

Letters to the Editor

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Electric Resistivity of Polycrystalline Graphite and Carbons

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SINGLE graphite crystals are known to be good conductors along the graphite planes and very poor ones across (ratio of resistivities 10^4 or higher.¹) Coulson² and Wallace³ have shown that a graphite crystal along the planes is a semiconductor with zero activation energy, the only difference with a metal consisting in a decrease of the density of states to zero for both the filled and unfilled bands as one approaches their common limit. Although the tight binding approximation does not yield a completely satisfactory description of all the properties of a graphite crystal, the result mentioned seems well established. The theory is, however, developed only for the case of infinitely large crystals.

For the artificial carbons with width of graphitic crystallites varies from 25A (calcined cokes) up to 1000A or more (highly graphitized specimens), we have therefore a continuous transition from small crystals down to the realm of large aromatic molecules. In studying the change of the electric resistivity ρ with temperature T , information is obtained only concerning the flow of electric current along the molecular planes, due to the high anisotropy of the crystallites. The curves ρ versus T reveal each a minimum, the position of the minimum shifting toward lower temperatures as the size of crystallites increases. The existence of the minimum is a direct consequence of an intrinsic semiconductivity of crystallites of graphite. In distinction to the infinitely large crystal it may be assumed that they possess a finite energy gap between the full and empty bands, the activation energy ΔE increasing with decrease of the crystallite size. The existence of an energy gap at the boundary of the Brillouin zone was found necessary for the explanation of the quantitative relations found in absorption spectra of aromatic molecules.⁴ For each carbon a negative temperature coefficient of resistance typical for semiconductors is observed at lower temperatures; for temperatures $T \gg \Delta E/k$ the presence of the gap becomes immaterial and a typical metallic behavior is obtained (linear increase). For commercial graphites an activation energy of about 25×10^{-3} ev is calculated from the position of the minimum (500°C).

For calcined cokes the effective energy gap is probably at least as large as 0.1 ev or greater, and the small crystals should consequently be very poorly conducting at room temperature. The relatively low electric resistivity of such carbons (five to seven times that of graphitized material at room temperature) must be due therefore to an extrinsic semiconductivity. The existence of such a semiconductivity is revealed by an analysis of the resistivity curves even for commercial graphite in the region 600°–150°K. The curves can be explained by assuming the presence of a constant number of free electrons in the conduction band in addition to the variable number of electrons activated from the lower band. Since in highly graphitized specimens there are practically no impurities present, the excess electrons must be due to the carbon atoms themselves. In the polycrystalline graphite some carbons on the periphery of the crystals are bound by valence forces to carbons belonging to neighboring crystallites.

A great number, however, of peripheral carbons (and also carbons around lattice imperfections) have free valence electrons which drop down from their energy states into the bottom of the conduction band (at very low temperatures into bound states around their original atoms with low activation energies ϵ), thus becoming the free excess electrons required for the conduction.

In conclusion, the polycrystalline carbons are true semiconductors with an activation energy depending on the size of crystallites. They show a metallic conductivity for $kT \gg \Delta E$, an intrinsic semiconductivity for $kT \sim \Delta E$ gradually changing as the temperature is lowered into a free electron excess semiconductivity. Finally, a fourth region should be reached at the lowest temperatures ($kT \sim \epsilon$) where a sharp increase in the resistivity is expected. A paper containing a more complete discussion of the problem and of all the experimental evidence available at present is in preparation. Experimental studies of the resistivity and Hall effect to check further the proposed explanation are in preparation.

Recently Bowen⁵ succeeded in reproducing the shape of the resistivity curve for the polycrystalline graphite by simply adding the scattering due to crystal boundaries to the temperature scattering for infinite crystals. It is believed, however, that Bowen overestimated the magnitude of the boundary effect.

¹ K. S. Krishnan and N. Ganguli, *Nature* **144**, 667 (1939).

² C. A. Coulson, *Nature* **159**, 265 (1947).

³ R. P. Wallace, *Phys. Rev.* **71**, 622 (1947).

⁴ H. Kuhn, *J. Chem. Phys.* **16**, 840 (1948); J. R. Platt, *J. Chem. Phys.* **17**, 484 (1949).

⁵ D. Bowen, *Phys. Rev.* **76**, 1878 (1949).

Element 97*

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GOOD evidence has been obtained for the identification of an isotope of the element with atomic number 97 through the irradiation of americium-241 with helium ions in the Berkeley 60-inch cyclotron. The particular isotope discovered is thought to be 97^{243} , or possibly 97^{244} , decaying with a 4.8-hour half-life by electron capture with approximately 0.1 percent alpha-decay branching. There seem to be three alpha-particle groups associated with the activity, the highest energy being 6.72 Mev.

The chemical separation of element 97 from the target material and other reaction products was made by combinations of precipitation and ion exchange adsorption methods making use of its anticipated properties of having III and IV oxidation states and its position as a member of the actinide transition series. The distinctive chemical properties made use of in its separation and the equally distinctive decay properties of the particular isotope constitute the principal evidence for the new element.

It is suggested that element 97 be given the name berkelium (symbol Bk), after the city of Berkeley, in a manner similar to that used in naming its chemical homologue terbium (atomic number 65) whose name was derived from the town of Ytterby, Sweden, where the rare earth minerals were first found.

We wish to acknowledge the assistance in the early phases of the problem and the continuous advice of Professor B. B. Cunningham, whose help contributed greatly to the success of the work.

It is a pleasure to acknowledge the special help in the bombardment of the americium samples of Professor J. G. Hamilton, G. B. Rossi, T. M. Putnam, Jr., M. T. Webb, and the operating crew of the 60-inch cyclotron in the Crocker Laboratory. The successful handling in a safe manner of the radioactivity involved was made possible through the use of the excellent protective equipment provided by Nelson Garden and the members of his Health Chemistry Group, particularly C. M. Gordon, W. G. Ruehle, and J. M. Davis. We wish also to thank E. K. Hulet and G. H. Higgins for their assistance in some of the experiments and to express our appreciation to Dr. Kenneth Street, Jr., for valuable