

## 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 (stoicheion) 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  $\Delta n_i$  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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**Roman Wölfel**



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## **Table of Contents Algorithms For Chemical Computations**

1. Understanding the eBook Algorithms For Chemical Computations
  - The Rise of Digital Reading Algorithms For Chemical Computations
  - Advantages of eBooks Over Traditional Books
2. Identifying Algorithms For Chemical Computations
  - Exploring Different Genres
  - Considering Fiction vs. Non-Fiction
  - Determining Your Reading Goals
3. Choosing the Right eBook Platform
  - Popular eBook Platforms
  - Features to Look for in an Algorithms For Chemical Computations
  - User-Friendly Interface
4. Exploring eBook Recommendations from Algorithms For Chemical Computations
  - Personalized Recommendations
  - Algorithms For Chemical Computations User Reviews and Ratings
  - Algorithms For Chemical Computations and Bestseller Lists
5. Accessing Algorithms For Chemical Computations Free and Paid eBooks

- Algorithms For Chemical Computations Public Domain eBooks
  - Algorithms For Chemical Computations eBook Subscription Services
  - Algorithms For Chemical Computations Budget-Friendly Options
6. Navigating Algorithms For Chemical Computations eBook Formats
    - ePub, PDF, MOBI, and More
    - Algorithms For Chemical Computations Compatibility with Devices
    - Algorithms For Chemical Computations Enhanced eBook Features
  7. Enhancing Your Reading Experience
    - Adjustable Fonts and Text Sizes of Algorithms For Chemical Computations
    - Highlighting and Note-Taking Algorithms For Chemical Computations
    - Interactive Elements Algorithms For Chemical Computations
  8. Staying Engaged with Algorithms For Chemical Computations
    - Joining Online Reading Communities
    - Participating in Virtual Book Clubs
    - Following Authors and Publishers Algorithms For Chemical Computations
  9. Balancing eBooks and Physical Books Algorithms For Chemical Computations
    - Benefits of a Digital Library
    - Creating a Diverse Reading Collection Algorithms For Chemical Computations
  10. Overcoming Reading Challenges
    - Dealing with Digital Eye Strain
    - Minimizing Distractions
    - Managing Screen Time
  11. Cultivating a Reading Routine Algorithms For Chemical Computations
    - Setting Reading Goals Algorithms For Chemical Computations
    - Carving Out Dedicated Reading Time
  12. Sourcing Reliable Information of Algorithms For Chemical Computations
    - Fact-Checking eBook Content of Algorithms For Chemical Computations
    - Distinguishing Credible Sources
  13. Promoting Lifelong Learning
    - Utilizing eBooks for Skill Development

- Exploring Educational eBooks
14. Embracing eBook Trends
- Integration of Multimedia Elements
  - Interactive and Gamified eBooks

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