

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 (molecule) 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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Computational Chemistry and Interactive Modeling Maël Bosson,2012 At the atomic scale interactive physically based modeling tools are more and more in demand Unfortunately solving the underlying physics equations at interactive rates is computationally challenging In this dissertation we propose new algorithms that allow for interactive modeling of chemical structures We first present a modeling tool to construct structural models of hydrocarbon systems The physically based feedbacks are based on the Brenner potential In order to be able to interactively edit systems containing numerous atoms we introduce a new adaptive simulation algorithm Then we introduce what we believe to be the first interactive quantum chemistry simulation algorithm at the Atom Superposition and Electron Delocalization Molecular Orbital ASED MO level of theory This method is based on the divide and conquer D C approach which we show is accurate and efficient for this non self consistent semi empirical theory We then propose a novel Block Adaptive Quantum Mechanics BAQM approach to interactive quantum chemistry BAQM constrains some nuclei positions and some electronic degrees of freedom on the fly to simplify the simulation Finally we demonstrate several applications including one study of graphane formation interactive simulation for education purposes and virtual prototyping at the atomic scale both on desktop computers and in virtual reality environments

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