Properties of Helium, Nitrogen, and He–N₂ Binary Gas Mixtures

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An extensive review is conducted and a comprehensive database is compiled for the transport and thermodynamic properties of helium and nitrogen and their binary mixtures at pressures up to 40 MPa and temperatures from 200 to 1500 K. These gases are attractive working fluids in a closed Brayton cycle for energy conversion in space reactor power systems and terrestrial power plants. Semi-empirical correlations are developed for calculating the properties of the He–N₂ binary mixtures based on the Chapman–Enskog kinetic theory for dilute gases and the application of the law of corresponding states. The correlations accurately account for the effects of pressure and temperature and are in excellent agreement with the compiled properties database.

Nomenclature

A_{12}^*	=	ratio of collision integrals, $\Omega^{(2,2)*}/\Omega^{(1,1)*}$
a_k	=	polynomial coefficients
B, B_{ii}	=	second virial coefficient of pure gas, m³/mol
$\bar{B}$	=	second virial coefficient of the gas mixture, m³/mol
B_{ij}	=	second virial interaction coefficient ($i \neq j$), m³/mol
B_r	=	reduced second virial coefficient, B/V^*
B_{12}^*	=	ratio of collision integrals, $[5\Omega^{(1,2)*} - 4\Omega^{(1,3)*}]/\Omega^{(1,1)*}$
C, C_{iii}	=	third virial coefficient of pure gas, m⁶/mol²
$\bar{C}$	=	third virial coefficient of the gas mixture, m⁶/mol²
C_{ijj}	=	third virial interaction coefficient ($i \neq j$), m⁶/mol²
$\hat{C}_P$	=	molar specific heat at constant pressure, J/mol · K
C_r	=	reduced third virial coefficient (C/V^{*2})
$\hat{C}_v$	=	molar specific heat at constant volume, J/mol · K
c	=	speed of sound, m/s
f_{12}	=	higher-order correction factor
$\hat{H}$	=	molar enthalpy, J/mol
k	=	Boltzmann constant, 1.380662×10^{-23} J/K
M	=	molecular weight, kg/mol
m	=	mass of one gas molecule (M/N_A), kg
N	=	number of components in a gas mixture
N_A	=	Avogadro number, 6.022045×10^{23} mol⁻¹
P	=	pressure, Pa
R_g	=	perfect gas constant, 8.31441 J/mol · K
$\hat{S}$	=	molar entropy, J/mol · K
T	=	temperature, K
T_{cr}, T_{ii}^*	=	critical temperature of pure gas, K
T_o	=	reference temperature, K
$\bar{T}^*$	=	pseudocritical temperature, K
T_{ij}^*	=	interaction temperature, $i \neq j$, K
$\hat{U}$	=	molar internal energy, J/mol
$\bar{V}^*$	=	characteristic molar volume of the gas mixture, m³/mol
V_{ii}^*	=	characteristic molar volume of pure gas $[V^* = R_g (T_{cr}/P_{cr})_i]$, m³/mol
V_{ij}^*	=	characteristic interaction molar volume ($i \neq j$), m³/mol
x_i	=	molar fraction of component i in the gas mixture
Z	=	compressibility factor
$\Delta\lambda$	=	excess conductivity ($\lambda(T, P) - \lambda^o(T)$), W/m · K
$\Delta\lambda_r$	=	normalized excess conductivity, $\Delta\lambda/(\lambda_{cr} - \lambda^o(T_{cr}))$
$\Delta\mu$	=	excess viscosity ($\mu(T, P) - \mu^o(T)$), Pa · s
$\Delta\mu_r$	=	normalized excess viscosity, $\Delta\mu/(\mu_{cr} - \mu^o(T_{cr}))$
ε_{ij}	=	depth of molecular potential well, J
θ	=	reduced temperature, (T/T_{cr})
λ	=	thermal conductivity, W/m · K
$\bar{\lambda}^*$	=	pseudocritical conductivity, W/m · K
μ	=	dynamic viscosity, Pa · s
$\bar{\mu}^*$	=	pseudocritical viscosity, Pa · s
v	=	gas molar volume, m³/mol
ρ	=	density, kg/m³
$\hat{\rho}$	=	molar density (ρ/M), mol/m³
ρ_r	=	reduced density (ρ/ρ_{cr})
σ_{ij}	=	distance for which the molecular potential is zero, m
Ψ	=	dimensionless function of reduced density
$\Omega^{(k,l)*}$	=	dimensionless collision integral

Subscripts

cr	=	critical
exp	=	experimental
o	=	dilute-density value, reference value at 0.1 MPa
r	=	reduced (dimensionless)
λ	=	thermal conductivity
μ	=	dynamic viscosity
1	=	heavier gas component in mixture (nitrogen)
2	=	lighter gas component in mixture (helium)

Superscripts

o	=	dilute-density value, reference value at 0.1 MPa
‾	=	gas-mixture property
*	=	critical or pseudocritical

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I.   Introduction

NOBLE gases and binary mixtures are being considered as working fluids for nuclear reactor power systems employing a closed Brayton cycle (CBC) for energy conversion. The operating pressure in space nuclear reactor power systems is ∼2.0 MPa versus 3.0–10 MPa for terrestrial nuclear power plants. The temperatures at the exit of the nuclear reactor and the inlet of the CBC compressor in space and terrestrial power applications are typically 1200–1400 K and 300–500 K, respectively.

The binary mixtures of He with krypton (Kr) and xenon (Xe) up to a molecular weight of 22 and 40 g/mol, respectively, have higher heat transfer coefficients than pure helium and significantly decrease the aerodynamic loading and, in turn, the size and mass of the CBC turbomachines [1]. A He–Xe binary mixture with a molecular weight

$M = 40$ g/mol is the working fluid of choice in space reactor power systems. This is because it reduces the size and mass of the turbomachines, despite the increase in the CBC pressure losses, which decreases the overall system efficiency by a few percentage points [1,2]. This binary mixture is not an attractive working fluid for terrestrial nuclear power plants in which the gas-cooled reactor with a thermal neutron spectrum is directly coupled to the CBC. The high neutron-capture cross section of xenon not only reduces the initial excess reactivity in the reactor, shortening the length of the operation cycle, but also generates radioactive decay species that would activate the structure and turbomachines.

Recent analyses [2,3] have shown that helium is relatively transparent to neutron irradiation. By contrast, the isotopes of $_{131}$Xe (21.1 at.% abundance) and $_{129}$Xe (26.4 at.% abundance) with thermal neutron-absorption cross sections of 110 and 21 barns and the $_{135}$Xe produced as a result of irradiation in the reactor are sources of high radioactivity. The absorption of neutrons by these Xe isotopes and their transmutation to radioactive nuclides of cesium, barium, iodine, and tellurium in the CBC loop represent a potential gamma radiation hazard to the power plant personnel in the vicinity of unshielded pipes. It is worth noting, however, that the activation of Xe is not much of a concern in space nuclear reactors with fast neutron energy spectra; thus, He–Xe could be used as working fluid in CBCs that are directly coupled to a fast-spectrum nuclear reactor heat source [2].

An alternative binary mixture working fluid is that of helium and nitrogen. Nitrogen is generally chemically inert, experiences little neutron activation in the nuclear reactor, and its use in CBC space power systems and terrestrial power plants can benefit from existing experience with air and nitrogen closed and open Brayton cycles. The $_{14}$N isotope has a thermal neutron-absorption cross section of only 1.8 barns, producing $_{14}$C and tritium, which do not emit gamma rays when decaying [3]. In addition to its abundance, the cost of acquiring nitrogen is significantly lower than noble gases. Nitrogen is produced on a massive scale with very high purity by liquefaction of air and fractional distillation. Therefore, there is a need to survey the literature and compile the existing thermodynamic and transport properties of the binary mixtures of helium and nitrogen to validate the development of reliable correlations for future design and operation optimization of the CBC turbomachines. The transport and thermodynamic properties of these two gases are quite different. In addition, at the pressures and temperatures of interest in space reactor power systems and terrestrial nuclear power plants, the theory of diluted gases based on the Chapman–Enskog kinetic theory is not accurate enough for predicting the gas properties [4]. Helium exhibits strong quantum effects, and the diatomic molecules of nitrogen have two rotational and one vibrational degrees of freedom in addition to the three translation degrees of freedom, making the application of present theories challenging for predicting the transport properties of nitrogen and its binary mixtures with helium.

Recently, accurate semi-empirical correlations for the properties of the 5 noble gases (He, Ne, Ar, Kr, and Xe) and their 10 binary mixtures have been developed [4]. These correlations are based on the Chapman–Enskog kinetic theory for dilute gases [5,6] and the application of the law of corresponding states [7], and they accurately account for the effects of pressure and temperature.

This work extends this approach to develop accurate correlations of the thermodynamic and transport properties of nitrogen and its binary mixtures with helium as functions of pressure and temperature. Helium behaves very differently from a perfect fluid and, like hydrogen, exhibits strong quantum effects. A well-known peculiarity is that the helium viscosity is independent of pressure and/or density, whereas the viscosity of other gases increases with density to various degrees, depending on the molecular weight. As a result, the law of corresponding states applied to the mixtures of the noble gases of Ne, Ar, Kr, and Xe do not apply to helium without some modification. Unlike nitrogen, helium and other noble gases are monoatomic, with only three translation degrees of freedom. These differences cause the molar heat capacity and specific heat ratio of nitrogen and its binary mixtures with helium to vary with temperature (and pressure), even at a dilute density, and they cause

the thermal conductivity to increase above that predicted by the kinetic theory for monoatomic gases.

The semi-empirical correlations developed in this work are validated using an extensive properties database compiled for He and N_2 and their binary mixtures, for temperatures from 200 to 1500 K and pressures up to 40 MPa. The predictions of these correlations are also compared with the values recommended by the National Institute of Standards and Technology (NIST) [8]. The following section reviews the major theoretical approaches used for calculating the transport and thermodynamic properties of monoatomic (e.g., helium) and polyatomic (e.g., nitrogen) gases and their binary mixtures.

II. Chapman–Enskog Kinetic Theory of Dilute Gases

The popular approach for determining the transport properties of dilute monoatomic gases with spherical nonpolar molecules and no internal degrees of freedom uses a perturbation technique to obtain an approximate solution [5] of Boltzmann's equation. This approach [6], developed before World War I by Chapman in England and independently by Enskog in Sweden, assumes that gases and their mixtures are sufficiently dilute, such that the mean free path of the molecules is much larger than their size. Thus, the molecules move freely and interact only through binary collisions, and the effect of the collisions with the wall is neglected.

The thermodynamic state of dilute gases is represented by a virial expansion with up to two coefficients, and the motion of the molecules is described by the classical Newtonian mechanics. The macroscopic behavior of these gases is thus described accurately using a single distribution function in the phase space of pairs of molecules [5], and the equilibrium properties are expressed solely in terms of the pair distribution function. The treatment of the kinetic theory of gases was extended to handle polyatomic molecules or molecules with internal quantum states by writing the collision integrals in terms of the differential cross sections representing the probability of a collision in which two molecules, initially in states i and j, end up in states k and l. Although the viscosity is a weak function of the internal degrees of freedom of the molecules, the thermal conductivity strongly depends on the rotational degrees of freedom, and the theoretical developments lead to the introduction of the Eucken correction [7].

Based on the available measurements of the second virial coefficient, viscosity, and the self- and binary-diffusion coefficients of noble gases and their mixtures, Kestin et al. [9] successfully developed a consistent set of parameters ε_{ij} and σ_{ij} and the universal shapes of the collision integrals to accurately fit the low-density data using the kinetic theory. Najafi et al. [10] and Kestin et al. [11] extended the principle of corresponding states to five parameters and extended the range of applicability from absolute zero temperature to the onset of ionization. The three new parameters characterize the shape of the molecular potential: one for the long-range potential tale and two for the short-range repulsive wall. The values of these parameters are independently available from theory, experiments involving scattering of noble gases in the thermal and high-energy ranges and dielectric and optical data. Using this approach, Kestin et al. [11] achieved remarkable success in predicting the equilibrium and transport properties of noble gases and their binary mixtures at dilute densities over a wide range of temperatures.

The treatment of the kinetic theory has been extended to predicting the equilibrium and transport properties of a number of polyatomic gases at low density, including nitrogen [12–14] and the transport properties (except thermal conductivity) of multicomponent mixtures of 11 polyatomic gases and 5 noble gases [15]. The nonspherical contributions of the polyatomic molecules are accounted for using additional dimensionless parameters for each gas component in the mixtures and mixing rules for the binary interactions. Although extremely useful whenever experimental data are not available, the relationships developed for the gas-mixture properties and the collision integrals [11,15] are cumbersome and complex functions involving many different parameters for each component in the mixture that are often unknown. These models also

neglect the effects of pressure. These limitations have led to the development of the Enskog modified kinetic theory (EMKT) for dense gases, briefly discussed next.

III. Enskog Modified Kinetic Theory for Dense Gases

The EMKT for dense gases [16], an extension of the dilute-gas kinetic theory, assumes that the gas molecules behave as rigid spheres and accounts for the multibody collisions (ternary and above) in the intermediate-density range. For dense gases, the mean free path of the molecules is not much larger than the molecular diameter, and is thus treated by including the momentum transport and the kinetic energy transfer between any two rigid spheres at collision. This collisional transfer is the principal mechanism for energy transport between gas molecules at high density.

The transport properties of a dense gas can be estimated using the EMKT from a priori knowledge of the transport properties of the dilute gas, the compressibility data of the dense gas, and the effective collision diameter of the molecules. This treatment has been applied successfully to helium and neon in the moderate-density range [16,17]. These gases have very low critical temperatures, and at high reduced temperatures, their molecules behave like hard spheres. However, the EMKT has not been as successful in predicting the properties of heavier noble gases such as argon [16,17], but predicts a larger increase in thermal conductivity with density than found experimentally and a strong dependence on temperature, which has not been observed experimentally. Though the forces considered in the EMKT cause inconsistent predictions of the thermodynamics of irreversible processes and this approach is only accurate to the first order in density, the EMKT is a good approach for estimating the transport properties of gases in the intermediate-density range when experimental data are not available.

The EMKT has been used to predict the properties of the binary mixtures of He–Ne, Ne–Ar, and He–Ar with some success [18] and has been extended to predict the properties of binary gas mixtures of dense polyatomic gases such as N_2–CO [19]. Despite these successes, the relationships in the EMKT are difficult to use in practice, due to the large number of parameters that need to be determined for both the pure gases and the unlike-molecule interactions.

This work conducted an extensive review and compiled an extensive database of the measured transport and thermodynamic properties of He and N_2 and their binary mixtures. This database is used to validate the semi-empirical correlations developed in this work for predicting these properties at temperatures between 200 and 1500 K and pressures up to 40 MPa. The next section presents and discusses the compiled database of the thermodynamic and transport properties of helium and nitrogen and their binary mixtures.

IV. Compiled Properties Database

An extensive review of the literature is conducted to compile reported measurements of the transport and thermodynamic properties of helium and nitrogen and their binary mixtures (He–N_2) as functions of temperature and pressure. The values of the critical parameters recommended by NIST [8] and listed in Table 1 are used in this work. The compiled database for the dynamic viscosity and thermal conductivity are summarized in Tables 2 and 3. The compiled experimental data are organized into four groups: 1) pure gases at dilute densities (i.e., pressures less than 0.1 MPa), 2) He–N_2 binary mixtures at dilute densities, 3) pure dense gases ($0.1 < P < 130$ MPa), and 4) dense He–N_2 binary mixtures. Tables 2 and 3 list the source references; the number of data points reported in each reference and considered in this work; and the temperature, pressure, and range of the reduced density, when relevant. The last columns in these tables give the maximum deviation between the predictions of the semi-empirical correlations

Table 1   Fundamental and critical properties of the pure noble gases and of nitrogen

Property	Helium	Neon	Argon	Krypton	Xenon	Nitrogen
M, kg/mol [8]	0.004003	0.020179	0.039948	0.0838	0.13129	0.028013
T_{cr}, K [8]	5.2	44.5	150.7	209.5	289.7	126.2
P_{cr}, MPa [8]	0.2275	2.678	4.863	5.51	5.84	3.396
ρ_{cr}, kg/m^3 [8]	69.64	481.9	535.6	908.4	1110.	313.3
V_{cr}, cm^3/mol [8]	57.48	41.87	74.59	92.25	118.28	89.41
$0.291V^*$, cm^3/mol	55.30	40.20	74.98	91.99	120.02	89.91
Deviation, %	−3.781	−3.987	+0.526	−0.278	+1.473	+0.561
V^*, cm^3/mol	190.04	138.16	257.66	316.13	412.45	308.97
$\Delta\mu_{cr} \times 10^6$, Pa·s	——	8.9	16.0	23.3	29.7	9.99
$\mu^o(T_{cr}) \times 10^6$, Pa·s	1.428	7.257	12.34	18.39	22.55	8.60
$\mu_{cr} \times 10^6$, Pa·s	——	16.157	28.34	41.69	52.25	18.59
$\mu_{cr}^* \times 10^6$ deviation, %	——	−0.786	−0.329	0.417	−0.076	2.80
Ratio $\mu_{cr}/\mu^o(T_{cr})$	——	2.226	2.297	2.267	2.317	2.162
$\Delta\mu_{cr}^*$ deviation, %	——	+1.803	−0.215	+1.554	−0.639	+8.10
$\Delta\lambda_{cr}$, mW/m·K	24.0	24.5	18.62	12.96	10.55	20.10
$\lambda^o(T_{cr})$, mW/m·K	10.32	10.97	9.603	6.868	5.416	11.74
λ_{cr}, mW/m·K	34.32	35.47	28.223	19.828	15.966	31.84
λ_{cr}^* deviation, %	6.5	0.47	−0.50	0.99	−0.28	−8.16
Ratio $\lambda_{cr}/\lambda^o(T_{cr})$	3.326	3.233	2.939	2.887	2.948	2.712
$\Delta\lambda_{cr}^*$ deviation, %	+0.490	−4.014	−0.484	+1.958	−0.422	−4.01

Table 2   Experimental data of dynamic viscosity of helium, nitrogen, and their binary mixtures

Gas	References	Number of data points	T, K	P, MPa	Reduced density	Model equations	Deviation from data, %
		Dilute gases					
Helium	[8,9,11,25,27]	94	100/1600	≤ 0.1	——	Eqs. (2a) and (6)	−1.8/ + 1.7
Nitrogen	[8,9,26,49,50]	65	100/2150	≤ 0.1	——	Eqs. (2b) and (5)	−0.9/ + 0.7
He–N_2	[9,51]	66	293/973	≤ 0.1	——	Eqs. (37) and (40)	−0.6/ + 0.6
		Dense gases					
Helium	[21,22,30,31,52–55]	139	223/523	≤ 21	$\rho_r \leq 0.56$	Eq. (6)	−1.0/ + 1.3
Nitrogen	[21,22,30,31,52,53,56,57]	200	223/538	≤ 40	$\rho_r \leq 1.10$	Eq. (5)	−2.5/ + 4.1
He–N_2	[21,22,58,59]	110	273/902	≤ 40	$\rho_r \leq 1.10$	Eq. (37)	−2.5/ + 1.6

Table 3 Experimental data of thermal conductivity of helium, nitrogen, and their binary mixtures

Gas	References	Number of data points	T, K	P, MPa	Reduced density	Model equations	Deviation from data, %
		Dilute gases					
Helium	[8,11,27,33]	73	200/1500	≤ 0.1	——	Eqs. (10) and (11)	$-0.9/ + 1.1$
Nitrogen	[28,33,50,60–62]	65	200/1500	≤ 0.1	——	Eqs. (10) and (12)	$-3.0/ + 2.1$
He–N_2	[63–65]	37	273/591	≤ 0.1	——	Eqs. (43) and (48)	$-3.5/ + 3.1$
		Dense gases					
Helium	[33,66,67]	210	250/1500	≤ 130	$\rho_r \leq 1.60$	Eq. (10)	$-2.6/ + 2.5$
Nitrogen	[19,28,33,60–62,68–75]	330	200/1173	≤ 101	$\rho_r \leq 2.00$	Eq. (10)	$-2.6/ + 3.1$
He–N_2	[76]	42	301	≤ 13	$\rho_r \leq 0.25$	Eq. (43)	$-3.8/ + 0.7$

Table 4 Experimental data of virial coefficients of helium, nitrogen, and their binary mixtures

Gas	References	Number of data points	Temperature, K	Model equations	Deviation from data
		Second virial coefficient B of pure gases			
Helium	[38,43,77,78]	52	35/1473	Eq. (15)	± 1.0 cm^3/mole
Nitrogen	[38,44,79,80]	53	123/1400	Eq. (16)	± 1.3 cm^3/mole
		Interaction second virial coefficient B_{12} of binary gas mixtures			
He–N_2	[43,44]	24	103/748	Eq. (36)	± 1.5 cm^3/mole
		Third virial coefficient C of pure gases			
Helium	[43]	9	123/323	——	——
Nitrogen	[38,43]	20	160/500	Eq. (17)	$-5.3\%/ + 10.0\%$

developed in this work and the reported experimental data and the values recommended by NIST for the pure gases.

The densities of He and N_2 and their binary mixtures are calculated using the virial equation of state, which requires knowledge of the second (B) and third (C) virial coefficients of the pure gases and the interaction (second virial coefficients B_{12} of the binary mixtures), which are all functions of temperature. The compiled database for these virial coefficients is listed in Table 4. Although many data are available for pure gases, few could be found for the interaction virial coefficient B_{12}. The maximum deviation in predicting the second virial coefficients is reported in Table 4 in cm^3/mol, because these coefficients span a wide range of negative and positive values.

Many investigators who measured the thermal conductivity and dynamic viscosity of dense pure gases and binary mixtures (Tables 2 and 3) also measured the pressure and density. This information is used directly to quantify the accuracy of the virial equation of state developed in this work. This equation agrees with the reported density data and the values recommended by NIST [8] to within $\pm 2\%$, when $T > (T_{cr} + 40$ K), $P < 130$ MPa, and $\rho < \rho_{cr}$. Outside the critical region, the virial equation of state predicts the density of helium and nitrogen to less than 0.4% of the values recommended by NIST [8]. As expected, the accuracy of this equation decreases when approaching the critical point and thus should only be used at temperatures higher than the critical value (i.e., $T > T_{cr}$).

The validated equation of state is used to calculate the thermodynamic properties (i.e., enthalpy, specific heat capacities $\hat{C}_P$ and $\hat{C}_v$, specific heat ratio γ, entropy, and speed of sound) of He, N_2, and their binary mixtures using fundamental thermodynamic relationships. Although no experimental data could be found for these thermodynamic properties, relationships developed in this work are successfully benchmarked using the values recommended by NIST [8] for He and N_2 at pressures from 0.1 to 20 MPa and temperatures up to 1500 K.

## V. Law of Corresponding States for Dense Gases

The thermodynamic and transport properties of helium and nitrogen gases at low temperature and high pressure (i.e., high density) are correlated using the virial state equation and the law of corresponding states [20], in terms of the critical temperature T_{cr} and critical density ρ_{cr}, instead of the potential parameters ε and σ in the Chapman–Enskog approach for dilute gases. Because accurate values of the critical temperature and density of pure gases are readily available [8], the present model is easily extended to multicomponent mixtures in the moderate- to high-density regions. The law of

corresponding states had generated little interest in the past, due to the lack of experimental data and association with either inaccurate equations of states, especially van der Waal's state equation [6] or approximate models for the intermolecular potential [i.e., the Lennard–Jones (6, 12) model] [5,20]. However, the use of the law of corresponding states in this work capitalizes on the large database compiled for the virial coefficients and transport properties of helium and nitrogen and their binary mixtures as functions of both temperature and pressure.

A. Dynamic Viscosity of Pure Gases

The large experimental database delineated in Figs. 1–5 shows that when $T > T_{cr}$ and $\rho < 2 \rho_{cr}$, the differences between the actual and diluted values of the viscosity and thermal conductivity of noble gases and nitrogen are solely functions of the density and not the temperature [21–23]. When plotted versus the reduced density (ρ/ρ_{cr}), the excess viscosity $\Delta\mu$ is described by a single curve (Fig. 1), which is used in this work to determine the critical viscosity of nitrogen gas. The critical viscosity of nitrogen, μ_{cr}, from the least-squares fit of the compiled experimental data in Fig. 1 and at a reduced density of unity can be expressed as

$$\mu_{cr} = \Delta\mu_{cr} + \mu^o(T_{cr}) = 9.99 \times 10^{-6} + 8.60 \times 10^{-6}$$
$$= 18.59 \times 10^{-6} \text{ Pa} \cdot \text{s} \tag{1}$$
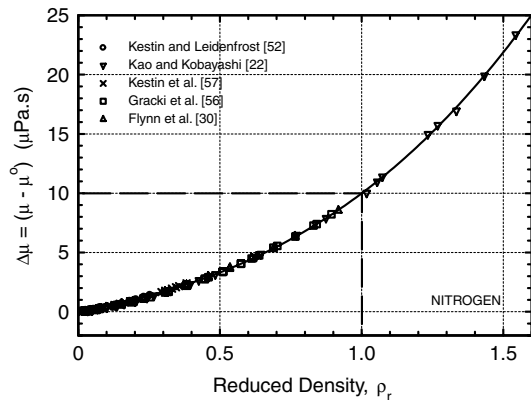
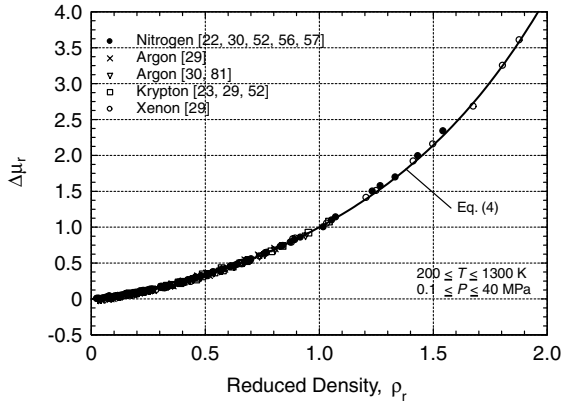**Fig. 1 Excess viscosity of nitrogen as a function of reduced density.**

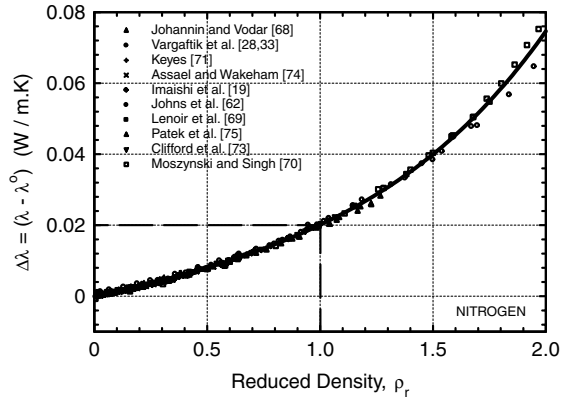**Fig. 2 Reduced, excess viscosity of nitrogen and the heavier noble gases.**

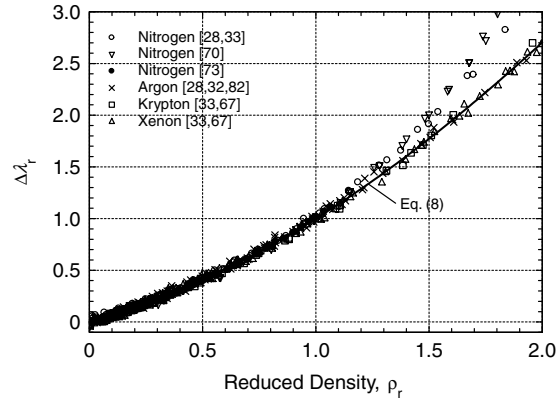**Fig. 3 Excess conductivity of nitrogen as a function of reduced density.**

Fig. 4 Reduced, excess conductivity of nitrogen and the heavier noble gases.

Calculating the critical viscosity requires accurate knowledge of the dilute-gas value at the critical temperature $\mu^o(T_{cr})$, which is 8.60×10^{-6} Pa · s for nitrogen. The determined critical viscosities for Xe, Kr, Ar, Ne [4], and N_2 using the same approach are listed in Table 1; they are 3–7% higher than those reported in [24].

The compiled experimental [25,26] and recommended tabulated values [11,26–29] of the viscosities of dilute helium and nitrogen for $100 \text{ K} < T < 1400 \text{ K}$ (Table 2) are correlated using the following expressions: For helium:

$$\mu^o(T) = 3.0629 \times 10^{-7} \times (T + 21.33)^{0.7243} \text{ Pa} \cdot \text{s} \quad (2a)$$

For nitrogen:
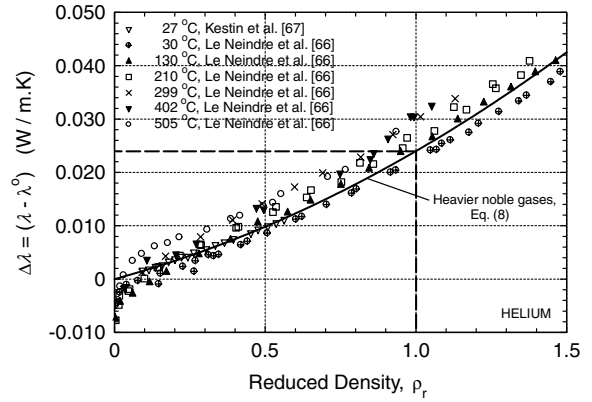
Fig. 5 Excess conductivity of helium as a function of reduced density.
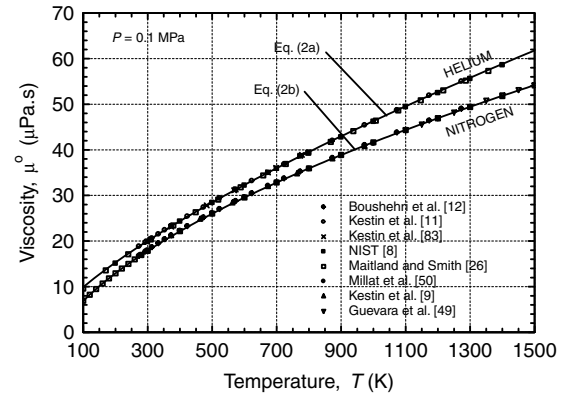
Fig. 6 Dynamic viscosity of helium and nitrogen at dilute-density/ atmospheric pressure.

$$\mu^o(T) = 9.9 \times 10^{-7} \times (T + 16.8)^{0.5646} - 77 \times 10^{-7} \text{ Pa} \cdot \text{s} \quad (2b)$$

Equation (2a) is in agreement with the Chapman–Enskog kinetic theory for dilute gases [11] to within ±1.3% (Fig. 6). The predictions of the viscosity of nitrogen using the extended kinetic theory developed by Boushehri et al. [12] are also in good agreement with Eq. (2b), with a maximum deviation of 1% (Fig. 6). Plotting the reduced excess viscosity, defined as

$$\Delta \mu_r = \frac{\mu(T, P) - \mu^o(T)}{\mu_{cr} - \mu^o(T_{cr})} \quad (3)$$

versus the reduced density ρ_r, the following unique relationship is obtained from the least-squares fit of the compiled experimental data for Ne, Ar, Kr, and Xe (Fig. 2):

$$\Delta \mu_r = \Psi_\mu(\rho_r) = 0.221\rho_r + 1.062\rho_r^2 - 0.509\rho_r^3 + 0.225\rho_r^4 \quad (4)$$

This equation clearly illustrates the law of corresponding states. The viscosity of nitrogen and noble gases is calculated as a function of temperature and pressure using the following general relationship (Fig. 2):

$$\mu(T, P) = \mu^o(T) + \Delta\mu_{cr} \times \Psi_\mu(\rho_r) \quad (5)$$

As shown in Table 2 and Fig. 7, this equation accurately reproduces the 200 experimental data points compiled for $T > 223 \text{ K}$ and pressures up to 40 MPa, with a maximum deviation ranging from −2.5 to +4.1%. Unlike other noble gases, the dynamic viscosity of helium is essentially independent of pressure and is solely a function of temperature [22,30,31]. Therefore, in this work, the viscosity of helium is calculated using a simplified form of Eq. (5):
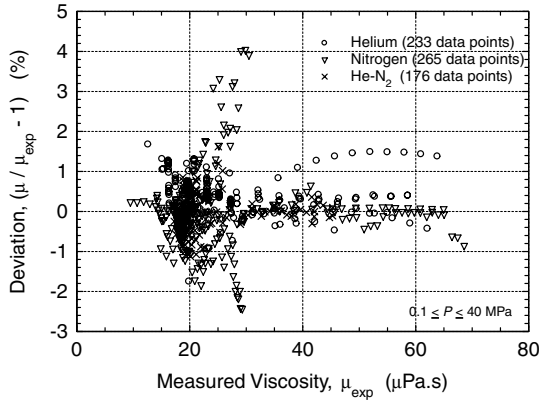
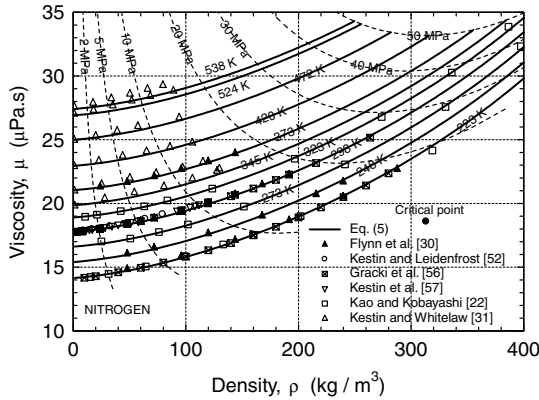Fig. 7 Percent deviation between predicted and measured viscosities.

Fig. 8 Viscosity of nitrogen as a function of pressure and temperature.

$$\mu(T, P) = \mu^o(T) \qquad (6)$$

Table 2 summarizes the compiled experimental database for the dynamic viscosities of helium and nitrogen. The maximum deviation of the predictions of Eq. (6) from the experimental data of the viscosity of helium at pressures up to 21 MPa is less than 1.8% (Fig. 7). The effects of temperature and pressure on the viscosity of nitrogen are also shown in Fig. 8, along with some of the compiled experimental data. Equation (5) compares very well with the 200 experimental data points compiled for nitrogen at temperatures up to 538 K, pressures to 40 MPa, and densities up to 1.10 (the critical value), with an absolute error of less than 4.1% (Table 2).

B. Thermal Conductivity of Pure Gases

A semi-empirical expression is developed for the thermal conductivity of dense noble gases using the same procedure outlined in the previous section for the dynamic viscosity. When plotted versus the reduced density [32], a unique curve is obtained for the excess thermal conductivity of nitrogen, $\Delta\lambda$ (Fig. 3). The critical conductivity of nitrogen is then calculated as

$$\lambda_{cr} = \Delta\lambda_{cr} + \lambda^o(T_{cr}) = 0.0201 + 0.01174 = 0.03184 \text{ W/m} \cdot \text{K} \qquad (7)$$

The dilute-gas thermal conductivity $\lambda^o(T_{cr})$ is obtained from the tables developed by a number of investigators [28,29,33,34], and the critical thermal conductivity is listed in Table 1. The prediction of the nitrogen critical conductivity is within 12% of that reported by Owens and Thodos in 1957 [35] using the same technique. An empirical relationship for calculating the reduced excess thermal conductivity of heavier noble gases (Ar, Kr, and Xe) has also been obtained from the least-squares fit of the compiled experimental data (Fig. 4) and is given as [4]

$$\Delta\lambda_r = \Psi_\lambda(\rho_r) = 0.645\rho_r + 0.331\rho_r^2 + 0.0368\rho_r^3 - 0.0128\rho_r^4 \qquad (8)$$

The experimental data for nitrogen fall on the solid line [Eq. (8)] for the noble gases, up to a reduced density of 1.2. At higher reduced densities, the excess thermal conductivity for nitrogen is progressively higher than that for noble gases (Fig. 4). However, because the excess thermal conductivity is a small fraction of the dilute-gas conductivity, the relationship for noble gases [Eq. (8)] could be used for nitrogen with a deviation of less than 3.1% from the experimental data, up to a reduced density of 2 (Table 3). The experimental data for the excess thermal conductivity of helium (Fig. 5) also follow closely the noble-gas relationship (8), when $\Delta\lambda_{cr}$ is taken equal to 0.024 W/m · K.

Using the two-parameter law of corresponding states to predict the thermal conductivities of binary gas mixtures as functions of pressure and temperature requires developing a semi-empirical expression for the critical thermal conductivity as a function of M, T_{cr}, and V_{cr}, as well as a relationship between $\lambda^o(T_{cr})$ and λ_{cr}. A general expression for the critical thermal conductivity of binary mixtures is developed, following the approach of Owens and Thodos [35], of the general form

$$\lambda_{cr}^* = aM^c T_{cr}^d (0.291 \times V^*)^g \qquad (9a)$$

The values listed in Table 1 for Ne, Ar, Kr, and Xe are used to evaluate the coefficient a and exponents c, d, and g. The final expression is [4]

$$\lambda_{cr}^* = 0.304 \times 10^{-4} \frac{T_{cr}^{0.277}}{M^{0.465}(0.291 \times V^*)^{0.415}} \qquad (9b)$$

The predictions of this expression are in excellent agreement with the compiled experimental data to within less than 1% for noble gases of Ne, Ar, Kr, and Xe [4], but are within +6.5% for He and −8.2% for nitrogen (Table 1). For the heavier noble gases of Ar, Kr, and Xe, the ratio $\lambda_{cr}/\lambda^o(T_{cr})$ is nearly constant and equal to 2.94 (Table 1 [4]). The thermal conductivities of the pure noble gases were calculated using the following semi-empirical relationship [4]:

$$\lambda(T, P) = \lambda^o(T) + \left(1 - \frac{1}{2.94}\right)\lambda_{cr}^* \times \Psi_\lambda(\rho_r) \qquad (10)$$

The values of $\lambda_{cr}/\lambda^o(T_{cr})$ for He (3.326) and N$_2$ (2.712) are higher and lower than 2.94, respectively. The differences in these ratios compensate for the errors in using Eq. (9b) to predict the critical thermal conductivities of these gases. As shown in Table 1, the critical excess conductivity $\Delta\lambda_{cr}$ is well predicted by $(1 - 1/2.94) \times \lambda_{cr}^*$, with a maximum deviation from experimental values of +0.49% for helium and −4.01% for nitrogen. Because the thermal conductivities of these gases exhibit a relatively weak dependence on pressure, these small errors in $\Delta\lambda_{cr}$ are inconsequential.

The thermal conductivity of the monoatomic dilute noble gases, such as helium, is calculated accurately in terms of the dynamic viscosity using the well-known kinetic-theory relationship [6]:

$$\lambda^o = \frac{15}{4}\frac{R_g}{M}\mu^o = \frac{15}{4}\frac{k}{m}\mu^o \qquad (11)$$

The higher-order correction factor, not included herein, is very close to unity [7] and amounts to less than 0.4%. Equation (11) agrees with the tabulated values [27,33] and with those calculated using the Chapman–Enskog approach for dilute gases [11]. The predictions are within ±2.6% of the data for the heavier noble gases at temperatures from 100 to 1400 K [4] and to within ±0.5% of the helium data for temperatures from 273 to 1273 K (Fig. 9). For the diatomic nitrogen, Eq. (11) underpredicts the thermal conductivity at dilute densities; thus, the following polynomial fit is developed instead:
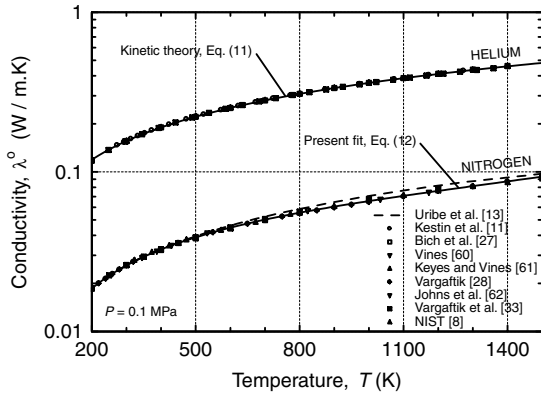
Fig. 9 Thermal conductivity of helium and nitrogen at dilute-density/atmospheric pressure.

$$\lambda^o(T) = 1.4964 \times 10^{-11} T^3 - 4.3052 \times 10^{-8} T^2 + 9.1894$$
$$\times 10^{-5} T + 1.839 \times 10^{-3} \text{ W/m} \cdot \text{K} \quad (12)$$

The predictions of the thermal conductivity of nitrogen using the extended kinetic theory developed by Uribe et al. [13] for molecular gases are shown in Fig. 9 by a dashed line. These predictions are higher than the experimental data by as much as 8% in the high temperature range shown.

The predictions of Eq. (10) are compared with the compiled experimental data for the thermal conductivities of helium and nitrogen (Table 3 and Fig. 10). The errors in predicting these thermal conductivities are less than 3.1% for temperatures greater than 200 K and for pressures as high as 130 and 101 MPa, respectively, (Table 3). The present predictions of the thermal conductivities of nitrogen and helium are shown in Figs. 11 and 12 as functions of pressure and temperature, along with some of the compiled experimental data. Equation (10) agrees well with the nitrogen data for densities up to 2.0 times the critical value, to within less than 3.1%, and with the helium data for densities up to 1.6 times the critical value, to within less than 2.6% (Table 3). The gas densities in Eqs. (4), (5), (8), and (10) are calculated using the procedure delineated in the next subsection.

C. Density of the Pure Gases

The densities of pure gases are calculated by solving the virial state equation, expressing the compressibility factor Z as a second-order polynomial in the molar density $\hat{\rho}$ as

$$P = (R_g T)(\hat{\rho} Z) = R_g T \times \hat{\rho}(1 + B\hat{\rho} + C\hat{\rho}^2) \quad (13)$$

This equation, shown to be accurate up to the critical density [36], is inverted analytically [37] to obtain an explicit expression for the gas density. The virial coefficients B and C in Eq. (13) are related to the
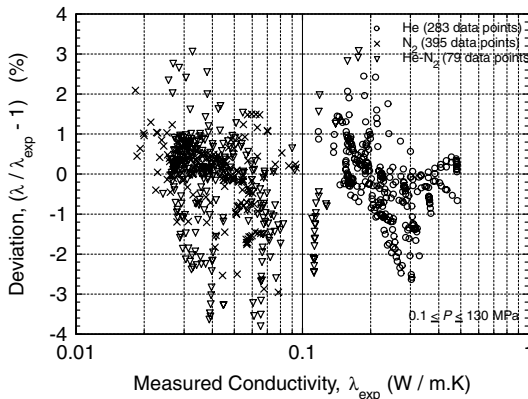

Fig. 10 Percent deviation between predicted and measured conductivities.
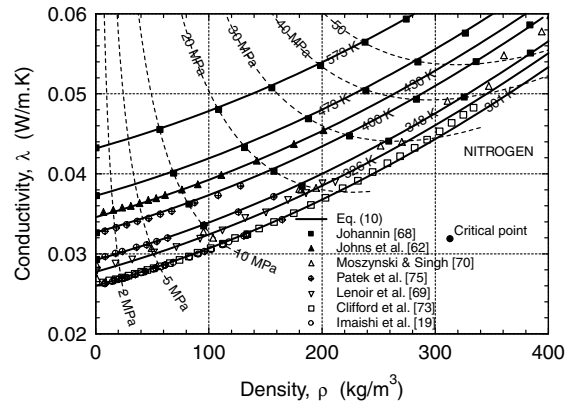

Fig. 11 Thermal conductivity of nitrogen as a function of pressure and temperature.
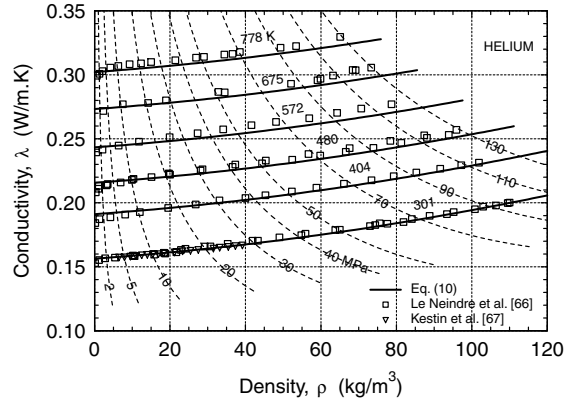

Fig. 12 Thermal conductivity of helium as a function of pressure and temperature.

intermolecular potentials [5] and have been experimentally determined for many gases (Table 4). The developed expression for the normalized second virial coefficient B_r, based on the compiled experimental data of noble gases (except He), is given as [4]

$$B_r = \left[-102.6 + \left(102.732 - 0.001 \times \theta - \frac{0.44}{\theta^{1.22}} \right) \times \tanh(4.5\sqrt{\theta}) \right] \quad (14)$$

This expression, valid for $0.5 < \theta < 22$, agrees with 95% of the compiled data to within ± 2 cm^3/mol [4]. The following relationship was developed for the second virial coefficient of helium [4]:

$$B(\text{He}) = 8.4 - 0.0018 \times T + \frac{115}{\sqrt{T}} - \frac{835}{T} \text{ cm}^3/\text{mol} \quad (15)$$

This expression agrees with the experimental data of Levelt Sengers et al. [38] and others (Table 4) to within ± 1.0 cm^3/mol (Fig. 13). For nitrogen, the best fit of the compiled experimental data gives the following correlation for the normalized virial coefficient, which is accurate to within ± 1.3 cm^3/mol (Fig. 13):

$$B_r(\text{N}_2) = 0.1366 - \frac{0.2788}{\theta} - \frac{0.1937}{\theta^2} \quad (16)$$

Compared with Eq. (16) and the compiled database in this work, Eq. (14) (shown as a dotted curve in Fig. 13) underpredicts the second virial coefficient for nitrogen.

The compiled experimental data for the normalized third virial coefficient of Ne, Ar, and Kr for $1.0 < \theta < 45$ has been correlated [4] as
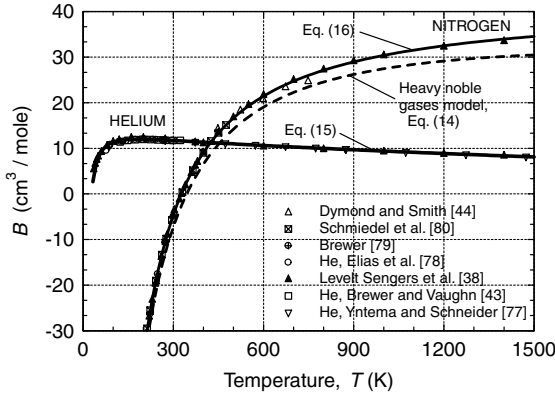
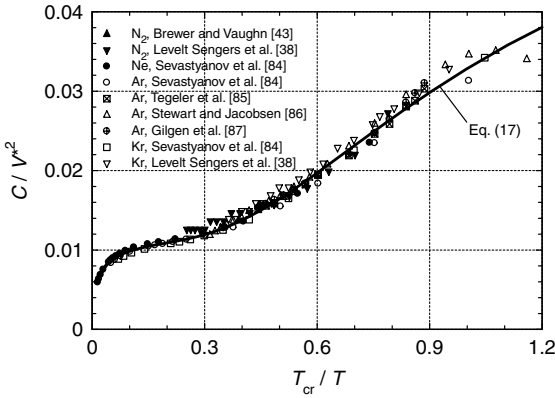**Fig. 13 Second virial coefficients of helium and nitrogen versus reduced temperature.**

Fig. 14 Reduced third virial coefficients of nitrogen and the noble gases.

$$C_r = 0.0757 + \left(-0.0862 - 3.6 \times 10^{-5} \times \theta + \frac{0.0237}{\theta^{0.059}}\right)$$
$$\times \tanh(0.84 \times \theta) \qquad (17)$$

This empirical expression has been shown to agree with more than 90% of the compiled experimental data for Ne, Ar, and Kr to within $\pm 5\%$ [4]. As shown in Fig. 14, Eq. (17) appropriately fits the experimental data for the third virial coefficient of nitrogen (Table 4). Because helium has a very low critical temperature [(5.2 K) see Table 1], its behavior closely mimics that of a dilute perfect gas at temperatures greater than 200 K and pressures less than 130 MPa; thus, the third virial coefficient for helium is neglected.

Equation (13) accurately predicts the compiled density data and the values recommended by NIST [8] for pure noble gases to within $\pm 2\%$ when $T > (T_{cr} + 40 \text{ K})$ and $\rho \leq \rho_{cr}$, but the accuracy decreases when approaching the critical point (Fig. 15). Outside the critical region, Eq. (13) predicts the density of helium and nitrogen gases to within less than 0.4% of the values recommended by NIST [8], but overpredicts the high densities, with a deviation that increases with increasing density (Fig. 15). Equation (13) agrees with the recommended values of the nitrogen densities by NIST [8] to within less than 2%, up to the critical values, temperatures as low as 200 K, and pressures up to 20 MPa (Fig. 15). These ranges of applicability more than cover those of interest in the potential applications to space reactor power systems and terrestrial nuclear power plants with gas-cooled reactors and CBC for energy conversion ($300 < T < 1200 \text{ K}$ and $1.0 < P < 10$ MPa).

D. Enthalpy and Specific Heats of Pure Gases and Mixtures

The thermodynamic properties of enthalpy, heat capacities, and entropy are calculated using fundamental thermodynamic relationships [39–41]. The molar enthalpy at $[T, P]$ is expressed as
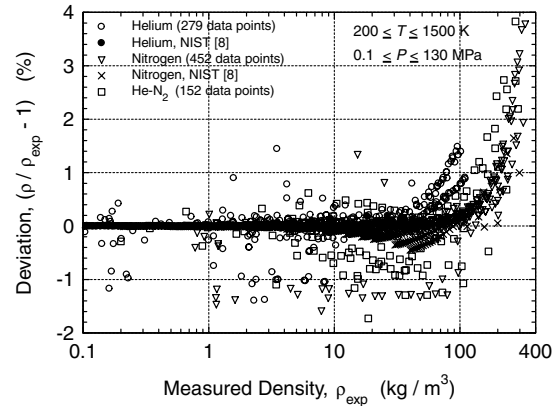

Fig. 15 Percent deviation between predicted and measured densities.

$$\hat{H}(T, P) = \hat{H}^o(T_o) + \int_{T_o}^T \hat{C}_P^o \, dT + R_g T(Z - 1)$$
$$+ \int_0^{\hat{\rho}} \left\{P - T\left[\frac{\partial P}{\partial T}\right]_{\hat{\rho}}\right\} \frac{d\hat{\rho}}{\hat{\rho}^2} \qquad (18)$$

The last integral is expressed in terms of the virial coefficients and their temperature derivatives using Eq. (13), where B and C are functions of temperature only:

$$P - T\left[\frac{\partial P}{\partial T}\right]_{\hat{\rho}} = -R_g \hat{\rho}^2 T^2 \left[\frac{dB}{dT} + \hat{\rho}\frac{dC}{dT}\right] \qquad (19)$$

When substituted into Eq. (18), the final expression for the molar enthalpy is

$$\hat{H}(T, P) = \hat{H}^o(T_o) + \int_{T_o}^T \hat{C}_P^o \, dT + \hat{\rho} R_g T\left[\left(B - T\frac{dB}{dT}\right)\right.$$
$$\left. + \hat{\rho}\left(C - \frac{T}{2}\frac{dC}{dT}\right)\right] \qquad (20)$$

The molar heat capacity at constant pressure is given by

$$\hat{C}_P = \left[\frac{\partial \hat{H}}{\partial T}\right]_P = \hat{C}_P^o + \hat{\rho} R_g\left[\left(B - T\frac{dB}{dT} - T^2\frac{d^2B}{dT^2}\right)\right.$$
$$+ \hat{\rho}\left(C - \frac{T^2}{2}\frac{d^2C}{dT^2}\right)\right] + R_g T\left[\left(B - T\frac{dB}{dT}\right)\right.$$
$$\left. + \hat{\rho}\left(2C - T\frac{dC}{dT}\right)\right] \times \left[\frac{\partial \hat{\rho}}{\partial T}\right]_P \qquad (21)$$

where

$$\left[\frac{\partial \hat{\rho}}{\partial T}\right]_P$$
$$= -\frac{(1 + B\hat{\rho} + C\hat{\rho}^2)(\hat{\rho}/T) + (dB/dT)\hat{\rho}^2 + (dC/dT)\hat{\rho}^3}{1 + 2B\hat{\rho} + 3C\hat{\rho}^2}$$
$$(22)$$

The molar heat capacity at constant volume is obtained using the following expression:

$$\hat{C}_v = \left[\frac{\partial \hat{H}}{\partial T}\right]_{\hat{\rho}} - \left[\frac{\partial}{\partial T}\left(\frac{P}{\hat{\rho}}\right)\right]_{\hat{\rho}}$$
$$= \hat{C}_v^o - \hat{\rho} R_g T\left[\left(2\frac{dB}{dT} + T\frac{d^2B}{dT^2}\right) + \hat{\rho}\left(\frac{dC}{dT} - \frac{T}{2}\frac{d^2C}{dT^2}\right)\right] \qquad (23)$$

Equations (21) and (23) give the specific heat ratio $\gamma = \hat{C}_P/\hat{C}_v$ for the various gases and their binary mixtures. The molar heat capacities are related by the relationship [41]

$$\hat{C}_P - \hat{C}_v = MT \left[\frac{\partial \upsilon}{\partial T} \right]_P \times \left[\frac{\partial P}{\partial T} \right]_v \qquad (24)$$

At a dilute density, the virial equation of state (13) reduces to the perfect gas law, and Eq. (24) gives

$$\hat{C}_v^o = \hat{C}_P^o - R_g \qquad (25)$$

According to classical mechanics and the theorem of equipartition of energy [42], the molar heat capacity $\hat{C}_P^o$ for dilute monoatomic noble gases on the right-hand side of Eq. (21) can be expressed as

$$\hat{C}_P^o = \tfrac{5}{2} R_g \qquad (26)$$

The molar heat capacity of dilute monoatomic noble gases is constant and independent of temperature. By contrast, the additional degrees of freedom of diatomic molecules [42] cause the molar heat capacities and the specific heat ratio of nitrogen (Fig. 16) and He–N$_2$ binary mixtures to vary with temperature, even at a dilute density. For nitrogen, the values of the molar heat capacity recommended by NIST [8] at atmospheric pressure and temperatures between 200 and 1500 K are fitted with a maximum deviation less than 0.04% using the following polynomial relationship (Fig. 16):

$$\hat{C}_P^o = R_g \times \sum_{k=0}^{6} a_k T^k \qquad (27)$$

The polynomial coefficients are $a_o = 3.5094$, $a_1 = 5.3927 \times 10^{-4}$ K^{-1}, $a_2 = -4.5995 \times 10^{-6}$ K^{-2}, $a_3 = 1.2879 \times 10^{-8}$ K^{-3}, $a_4 = -1.3955 \times 10^{-11}$ K^{-4}, $a_5 = 6.8333 \times 10^{-15}$ K^{-5}, and $a_6 = -1.2719 \times 10^{-18}$ K^{-6}.
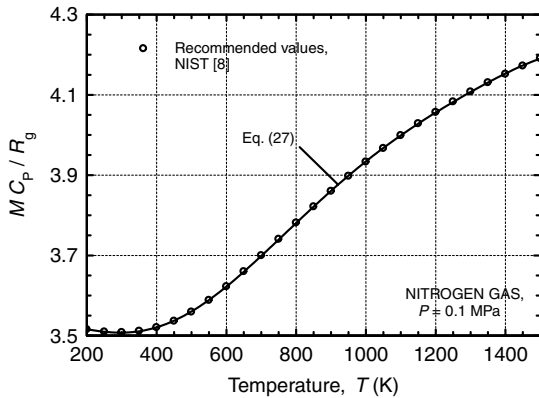

Fig. 16   Specific heat at constant pressure of nitrogen at atmospheric pressure.
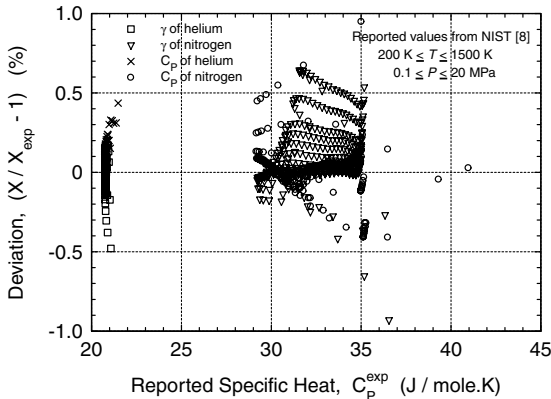

Fig. 17   Percent deviation between predicted and recommended values of thermodynamic properties.
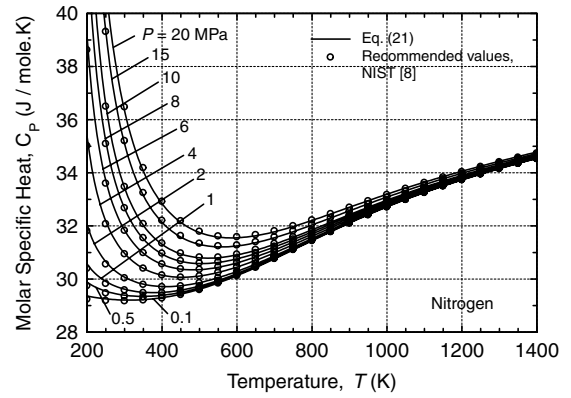

Fig. 18   Specific heat of nitrogen as function of pressure and temperature.
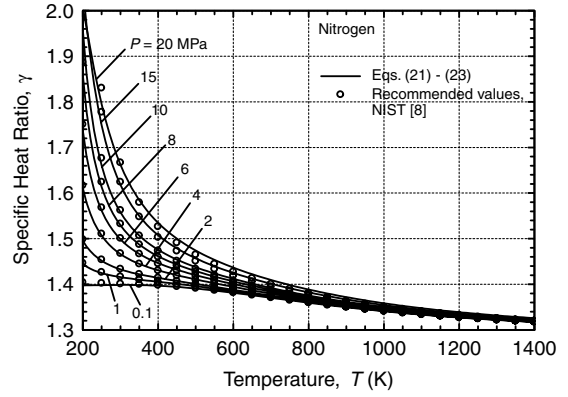

Fig. 19   Specific heat ratio of nitrogen as function of pressure and temperature.

For helium–nitrogen binary mixtures, the molecular weight is given by $\bar{M} = x_1 M_{N_2} + x_2 M_{He}$ and the molar heat capacity at a dilute density is obtained from the relationship

$$\hat{C}_P^o = x_1 \left(\hat{C}_P^o \right)_{N_2} + x_2 \left(\hat{C}_P^o \right)_{He} = x_1 R_g \times \sum_{k=0}^{6} a_k T^k + x_2 \times 2.5 R_g \qquad (28)$$

Integrating Eq. (28) and substituting into Eq. (20) gives

$$\int_{T_o}^{T} \hat{C}_P^o \, dT = x_1 R_g \times \sum_{k=0}^{6} a_k \frac{T^k - T_o^k}{k+1} + x_2 \times 2.5 R_g (T - T_o) \qquad (29)$$

The calculated values of the molar enthalpy, molar heat capacity, and the specific heat ratio using the preceding relationships are in good agreement with those recommended by NIST [8] for helium and nitrogen at pressures up to 20 MPa and temperatures between 200 and 1500 K (Fig. 17). The maximum deviation of the predicted values of the molar specific heat and specific heat ratio for helium is 0.5%. For nitrogen, the maximum deviation of the molar specific heat compared with the reported values by NIST is 1.2%, with 2% for the specific heat ratio. The largest deviations occur below 400 K and beyond 10 MPa.

The present predictions of the molar heat capacity and specific heat ratio for nitrogen are shown in Figs. 18 and 19 as functions of both pressure and temperature, along with the values recommended by NIST [8]. Unlike helium, which exhibits a nearly constant specific heat over the operating conditions of interest, nitrogen exhibits significant variations (up to 40%) with both pressure and temperature. These variations are well predicted by Eqs. (20–29) (Figs. 18 and 19).

E. Entropy and Speed of Sound of Pure Gases and Binary Mixtures

The gas entropy is calculated using the following expression:

$$\hat{S} = \hat{S}^o(T_o) + \int_{T_o}^{T} \frac{\hat{C}_P^o}{T}\, dT - R_g \left[\ln\left(\frac{\hat{\rho}}{\hat{\rho}_o}\right) + B(\hat{\rho} - \hat{\rho}_o) \right.$$
$$\left. + \frac{C}{2}\left(\hat{\rho}^2 - \hat{\rho}_o^2\right) \right] - R_g T \left[\frac{\partial B}{\partial T}(\hat{\rho} - \hat{\rho}_o) + \frac{1}{2}\frac{\partial C}{\partial T}\left(\hat{\rho}^2 - \hat{\rho}_o^2\right) \right]$$

(30)

The density $\hat{\rho}_o = \hat{\rho}(T, P_o)$ is evaluated at the operating temperature and at the reference dilute pressure P_o, and the density $\hat{\rho} = \hat{\rho}(T, P)$ is evaluated at the operating pressure and temperature. For the helium binary mixtures with nitrogen, integrating the expression for the mixture heat capacity at dilute density in the entropy Eq. (30) gives

$$\int_{T_o}^{T} \frac{\hat{C}_P^o}{T}\, dT = x_1 R_g \times \left[a_o \times \ln\left(\frac{T}{T_o}\right) + \sum_{k=1}^{6} a_k \frac{T^k - T_o^k}{k} \right]$$
$$+ x_2 \times 2.5 R_g \times \ln\left(\frac{T}{T_o}\right)$$

(31)

The entropies calculated using Eq. (30) are compared successfully with those recommended by NIST [8] for He, Ne, Ar, Kr, Xe, and nitrogen at pressures up to 20 MPa and temperatures between 200 and 1500 K, with a maximum deviation of less than 0.05%.

The speed of sound is calculated from the relationship [41]

$$c = \frac{\gamma}{M}\left[\frac{\partial P}{\partial \hat{\rho}}\right]_T = \left[\frac{\gamma R_g T}{M} \times (1 + 2B\hat{\rho} + 3C\hat{\rho}^2)\right]^{1/2}$$

(32)

The calculated values are compared successfully with those recommended by NIST [8] for He, Ne, Ar, Kr, Xe, and nitrogen for pressures up to 20 MPa and temperatures in the range of 200–1500 K. The maximum deviation at the largest density (200 K and 20 MPa) is 0.4% for the speed of sound in helium, and at temperatures greater than 300 K and pressures less than 20 MPa, the maximum deviation is 2.3% for the speed of sound in nitrogen. These errors, however, decrease very rapidly, becoming insignificant as the temperature increases and/or the pressure decreases. For example, above 400 K, the error in calculating the nitrogen speed of sound using Eq. (32) is less than 0.8% for pressures up to 20 MPa. The molar density $\hat{\rho}$ of the He–N$_2$ binary gas mixtures in Eqs. (20–23), (30), and (32) is calculated next.

VI. Law of Corresponding States for Dense Gas Mixtures

In this section, the principle of corresponding states is extended to predict the thermophysical properties of dense binary gas mixtures and is applied to He–N$_2$ gas mixtures. The mixture's thermodynamic properties are determined using Eqs. (20–23), (30), and (33) by employing the expressions developed in this section for the virial coefficients $\bar{B}$ and $\bar{C}$, and the density $\hat{\rho}$ of the mixtures [4].

A. Density of Binary Gas Mixtures

The virial equation of state (13) for a pure gas applies to the He–N$_2$ binary mixtures when using the virial coefficients of the mixture, given by the kinetic theory of gases, on the basis of statistical mechanics [5,36] as

$$\bar{B} = x_1^2 B_{11} + 2x_1 x_2 B_{12} + x_2^2 B_{22} \tag{33a}$$

$$\bar{C} = x_1^3 C_{111} + 3x_1^2 x_2 C_{112} + 3x_2^2 x_1 C_{122} + x_2^3 C_{222} \tag{33b}$$

The coefficients B_{ii} and C_{iii} are those of the gas component i, and the interaction coefficients B_{ij} and C_{ijk} characterize the interaction among the unlike molecules ($i \neq j$) in binary and ternary collisions. The unknown third virial interaction coefficients are usually expressed as functions of those of the pure gases using the following combination rules [43]:

$$C_{112} = \left(C_1^2 C_2\right)^{1/3}, \qquad C_{122} = \left(C_1 C_2^2\right)^{1/3} \tag{34}$$

The second virial coefficients B_{11} and B_{22} for nitrogen and helium are given by Eqs. (15) and (16), respectively. For the helium–nitrogen mixtures, the following relationship is developed for the second virial interaction coefficient:

$$B_{12} = 34.39 - 0.01395 \times T - \frac{2585}{T} \ \text{cm}^3/\text{mol} \tag{35}$$

This expression agrees with the experimental data reported by Brewer and Vaughn [43] and Dymond and Smith [44] (Table 4) to within ± 1.5 cm^3/mol in the temperature range 100–800 K (Fig. 20). The predictions of the virial equation of state (13) agree with the compiled experimental database in this work for the density of He–N$_2$ mixtures at temperatures greater than 273 K, pressures up to 40 MPa, and densities up to the critical value, with a maximum deviation of 2% (Fig. 15). For reduced densities in the range of 1.0–1.1, the deviation between the present density predictions and the experimental data of the He–N$_2$ binary mixtures increases to a maximum of 4%.

B. Dynamic Viscosity of Dense Helium–Nitrogen Gas Mixtures

The two-parameter law of corresponding states for nitrogen [Eq. (5)] is extended to predict the viscosity of the He–N$_2$ binary gas mixtures as

$$\bar{\mu}(T, P) = \bar{\mu}^o(T) + x_1 \times (\Delta\mu_{cr})_1 \times \Psi_\mu\left(\frac{0.291 \times \bar{V}^*}{\bar{M}}\rho\right) \tag{36}$$

This form accounts for the fact that the viscosity of helium is essentially independent of pressure [22,30,31] [see Eq. (6)]. In Eq. (36), the density ρ, molecular weight $\bar{M} = x_1 M_1 + x_2 M_2$, and characteristic molar volume $\bar{V}^*$ are those of the He–N$_2$ gas mixture. The latter is calculated using the van der Waals mixing rule [45]:

$$\bar{V}^* = \sum_{i=1}^{N}\sum_{j=1}^{N} x_i x_j V_{ij}^* = x_1^2 V_{11}^* + 2x_1 x_2 \left(\frac{V_{11}^* + V_{22}^*}{2}\right)$$
$$+ x_2^2 V_{22}^* = x_1 V_{11}^* + x_2 V_{22}^* \tag{37}$$

This equation uses the linear average of the characteristic volumes, $V_{ij}^* = (V_{ii}^* + V_{jj}^*)/2$. As shown in Table 2, Eq. (36) predicts the compiled data for the viscosity of the binary mixtures of He–N$_2$ at pressures up to 40 MPa and reduced densities up to 1.10, to within 2.5%. In this equation, $\bar{\mu}^o(T)$, the viscosity of the dilute binary gas mixture, is calculated using the Chapman–Enskog kinetic-theory approach [6] as

$$\bar{\mu}^o = \left[\frac{x_1^2}{H_{11}} - \frac{2x_1 x_2 H_{12}}{H_{22} H_{11}} + \frac{x_2^2}{H_{22}}\right] \times \left[1 - \frac{H_{12}^2}{H_{22} H_{11}}\right]^{-1} \tag{38}$$
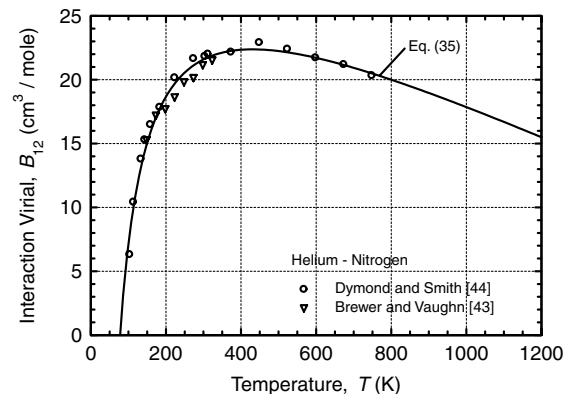

Fig. 20 Second virial interaction coefficient of helium–nitrogen gas mixtures.

The interaction coefficient $H_{12} \ll \sqrt{H_{11}H_{22}}$ is negligible for all 10 noble-gas binary mixtures [4] and for He–N$_2$ gas mixtures; thus, Eq. (38) reduces to the Sutherland and Wassiljewa expression, as detailed in [7]:

$$\bar{\mu}^o = \frac{\mu_1^o}{1 + \phi_{12}(x_2/x_1)} + \frac{\mu_2^o}{1 + \phi_{21}(x_1/x_2)} \quad (39)$$

The interaction coefficients ϕ_{ij} in Eq. (39) are given by [6]

$$\phi_{12} = \frac{\mu_1^o}{\mu_{12}}\left[\frac{2m_1 m_2}{(m_1+m_2)^2}\right] \times \left[\frac{5}{3A_{12}^*} + \frac{m_2}{m_1}\right]$$
$$\phi_{21} = \frac{\mu_2^o}{\mu_{12}}\left[\frac{2m_1 m_2}{(m_1+m_2)^2}\right] \times \left[\frac{5}{3A_{12}^*} + \frac{m_1}{m_2}\right] \quad (40)$$

In these equations, $A_{12}^* = \Omega^{(2,2)*}/\Omega^{(1,1)*}$, a very-slow-varying function of $T_{12}^* = kT/\varepsilon_{12}$, is taken as constant and equal to 1.10. For the Lennard–Jones (6,12) potential [5], $1.094 < A_{12}^* < 1.119$ in the temperature range of $1.0 < T_{12}^* < 20$.

In Eqs. (40), μ_{12}, a theoretical function of temperature only, is the viscosity of a hypothetical gas with a molecular mass: $m_{12} = [2m_1 m_2/(m_1+m_2)]$. Based on the compiled experimental data of the viscosity of dilute He–N$_2$ binary mixtures at different temperatures (Table 2), the following expression for the interaction viscosity, with a maximum deviation of 0.3% in the temperature range 293–973 K, is obtained:

$$\mu_{12}(T) = 0.5131 \times 10^{-6} \times (T - 29.7)^{0.6266} \text{ Pa}\cdot\text{s} \quad (41)$$

The predictions of Eqs. (39–41) are in excellent agreement with the compiled dilute viscosity data for He–N$_2$ mixtures (Table 2) to within $\pm 0.6\%$ (Figs. 7 and 21) and to within $\pm 3.7\%$ of the data for the 10 noble-gas binary mixtures at temperatures up to 1553 K [4]. The predictions of the dynamic viscosity of the dense He–N$_2$ binary mixtures using Eq. (36) are in excellent agreement with the compiled experimental data (Figs. 22 and 23), with a maximum deviation of 2.5% for pressures up to 40 MPa, reduced densities up to 1.10, and temperatures in the range of 273–973 K (Table 2).

C. Thermal Conductivity of Dense Helium–Nitrogen Gas Mixtures

The two-parameter law of corresponding states for pure gases [Eq. (10)] is extended to predict the thermal conductivity of binary gas mixtures [4]:

$$\bar{\lambda}(T,P) = \bar{\lambda}^o(T) + \left(1 - \frac{1}{2.94}\right)\bar{\lambda}^* \times \Psi_\lambda\left(\frac{0.291 \times \bar{V}^*}{\bar{M}}\rho\right) \quad (42)$$

The pseudocritical conductivity of the mixture $\bar{\lambda}^*$ is calculated using the form given by Eq. (9b):

$$\bar{\lambda}^* = 0.304 \times 10^{-4}\frac{(\bar{T}^*)^{0.277}}{\bar{M}^{0.465}(0.291 \times \bar{V}^*)^{0.415}} \quad (43)$$
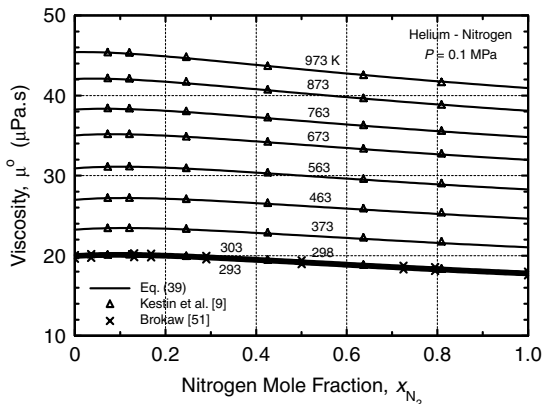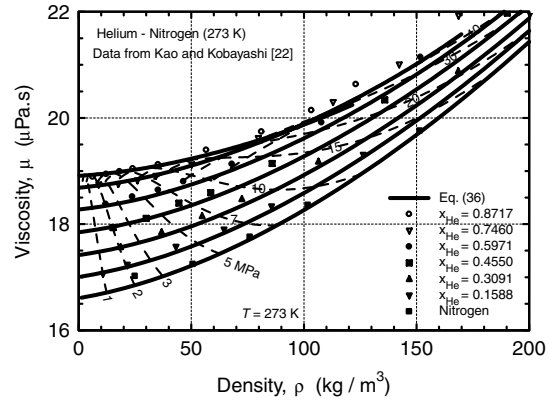

Fig. 22 Viscosity of helium–nitrogen binary gas mixtures at 273 K and different pressures.
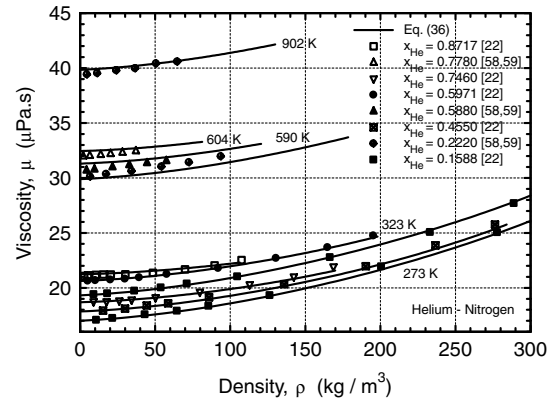

Fig. 23 Viscosity of helium–nitrogen binary gas mixtures at different pressures and temperatures.

The pseudocritical volume $\bar{V}^*$ of the binary mixtures is calculated using the first van der Waals mixing rule [Eq. (37)], and the pseudocritical temperature $\bar{T}^*$ of the binary gas mixtures is calculated using the second van der Waals mixing rule [45]:

$$\bar{V}^*\bar{T}^* = \sum_{i=1}^{N}\sum_{j=1}^{N} x_i x_j V_{ij}^* T_{ij}^* = x_1^2 V_{11}^* T_{11}^*$$
$$+ 2x_1 x_2 V_{12}^* T_{12}^* + x_2^2 V_{22}^* T_{22}^* \quad (44)$$

Among the various mixing rules reported in the literature [46–48] and examined in this work, these equations were the most accurate when compared with the compiled experimental data for the thermal conductivities of the dense binary mixtures of He–Ne, He–Ar, and Ne–Ar, with a maximum deviation of $\pm 5.0\%$ [4]. The interaction mixing rules used in this work are the linear average of the characteristic volumes and the characteristic temperatures based on the London theory of dispersion forces [36]:

$$V_{ij}^* = \frac{V_{ii}^* + V_{jj}^*}{2} \quad (45)$$

$$T_{ij}^* = \frac{4\beta}{(1+\beta)^2}\sqrt{T_{\text{cr}_i}T_{\text{cr}_j}} \quad (46)$$

where

$$\beta = \frac{V_{ii}^*}{V_{jj}^*}$$

Fig. 21 Viscosity of helium–nitrogen binary gas mixtures at low density ($P < 0.1$ MPa).

These mixing rules, which worked well for the binary mixtures of Ar–Kr, Ar–Xe, Kr–Xe, Ne–Ar, Ne–Kr, and Ne–Xe [4] are used herein for the He–N$_2$ binary mixtures.

In Eq. (42), the thermal conductivity of the dilute binary mixture is calculated using the Chapman–Enskog kinetic-theory approach [6]:

$$\bar{\lambda}^o = \left[\frac{x_1^2}{L_{11}} - \frac{2x_1x_2L_{12}}{L_{22}L_{11}} + \frac{x_2^2}{L_{22}}\right] \times \left[1 - \frac{L_{12}^2}{L_{22}L_{11}}\right]^{-1} \quad (47)$$

where

$$L_{11} = \frac{x_1^2}{\lambda_1^o} + \frac{x_1x_2}{2\lambda_{12}}$$
$$\times \frac{(15/2)m_1^2 + (25/4)m_2^2 - 3m_2^2B_{12}^* + 4m_1m_2A_{12}^*}{(m_1+m_2)^2A_{12}^*} \quad (48a)$$

$$L_{22} = \frac{x_2^2}{\lambda_2^o} + \frac{x_1x_2}{2\lambda_{12}}$$
$$\times \frac{(15/2)m_2^2 + (25/4)m_1^2 - 3m_1^2B_{12}^* + 4m_1m_2A_{12}^*}{(m_1+m_2)^2A_{12}^*} \quad (48b)$$

$$L_{12} = -\frac{x_1x_2}{2\lambda_{12}} \times \frac{m_1m_2}{(m_1+m_2)^2A_{12}^*} \times \left(\frac{55}{4} - 3B_{12}^* - 4A_{12}^*\right) \quad (48c)$$

Unlike the dynamic viscosity, the interaction coefficient L_{12} cannot be neglected. In Eqs. (48a–48c), the coefficient B_{12}^* is given by

$$B_{12}^* = \frac{5\Omega^{(1,2)*} - 4\Omega^{(1,3)*}}{\Omega^{(1,1)*}} \quad (49)$$

This coefficient, a slow-varying function of $T_{12}^* = kT/\varepsilon_{12}$, is taken as constant and equal to 1.10. For the Lennard–Jones (6,12) potential [5], $1.090 < B_{12}^* < 1.119$ in the temperature range of $2.0 < T_{12}^* < 20.0$. The interaction thermal conductivity λ_{12} in Eqs. (48a–48c), a theoretical function of temperature only, is related to the interaction viscosity μ_{12} by the following relationship:

$$\lambda_{12} = \frac{15}{4}\frac{k}{m_{12}}\mu_{12}f_{12} \quad (50)$$

The correction factor f_{12} compensates for the error resulting from using only the first-order kinetic-theory formula to determine $\bar{\lambda}^o$ [Eq. (47)]. A value $f_{12} = 0.927$ is determined from the compiled experimental data for the thermal conductivities of dilute He–N$_2$ binary mixtures (Table 3), with a maximum deviation of 3% for λ_{12}. Equation (47) predicts the thermal conductivity of the dilute He–N$_2$ mixtures with a maximum deviation of 3.5% from the reported data (Figs. 10 and 24) in the temperature range of the data, 273–591 K (Table 3).

The predictions of the thermal conductivity of the dense He–N$_2$ mixtures using Eq. (42) are also compared with the experimental data
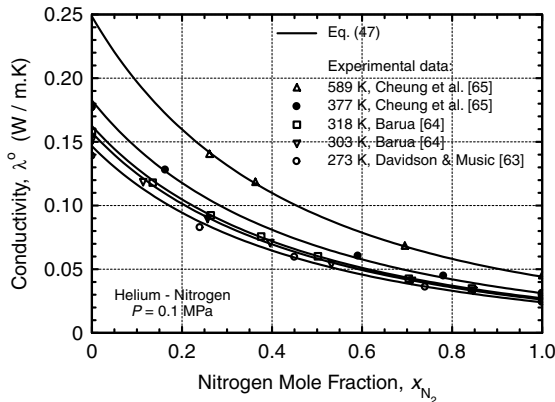

Fig. 24 Conductivity of helium–nitrogen binary gas mixtures at low density ($P < 0.1$ MPa).
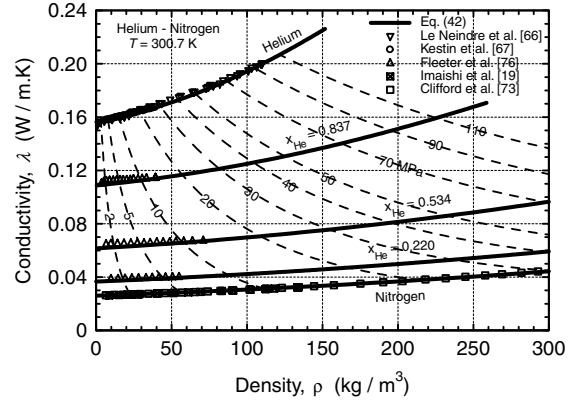

Fig. 25 Conductivity of helium–nitrogen binary gas mixtures at 301 K and different pressures.

at 301 K in Fig. 25. The predictions are in good agreement with the compiled experimental data up to 13 MPa and for reduced densities up to 0.25, with a maximum deviation of 3.8% (Fig. 10 and Table 3). Equation (42) was shown to accurately predict the thermal conductivity data of the five noble gases and the dense binary mixtures of He–Ne, He–Ar, and Ne–Ar at pressures up to 20 MPa in the temperature range of 283–603 K (corresponding to reduced densities as high as 0.53), with a maximum deviation of 5% [4].

VII. Conclusions

This work conducted a comprehensive literature review and compiled an extensive database of the measured transport and thermodynamic properties of helium and nitrogen gases and their binary mixtures at pressures up to 40 MPa and temperatures between 200 and 1500 K. In addition, semi-empirical correlations are developed for calculating the properties of these gases based on the Chapman–Enskog kinetic theory for dilute gases and the application of the law of corresponding states to account for the effects of temperature and pressure. The developed correlations are in good agreement with the compiled database of experimental measurements for helium, nitrogen, and He–N$_2$ mixtures and with thermodynamic property values recommended by NIST for helium and nitrogen.

The developed correlation for predicting the dynamic viscosity of helium is in agreement to within $\pm 1.3\%$ with that calculated based on the Chapman–Enskog kinetic theory for dilute gases and to within $\pm 1.8\%$ with the compiled data. The predictions of the thermal conductivity of nitrogen using an extended kinetic theory for molecular gases are higher than the experimental data by as much as 8% at high temperatures. The predictions of the developed correlation for the thermal conductivity of dilute He–N$_2$ gas mixtures agree with the compiled data to within 3.5% in the temperature range of the data, 273–591 K. For the dense He–N$_2$ gas mixtures, the present predictions of the thermal conductivity are in good agreement with the compiled experimental data for pressures up to 13 MPa and reduced densities up to 0.25, with a maximum deviation of 3.8%. This correlation also predicts the thermal conductivity data of the five noble gases and the dense binary mixtures of He–Ne, He–Ar, and Ne–Ar at pressures up to 20 MPa and temperatures of 283–603 K, with a maximum deviation of 5%.

The present predictions of the gas density agree with the values recommended by NIST to within less than 2%, up to the critical density, temperatures as low as 200 K, and pressures up to 20 MPa. The maximum deviation occurs in the critical region and decreases rapidly with increasing temperature and/or decreasing pressure. These ranges of applicability more than cover those of interest in potential applications to space reactor power systems and terrestrial nuclear power plants with gas-cooled reactors and CBC for energy conversion ($300 < T < 1200$ K and $1.0 < P < 10$ MPa).

The calculated values of the enthalpy, molar heat capacity, and specific heat ratio using the developed correlations in this work are in

good agreement with those recommended by NIST for helium and nitrogen at pressures up to 20 MPa and temperatures between 200 and 1500 K. The maximum deviation between predicted and recommended values of the molar heat capacity and specific heat ratio for helium is 0.5%. For nitrogen, the maximum deviation between the predicted molar heat capacity and that reported by NIST is 1.2%, with 2% for the specific heat ratio. The thermodynamic relations for calculating the entropy and speed of sound compare successfully with the values recommended by NIST for the noble gases of He, Ne, Ar, Kr, and Xe, and for nitrogen, up to 20 MPa and temperatures between 200 and 1500 K. The maximum deviation for the speed of sound in this range is 0.4% for helium, and at temperatures greater than 300 K and pressures less than 20 MPa, the maximum deviation for nitrogen is 2.3%. The error, however, decreases rapidly, becoming insignificant as the temperature increases and or the pressure decreases. Above 400 K, the error in the calculated speed of sound in nitrogen is less than 0.8% for pressures up to 20 MPa.

Acknowledgment

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