

CHAPTER TWO

The Closed-System Constraint and Chemical Stoichiometry

Here we develop the basis for the constraint on the computation of chemical equilibrium that is due to the requirement for conservation of elements in a closed system undergoing chemical change, a special form of the law of conservation of mass. This constraint is intimately bound up with what is usually called *chemical stoichiometry*,* whether it is expressed directly in terms of conservation equations or indirectly in terms of chemical equations. The specific purpose of this chapter is to develop chemical stoichiometry for a closed system in a form suitable for incorporation in an equilibrium-computation algorithm. Useful tools for this are provided by linear algebra since the conservation equations are themselves linear algebraic equations, and in what follows we make use of vector-matrix notation. The end result is a stoichiometric-coefficient algorithm, computer programs for which are contained in Appendix A.

2.1 THE APPROACH

We first define a closed system and then develop a method (Smith and Missen, 1978) for treating chemical stoichiometry that involves generating, *a priori*, an appropriate set of chemical equations. Since another approach involves starting with a set of such equations, we discuss the implications of this and finally consider some special stoichiometric situations. In this sense the treatment is more general than, and goes beyond, the specific purpose indicated previously.

*The word "stoichiometry," of Greek origin, strictly means measurement (unity) of an element (stoichion) in chemical stoichiometry the element is a chemical element.

2.2 THE CLOSED-SYSTEM CONSTRAINT

2.2.1 The Element-Abundance Equations

A closed system has a fixed mass, that is, it does not exchange matter with its surroundings, although it may exchange energy. It may consist of one or more than one phase and may undergo reaction and mass transfer internally. Its importance in equilibrium computations is that the equilibrium conditions of thermodynamics (Chapter 3) apply primarily to such a system.

In the laboratory and in chemical processing, the concept of a closed system obviously applies to a batch system. Perhaps less obviously, it also applies to a fluid system undergoing "plug" flow in which there is no mixing or dispersion in the direction of flow and in which all elements of fluid have the same residence time in a particular vessel or conduit (Levenspiel, 1972, p. 97). In such a case each portion of fluid, of arbitrary size, acts as a batch system in moving through the vessel. This description is most suitably applied to a fluid flowing at a relatively high velocity in a conduit of uniform cross section.

Operationally, any description of a closed system is an expression of the law of conservation of mass. A closed system can be defined by a set of element-abundance equations expressing the conservation of the chemical elements making up the species of the system. There is one equation for each element, as follows:

$$\sum_{i=1}^N a_{ij} n_i = b_j, \quad j = 1, 2, \dots, M, \quad (2.2-1)$$

where a_{ij} is the subscript to the i th element in the molecular formula of species i , n_i is the number of moles of i (in some basis amount of system); b_j is the fixed number of moles of the j th element in the system; M is the number of elements; and N is the number of species. Alternatively, equation (2.2-1) may be written so as to express the change from one compositional state to another:

$$\sum_{i=1}^N a_{ij} \Delta n_i = 0, \quad j = 1, 2, \dots, M, \quad (2.2-2)$$

where Δn_i is the change in the number of moles of the i th species between two compositional states of the system.

In vector-matrix form, the element-abundance equations 2.2-1 and 2.2-2 are, respectively,

$$A n = b, \quad (2.2-3)$$

and

$$A \Delta n = 0, \quad (2.2-4)$$

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