

NASA CR 71313

NASA Grant NGR-21-003-053

Semiannual Status Report No. 1

July 1, 1965 to December 31, 1965

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University of Maryland
College Park, Maryland

GPO PRICE \$ _____

CFSTI PRICE(S) \$ _____

Hard copy (HC) \$ 1100

Microfiche (MF) 150

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853 July 65

FACILITY FORM 602

N66 23775

(ACCESSION NUMBER)

(THRU)

(PAGES)

(CODE)

CR 71313

18

(NASA CR OR TMX OR AD NUMBER)

(CATEGORY)

Statement of Work.

The purpose of this program is to compile and critically evaluate diffusivity and viscosity data on gas, liquid, solid and turbulent flow systems. The program was begun during the past six months. Progress in the various areas of the program is discussed separately in the following sections.

(a) Diffusion in Gases.

The objectives during the last six months were: (1) to prepare a comprehensive literature survey of experimental measurements of dilute gas binary diffusion coefficients; and, (2) to initiate a program for the evaluation and correlation of diffusion measurements. The literature search is essentially complete. Diffusion coefficients of the best-known binary systems, helium-argon, are being correlated. This review of He-Ar data indicates that the correlated experimental results will be satisfactory for a reference system.

The literature survey obtained copies of published papers beginning with the earliest useful diffusion measurements (1870). Approximately 200 references have been obtained. It remains to complete the cross-checking of references cited within each paper and to continue the retrieval of new diffusion measurements. An annotated bibliography will be prepared.

The diffusion measurements for helium-argon are the most comprehensive of all binary gas systems. Published data from 1904 (Schmidt) to 1966 (Malinauskas) are available. A total of 15 scientists have measured the diffusion coefficient (D_{Ar-He})

at low pressures, approximately 1 atm. The direct measurements are between the temperatures of 273° and 1063°K. Indirect measurements involving mixture viscosity, thermal diffusion, and molecular beam scattering extend the available temperature range over which $D_{\text{He-Ar}}$ can be obtained down to 72°K and up to 15,000°K.

A graph of $\log D_{\text{Ar-He}}$ versus $\log T$ (72°K- T - 15,000°K) indicated that a correlation of the form $D = aT^5$ was most applicable. Since measurements had been made at various compositions, the experimental values of $D_{\text{He-Ar}}$ were corrected to a fixed composition by a semi-empirical correlation.¹ At a maximum the effect 4% between a helium trace in argon and an argon trace in helium. The data were reduced to argon trace composition. The diffusion data are now being least-squared with various weights and in various temperature ranges. This numerical experimentation is designed to give us some estimate of the reliability and sensitivity to error of our final summary of all measurements.

A theoretical investigation of higher kinetic theory approximations has been made to assess the error involved in calculating D_{12} and to help decide what composition such a calculated D_{12} refers to.² The results are very encouraging.

When the helium-argon system is finished, we will proceed to other binary systems for which large quantities of data exist.

¹Mason, E. A., S. Weissman and R. P. Wendt, "Composition Dependence of Gaseous Thermal Diffusion Factors and Mutual Diffusion Coefficients," *Physics of Fluids*, **7**, (2), 174-179 (1964).

²Storvick, T. S. and E. A. Mason, to be published.

Liquids.

Data on the liquid phase molecular transport coefficients is being compiled and evaluation of some systems will begin shortly. Both aqueous and non-aqueous solvent systems with ionizing solutes are included in the program. Excluded initially from this compilation and evaluation survey are those systems in which the solvent or the solute represents a heterogeneous chemical species that can not be readily characterized on the basis of pure component properties--for example, a liquid tar or broad spectrum liquid petroleum of generally variable composition and properties would not be included, but a binary, ternary or multicomponent mixture (either solvent or solute) of pure components with a definable composition is to be included.

For the purpose of providing a convenient format for the initial characterization of liquid systems, the following classification system has been tentatively adopted:

- I. Polar Solvents with Ionizing or Non-Ionizing Solutes.
- II. Non-Polar Organic Solvents with Ionizing or Non-Ionizing Solutes.
- III. Inorganic and Liquid Metal Solvents with Organic or Inorganic Solutes.

The general plan of operation is as follows: 1) establish the defining bounds and the inter-relationships of the various molecular transport coefficients and classify the prevailing experimental techniques generally used; 2) survey the literature to determine the available information and decide the most

appropriate informational format for the compilation and presentation of the information; and (3) critically evaluate the collated information.

In the initial phase of the project the literature has been surveyed to permit a general classification and delineation of the experimental techniques and the theories of liquid phase molecular transport have been reviewed and collated to enable the definition of the various possible transport coefficients---integral, differential, etc.

Because of the large scope of each of the above areas, the initial phases of the literature survey have been directed primarily at the last five years (1960-1965). One of the reasons for this initial limitation was that this period would afford a barometer for the sources of current information and serve as a guide for continuing efforts; also, the current literature is least likely to be available in compendiums available to researchers in the field. Some of the more significant earlier collections have also been included in these initial compilations.

Turbulent Transport Coefficients.

Data on the transport of chemical species, heat, and momentum in turbulent flow fields is being compiled and evaluated. Both confined and free flow fields are being surveyed. Some of the types of transport in confined flow are: heat, mass and momentum transport in conduits; porous media, and boundary layers. Free or unconfined flow includes; jets and streams issuing into

large bodies; meteorological, and oceanographic flows.

Only systems for which the molecular transport coefficients are well established are being considered. Excluded from the survey are: non-Newtonian fluids; systems at extremely high or low temperatures; magneto hydrodynamic systems; and, very low pressure gases. The major emphasis at present is on subsonic turbulent flow of gases and liquids. Some information is becoming available on turbulence in supersonic flow of gases. As more information becomes available super-sonic flow fields will be given increased consideration.

The plan of operation follows a three step sequence: (1) define the quantities to be measured and the interrelationship between the basic turbulent properties and the turbulent transport; (2) survey the literature in each of the above areas to determine what information and data are available; (3) evaluate the data and present it in convenient form for use by workers in the field.

In the initial phase of the project the literature survey has been directed toward obtaining a comprehensive coverage of the above areas rather than an all inclusive survey. In this regard attention has been directed primarily to the literature of the past five years.

It has been our experience that the nomenclature and methods of analysis in the various areas are sufficiently different to warrant the preparation of a brief statement that outlines the subject and provides a means of unifying and integrating the

data to a common format. Such a statement has been drafted and will be included in the annual report along with the data obtained from the literature survey.

Solids and Penetrant - Polymers Systems.

Work on these areas was not begun during the first six months of the program. The main reason for lack of activity in the solids area was that Professor Schamp was appointed Dean of Faculty at the new Baltimore Campus of the University of Maryland.

Personnel.

The investigation of diffusion in gases is directed by Dr. E. A. Mason. He has been assisted by one graduate student, Mr. T. R. Marrero. The liquid diffusion survey is directed by Dr. R. B. Beckmann, with the assistance of two graduate students, Mr. W. B. Ellis and Mr. P. N. Vashist. Dr. J. M. Marchello is directing the turbulent transport survey and was assisted by one graduate student, Mr. S. D. Cramer.

Mr. Cramer and Mr. Ellis leave the project in February and will be replaced with other graduate students.