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Two-dimensional diffusion in a Stefan tube: The classical approach

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HIGHLIGHTS

- ► Classical approach is used to model two-dimensional mass transport in a Stefan tube.
- ► The Navier–Stokes equations are solved numerically using a finite difference method.
- ► A diffusion creep (slip) boundary condition is used on the side walls.
- ► Comparison with a published solution using the Kerkhof–Geboers transport equations.
- ► Difficulties with the Kerkhof–Geboers solution are discussed.

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ABSTRACT

Contrary to claims made in the recent literature, we show that the classical approach to mass transport in gas mixtures can be used to model two-dimensional Stefan tube diffusion. Numerical solutions are obtained for water evaporating into air, using the Kramers–Kistemaker diffusion slip velocity as the boundary condition on the side walls. A comparison is made with the analysis of Salcedo-Diaz et al. (2008 Velocity profiles and circulation in Stefan-diffusion. Chem. Eng. Sci. 63, 4685–4693), in which the classical approach is rejected in favor of the Kerkhof–Geboers equations. Difficulties inherent in this latter approach are discussed in detail.

1. Introduction

Salcedo-Diaz et al. (2008) have revisited the familiar Stefan-tube diffusion problem as a showcase for demonstrating the use of the Kerkhof–Geboers “Unified Theory of Isotropic Molecular Transport Phenomena” (Kerkhof and Geboers, 2005a,b). In the 2005 papers, one-dimensional applications were demonstrated: the objective of the 2008 paper was to obtain two-dimensional solutions for the Stefan-tube problem, and thereby evaluate the adequacy of the well known one-dimensional solution for diffusion with one component stationary. In addition, some emphasis is given to use of the model results to illuminate the essential physics of the problem. Results are presented for continuum conditions, as well as for a Knudsen number ~ 3. In this paper our focus is on the continuum limit. In order to provide a rationale for the use of the Kerkhof–Geboers theory, Salcedo-Diaz et al. (2008) argued that the “classical approach” cannot be used for this (and some other) diffusion problems: thus it is appropriate that we first critically examine the reasons given for this rejection.

Salcedo-Diaz et al. (2008) describe the classical approach as one in which the mixture motion is described by the Navier–Stokes equations, and species transport by $(n-1)$ Maxwell–Stefan equations. For a binary mixture, species transport is then described by a single convection–diffusion equation in terms of Fick’s law. They claim that the classical approach cannot hold for the Stefan tube problem and refer to a “thought experiment” described in an earlier paper (Kerkhof and Geboers, 2005a). This thought experiment concerns isobaric diffusion as experimentally investigated by Graham (1833), Remick and Geankoplis (1973), and others. In attempting to obtain a solution using classical theory, they integrate the momentum equation to show that the mass average axial velocity is constant across the tube cross-section. They then apply a no-slip wall boundary condition to conclude that the axial velocity is zero across the tube in violation of Graham’s law. This use of the no-slip boundary condition is quite incorrect. It is most puzzling why the authors did so, given that their literature citations contain key papers that show the no-slip boundary condition to be invalid when there is an axial wall concentration gradient. The correct boundary condition is the diffusion slip (creep)

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velocity. The diffusion slip velocity was first derived by Kramers and Kistemaker (1943) in connection with equimolar counter-diffusion, and subsequently used by Hoogschagen (1955) and Kruger (1976) for isobaric diffusion. Kerkhof and Geboers (2005a) also cite the monograph by Jackson (1977) in which the diffusion slip velocity is again derived. Interestingly, Jackson states clearly that use of the no-slip boundary condition can lead to substantial errors in the fluxes, and "Nevertheless this inappropriate condition is still invoked in solving diffusional problems." In recent years rigorous kinetic theory models have been used to obtain improved data for the diffusion slip velocity: Mills (2007) presents a review of this work and its implications.

In what follows we will first demonstrate the use of the classical approach for the two-dimensional Stefan-tube problem. Subsequently we will make comparisons with the results obtained by Salcedo-Diaz et al. (2008) using the Kerkhof–Geboers theory.

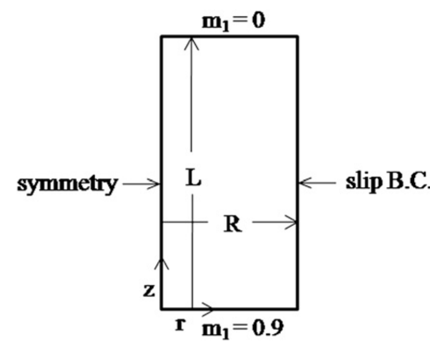

Fig. 1. Schematic of the Stefan tube.

2. Classical analysis of the Stefan-tube

2.1. Prior work

One-dimensional analysis of the Stefan-tube (and related heat-pipe problem) is found in standard texts and generally referred to as "diffusion with one component stationary". The Stefan tube has been used for measurement of diffusion coefficients in vapor-gas mixtures for many years. Early on, there was concern that the 1-D analysis might not be adequate due to end effects (Lee and Wilke, 1954; Heinzelmann et al., 1965). Thus there was the motivation to explore 2-D effects. With advances in numerical methods, the required solution of the governing elliptic nonlinear partial differential equations became feasible in the late 1960s: 2-D results for the related heat-pipe problem were obtained by McDonald et al. (1971) and for the Stefan-tube by Meyer and Kostin (1975). However, in both studies the no-slip boundary condition was used on the tube walls because these workers were unaware of the significance of diffusion slip. Nevertheless, the results were useful in showing that the gas circulated, rather than being stationary, and that the 2-D flow features did not have a significant effect on the radial concentration profiles and evaporation rates.

In the case of water evaporating into air, the diffusion slip velocity is directed toward the water surface and is at most about 25% of the bulk velocity (Mills, 2007). Accounting for the diffusion slip velocity rather than using a no-slip condition is not expected to substantially alter the conclusion of McDonald et al. (1971) that 2-D features have but a small effect on the evaporation rate. Nevertheless, in order to counter the claims of Kerkhof and Geboers (2005a), we have obtained numerical solutions for the 2-D continuum problem.

2.2. Analysis

The governing conservation equations for isothermal, steady flow of an incompressible binary ideal gas mixture, in the absence of body forces and chemical reacting, are written in cylindrical coordinates with axial symmetry. For the coordinate system shown in Fig. 1,

mass:

$$\frac{\partial}{\partial r}(\rho r v_r)+r\frac{\partial}{\partial z}(\rho v_z)=0 \tag{1}$$

radial momentum:

$$\rho\left(v_r\frac{\partial v_r}{\partial r}+v_z\frac{\partial v_r}{\partial z}\right)=-\frac{\partial p}{\partial r}+\frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rr})+\frac{\partial \tau_{rz}}{\partial z} \tag{2}$$

where

$$\tau_{rr}=\mu\left(2\frac{\partial v_r}{\partial r}-\frac{2}{3}\nabla\cdot v\right) \tag{3a}$$

$$\nabla\cdot v=\frac{1}{r}\frac{\partial}{\partial r}(r v_r)+\frac{\partial v_z}{\partial z} \tag{3b}$$

$$\tau_{rz}=\mu\left(\frac{\partial v_z}{\partial r}+\frac{\partial v_r}{\partial z}\right) \tag{3c}$$

axial momentum:

$$\rho\left(v_r\frac{\partial v_z}{\partial r}+v_z\frac{\partial v_z}{\partial z}\right)=-\frac{\partial p}{\partial z}+\frac{1}{r}\frac{\partial}{\partial r}(r\tau_{rz})+\frac{\partial \tau_{zz}}{\partial z} \tag{4}$$

where

$$\tau_{zz}=\mu\left(2\frac{\partial v_z}{\partial z}-\frac{2}{3}\nabla\cdot v\right) \tag{5}$$

species conservation:

$$\rho\left(v_r\frac{\partial m_1}{\partial r}+v_z\frac{\partial m_1}{\partial z}\right)=\frac{1}{r}\frac{\partial}{\partial r}\left(r\rho\mathcal{D}_{12}\frac{\partial m_1}{\partial r}\right)+\frac{\partial}{\partial z}\left(\rho\mathcal{D}_{12}\frac{\partial m_1}{\partial z}\right) \tag{6}$$

for negligible pressure diffusion. The boundary conditions imposed on these equations are

$r=0$:

$$v_r=0;\quad \frac{\partial v_z}{\partial r}=0 \tag{6a, b}$$

$r=a$:

$$v_r=0;\quad v_z=v_w \tag{7a, b}$$

$z=0$:

$$v_r=0;\quad m_1=m_{1,s};\quad v_z=-\frac{\mathcal{D}_{12}}{1-m_{1,s}}\left.\frac{\partial m_1}{\partial z}\right|_0 \tag{8a, b, c}$$

$z=L$:

$$\frac{\partial v_z}{\partial z}=0;\quad m_1=0 \tag{9a, b}$$

Also a total pressure is specified at $r=0$, $z=L$. Eq. (7b) requires a specification of the diffusion slip (creep) velocity; using the Kramers and Kistemaker (1943) model

$$v_w=\frac{\left(1/M_1^{1/2}\right)-\left(1/M_2^{1/2}\right)}{\left(m_1/M_1^{1/2}\right)+\left(m_2/M_2^{1/2}\right)}\mathcal{D}_{12}\frac{\partial m_1}{\partial z} \tag{10}$$

Mills (2007) has evaluated the accuracy of this model using experimental data and recent rigorous kinetic theory results and found that the model gives reasonable results, particularly when the molecular weights of the two species are substantially different: for our present purpose Eq. (10) is quite adequate. Eq. (8c) expresses