Brief Review of the Previous Theoretical and Experimental Works

Since 1911 onward a large number of workers had devoted themselves to the theoretical and experimental developments on the phenomenon of thermal diffusion in a thermal diffusion column. As a result, the literature of the subject has now become so vast that a separate monograph is required to review it properly. Here, an attempt has been made to accumulate some of the essential and important experimental and theoretical works on thermal diffusion column by different authors in different times.

The effect of thermal diffusion in liquid was discovered by Dufour[1)] and Soret[2)] in the middle of the 19th century. But thermal diffusion in gases was theoretically predicted in 1911 by Enskog[3)] and Chapman[4)]. In 1917 Chapman and Dootson[5)] established the existence of thermal diffusion in gases with H_2-CO_2 gas mixture. Interests on thermal diffusion was largely increased when Clusius and Dickel[6)] invented thermal diffusion column (TDC) in which the elementary effect of separation could be multiplied in a simpler way. This discovery, however, made it possible to enrich rare isotopes in amount of gram per day in multistage installation of TD columns called cascade designed by Väsaru et al[7)].

The analysing technique of the two bulb apparatus were much more refined during the middle of the twentith century. With a two bulb apparatus designed by Mason[8)] it becomes possible to get reliable results of thermal diffusion coefficients from the rate of approach to the steady state. In 1955 Clusius and Huber[9)] have invented the swing separator in which the elementary effects of thermal diffusion can be multiplied with a known factor. However, to avoid errors much care might be taken in such measurements.

Thermal diffusion column on the other hand is easy to construct and can produce enriched isotopes in a relatively short time. With special techniques Clusius[10)] had produced highly enriched rare stable isotopes by TDC. The most striking theoretical work on thermal diffusion column was done by Furry, Jones and Onsager[11)], Furry and Jones[12)] and Jones and Furry[13)] in their classic papers. Subsequently the theory was extended by different authors [14-15)]. But only a qualitative agreement with the experimental data was observed.

Meanwhile using spherically symmetric intermolecular potential, Chapman and Enskog[16] succeeded very well in predicting the transport properties of the symmetrical molecules. Wang Chang, Uhlenbeck and de Boer[17] developed the formal kinetic theory in order to include the existence of internal degrees of freedom and inelastic collisions among molecules. Monchick, Yun and Mason[18] extended this theory to include the binary molecular interactions. In spite of its great theoretical significance, these advances failed to put forth the required improvement. Van de Ree[19] made an attempt to give a reason for such theoretical failure and contributed a more adequate calculation of the thermal diffusion factor of the asymmetric molecules.

Grew and Ibbs[20] had attempted to develop the theory of separation in a TD column. Still the theory of separation by the TD column is understood semiquantitatively perhaps due to involvement of both the convective and diffusion flow between the column surfaces. Further, discrepancies are caused due to deviation of the experimental column from the idealised one. Moran and Watson[21], Corbett and Watson[22] had however, shown that many of the discrepancies disappeared in a carefully constructed column.

The characteristics of separation of the gas mixtures like Ne-Ar, He-Ar etc. in a thermal diffusion column was investigated by Saxena and Saxena[23], Raman and Saxena[24] had computed the thermal diffusion column shape factors to explain the proper behaviour of gas mixtures in a TD column. Rutherford et al [25,26] had constructed a large number of TDC with various column geometries to enrich rare isotopic binary molecular mixtures. Many precautions had been taken into account in performing their experiments so that preliminary experimental errors could be removed. Column experiments were performed with different binary monatomic or polyatomic gas mixtures like He, Ne, Ar, Kr, CO, CH_4, N_2, CH_3Cl etc. The experimental results as obtained are of much importance to those who are investigating thermal diffusion column behaviours.

Saviron et al[27] had tried to give a simplified formulation for the steady state maximum separation factor and optimum pressure of a TD column. A flow pattern weighted average temperature as a reference temperature is introduced. Starting from the classical FJO theory and introducing the reference temperature attempts have been made to obtain the formulation in which the influence of the nature of the gas appears only through transport coefficients. A proportionality relationship of the logarithmic of maximum separation factor $\ln q_{max}$ with the thermal diffusion factor α_T of the mixture is obtained. The proportionality constant is supposed to be molecular model independent parameter.

The influence of thermodynamic parameter of any binary gas mixture in a TDC, the geometry of TDC on the degree of separation and the energy consumption of a gas in a TDC is interestingly investigated by Leyarovski et al[28]. To explain the gas behaviour under the influence of temperature gradient in a TD column an effective transfer unit height (TUH) is introduced. Evaluation of the degree of enrichment of the heavy to the light gas molecules of the mixture is made possible only if the initial concentration, the geometry of the column, the cold wall and the hot wall temperatures of the TD column are known.

An approximate formulation for the desired column constants are, however, derived by Yamamoto et al[29]. The constants are expressed explicitly in terms of the geometry of the column and physical properties of the gases to be separated. Attempts are made to estimate the optimum pressure of a gas mixture in the TD column. Theoretical predictions as obtained are systematically compared with experimental results for binary, multicomponent monatomic and for isotopically substituted polyatomic gas mixtures. Moreover, optimum pressures were studied for H_2-HT, D_2-HT binary molecular mixture where the mass difference between the components are practically nil. It is found that even for mixture of unlike gases or systems of low atomic weights the predicted optimum pressure are in good agreement with the experimental data.

The theory actually advanced by Furry-Jones and Onsager[11] was later on developed by different authors in different times. The theories are, however, useful for roughly predicting the column behaviour. The analysis is not so simple and it requires numerical calculations with computer.

Under such context, Acharyya et al[30] and Datta et al[31] introduced a column calibration factor F_s so that the thermal diffusion column behaviour can be understood in a simpler way. Thermal diffusion factors α_T of different isotopic, nonisotopic and monatomic molecules were evaluated by the CCF method and compared with experimental α_T by the existing methods. Role of the CCF in determining the temperature or composition dependence of α_T are carefully examined by Acharyya et al[30] and Datta et al[31]. The reliable α_T's for He - HT, He-HD, Ne, Ar, Kr, Xe isotopic mixtures and other like or unlike molecules are obtained. The application of the CCF in thermal diffusion column and its wide significance inspired the present author[32] to derive the functional relationship of F_s with the geometry of the TD column from the Navier-Stokes hydrodynamical equation.

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