

In the preparation of the new trihydroxypolyporic acids the reaction of 2,5-dichlorobenzoquinone with the appropriate *N*-nitrosoacetanilide gave 2,5-dichloro 3-phenyl 1,4-benzoquinone and 2,5-dichloro 3-(4-*t*-butylphenyl) 1,4-benzoquinone, which were then treated with diazotized 3,4,5-trimethoxyaniline. The dichloroterphenyl quinones thus prepared were hydrolysed to the corresponding dihydroxyquinones. Demethylation of the dihydroxy terphenyl quinones with a mixture of hydrobromic acid and acetic acid gave the pentahydroxyquinones directly; reductive demethylation followed by oxidation was not attempted.

The antioxidant activity of the compounds was tested in pure linoleic acid and in pure methyl linoleate at a temperature of 25° C. Uptake of oxygen was followed with conventional Warburg apparatus and this was correlated with peroxide formation, which was determined by means of an iodometric method<sup>4</sup>. A special test was used for the determination of antioxidant efficiency in the presence of metal catalysts, using ferrous phthalocyanine in ethyl benzoate solutions as standard. The results indicated that polyporic acid had no significant antioxidant activity, while atromentin and leucamelone were comparable with conventional antioxidants, for example, propyl gallate, for the inhibition of fat autoxidation. The new antioxidants 3,4,5-trihydroxypolyporic acid and 4'-*tert*-butyl 3,4,5-trihydroxypolyporic acid showed outstanding activity. In linoleic acid these compounds gave protection factors which were, respectively, about ten and twenty-five times that obtained with propyl gallate. It should be noted that the *tert*-butyl substituent substantially increases the solubility of the trihydroxypolyporic acid in higher fatty acids and their esters.

Anti-leukaemic activity of unsubstituted polyporic acid was reported recently<sup>5</sup>; it would be interesting to compare the polyhydroxypolyporic acids which show greater antioxidant activity.

The work was supported by the U.S. Department of Agriculture P.L.480 counterpart funds.

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<sup>1</sup> Uri, N., in *Autoxidation and Antioxidants*, edit. by Lundberg, W. O. (Interscience Publishers, Inc., New York) (in the press).

<sup>2</sup> Simpson, T. H., and Uri, N., *Chem. and Indust.*, 956 (1956). Uri, N., *Essential Fatty Acids*, 30 (Butterworths Scientific Publications, London, 1958).

<sup>3</sup> Akagi, M., *J. Pharm. Soc. Japan*, **62**, 202 (1942).

<sup>4</sup> Heaton, F. W., and Uri, N., *J. Sci. Food and Agric.*, **9**, 781 (1958).

<sup>5</sup> Burton, J. F., and Cain, B. F., *Nature*, **184**, 1326 (1959).

### Ionic-Covalent Bonding in Crystals

AN attempt<sup>1</sup> to resolve misunderstandings regarding ionic-covalent bonding in solids has been criticized in a recent article by Mooser and Pearson<sup>2</sup>. It should be pointed out, however, that their discussion of formal ionicity is limited to the simple and well-known case of atoms which each contribute one electron to a two-electron bond, and cannot be applied as it stands to 'dative bond' systems (such as are found in the zinc blende structure and in many other types of crystal, as well as in co-ordination complex molecules). In such systems formal covalency, defined as a state in which bonding electrons are equally shared between bonded atoms, can only be achieved by a donor-acceptor relationship with associated charge separation.

The main point in my communication<sup>1</sup> was the importance of the neutral-bond state (defined as that with zero charge separation) as the logical zero for the scale of electronegativity. (It should be noted that this is generally applicable; thus it holds for bonds in diatomic molecules, since for such molecules neutral-bonded and formally covalent states are identical.) This suggests that 'ideal covalency' (so termed for want of a better name) is associated with zero charge separation rather than with equal sharing of electrons. If this is so, the difficulty pointed out by Mooser and Pearson regarding the zero of the scale of bond ionicity used in my communication<sup>1</sup> can readily be resolved. It may well be that some revision of the accepted definition of the covalent bond on these lines is needed. However, in terms of the definitions used, the conclusions regarding charge separation presented in ref. 1 are still valid. In this connexion the concluding statement by Mooser and Pearson that "any attempts such as have been made to determine effective charges from formal bond ionicities are bound to fail" is certainly not a dogmatically required belief. It is my opinion, on the contrary, that useful qualitative information regarding charge separations in crystals can be obtained, although, for obvious reasons, some of which are referred to in my earlier communication, no quantitative estimates are as yet possible.

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<sup>1</sup> Goodman, C. H. L., *Nature*, **187**, 590 (1960).

<sup>2</sup> Mooser, E., and Pearson, W. B., *Nature*, **190**, 406 (1961).

GOODMAN's comments on our recent article<sup>1</sup> in no way affect our views on the subject of ionic-covalent bonding as outlined there. Since it is obvious, however, that Goodman is misinterpreting these views, we should like to stress once again the following points:

(1) The 'effective bond ionicity' is defined in terms of the actual charges associated with the constituents of a compound. It is, therefore, a physical quantity which can, at least in principle, be measured.

(2) The 'formal bond ionicity' is a purely theoretical concept. It is, in fact, an abbreviation with which to indicate to what extent various arbitrarily chosen valence structures are mixed to describe the bonding in a molecule or a solid. Contrary to Goodman's remark, the formal bond ionicity is defined also for 'dative' bonds.

(3) The covalent bond is defined as arising from the sharing of two electrons between two bonded atoms. It does not specify the charges associated with these atoms and a covalent valence structure cannot, therefore, in general, be taken as the zero of the bond-ionicity scale.

(4) All the definitions given by us<sup>1</sup> and repeated here are in agreement with chemical tradition. The intention of our article was merely to point out that two different meanings are currently attached to the term 'bond ionicity'. To distinguish between them, we introduced the names effective and formal bond ionicities.

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<sup>1</sup> Mooser, E., and Pearson, W. B., *Nature*, **190**, 406 (1961).