

INORGANIC CHEMISTRY.

1. NON-STOICHEIOMETRIC COMPOUNDS.

RECENT work in inorganic chemistry has raised the question of the validity of the law of constant proportions, as applied to solid compounds. The existence and classification of "Berthollide" compounds have been noted previously in these Reports,¹ and it appears opportune to review our present state of knowledge of the subject.

It is generally recognised that the intermediate phases in metallic systems may exist over a range of composition, not necessarily including a rational chemical formula; an idealised chemical formula can usually be assigned, however, based on the composition of the unit cell of the crystal.² Between intermetallic and ionic compounds there is a transition, rather than an abrupt demarcation,³ depending on the difference in electronegativity of the combining atoms, and whereas the elements of Groups VB, VIB, and VIIB form true salts with the most electropositive metals (Groups IA, IIA), their compounds with the transition and B-sub-group metals display a complete transition from the ionic to the quasi-metallic type. Variability of composition runs broadly parallel to sub-metallic properties, but is by no means limited to compounds of obviously sub-metallic character.

The distinction between solid solutions, interstitial compounds and non-stoicheiometric compounds is, in the last analysis, rather arbitrary. N. S. Kurnakow⁴ first proposed the term "Berthollide" (as distinct from "Daltonide") to describe homogeneous phases in systems where the maxima or minima of properties—melting point, conductivity, lattice order, etc.—do not coincide with a rational atomic ratio of the components. For the purpose of this report it is convenient to follow W. Schottky and C. Wagner⁵ in considering the familiar "Daltonide" type as a special case of the "ordered mixed phase", a 2-component (or multicomponent) system with statistically regular lattice array.

Our present knowledge of crystal structure confers precise meaning on the term "solid solution" as applied to crystals of atomic lattice types. In a crystal phase of ideal formula AB_n , a stoicheiometric excess of element B can be accommodated structurally in only three ways: (i) *Substitutional* solid solution: B atoms replace A atoms on lattice sites proper to A. (ii) *Interstitial* solid solution: additional B atoms are located in inter-lattice positions. (iii) *Subtractive* solid solution: all B atoms occupy proper B lattice sites, but a number of A lattice sites is left untenanted.

Since (ii) increases and (iii) decreases the average weight per unit cell, distinction between them is possible by combining density and X-ray cell

¹ *Ann. Reports*, 1933, **30**, 381; 1935, **32**, 211.

² A. Westgren, *Angew. Chem.*, 1932, **45**, 33.

³ Cf. E. Zintl, *ibid.*, 1939, **52**, 1.

⁴ *Z. anorg. Chem.*, 1914, **88**, 109.

⁵ *Z. physikal. Chem.*, 1930, **B**, **11**, 163.

Annual Reports On The Progress Of Chemistry Inorganic Chemistry V 91

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