

Figure I A plot of relative electric strengths against the boiling points for the various gaseous dielectrics listed in Table I; the numbers in this figure correspond to numbers identifying the various gases in Table I.

necting the electric strengths of a very wide range of dielectric gases with their fundamental physical, chemical, structural or compositional parameters seem to exist in the literature. Also such correlations as shown in Fig. 1 can perhaps provide a good starting point for the selection of new gaseous dielectric materials of use in power engineering, high voltage technology and industrial insulation.

Acknowledgement

Thanks are due to Dr A. Théorêt for help in locating sources of data on the electric strength of some gases.

References

- 1. A. K. VIJH, J. Mater. Sci. 11 (1976) 784.
- H. L. SAUMS and W. W. PENDLETON, "Materials for Electrical Insulating and Dielectric Functions", (Hayden, Rochelle Park, New Jersey,) p. 313.
- 3. L. PAULING, "College Chemistry", 3rd Edn. (Freeman, San Francisco, 1964) p. 445.
- 4. L. L. ALSTON (Ed.), "High Voltage Technology" (Oxford University Press, New York, 1968).
- P. J. HARROP, "Dielectrics" (Wiley, New York, 1972).
- 6. W. BRAKER and A. K. MOSSMAN, "Matheson Gas Data Book" (Matheson Gas Products, East Rutherford, New Jersey, 1971).
- 7. R. T. SANDERSON, "Inorganic Chemistry" (Reinhold, New York, 1967).

Received 5 February and accepted 24 February 1976

> ASHOK K. VIJH Hydro-Quebec Institute of Research, Varennes, P.Q., Canada

Physical characteristics of extruded carbon mixes

Carbon products are fabricated from a mixture of carbon filler and binder, such as coke and pitch respectively. This mixture is formed by moulding or extrusion and is then baked. The quality of the baked product is dependent upon the quality of the green carbon, which is considered as acceptable if free from surface cracks and containing an optimum amount of binder. Many investigations have been performed on the moulded carbon mixes [1-3], but little has appeared in the literature regarding the extruded carbon mixes [4-6]. A systematic study was made of the physical characteristics of extruded carbon mixes with regard to the processing conditions.

Fincle [4] reports some increase in viscosity of © 1976 Chapman and Hall Ltd. Printed in Great Britain.

carbon mixes when extruded at high speeds, but no account of the extruded irregularities has been given. An explanation for the viscosity increase in the carbon mixes is proposed in this note. Suitable experiments were performed with carbon mixes to detect the presence of such extrudate irregularities and the results are reported below.

The carbon mixes in the present investigation consisted of calcined petroleum coke as filler and coal tar pitch as binder. The characteristics of these materials are given in Table I. Seven mixes were made using different proportions of binder. The filler in the seventh mixture consisted of a different particle size distribution. The above mixtures were roll-mixed at 120° C and then ground to pass through 60 B.S. mesh. Slugs, 12.5 cm diameter, were made from these mixtures which were extruded at speeds of either 2 to 4 m or 25 to 40 m min^{-1} in a 200 tonne extrusion press. The extruded rods were collected and then checked for any extrudate irregularities.

The apparent density of the green rods (thirty taken at random from the batch) was determined by measuring their mass, length and diameter. These green rods were then baked in an electric muffle furnace to 950° C in a 4 day baking cycle. The baked rods were cut to standard lengths and subjected to baked density, electrical resistivity and transverse strength measurements. The electrical resistivity was determined by passing a suit-

TABLE I

| Material | Characteristics | | | | |
|--------------------------|--------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------|--|--|--|
| Coal tar pitch binder | Ring and ball softening point 78° C Coking value = 36% Apparent density = 1.25 g cm^{-3} | | | | |
| Petroleum coke filler | Calcination temperature = 1300° C Ash content = 0.5 wt % Bulk density = 0.82 g cm ⁻³ [7] Sieve analysis (B.S. sieves) | | | | |
| | $\begin{array}{r} + & 60 \\ - & 60 + 100 \\ - & 100 + 200 \\ - & 200 + 300 \\ - & 300 \end{array}$ | 5% 10% 34% 10% 38% | | | |

able current through the cross-section of the rod held between two thick brass plates and measuring the voltage drop between two probes placed along the length of the rod. The transverse strength was measured by placing the rod on two knife edges and applying an increasing load at its centre with a third knife edge until the rod broke. The results of these experiments have been summarized in Table II.

It is seen from this table that carbon mixes extruded at high speeds have higher apparent density, greater strength and lower electrical resistivity than those extruded at low speeds. The reason for these improved properties is the pressure which causes the binder to penetrate into those places which it could not reach in the mixing or moulding operations. Such a product after baking will obviously have high density, more strength and less resistivity. The speed of extrusion may also be increased by placing a hot flame over the extruding die; however, this leads to cracks in the green as well as baked carbons. This suggests the importance of pressure over speed in the extrusion of the carbon mixes.

It is further indicated in Table II that good carbon mixes corresponds to batch II containing 38% binder. The level of binder above or below this

TABLE II Properties of extruded carbon mixes at low and high speeds

| Batch | Binder (%) | Comparative speed | Green density (9 cm ⁻³) | Mean baked density (9 cm ⁻³) | Mean resistivity (μΩm) | Mean transverse strength (N mm ⁻²) | Hydraulic pressure (10 ⁻⁶ X dyn cm ⁻² | Speed of extrusion) (m min ⁻¹) | Green rod diameter (mm) |
|-------|---------------|----------------------|-------------------------------------------|---------------------------------------------------|------------------------------|---------------------------------------------------------|-------------------------------------------------------------------|---------------------------------------------------|-------------------------------|
| 1 | 36 | low | 1.766 | 1.683 | 36.5 | 49.4 | 11.3 | 1.8 | 8.28 ± 0.02 |
| | | high | 1.763 | 1.686 | 36.0 | 51.6 | 20.7 | 12 - 15 | 8.36 ± 0.02 |
| 2 | 38 | low | 1.778 | 1.697 | 34.0 | 59.3 | 10.4 | 3 - 4.5 | 8.30 ± 0.01 |
| | | high | 1.791 | 1.698 | 32.7 | 61.2 | 15.2 | 30 - 39 | 8.26 ± 0.005 |
| 3 | 40 | low | 1.787 | 1.656 | 36.0 | 52.3 | 5.5 | 3-4.5 | 8.30 ± 0.02 |
| | | high | 1.786 | 1.665 | 35.7 | 53.8 | 6.9 | 18 - 24 | 8.30 ± 0.02 |
| 4 | 42 | low | 1.775 | 1.654 | 35.3 | 54.8 | 6.2 | 3-6 | 8.29 ± 0.01 |
| | | high | 1.776 | 1.657 | 35.0 | 57.4 | 6.9 | Snaky | 8.29 ± 0.01 |
| 5 | 44 | low | 1.768 | 1.623 | 34.9 | 55.0 | 5.5 | Snaky | 8.31 ± 0.05 |
| | | high | 1.767 | 1.657 | 33.6 | 56.2 | 5.5 | Snaky | 8.34 ± 0.04 |
| 6 | 46 | low | 1.760 | 1.618 | 35.9 | 54.0 | 35 - 5.5 | Snaky | 8.18 ± 0.03 |
| | | high | 1.755 | 1.618 | 35.2 | 53.8 | 6.2 | Snaky | 8.30 ± 0.03 |
| 7 | 40 | low | 1.763 | 1.655 | 23.1 | 44.8 | 6.2 | 18 | 8.28 ± 0.01 |
| | | high | 1.764 | 1.663 | 22.8 | 50.6 | 8.3 | 36 | 8.29 ± 0.01 |

optimum level leads to comparatively inferior properties. A high speed of extrusion was not possible at the binder level of 42% or more, because of the resultant snake-like extrusion. This cannot be classified as an extrudate irregularity because it is caused by the excessive binder in the mix.

Fincle [4] reports an increase in viscosity of the green mix at a shear rate of about 100 sec^{-1} . There are two possible reasons for such an increase. One is due to the extrusion near the end of the batch of carbon when the speed of extrusion gradually becomes slower. In order to maintain the speed, the pressure of extrusion must be increased which results in an increased viscosity of the mix. The second reason may be the removal of binder from the green mix at such high pressures. This would leave a mix with comparatively less binder which would give rise to increased viscosity.

Tordella [8] reports the appearance of irregularities in the polymers extruded above a critical flow rate. Since carbon mixes behave as a viscoelastic material, the possibility of extrudate irregularities occurring in this case cannot be ruled out. However, from the results of the present study, it is concluded that extrudate irregularities do not occur at least up to shear rates of 650 sec^{-1} . The possibility of extrudate irregularities occurring at shear rates higher than this cannot be eliminated.

Acknowledgements

The author is grateful to Dr S.S. Chari for continued guidance and helpful discussions and to Dr G.C. Jain for his keen interest and encouragement in the study. Thanks are due to the Director, National Physical Laboratory for permission to publish the paper.

References

- S. MOROZOWSKI, Proceedings of the 1st and 2nd Carbon Conferences (University of Buffalo, New York, 1956) p. 195
- 2. E. J. SELDIN, Proceedings of the 3rd Carbon Conference, (Pergamon Press, Oxford, 1959) p. 675.
- J. OKADA and Y. TAKEUCHIE, Proceedings of the 4th Carbon Conference, (Pergamon Press, Oxford, 1961) p. 54.
- R. L. FINCLE, paper No. 153, Seventh Conference on Carbon, Abstract; *Carbon* 3 (1965) 370.
- 5. G. BHATIA, Carbon 11 (1973) 437.
- 6. Idem, Presented in Symposium on Large deformations, I.I.T., Delhi, November 1975.
- 7. J. N. BOHRA and G. BHATIA, 12th Biennial Carbon Conference Pennsylvania, in press.
- J. P. TORDELLA, Trans. Soc. Rheology 1 (1957) 203.

Received 28 November 1975 and accepted 16 February 1976

> G. BHATIA Division of Materials, Carbon Technology Unit, National Physical Laboratory, New Delhi - 110012, India

New polytypes in the Be-Si-O-N system

Huseby et al. [1] have recently shown the existence of a series of crystallographically similar phases at Be₃N₂-rich compositions in the Si₃N₄- $Be_3N_2-BeO-SiO_2$ system. These phases occur with ranges of homogeneity elongated along lines of constant metal: non-metal atom ratio M_{m+1} X_m where m has values 4, 5, 6, 7 and 9. The minimum value, m = 2, gives beryllium nitride (Be₃N₂), the crystallography of which is well known [2,3]. The maximum value, $m = \infty$, gives a range of compositions extending from beryllium-silicon nitride (BeSiN₂) to beryllium oxide (BeO). In the present communication, the unidentified powder diffraction patterns reported for the new phases [1] are indexed and interpreted in terms of wurtzite-type polymorphs containing excess metal atoms, the structures of © 1976 Chapman and Hall Ltd. Printed in Great Britain.

which are directly related to their compositions and also to recently discovered $M_m X_{m+1}$ polytypes in the Si-Al-O-N system.

The indexed diffraction patterns for compounds with $m \le 6$ are given in Table I. In every case the unit cell is hexagonal with $a \sim 2.8$ Å and with c large and varying from compound to compound in a manner characteristic of a polytypic series [4]. The cell dimensions of all the unidentified phases reported by Huseby *et al.* [1] are given in Table II together with the appropriate polytype designations.

The crystal structures of both beryllia and beryllium-silicon nitride are of the wurtzite type with a hexagonal close-packing of nonmetal atoms and with the metal-atoms occupying all of the upward-pointing tetrahedra. The crystal structure of β -beryllium nitride, although originally thought to consist of alternate layers of upward