rapid assessment of a large number of different materials, it is now widely appreciated that considerable uncertainties arise when attempting to extrapolate results to other, more practical, conditions of sliding. It is therefore essential to supplement pin and ring tests by trials of a limited number of materials in more realistic conditions. Such practical trials are inevitably very time-consuming, and at present only a start has been made. Thrust-bearing tests at a PV of 12500 (131 ft/min and 93 lb/in²) have confirmed the potential value of PTFE reinforced with carbon fibre as an alternative to glass¹⁷. When sliding against steel, wear rates were - PTFE, 3×10^{-8} cm³/cm kg; PTFE + 25% glass, 3×10^{-9} cm³/cm kg; PTFE + 25% carbon fibre (type I), 1.5×10^{-10} cm³/cm kg. Other applications in which carbon fibre reinforced polymers are being tried include self-aligning spherical bearings, cages for rolling element bearings, gears for use in ultra-high vacuum, gas bearing components (where high stiffness, dimensional stability and good start-stop performance are essential) and dynamic seals in hydraulic systems. It is hoped to report on the results of these tests at some future date.

A wide range of polymers, reinforced with carbon fibres, shows essentially the same wear rate, irrespective of the inherent wear rate of the parent polymer. Thus, materials which have, as yet, found no application because of their high rates of wear, may now be considered potentially useful in the reinforced state.

One of the most important advantages of type I carbon fibres as an alternative to glass is their considerably lower abrasiveness. This aspect has been discussed in detail elsewhere⁴. Although carbon fibres are intrinsically more expensive than glass, this need not necessarily limit their use in dry bearing composites. The amount of fibre used in a bearing will be relatively small, but, perhaps of greater importance, the lower abrasiveness of the composite obviates the use of a hardened counterface, thus reducing the overall cost of the component. A further advantage of carbon fibres is their good

electrical conductivity¹³. This feature is relevant to bearing applications where charge accumulation may be encountered, e.g. in airframe components, and to electrical contact applications generally.

Acknowledgements

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A

1. Acrylonitrile - butadiene - styrene copolymer (ABS, 'Lustran')
2. Friedel - crafts resin (mk II)
3. Polyvinylchloride
4. Polytetrafluoroethylene (PTFE, 'Fluon G4')
5. Polytetrafluoroethylene (PTFE, 'Kel-F81')
6. Polyvinylidene chloride ('Saran')
7. Polyvinylchloride - polyvinylacetate copolymer ('Vinylite')
8. Polypropylene (HM 20/70)
9. Epoxy resin ('815')
10. Polymethylmethacrylate ('Maklon')
11. Polyester ('SR 17449')
12. Polysulphone
13. Polyphenylene oxide
14. Polycarbonate ('Makrolon')
15. Phenolic resin
16. Nylon 6.6 ('A100')
17. Polyethylene (low density)
18. Acetal copolymer ('Kematal')
19. Polyimide
20. Polyformaldehyde ('Delrin')
21. Polyethylene (high density, 'Rigidex')

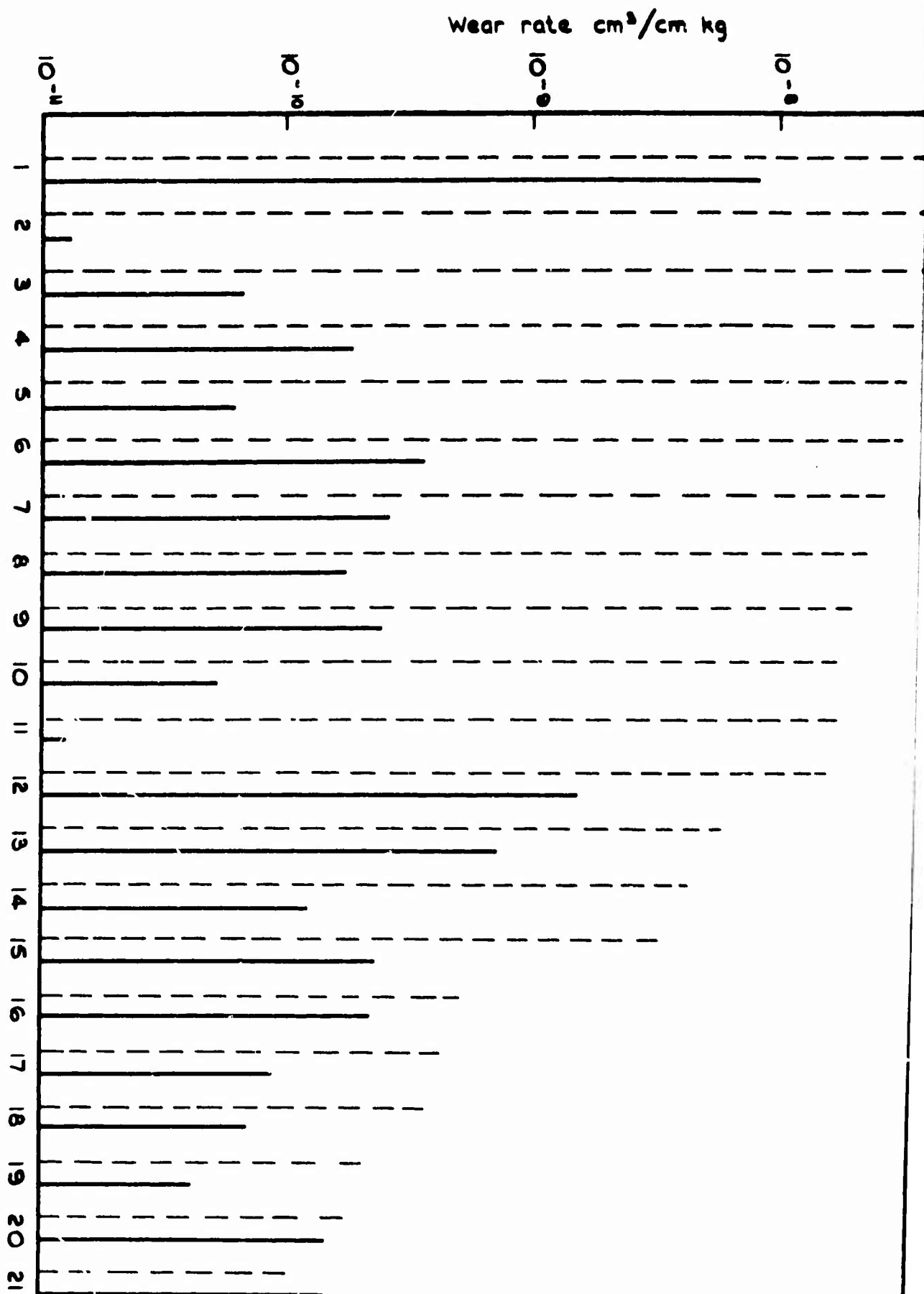
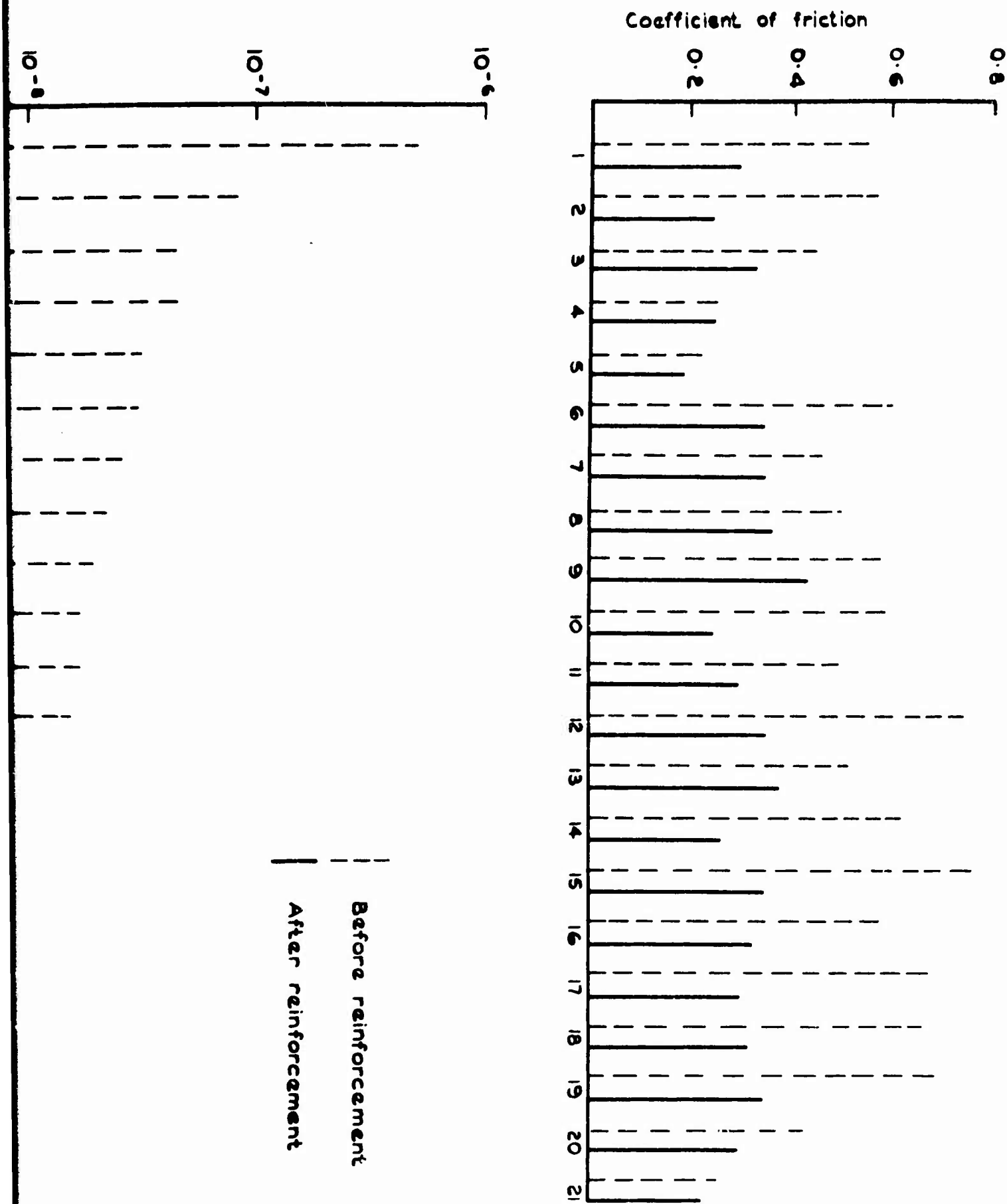


Fig.1 Friction and wear rates of polymers before and

B

Fig.1



Before and after carbon fibre reinforcement (25 - 30% wt)

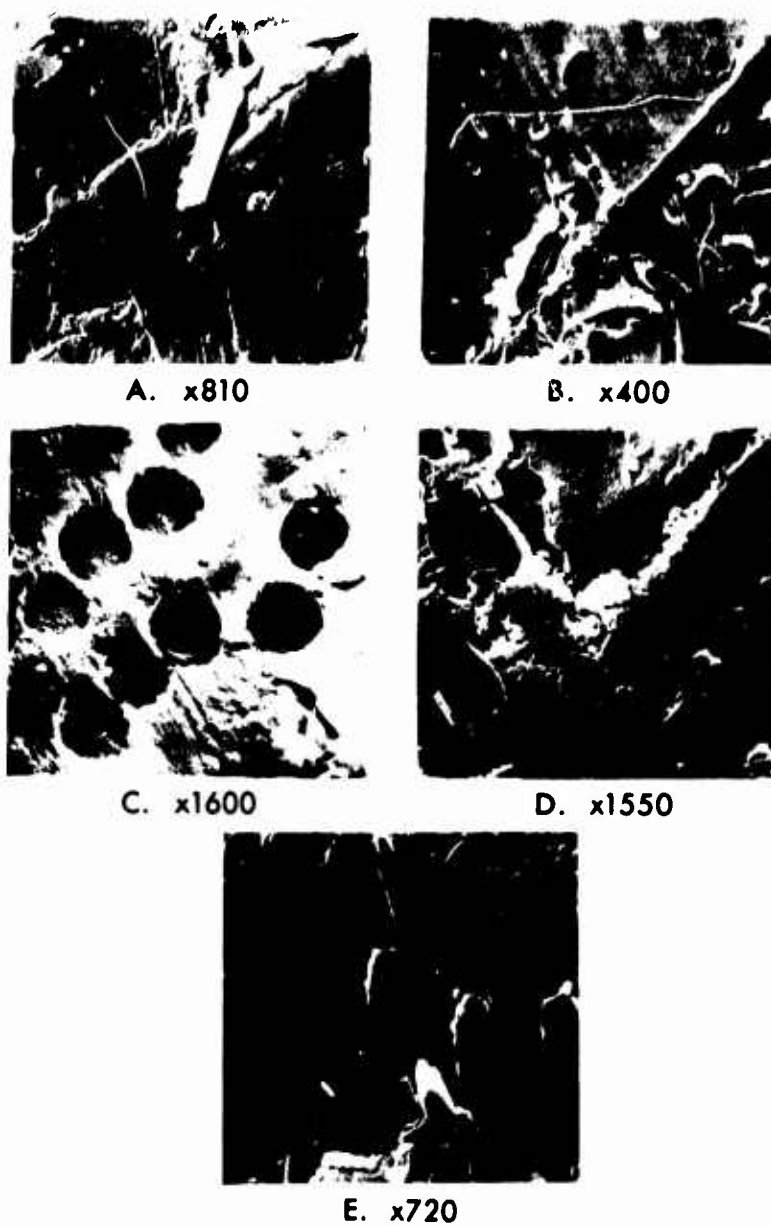
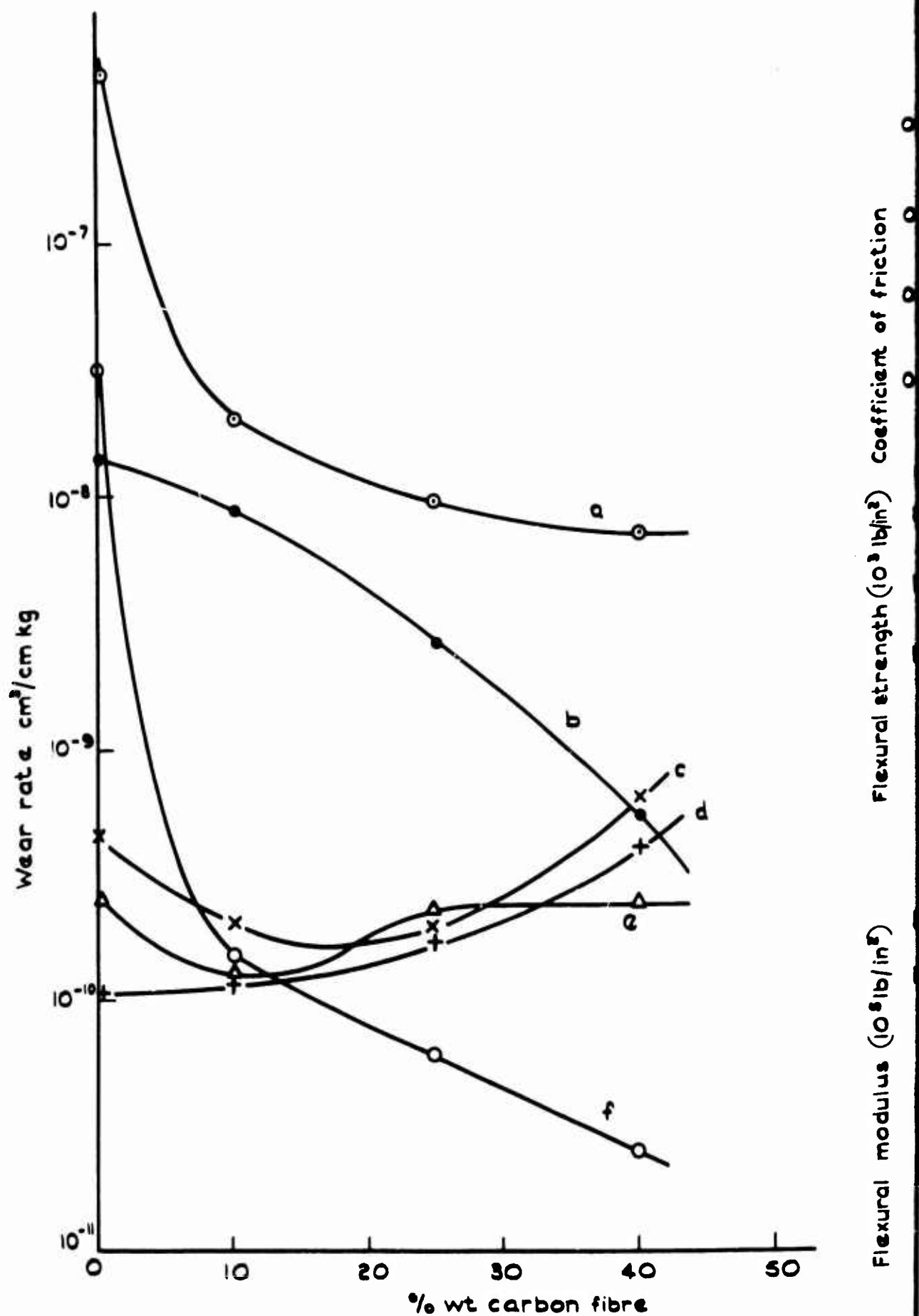


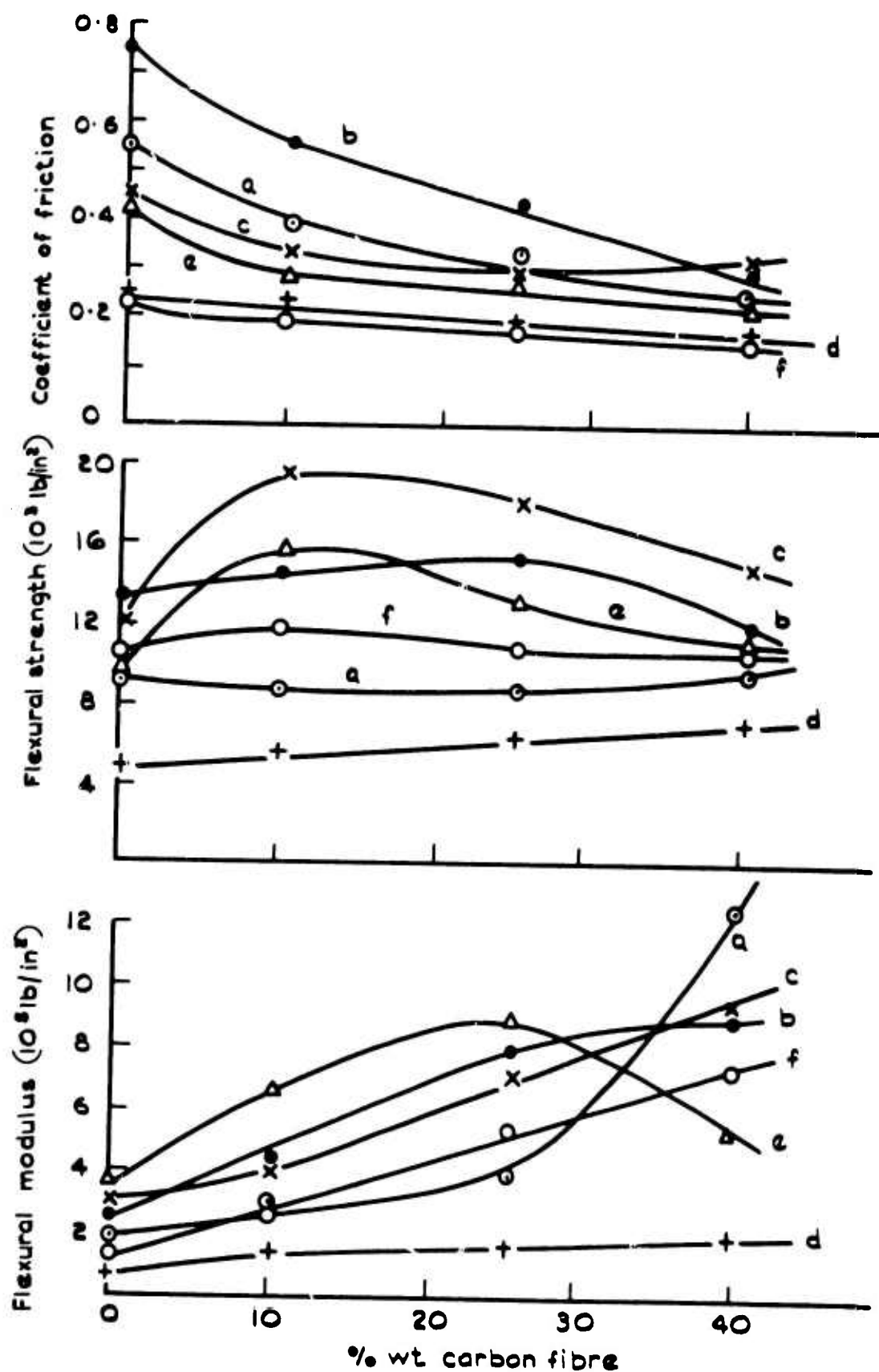
Fig.2. Scanning electron micrographs of worn composites with 25% WT. carbon fibre. A-Polypropylene, B-P.T.F.E., C-Polyimide, D-P.T.F.E., E-Friedel - Crafts Mk.2



a ABS, b Polysulphone, c Nylon 6.6, d Polyethylene (high density), e

Fig.3 Effect of carbon fibre concentration on and flexural modulus for

Fig. 3



density), a 'Delrin' f PTFCE

ation on wear rate, friction, flexural strength
modulus for 6 polymers

Fig. 4

014 900490

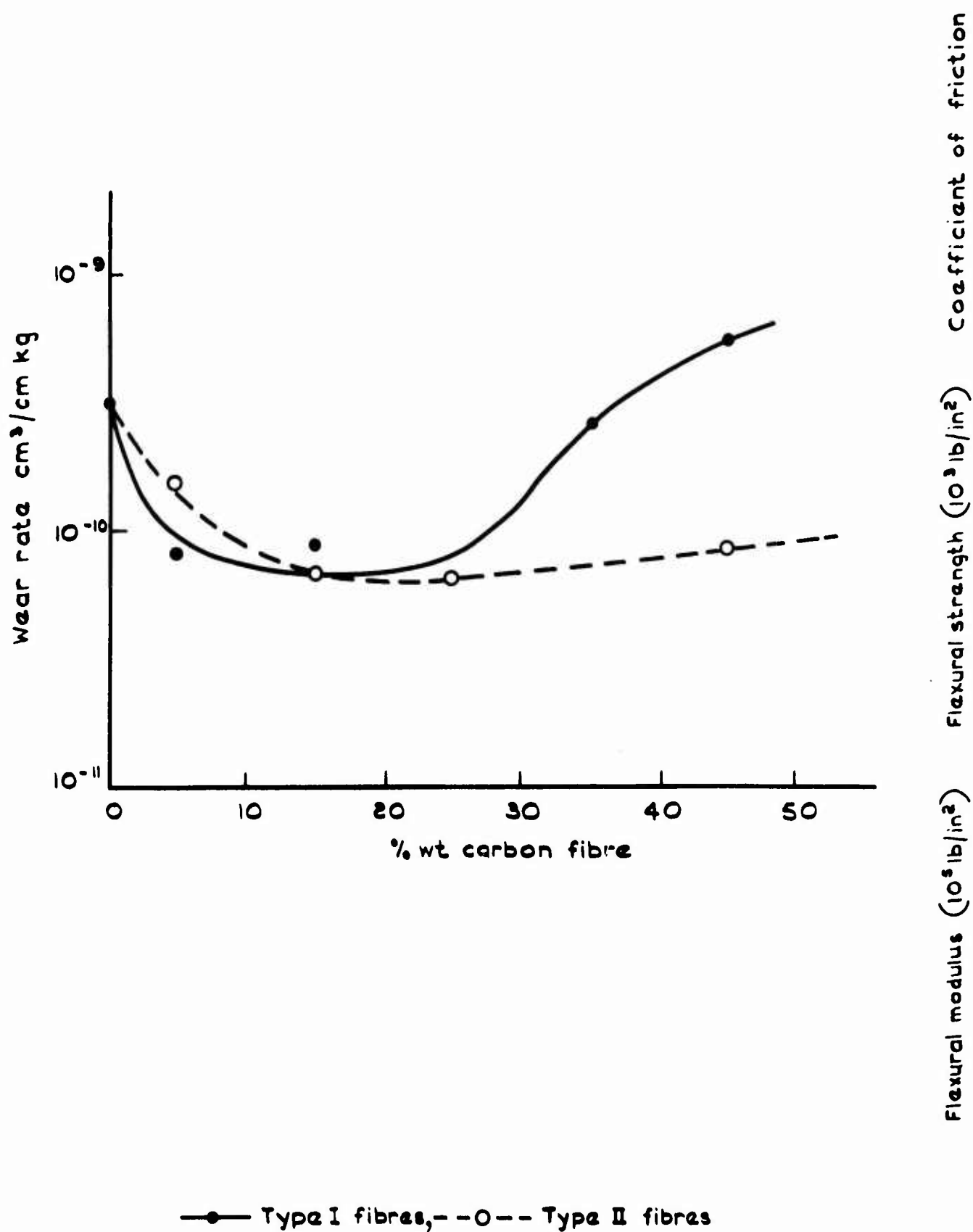
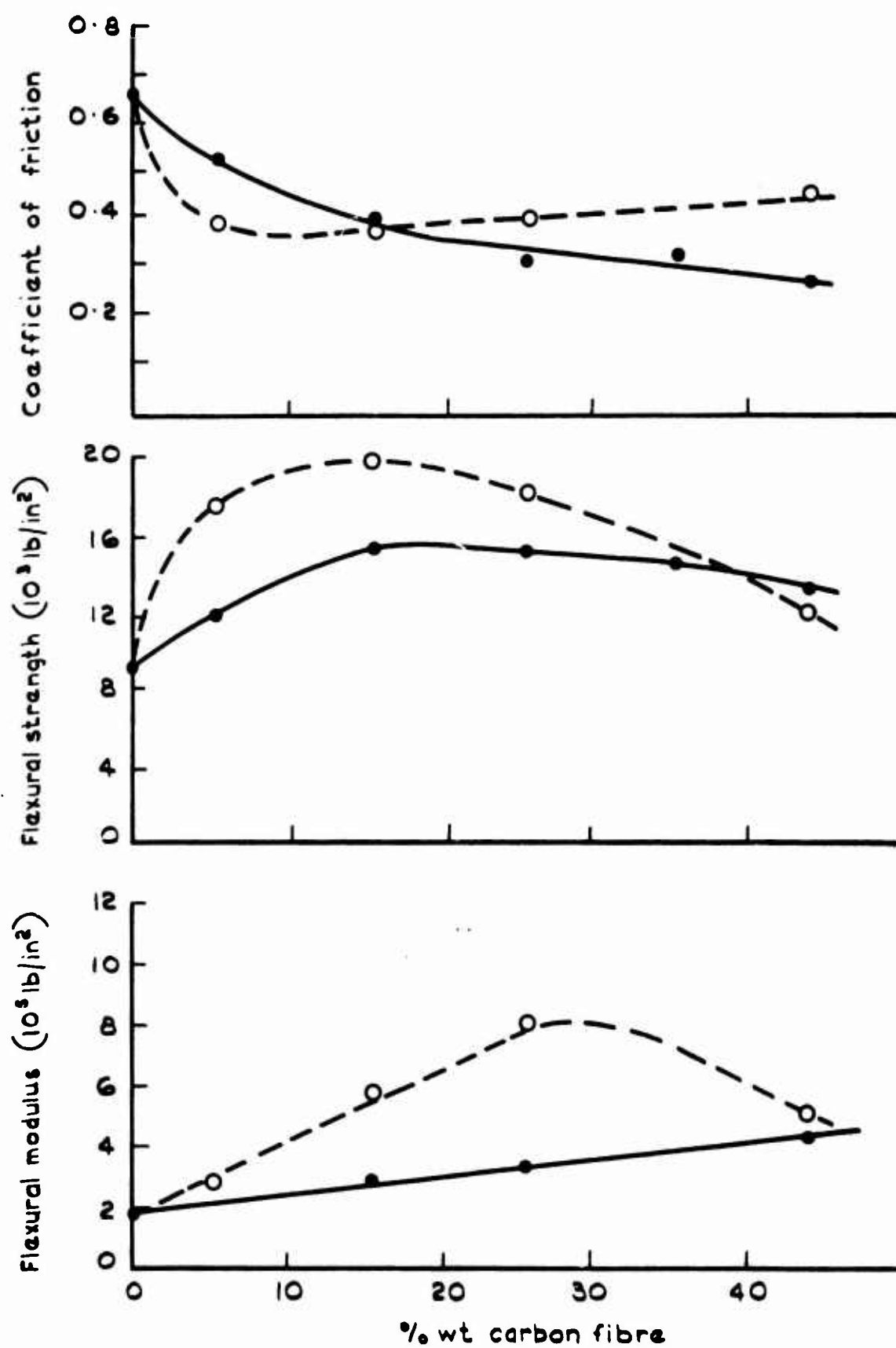


Fig. 4 Effect of carbon fibre concentration in



tration in acetal copolymer ("Kematal")

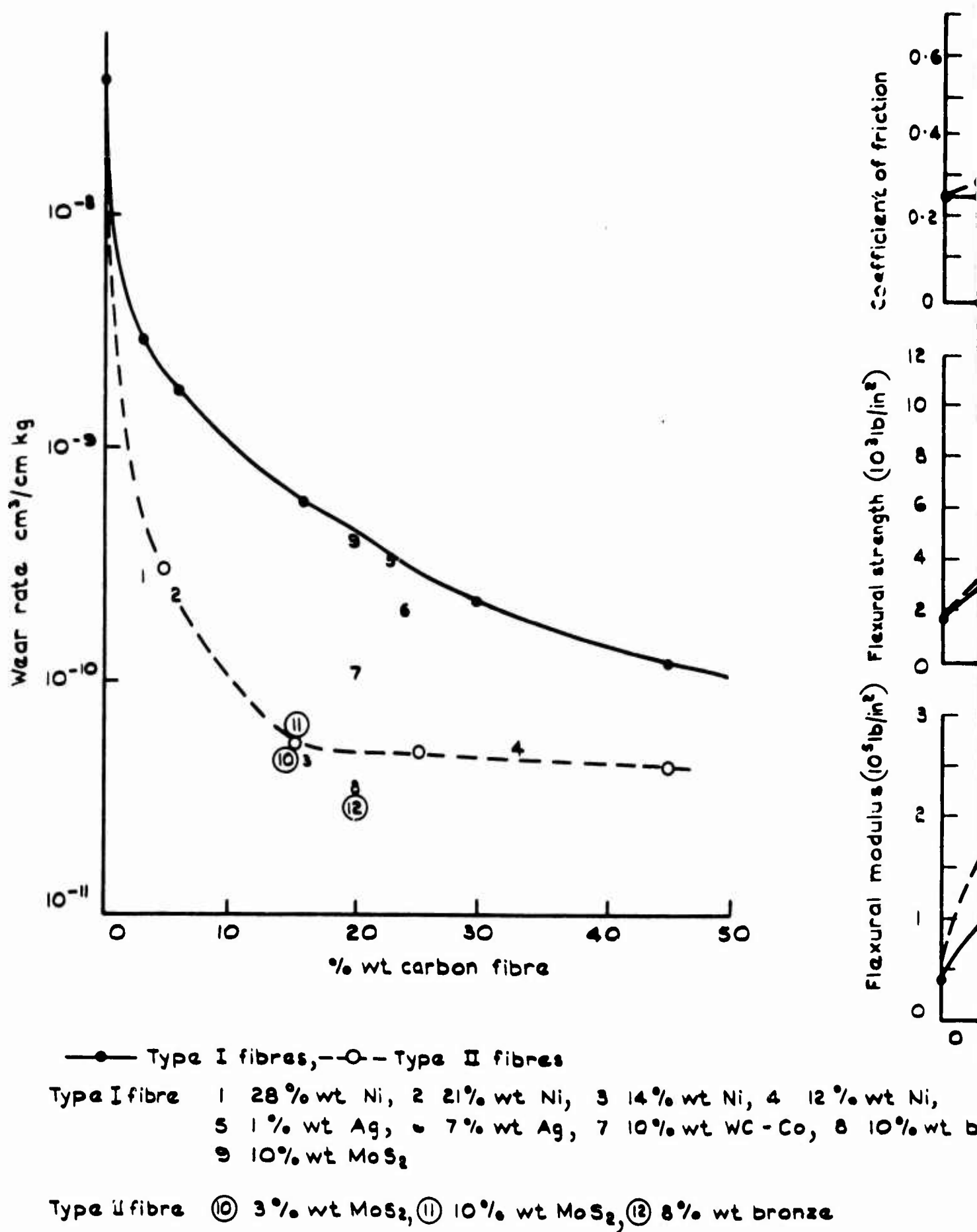
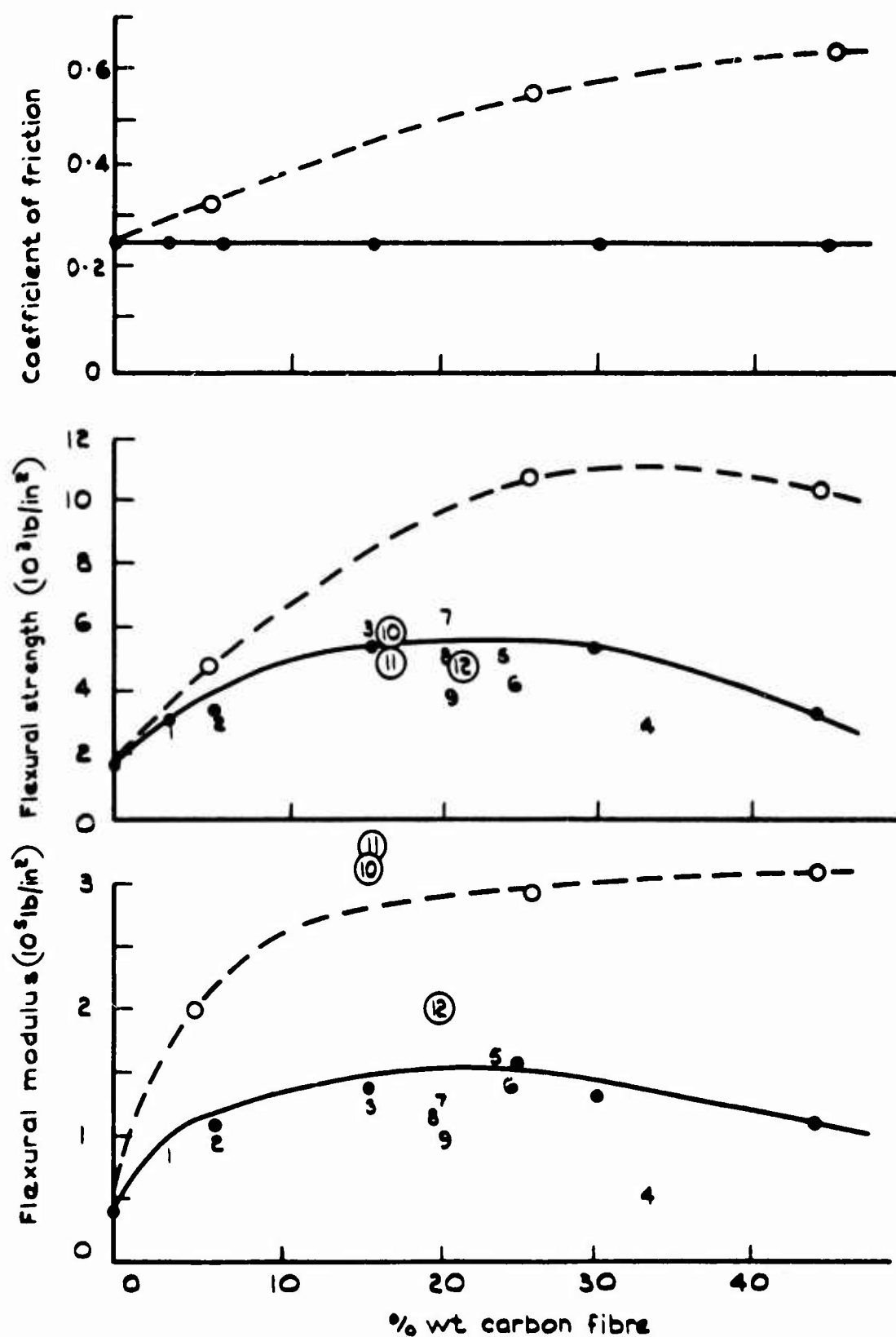


Fig.5 Effect of carbon fibre concentration and additions to PTFE



1, 4 12% wt Ni,
 2, 5 10% wt bronze

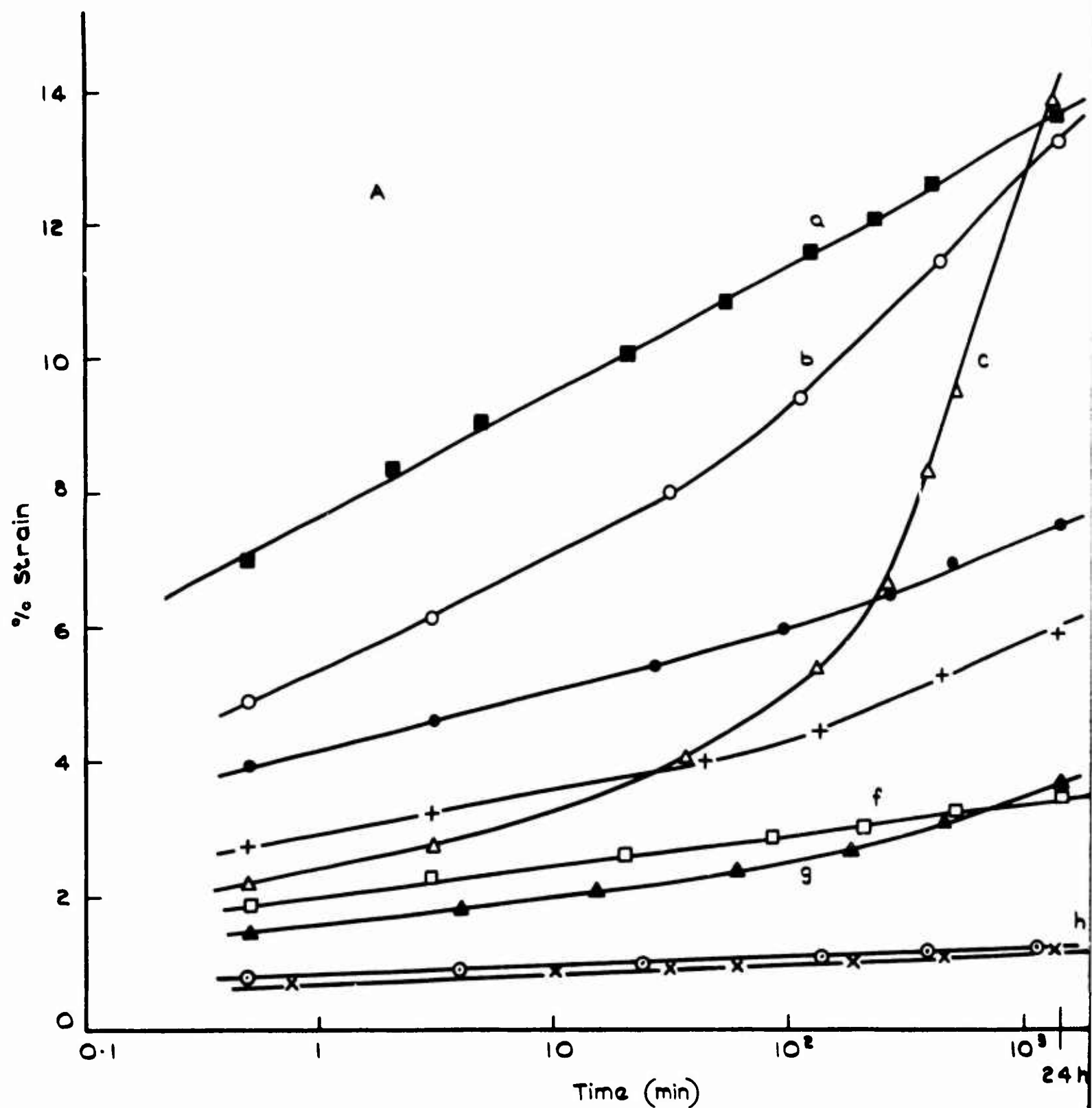
Speed = 270cm/s

bronze

concentration and third component
 ions to PTFE

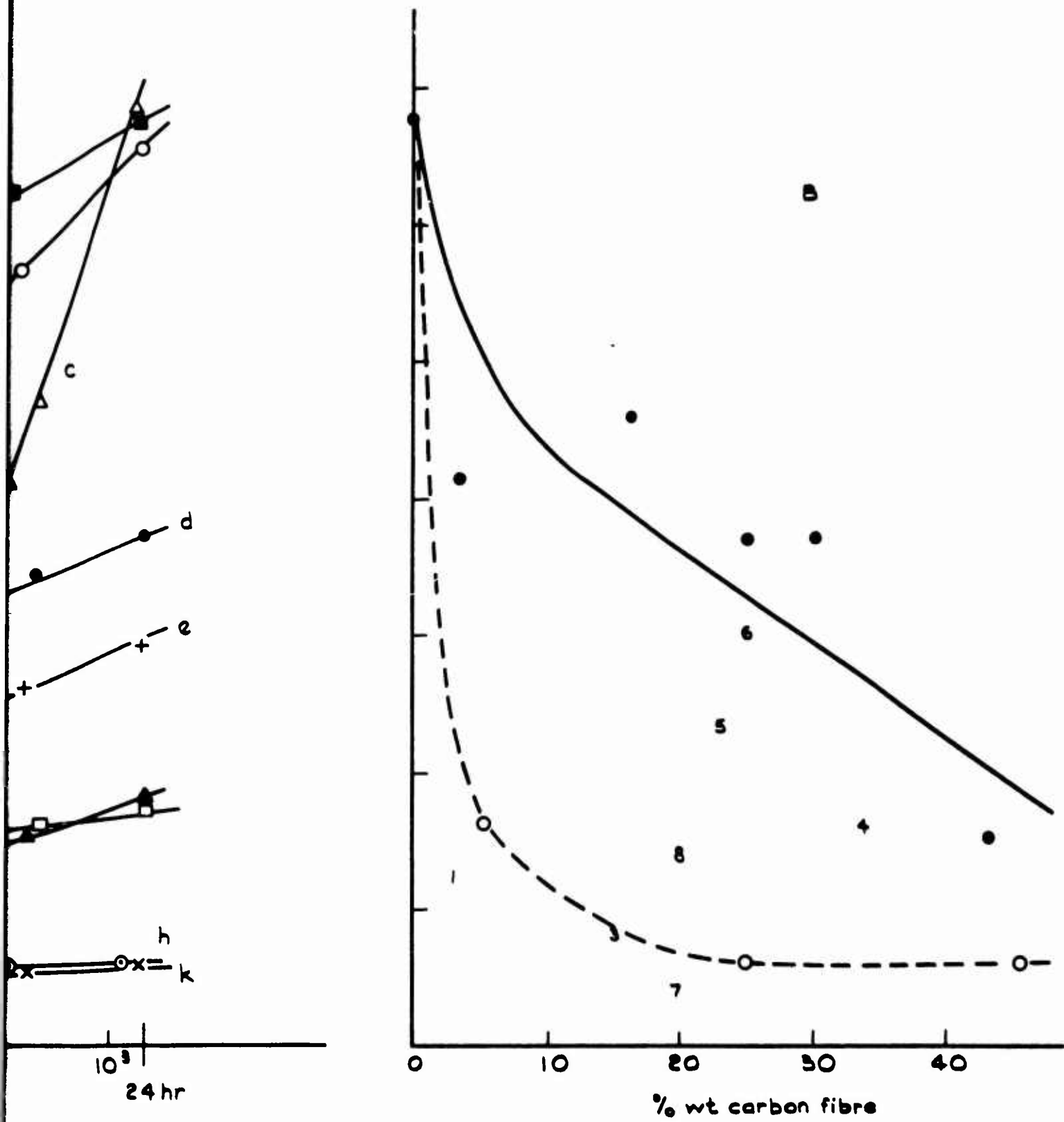
Fig. 6

014 900492



- A Variation with time a PTFE, b 25% wt glass, c 25% wt asbestos, d
f Bronze - graphite ('Glacier DQ'), g 25% wt coke,
- B Carbon fibre reinforced composites after 24 hr —●— Type I fibre —○— Type II fibre

Fig. 6 Deformation under load for various filled



Asbestos, d 25% wt carbon fibre, (type I), e 33% wt graphite
10% wt coke, h 25% wt carbon fibre (type II), k Mica (Fluorocint)
Type II fibre. Numbers as in the legend to Fig. 5

Various filled and reinforced PTFE materials

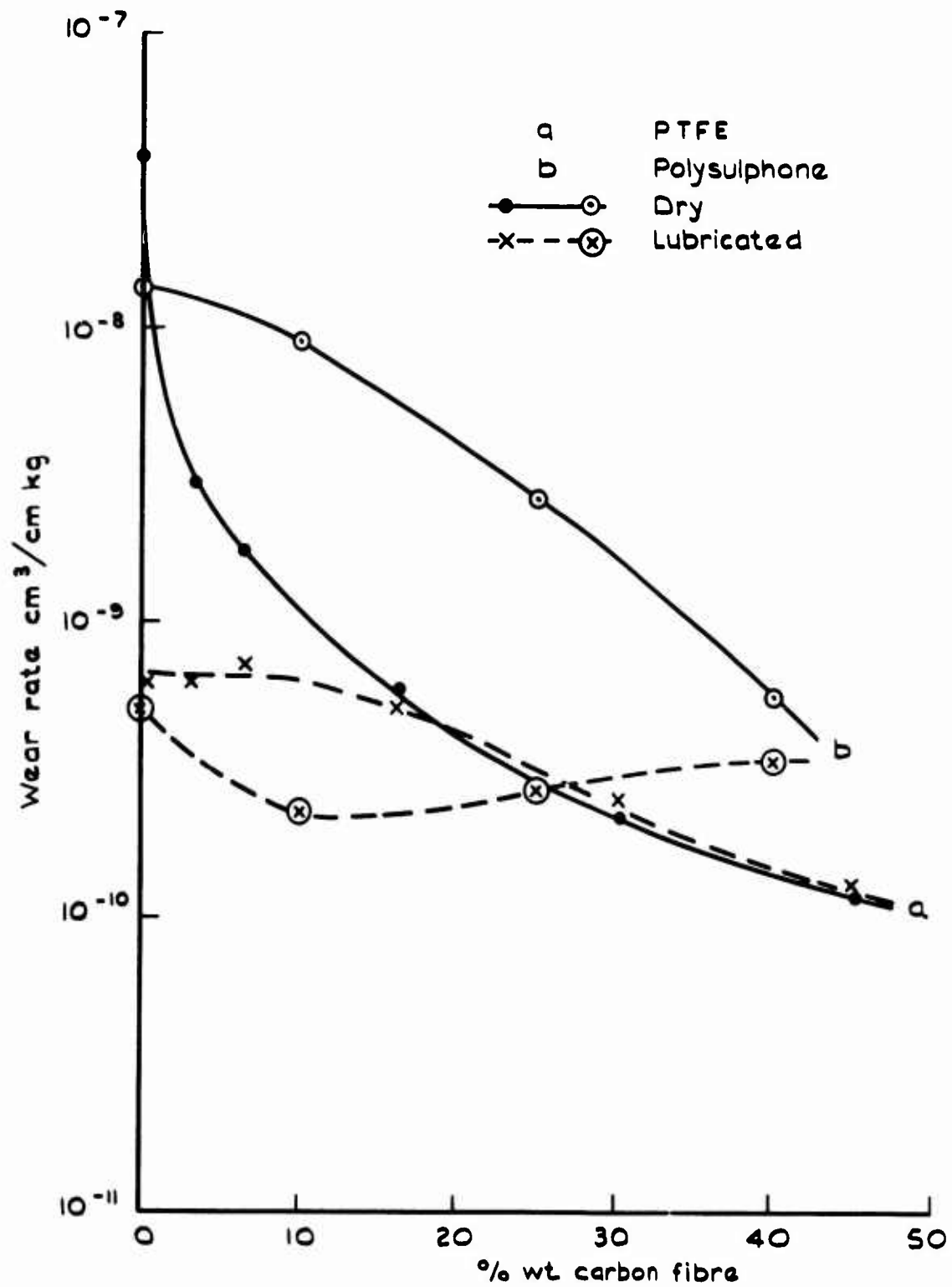


Fig. 7 Effect of lubrication by n-hexadecane on the wear rate of two carbon fibre composites