HIGH PRESSURE, TRANSPORT PROPERTIES OF FLUIDS: THEORY AND DATA FROM LEVITATED FLUID-DROPS AT COMBUSTION-RELEVANT TEMPERATURES

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OBJECTIVES

The objective of this investigation is to derive a set of consistent mixing rules for calculating diffusivities and thermal diffusion factors over a thermodynamic regime encompassing the subcritical and supercritical ranges. These should serve for modeling purposes, and therefore for accurate simulations of high pressure phenomena such as fluid disintegration, turbulent flows and sprays. A particular consequence of this work will be the determination of effective Lewis numbers for supercritical conditions, thus enabling the examination of the relative importance of heat and mass transfer at supercritical pressures.

TECHNICAL DESCRIPTION

This investigation addresses some crucial issues in transport property calculations that lie at the core of understanding supercritical fluid behavior for combustion applications. Although the high pressure multicomponent fluid conservation equations have already been derived and approximately validated for binary mixtures by this PI (see Figs. 1 and 2 below, and Harstad and Bellan, 2000), the validation of the multicomponent theory is hampered by the lack of existing mixing rules for property calculations. Classical gas dynamics theory can provide property mixing rules at low pressures exclusively. While thermal conductivity and viscosity high-pressure mixing rules have been documented in the literature, there is no such equivalent for the diffusion coefficients and the thermal diffusion factors.

Therefore, the primary goal of this investigation is to extend the low pressure mixing rule theory to high pressures and validate the new theory with experimental data from levitated single drops. The two properties that will be addressed are the diffusion coefficients and the thermal diffusion factors. To validate the property calculations, ground-based experiments from levitated drops will be conducted. The levitated drops will be obtained using an electrostatic technique having the particular advantage of creating nearly spherical drops whose internal flow is minimized. The apparatus is sketched in Fig. 3. Additionally, it is planned to build a laboratory mini drop tower so as to allow the performance of initial microgravity experiments without traveling to NASA facilities; thus, saving time and expense. More importantly, this mini drop tower will allow the performance of time consuming on site calibrations which are necessary until a standard experimental procedure is established. The primary output of the experimental data will be the measurement of the drop diameter which will then be compared with the theoretical predictions.

Part of the database will be used to construct the mixing rule models while the remaining part will be used to validate the derived models. Such a strategy was already used by the PI in comparing theoretical predictions of free-floating drops with the suspended drop experiments of

Nomura et al., 1996. While these comparisons were promising (see Harstad and Bellan, 2000), they embedded the uncertainties of comparing numerical results from free drops with data from suspended drops. This uncertainty will be mitigated by the present experimental technique.

The regime of investigation will include subcritical as well as supercritical pressures at combustion relevant temperatures. The focus will be on evaporation/mass-emission experiments and on drop sizes varying from the large ones used in suspended drop experiments to small ones of size relevant to combustion engines. This range of parameters will bridge the gap between the current high pressure microgravity data which encompasses only combustion experiments for which there are no measurements of the flame temperature, and the data needed for model validation which requires the knowledge of drop effective-far-field conditions. The levitated drop experiments may also allow the utilization of drops that are about one order of magnitude smaller than those employed in suspended drop experiments, thereby further reducing buoyancy as well as making the range of parameters more relevant to combustion devices.

This study is in the spirit of a short exploratory program, focusing exclusively on binary mixtures. The plan of this exploratory investigation is to accomplish the following:

1. Accurate binary diffusion coefficients calculations – monoatomic species.

A formalism will be derived for the binary diffusion coefficients at all pressures. Calculations will be first performed for monoatomic compounds to obtain a baseline verification of our formalism; the predicted results will be compared with experimental measurements (Balenovic et al., 1970) for self diffusion in Ar, and binary diffusion in Ar-Kr, Ar-He and He-Ar. One of the important issues to be determined is the meaning of the quantities experimentally measured: (i) the diffusion coefficient, **or** (ii) the product of the diffusion coefficient by the mass diffusion factor (i.e. the effective coefficient of the Fick term). This issue is crucial at high pressures since mixtures show increasing non-ideality with increasing pressure, reducing the mass diffusion factor below the unity value characteristic of ideal mixtures at low pressures. Part of this study will include an exploration of the sensitivity of the prediction of the drop diameter to the uncertainties in the diffusion coefficient.

2. Accurate binary diffusion coefficients calculations – general species

The formalism derived in the first year of study will be here applied to a variety of binary systems such as: He-N_2, Ar-N_2, N_2-He, N_2-H_2, He-CH_4, He-C_2H_6 and He-C_3H_8. Experimental data for verification exists for all these mixtures (Balenovic et al., 1970). Additionally, there is also experimental data quantifying the self diffusion in CO_2, CH_4, C_2H_4 and H_2O (Liu and Macedo, 1995). Maynard and Grushka, 1975, present extensive tables of measured (using gas-chromatography broadening techniques) infinite dilution binary diffusion coefficients, some at high pressures; comparisons with this data will be performed as well. Recently, binary diffusion coefficients of naphthalene and biphenyl in CO_2 at infinite dilution at 330 K and as a function of the CO_2 density were published by Schneider, 1998. The diffusion coefficient of toluene in supercritical CO_2 was plotted also as a function of the CO_2 density by Bruno, 1993. During the course of this work, EOSs for all these compounds will be determined by using either data, or our own method for obtaining computationally simple, yet accurate EOSs (Harstad et al., 1997), or cubic EOSs, as appropriate. At the suggestion of the reviewers, part of this study will include an exploration of the sensitivity of the prediction of the drop diameter to the uncertainties in the diffusion coefficient.

3. Thermal diffusion factors for binary mixtures

During the last part of this exploratory study, the fluid drop model of Harstad and Bellan, 2000, embedding the validated diffusion coefficient model determined in the first two years of this investigation, will be applied to some binary mixtures of interest to combustion science. Calculations will be performed for hydrocarbon fluid-drops in nitrogen, and the calculated drop boundary as function of time will be compared to that measured during the levitated drop experiments conducted by the co-PI. The value of the thermal diffusion factor will be backed-up from this comparison by performing calculations for several values of the thermal diffusion factor and observing which one matches better the experiment. The experimental margin of error (error bars) will be transferred by the model into error bars on the determination of the thermal diffusion factor. Part of the data will be used for determining the value of the thermal diffusion factor, and the remaining data will be used for model validation. Because of this strategy, coordination with the experiment will be initiated as soon as the feasibility of the experiment has been ascertained (in the previous years of this exploratory study) to ensure that data are obtained at conditions encompassing both the determination and the validation aspects.

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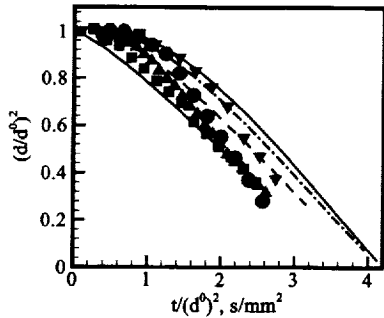
Figure 1. Comparison of moderate (~650K) temperature model predictions of Harstad and Bellan, 2000, with the data of Nomura et al., 1996. Details in Harstad and Bellan 2000.
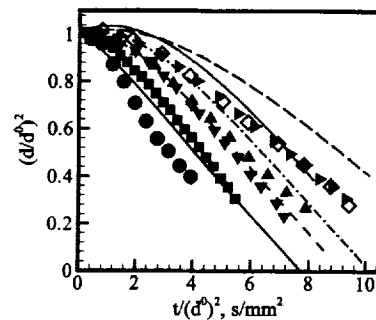
Figure 2. Comparison of low (~450K) temperature model predictions of Harstad and Bellan, 2000 with the data Nomura et al., 1996. Details in Harstad and Bellan, 2000.
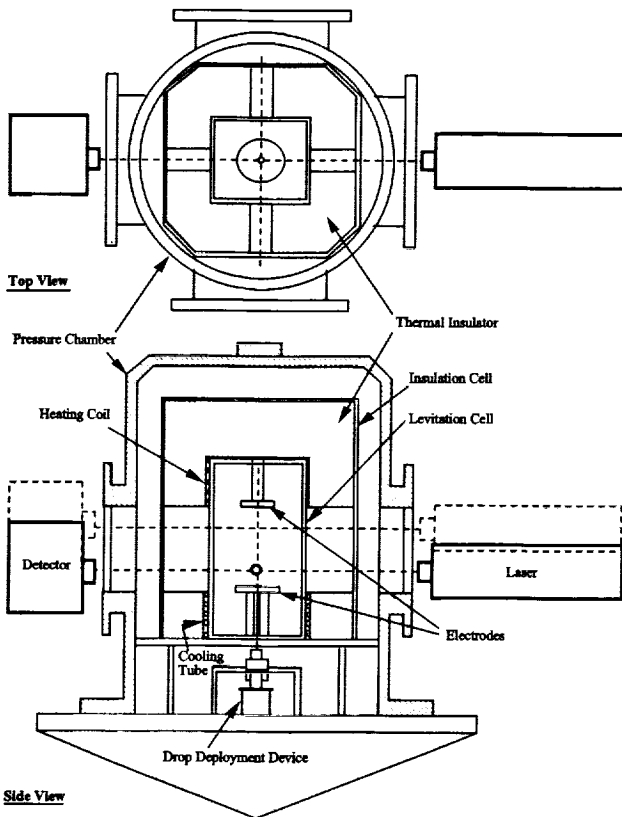

Figure 3. Schematics of the apparatus. A pair of electrodes is placed inside of the levitation cell made from quartz glass. The cell has a square cross section (as shown in the top view) for an undistorted view of a levitated drop. The top and bottom portions of the cell are wound by a heating coil and a cooling tube, respectively. These elements are used to create temperature gradients within the cell along the vertical axis at elevated temperatures. The cell is covered by thermal insulators except for 4 viewing ports. A drop deployment device is located under the levitation cell. The device consists of a syringe with a fine needle and two linear actuators. The drop deployment device and the insulation cell which houses the levitation cell are placed inside of a pressure chamber. A laser source and a detector are not a part of the dropping module but they are depicted here to show their functions.