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Author(s)		
Name	**Organisation**	**E-mail**
Anders Austegard	SINTEF Energy research	Anders.Austegard@sintef.no
Jacob Stang	SINTEF Energy research	Jacob.Stang@sintef.no
Geir Skaugen	SINTEF Energy research	Geir.Skaugen@sintef.no

Abstract

Several models for transport properties that are suggested in the literature are investigated and compared with collected experimental data on mixtures of CO_2. Binary mixtures of CO_2 with the impurities Ar, CH_4, CO, H_2, N_2, N_2O, O_2 or SO_2 in temperature, pressure and concentration range relevant for transport, capture and storage. Models for viscosity, thermal conductivity and diffusivity are compared. The standard models available in the NIST Reference Fluid Thermodynamic and Transport Properties Database (REFPROP Version 9.0) are also included in the comparisons.

New regressed parameters for use with existing models by Mason and Saxena model for mixture thermal conductivity at low pressure are also presented.

The models for that have been evaluated are based on pure components estimates added for non-ideal effects of mixing at low pressure and additional effects of operating at high pressure. The recommended models from this evaluation are the use of TRAPP extended corresponding state method with Propane as the reference fluid. For the low pressure mixture term in the TRAPP method it is recommended to use the model by Reichenberg for dynamic viscosity and the model by Mason and Saxena for thermal conductivity. New regressed binary interaction coefficients for the Mason and Saxena model are presented for the mixtures CO_2-Ar, CO_2-CH_4, CO2-H_2 and CO_2-H_2O.
For prediction of thermal diffusivity at low pressure the model by Wilke and Lee.

In this work evaluation of models for pure components at higher pressure.
Very little experimental data is available for mixtures in the liquid or dense phases at higher pressure for CO_2-mixtures to evaluate the models against.

For diffusivity at high pressure, no data is found in the literature.

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1 INTRODUCTION

The transport properties measured through viscosity, thermal conductivity and diffusivity give details about transport of momentum, thermal energy and components. The physical processes are quite equal and of that reason they are treated with the same modelling approach.. For calculation of heat transfer it is important to have an accurate value of these properties, and it will help modelling and designing process units like heat exchangers and separators and to estimate friction and heat losses and the risk of corrosion during pipeline transport of CO_2 containing residual components.
The basis for this assessment are the models for prediction of transport properties that are summarized in the book by Poling, Prausnitz and O'Connell[1]. Some of the prediction models can be improved by new regression of some of the model parameters. In additions theses models, the standard prediction models from the NIST Reference Fluid Thermodynamic and Transport Properties Database (REFPROP Version 9.1) [2, 3] will be evaluated. These methods are based on extended corresponding state principles. An implementation of such methods known as TRAPP [4] are the preferred choice in commercial process simulators like Aspen Hysys and Pro/II for light hydrocarbon systems, although with their own in-house modifications to the pure component equations.[5]

The experimental data used in this study is the data that were collected by
Li et. al [6] for their review on transport property for CO_2 mixtures.

1.1 Gases of interest

This study focuses on relevant mixtures for the transport of CO_2 coming from various CO_2 capture processes. According to Li et. al. [7], a pressure range from 0.5 bars to 500 bars and a temperature range from -55°C to 1350°C are interesting,

Table 1 Temperature and pressure range of interest.

Temperature area	Max pressure	Application
-55 °C to 40°C	200	Transport and purification
4 °C to 150 °C	500	Storage
150 °C to 1350 °C	80	CO_2 capture, pre-combustion

According to Li et. al. [6] max impurity is given in Table 2

Table 2 Maximum mole fraction of impurities.

Component	N_2	O_2	Ar	SO_2	H_2S + COS	NO_x	CO	H_2	CH_4	H_2O	Amines	NH_3
Max impurity in mol %	10%	5%	3.5%	1.5%	1.5%	0.3%	0.2%	4%	4%	6.5%	0.01%	3%

In this study, pure component and binary mixtures of CO_2 and the following gases are assessed: N_2, O_2, Ar, SO_2, N_2O, CO, H_2, CH_4, H_2O and some Amines.

2 MODEL FOR VISCOSITY

Viscosity gives the shear stress $\tau = \eta \frac{dv}{dz}$ with symbol η and unit Ns/m^2 = kg/ms eventually poise (P) 1P = 10^{-7} Ns/m^2

The value is

$$\eta = \frac{\rho' vL}{3} = (const)\frac{T^{\frac{1}{2}}M^{\frac{1}{2}}}{\sigma^2} \qquad (1)$$

where ρ is density (kg/m^3), v is the average molecule velocity, L is the mean free path and σ is the collision diameter. When the temperature increases the molecules come closer to each other and the collision diameter reduce. Of that reason viscosity increases more than T$^{1/2}$.

This model is valid for gases, also at high pressure, but it is not valid for liquids that have high viscosity even when the free path (L) is low.

There are models for single components at low pressure, for mixture at low pressure, single components at high density, mixtures at high density and liquids. We are here particularly interested in models for mixtures at high density. High density means that the gas is compressed to a pressure where ideal gas no longer can be assumed. This goes into the liquid area, but it is not made for liquids. Here usually the word high pressure is used instead of high density.

In addition to the methods described here, models in REFPROP[3] also predict viscosity for mixtures at high pressure.

2.1 Viscosity models for single components at low pressure

2.1.1 Curve fitting

For single component at low pressure the viscosity is only a function of temperature, and one can do direct curve fitting of experimental data of viscosity as function of temperature. Zeberg-Mikkelsen[8] have done curve fitting for argon, hydrogen, methane, neon, nitrogen and oxygen.

2.1.2 Statistical mechanics

For ideal gas with spheres, the viscosity can be predicted from statistical mechanics. In accordance with the Chapman-Enskog model[9], it will become

$$\eta = 2.669 \times 10^{-6}\frac{(MT)^{1/2}}{\sigma^2\Omega_v},$$

$$\Omega_v = f(T^*) = [A(T^*)^{-B}] + C[\exp(-DT^*)] + E[\exp(-FT^*)], T^* = \frac{T}{\varepsilon/k} \qquad (2)$$

where Ω_v is the Lennard-Jones viscosity collision integral. It tells something about how close the molecules get before they influence each other's and how it reduces with temperature. It is one for no attacking hard spheres. This formula gives $\Omega_v \approx 1.6(T^*)^{-0.5}$. This formula is similar to Eq(1).

Hydrogen is also adapted to measurements of McCarty[10] trough NIST[2]. Two types of hydrogen exists orthohydrogen and parahydrogen. In this assessment it is assumed normal hydrogen that consists of 75% orthohydrogen and 25% parahydrogen. The difference in prediction of viscosity for ortho- and parahydrogen is small. However for prediction of thermal conductivity the difference is quite large between the two states (approximate 20%) so the fraction between ortho- and para need to be known.

Chung et.al[11] have made a general model for viscosity. It requires critical temperature and volume, acentric factor and dipole moment (in Debyes) for all materials. For polar substances it requires a special correction depending on the number of –OH groups. It is not usable for quantum gases (He, H_2, D_2). Poling[1] give a table over deviation for given gases. There the average deviation for Chung' model is 1.9%. Because of the low deviation and that the required physical properties are available for the relevant components, the Chung's method is one of the models that is investigated for CO_2 mixtures.

In addition Lucas [12], Reichenberg[13] have proposed alternative models based on the corresponding state principle for pure components. Lucas has not included associate gases and Reichenberg is only focussing on organic gases.

Table 3 Overview of models for predicting viscosity for pure components

Modell	**Lucas[12]**	**Reichen-berg[13]**	**Chung[11]**	**Curve fitting of experiment**
Year	1984	1979	1988	
Takes quantum fluid (He, H_2, D_2)	Yes	No	No	Yes
Takes polar gases as water	No	Yes	Yes	Yes
Takes non-organic gases	Yes	No	Yes	Yes
Average deviation, Poling[i, 1]	3.0%	1.9%	1.9%	
Number of fluid not included for deviation (Total 29 fluids)	0	1	7	
Parameters required except critical parameters	Dipole momentum, factor for quantum gases	Dipole momentum, groups in the molecule	Dipole momentum, factor for polar molecules or –OH groups	Experimental data
Evaluated	No	No	Yes	Yes, for some gases
Default model			When single component is not available	When available
Comment	High deviation, does not take polar gases	Only for organic molecules	Can be used for all used components except H_2 where curve fitting can be used	Best choice when available

Chung's model can also be extended to include mixtures and gases at higher pressure.
The most used models for prediction the transport properties for mixtures, viscosity at higher pressure and the thermal conductivity are based on results from the pure component viscosity predictions. So if the viscosity for pure components is wrong, viscosity for mixture, and high pressure as well as conductivity also become wrong

2.1.3 Chung's[11] model

Chung's model uses the Chapman-Enskog model as shown in Eq(2) with critical volume and temperature to get $\epsilon/k(=T_c/1.2593)$ and with hard sphere diameter $\sigma = 0.809V_c^{1/3}$. Then the viscosity is multiplied with a factor that depends on the acentric factor, the dipole moment and an association factor for highly polar gases.

The viscosity is then:

$$\eta = 40.785\frac{F_c(MT)^{1/2}}{V_c'^{2/3}\Omega_v} \tag{3}$$

η =viscosity, μP
M = Molecular weight g/mol
T = temperature, K
T_c= Critical temperature
V'_c = Critical volume, cm^3/mol
Ω_v = Viscosity collision integral from (2) and $T^* = 1.2593\ T/T_c$

$$F_c = 1 - 0.2756\omega + 0.059035\mu_r^4 + \kappa$$

κ = Special parameter for polar components. Value is set by Chung and given by Polling [1]
κ = 0.076 for H_2O.

2.1.4 Viscosity models from curve fitting to experimental data

Where experimental data is available for a pure component is available, an alternative to using Chung's model is to fit the data to a chosen viscosity model dependent of the temperature alone. As seen from Eq(2), the term $T^{1/2}$ should be a part of the expression. One such model is proposed by Zeberg-Mikkelsen[8].

Zeberg-Mikkelsen model uses:

$$\eta = A_1T^{1/2} + A_2T^{A_3} \tag{4}$$

Here the constants A_1, A_2 and A_3 are found from regression. However, when trying to adapt the viscosity, the function

$$\eta = A_1T^{1/2} + A_2T^{2/2} + A_3T^{3/2} + .. \tag{5}$$

is easier to adapt and it is as good as the Zeberg-Mikkelsen's formula.

None of the models (Eq 4 or 5) can be extrapolated outside the (experimental) range for the curve fitting. Outside that range the Chung's model is used in this evaluation scaled to give equal values as the regressed model at the temperatures T_{min} and T_{max}.
For temperatures below T_{min} the following formula is used:

$$\eta(T) = \eta_{Chung}(T)\frac{\eta_{adapted}(T_{min})}{\eta_{\mathrm{Chung}}(\mathrm{T}_{\mathrm{min}})}$$

and similar formula is used above T_{max}.

For Hydrogen, the Zeberg-Mikkelsen formula is only fitted down to 200K, but in the evaluation, results for normal hydrogen from REFPROP are used below 200K. The results from REFPROP for hydrogen are from McCarty[10] (Prior to version 9.1)

A new viscosity model for Normal Hydrogen is available from REFPROP v9.1 from [14]

For all the components that have new regressed parameters, Appendix A shows the formula, parameter A_i and where the parameters are coming from.

2.2 Viscosity for mixtures at low pressure

For mixture one can use individual viscosity models for each of the components or find average values for the input parameters like the acentric factor and the critical parameters and use that in the chosen viscosity model. When the viscosities for each of the components in the mixture is calculated this will give more flexibility when choosing a specific prediction model for the components. A list of some of the suggested models for prediction of viscosity at low pressure is listed in Table 4.

Table 4 Models for viscosity at low pressure

Model	Lucas[12]	Reichenberg[13]	Chung[11]	Wilke[15]	Herning[16]
Year	1984	1979	1988	1950	1936
Need pure gas properties	No	Yes	No	Yes	Yes
Need except in addition to critical parameters	Dipole momentum	Dipole momentum	Dipole momentum, Kappa^1	No	No
Can use binary parameters			Yes		
Step in the function	Yes		No		
Max error Poling	13% CF – SF	4.8% NH_3-H_2	23% N_2-H_2	12% N_2-H_2	19% NH_3-H_2
Max error without H_2	13% CF-SF	4.6% CF_4-SF_6	11% NH_3-CH_3NH_2	6% N_2-C_7H_{16}	11% N_2-C_7H_{16}
Usable with hydrogen	Yes	Yes	No		
Evaluated	No	Yes	Yes	No	No
Default		Yes			
Computational issues	Have step in the value, not suitable for numeric use	Low deviation when viscosity for each component is known	Best model when viscosity for each component is not known.	High deviation	High deviation

^{1}: Special parameter for polar gases

All the methods are described by Poling[1]

2.2.1 Reichenberg model

Reichenberg uses the viscosity for the pure components at the system temperature as a parameter in the mixture model. That is a benefit since the viscosity for pure components is much easier to estimate compared to a mixture, The pure component low pressure viscosity can also be found directly by regression from available experiments.
Critical temperature, mole weight and dipole moment are used in the mixing rules to calculate the total mixture viscosity.

2.2.2 Chung's model

In opposite to Reichenberg model Chung do not use the individual pure component viscosity in the model. Instead the same model as for pure components is used with average values of $\sigma, \epsilon/k, \kappa, M$, w for the mixture depending of the composition.. The average values for the input parameters are calculated from quadratic mixing rules with specific combining rules for each input property. When pure component viscosity is calculated with the mixture model, the combining rules model simplifies this to be exactly like the model shown for pure components in, Eq. (2b).

2.3 Viscosity for mixtures at higher pressure

The viscosity for a mixture at higher temperature can be estimated by getting average values of the parameters like critical pressure and temperature and then use a model for a pure component to calculate viscosity. The other method is to use viscosity at low pressure for a mixture and then extend it to higher pressure.

Table 5 Models for viscosity at higher pressure

Model	**Lucas[12]**	**Chung[11]**	**TRAPP Hubert[17]**	**REFPROP V9.0, NIST[2]**
Year	1984	1988	1996	2013
Uses viscosity for mixture at low pressure	No	No	Yes	-
Applicable close to liquid region	No	No	Yes	Yes
Evaluated	No	No	Yes	Yes

Here only the TRAPP method is evaluated (besides the default method in REFPROP V9.0) since it can give viscosity close to the liquid region. It is also a good method when the viscosity at low pressure is known. Besides, it is the newest method.

2.3.1 The TRAPP method

The TRAPP[4] method is an extended corresponding state method for prediction of density and transport properties like viscosity and thermal conductivity for pure components and mixtures. The method is described by Poling[1] and Huber[17] and is shown in Eq.6

$$\eta_m(T,\rho,x) = \eta_m^0(T,x) + F_{\eta m}\big(\eta^R(T_0,\rho_0) - \eta^{R0}(T_0)\big) + \Delta\eta^{ENSKOG}(T,\rho,x) - \Delta\eta^{ENSKOG,0}(T,x) \tag{6}$$

Here the variables are:

η_m Viscosity for the mixture

η_m^0 Viscosity for the mixture at low pressure. Here one selects a model in section 2.2

η^R	Viscosity for the reference fluid, Propane is at a scaled temperature and density.
$T_0 = T/f_m$	The temperature is divided on a factor f_m to get the temperature of the reference fluid that gives similar properties.
$\rho_0 = \rho h_m$	The density is multiplied with a factor h_m. The factors h_m and f_m are based on the evaporating pressure.
η^{R0}	Viscosity for reference fluid at low pressure
$\Delta\eta^{ENSKOG}(T,\rho,x)$	Enskog addition that accounts for the situation where one molecule is much larger than the other molecule. $\Delta\eta^{ENSKOG}$=0 for pure components.
$\Delta\eta^{ENSKOG,0}(T,x)$	Enskog term at low pressure.

The basis for the TRAPP method is to relate the residual viscosity for a gas mixture (or pure fluid) to the residual viscosity of a reference fluid. The residual viscosity is the difference between the viscosity at the system state (T, P) and the viscosity at low pressure (T, P=1atm). The system temperature and pressure (or density) have to be scaled to corresponding reference fluid temperature T_0 and density ρ_0. For mixtures, mixture rules are applied to the "correspondence" factor.

It exist various variants of the TRAPP method. In this evaluation Propane is used as reference fluid. Polling [1] use then prediction model from Younglove [18] to calculate the viscosity for the reference fluid. However since the development of this model, more data has been collected and Vogel (1998)[19] has made a newer correlation for viscosity of Propane. The Vogel model is also used in REFPROP[3].

The Enskog term[20] accounts for the situation where the sizes of the molecules are different. It is zero for pure component, but it is not zero at low pressure. This effect is already included in the term η_m^0, and to avoid that the effect is included twice the term $\Delta\eta^{ENSKOG,0}$ needs to be subtracted. This is not done in the implementation shown by Polling[1], but it is done in the implementation done for the evaluations in this assessment.. The difference between including the Enskog term twice and not can be seen in Figure 1 for mixture H_2-CO_2.

The TRAPP model is described in detail by Polling[1]. The term $\Delta\eta^{ENSKOG,0}(T,x)$ is calculated with ρ = 0.33 mol/m^3. For component *i* the component scaling factors are calculated from:

$$\begin{aligned} f_i &= \frac{T_{c,i}}{T_c^R}\left[1+(\omega_i-\omega^R)\left(0.05203-0.7498\ln\left(\frac{T}{T_{c,i}}\right)\right)\right] \\ h_i &= \frac{\rho_c^R}{\rho_{c,i}}\frac{Z_c^R}{Z_{c,i}}\left[1+(\omega_i-\omega^R)\left(0.1436-0.2822\ln\left(\frac{T}{T_{c,i}}\right)\right)\right] \end{aligned} \tag{7}$$

where $T_{c,i}$, $\rho_{c,i}$, and $Z_{c,i}$are the critical properties for component *i* .

For the reference fluid, propane it is used:

$$T_c^R = 369.8\text{K}, \qquad \rho_c^R = 4.914\ \text{mol/L}, \qquad Z_c^R = 0.281, \qquad \omega^R = 0.152\ \text{Debye}$$

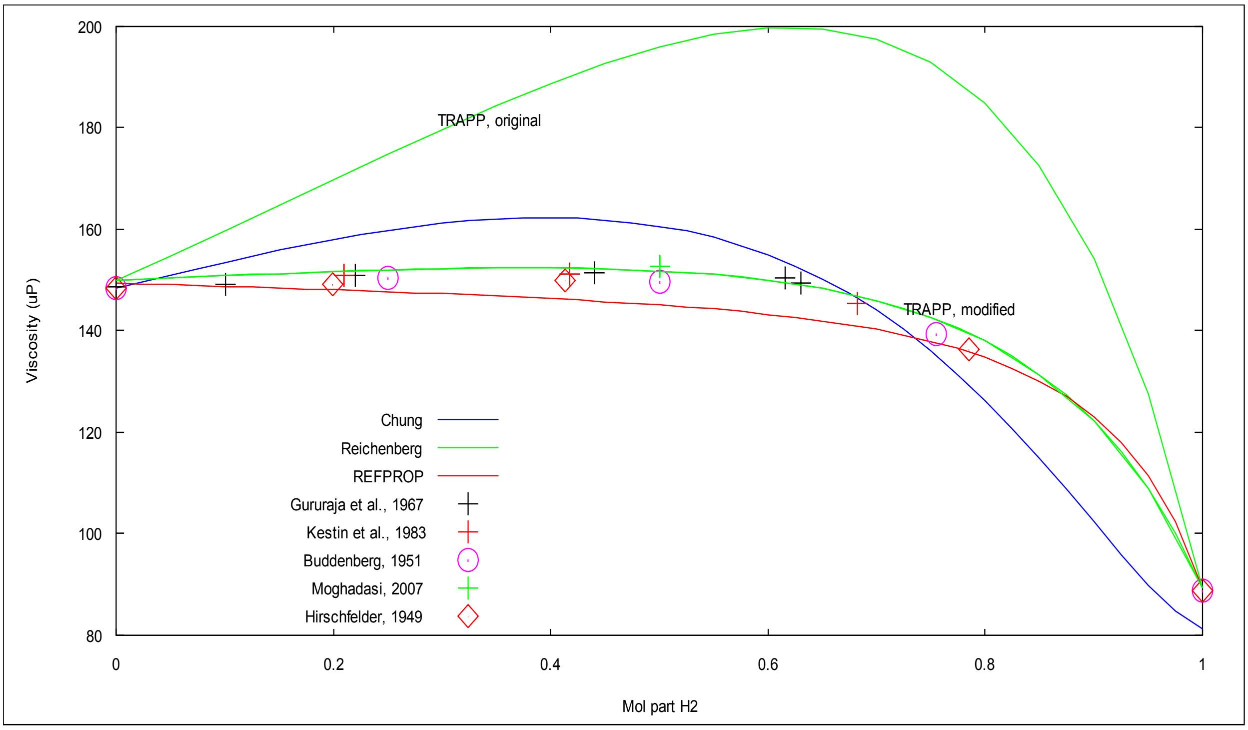

Figure 1 The TRAPP method applied with and without subtraction of the term $\Delta\eta^{ENSKOG,0}$ compared to experimental values and prediction from REFPROP v9.0

2.4 Liquid viscosity

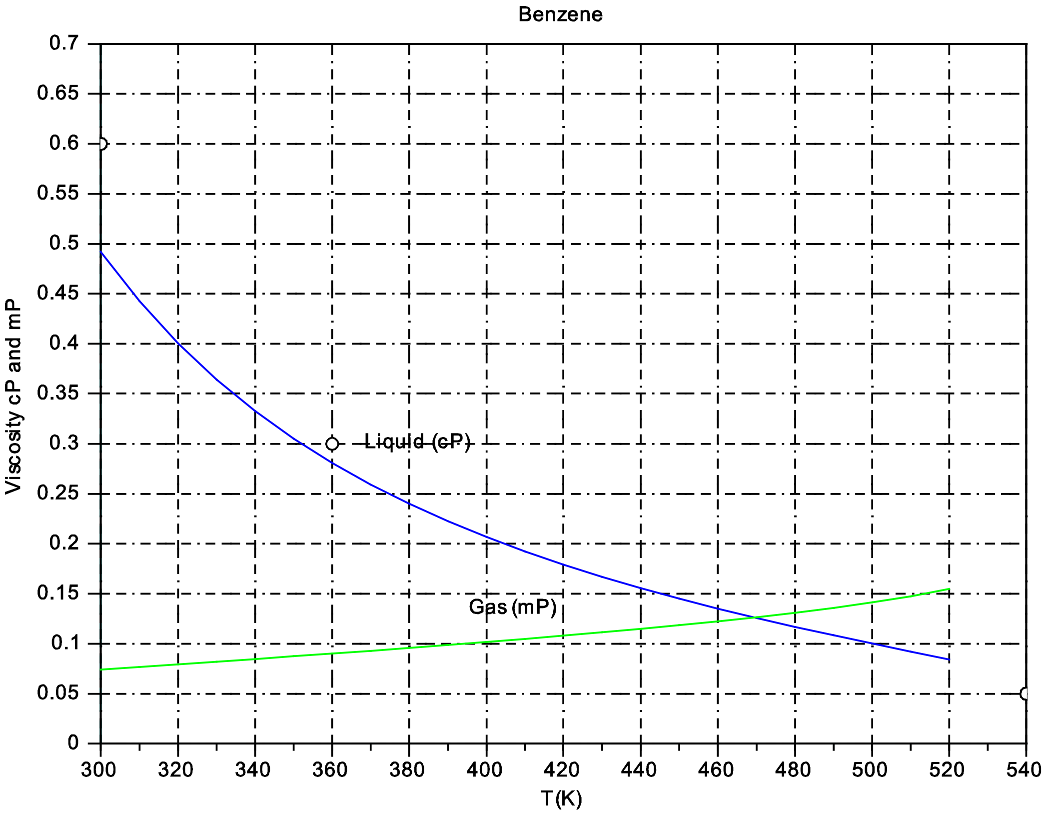

Figure 2 Liquid and gas viscosity calculated with the TRAPP method for Benzene. Measures from Polling[1] fig 9-9.

Figure 2 shows the gas and liquid viscosity for Benzene, calculated with TRAPP method in which the Chung method is used for ideal gas. The TRAPP method is based on Propane, but Benzene is quite equal Propane with the consequence that the results are good for this method although not intended for use in the liquid area. However, for water at 100 °C and 1 Bar., the TRAPP method predicted 17.7 mP where the measured value show 2.8 mP

3 MODEL FOR THERMAL CONDUCTIVITY

Thermal conductivity λ gives the heat transport $Q = A\lambda\ (dT)/(dz)$. It has the symbol λ and unit W/mK.

The value is:

$$\lambda = \frac{vLC_v\rho}{3} \tag{8}$$

where *n* is molar density (mol/m^3), v is the average molecular velocity and L is mean free path. This formula is very similar to the formula in Eq(1) for viscosity.

For monoatomic gases the formula can be written:

$$\lambda = \frac{25}{32}(\pi m_m kT)^{1/2}\frac{C_v/M}{\pi\sigma^2\Omega_v} \tag{9}$$

This is very similar to Eq(2) for viscosity. The dimensionless group$\frac{\lambda M}{\eta C_v}$ in Eq,(9) is called the Eucken factor:

$$\text{Eucken} = \frac{\lambda M}{\eta C_v} \tag{10}$$

Dividing Eq(9) with Eq(2) provides an Eucken factor very close to 2.5 which holds true for monoatomic gases,. For gases that are not monoatomic, one has rotation energy and vibration energy that reduce the Eucken number. For monoatomic gases one has a $C_{v} = 3/2$ R and it increases with the degrees of freedom. When C_v increases the Eucken number goes down.

This illustrates the close relationship between thermal conductivity and viscosity, and many models use the viscosity as part of the thermal conductivity model. The approach for modelling thermal conductivity of high pressure gas mixtures is constructed in the same way as for the viscosity. First we need a model for pure gases, then extend it to mixtures, and then extended it to mixtures at high pressure. High pressure means that the molecules are close together. This is also the case for the liquid phases in general but a high pressure model is not intended for use to calculate liquid conductivity and the error can become high when used for liquids.

3.1 Thermal conductivity for single components

Table 6 Model used for single components

Model	**Eucken**	**Modified Eucken[21]**	**Stiel and Thodos[22]**	**Roy and Thodos[23]**	**Chung et al[11]**	**Curve fitting of experiments**
Year	Early	1962	1964	1968	1988	
Uses viscosity	Yes	Yes	Yes	No	Yes	No
Calculate Eucken factor	Yes	Yes	Yes	No	Yes	No
Uses heat capacity	Yes	Yes	Yes	No	Yes	No
Other requirements					Acentric factor, special parameters for polar molecules	
Max error[1]	23%	33%	22%	17%	17%	
Max error CO_2[1]	13%	15%	10%	No data	11%	
Usable for polar components	No	No	No	No	Yes	Yes
Used in the evaluation	No	No	No	No	Yes	Yes, for some gases
Default model					When curve fitting is not available	When available
Comments	High deviation, not for polar			Require special parameters	Available for polar, few extra parameters	Best choice when available

[1]: From Poling[1]. They have collected data and done comparison with the models.

As seen in Table 6 most of the models use the Eucken factor. The three first models; Eucken, modified Eucken and Stiel and Thodos divide the internal energy into translation energy and an internal energy (Rotation energy and vibration energy in the molecules that increase C_v). The Eucken model reduces the Eucken factor quite much with an increase in C_v. It is less reduced with the modified Eucken model while the Stiel and Thodos model lies somewhere in between.

3.1.1 Chung's model

Chung et al. also use the Eucken number, but they make a regression based on C_v/R, acentric factor and temperature. They also extend this model to mixtures and to mixtures at higher pressure. They also use a factor β that is a function of an acentric factor, but this needs to be especially set for polar compounds. The problem, however, is that then it cannot be expanded to mixtures.

3.1.2 Curve fitting to experimental data

NIST[2] has collected data from numerous experiments and curve-fitted the experiments. Many components are tested with the model from Chung[9]. Those with considerable deviations are re-regressed,. The data is fitted to the formula:

$$\lambda = \frac{A_0 + A_1 x + A_2 x^2 + A_3 x^3}{1000} \qquad x = \left(\frac{T}{100}\right)^{1/2} \tag{11}$$

This improved the model, resulting in a difference of about 0.1%, max value 0.5% for H_2.

3.2 Thermal conductivity for mixtures at low pressure

Table 7 Thermal conductivity for mixtures at low pressure

Model	**Mason and Saxena[24] with viscosity**	**Mason and Saxena without viscosity**	**Corresponding state, Chung[11]**	**Mason and Saxena with adjusted $\varepsilon_{i,i}$**
Year	1958	1958	1988	
Need viscosity for components	Yes	No	No[1]	No
Need thermal conductivity for components	Yes	Yes	No	Yes
Work for polar components	No	No	No	Yes
Others				Need at least one measure of a mixture value
Evaluated	Yes	Yes	Yes[2]	Yes[3]
Default	No	No	No	Yes
Comment	Need viscosities	Low deviation	Depending on Chung's equation for components	Lower deviation than Mason and Saxena

[1]:The formula contains the viscosity but it consistently uses Chung's model for viscosity for mixture at low pressure.
[2]: Uses Chung's equation for high pressure
[3]: For binary parameters, see Appendix E

Table 7 shows models for thermal conductivity for low pressure. All of them are described by Poling[1]. Except Chung's model, all use Wassiljewa's equation for the mixture conductivity

$$\lambda_m = \sum_{i=1}^{n} \frac{(x_i \lambda_i)}{\left(\sum_{j=1}^{n} x_j A_{ij}\right)}, A_{ii} = 1 \tag{12}$$

In the Wasilijewa equation the binary interaction terms, $A_{ij,}$ need to be determined for each pair. Several correlations for this are proposed in the literature. Another alternative is to measure the conductivity of binary mixtures. Then the A_{ij} is known when the conductivity of the two components is known. When one of the components is polar, this is the recommended way to determine A_{ij} since models examined in the literature are not found to be particularly accurate. Next, some of the evaluated empirical models for calculations of A_{ij} will be discussed.

3.2.1 Mason and Saxena model with viscosity

Mason and Saxena[20] (sometimes called Mason S.) use the following value for A_{ij} in Eq(12)

$$A_{ij} = \frac{\varepsilon_{i,j}\left(1+\left(\frac{\lambda_{tr,i}}{\lambda_{tr,j}}\right)^{\frac{1}{2}}\left(\frac{M_i}{M_j}\right)^{\frac{1}{4}}\right)^2}{[8(1+M_1/M_2)]^{1/2}} \tag{13}$$

In Mason and Saxena's model $\varepsilon_{i,j}$ is a numerical constant close to unity, independent of components, and usually set to a value of 1.0 [1]. Different values have been proposed and for this assessment, a regression has been performed for relevant CO_2 binary mixtures as discussed in section 3.2.3

The relation between the conductivities, $\left(\frac{\lambda_{tr,i}}{\lambda_{tr,j}}\right)$ use the monoatomic values of thermal conductivities and can be calculated from the viscosity by:

$$\frac{\lambda_{tr,i}}{\lambda_{tr,j}} = \frac{\eta_i/M_i}{\eta_j/M_j} \tag{14}$$

3.2.2 Mason and Saxena model without viscosity

An alternative to Eq(14) for calculation of the relation between conductivities in Eq(13) to get A_{ij} is shown in Eq(15):

$$\frac{\lambda_{tr,i}}{\lambda_{tr,j}} = \frac{f(T_{r,i})/\Gamma_i}{f(T_{r,j})/\Gamma_j}, \qquad f(T_r) = e^{0.0464T_r} - e^{-0.2412T_r}, \qquad \Gamma_i = 210\left(\frac{T_{ci}M_i^3}{P_{ci}^4}\right)^{\frac{1}{6}} \tag{15}$$

In this way we are not depending on how the viscosity is calculated. The results on the prediction of conductivity from these two methods show only a small difference. For the mixture CO_2-H_2 with very large difference in the molecules, and over 10 times difference in viscosity, the maximum difference on the prediction of thermal conductivity was 4%

3.2.3 Mason and Saxena with regressed binary interaction coefficients

Here $\varepsilon_{i,j}$ is regressed for each binary system where experimental data is available and set to 1.0 otherwise. When $\varepsilon_{i,j}$ is regressed, this equation reduces to the Wassiljewa equation (Eq(12)), except for a week temperature dependence. Eq(15) is used to get $\lambda_{tr,i}/\lambda_{tr,j}$ and not the viscosity, to get an value independent of viscosity model. The resulting binary coefficients are shown in Table 8 and further elaborated in Appendix E.

Table 8 Regressed binary interaction coefficient. $\varepsilon_{i,j}$ for the Mason-Saxena model for A_{ij}

Mixture	CO_2-Ar	CO_2-CH_4	CO_2-H_2	CO_2-H_2O	All other binaries

Binary coefficient $\varepsilon_{i,j}$	1.054	1.110	1.268	0.911	1.0

3.2.4 Chung's model

This model is the same as for gas mixtures at high pressure described in 3.3.1

3.3 Thermal conductivity for mixtures at high pressure

The thermal conductivity increases with the pressure. In addition it is an enhancement close to the critical point for a pure component, at what point the conductivity increases several times. This effect is illustrated in Figure 3 for pure CO_2 and discussed further in section 3.3.3

This is different compared to the viscosity.

Table 9 Models used for thermal conductivity at high temperatures

Model	**Stiel and Thodos[22]**	**Chung et al[11]**	**TRAPP, Huber[17]**	**REFPROP, NIST[2]**
Year	1964	1988	1996	
Uses conductivity at low pressure for mixture	Yes	No	Yes	
Includes enhancement close to critical point	No	No	No	Yes
Other parameters needed except critical parameters and mole-weight	No	Acentric factor, dipole moment, own parameter for polar fluids	Acentric factor,	Include fitting of experiments
Good at single component high pressure	Ok	Ok	Ok	Very good
Good at mixture	Ok	Ok	Ok	Not
Evaluated	No	Yes	Yes	Yes
Default	No	No	Yes	No
Comments	To large deviation		Show best overall result	

3.3.1 Chung's model

Poling[1] have described the method in details. Firstly, on the heat capacity, critical parameters are calculated for the mixture. Then this is used to calculate viscosity for the mixture after Chung's formula. Subsequently, the conductivity is calculated for low pressure and finally it is extended to higher pressure. It uses the heat capacity, acentric factor, dipole moment and a factor κ for polar components. κ is described in section 2.1.1.

The benefit of Chung's model is that it does not need measured conductivities.

3.3.2 TRAPP model

The TRAPP model uses the same modelling approach as for viscosity. It is described in details by Poling[1]. It calculates the difference between thermal conductivity for gas mixture at low pressure and at given pressure. It uses Propane as a reference fluid and scales the density and temperature in the same way as for the viscosity. It does not use the Enskog correction. The formula becomes:

$$\lambda_m(T,\rho,x) = \lambda_m^0(T,x) + F_{\lambda m}(T,x)X_{\lambda m}(x)[\lambda^R(T/f_m,\rho h_m) - \lambda^R(T/f_m)] \qquad (16)$$

Here λ^0_m is the thermal conductivity for mixture at low pressure, x is the composition, and λ^R is the conductivity at the reference fluid at a scaled density and temperature. The scale factors f_m and h_m are calculated in the same way as for the viscosity.

3.3.3 Enhancement of thermal conductivity close to the critical point

The critical enhancement of the thermal conductivity is only accounted for in models for pure components. In the vicinity of the "pseudo-critical" point for mixtures this enhancement will not occur. As a consequence mixture models based on corresponding state principles do not include this enhancement term for the reference fluid. So using a corresponding state method also for a pure component will underestimate the thermal conductivity in the critical region. This is illustrated in Figure 3 for pure CO_2 when the model from [25] used in REFPROP is compared to the TRAPP method.

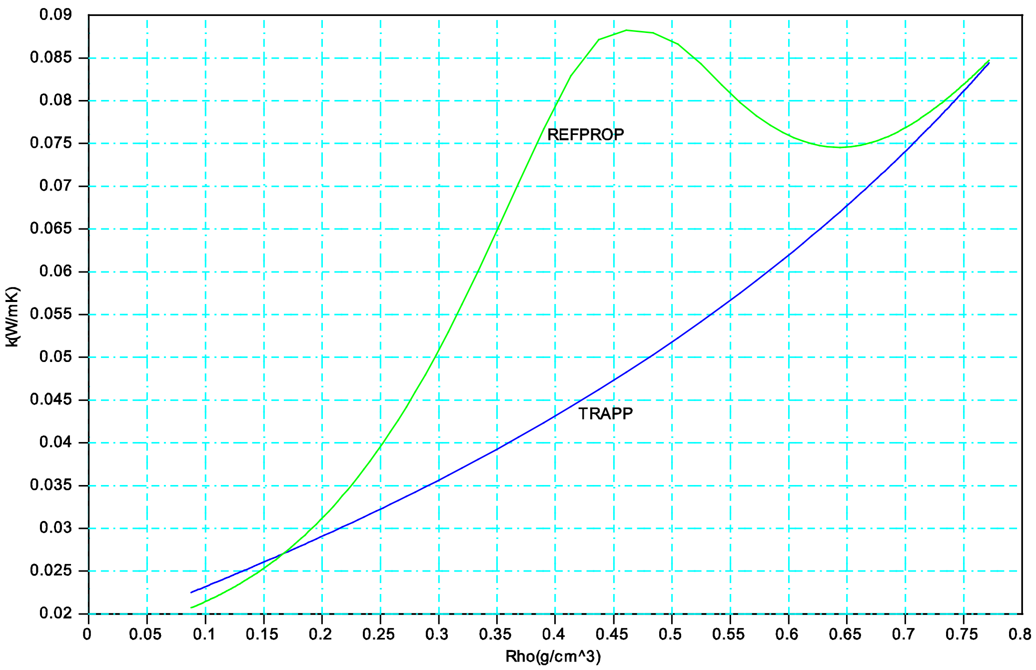

Figure 3: Comparison of TRAPP and REFPROP using the model from Vesovic [25] for CO_2 at 34.7 °C

3.4 Thermal conductivity for liquids

Table 10 Calculated liquid thermal conductivity (W/m K)

Liquid	Temperature(C)	Measured[26]	TRAPP	Chung	REFPROP
Benzene	20	0.154	0.143	0.142	0.143
n-Pentane	20	0.136	0.116	0.110	0.117
n-Octane	20	0.147	0.107	0.128	0.119
Water	0	0.558	1.76	0.691	0.598

Water	100	0.682	1.34	0.762	--[1]
Average error			72%	15%	
Average error Pentane/Octane			21%	16%	17%

Table 10 shows the calculated conductivity for the liquids. None of the methods intend to calculate conductivity deep into the liquid phase. One sees that Chung's method gives best results both for polar fluids as water and nonpolar hydrocarbons.

4 MODELS FOR DIFFUSIVITY

In a multi component system the evaporating is dependent on diffusivity. One example is evaporating at a temperature below 100 °C (and 1 atmosphere pressure), where the water pressure at the surface is the evaporating pressure. The evaporation velocity depends on transport of the vapour that again depends on the diffusion of vapour through the boundary layer between water and air.

Diffusion can occur due to of the temperature gradient, external force, pressure or concentration gradients. We will here concentrate on concentration gradient.

The mole flux of component *i* can be written:

$$J_i = N_i - x_i \sum_{j=1}^{n} N_j \tag{17}$$

This mean that diffusive flux of component *i* is a flux of component *i* minus flux of the mixture times concentration of component *i*.

Diffusive flux is then for a binary of component 1 and 2:

$$J_1 = -\rho D_{12}\frac{dx_1}{dz}, J_2 = -\rho D_{12}\frac{dx_2}{dz} \tag{18}$$

Here D_{12} is the binary diffusion flux between component 1 and 2.

The theory for diffusivity has the basis in the Chapmen-Enskog theory that is used for viscosity. The diffusivity can be expresses as shown Eq(19):

$$D = \frac{vL}{3} \tag{19}$$

where *L* is the mean free path, and *v* is the average molecular velocity

The theoretical equation for diffusion in binary gas mixtures developed from solving the Boltzmann equation is shown in Eq (20)

$$D_{AB} = \frac{3}{16}\frac{(4\pi kT/M_{AB})^{1/2}}{\rho N_A \pi \sigma_{AB}^2 \Omega_D} f_D = \frac{0.00266T^{3/2}}{PM_{AB}^{1/2}\sigma_{AB}^2\Omega_D} \tag{20}$$

Here mole weight M_{AB} (g/mol), atom diameter σ_{AB}(Å) and diffusion collision integral Ω_D are defined in Eq(21). The last part assumes $f_D=1$ and ideal gas.

The model is only valid for low to moderate pressures.

4.1 Diffusion of Binary gas mixtures at low pressure

Table 11 Models evaluated for binary gas mixtures at low pressure

Model	Wilke and Lee[27]	Fuller et al[28]	Theoretical ,Eq(20)	Broakaw[29]
Year	1955	1969		1969
Parameter needed except critical parameters and mole weight	Liquid density	Diffusion volume, based on chemical formula	Lennard-Jones energy	Polar momentum and Lennard Jones energy
Average error Polling [1]	10%	5%	8%	Only calculated for some binaries
Max error Polling	25%, Ar-O_2	25% Air –SO_2	25% Ethylene – water	
Evaluated	Yes	Yes	No	No
Default	When diffusion volume is not available	When diffusion volume is available		
Comment	Only method without other parameters	Lowest deviation	Requires Lenard Jones Energy	

Curve fitting of experiments for diffusion coefficients are not an alternative as for viscosity and thermal conductivity since individual regressions for each binary will be needed, and not only for each component. Alternatively one can use a model based on curve fitting of data for components.

Generally all models are based on the theoretical model. It is various ways to get the molecule diameter and some corrections

4.1.1 Wilke and Lee model[27]

This model is the only model that does not require special component parameters for calculating of diffusivity. It is based on the liquid volume. In the evaluation, the normal boiling point temperature (at 1 atm) is known for all components, and the corresponding volume is calculated with the Lee-Kesler equation of state. Since it does not require special parameters that are adapted, this is also the method that gives the largest deviations.

It is a variation of the theoretical model in Eq(20) and it provides the diffusivity:

$$\begin{gathered}
D_{\mathrm{AB}} = \frac{\left[3.03 - \left(0.98/M_{\mathrm{AB}}^{1/2}\right)\right](10^{-3})T^{3/2}}{P M_{\mathrm{AB}}^{1/2}\sigma_{\mathrm{AB}}^{2}\Omega_D} \\
M_{AB} = \frac{2}{1/M_A + 1/M_B}, \qquad \sigma_{AB} = \frac{\sigma_A + \sigma_B}{2}, \qquad \sigma = 1.18\, V_b^{1/3} \\
\Omega_D = \frac{1.06036}{(T^*)^{0.15610}} + \frac{0.1930}{\exp(0.47635T^*)} + \frac{1.03587}{\exp(1.52996T^*)} + \frac{1.76474}{\exp(3.89411T^*)} \\
T^* = \frac{T}{\varepsilon_{AB}/k}, \qquad \varepsilon_{AB} = (\varepsilon_A\varepsilon_B)^{1/2}, \qquad \frac{\varepsilon}{k} = 1.15\, T_b
\end{gathered} \tag{21}$$

Here M_A and M_B are molecular weights for component A and B, T_b is boiling temperature at 1 atm for each of the components and V_b is liquid volume at the normal boiling point for each of the components in cm^3/mol. The variable σ is hard sphere diameter in Å and Ω_D is the Lennard-Jones potential.

4.1.2 Fuller et al model[28]

This model calculates a diffusion volume for each atom, some groups and some of the components.

It provides the following diffusion coefficient:

$$D_{\mathrm{AB}} = \frac{0.00143 T^{1.75}}{P M_{\mathrm{AB}}^{1/2} \left[(\Sigma_v)_{\mathrm{A}}^{1/3} + (\Sigma_{\mathrm{v}})_{\mathrm{B}}^{1/3} \right]^2} \tag{22}$$

M_{AB} is defined in Eq(21), Σ_v is found by summing atomic diffusion volumes. The atomic diffusion volumes are tabulated either for simple molecules or for each atom for other more complex molecules. These are based on regression and tabulated by [28] and also listed by Poling[1] (Table 11.1)

4.2 Diffusion coefficient for binary gas mixtures at high pressure

Table 12 Models used for diffusion coefficient at high pressure

Model	Takahashi[30]	Riazi and Whitson[31]	He and Yu[32]
Year	1974	1993	1998
Use diffusivity at low pressure	Yes	Yes	No
Need except critical parameters and mole weight	No	Viscosity	Liquid volume and boiling temperature
Comment	Based on a figure		Limited range, $0.66 < T_r < 1.78$, $0.22 < \rho_r < 2.62$
Evaluated	No	Yes	No
Computational issues	Difficult to program a figure	Only usable	Have a limited range

All models are in some way based on reduced quantities, and none are expected to work when one gets into the liquid area.

4.2.1 Riazi and Whitson model

Riazi and Whitson relate diffusivity to viscosity at the following formula:

$$\frac{\rho' D_{\mathrm{AB}}}{(\rho' D_{\mathrm{AB}})^0} = 1.07 \left(\frac{\mu}{\mu^0} \right)^{b + c P_r}$$
$$b = -0.27 - 0.38\omega, \qquad c = -0.05 + 0.1\omega, \qquad \omega = y_A \omega_A + y_B \omega_B, \qquad P_r = P/P_c, \qquad P_c = y_A P_{cA} + y_b P_{cB} \tag{23}$$

Here superscript 0 means condition at low pressure.

4.3 Diffusivity with more than two components

The Stefani Maxwell equation gives:

$$\frac{dx_i}{dz} = \frac{1}{\rho}\sum_{j=1}^{n}\frac{x_i x_j}{D_{ij}}\left(\frac{J_j}{x_j}-\frac{J_i}{x_i}\right) \tag{24}$$

Often we would like a diffusion of one component into a mixture. Then we can look at a typical example with 3 components.

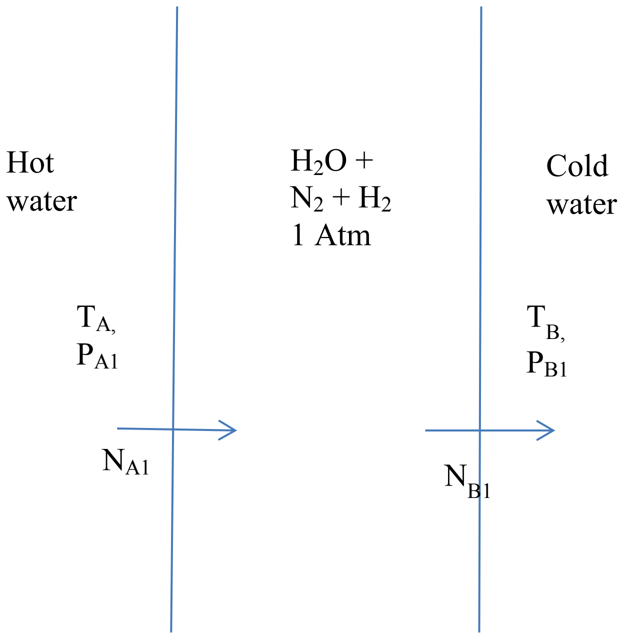

Figure 4: Diffusion in a multi component gas mixture

Figure 4 shows a space filled with nitrogen and hydrogen gas at one atmosphere between two layers of water, both below 100 °C. On the left side we have hot water with high temperature and then high evaporating pressure (but below 1 atm). On the right we have water with lower temperature and evaporating pressure. The water evaporates on left side and condenses on right side.

The nitrogen and hydrogen are kept at place so $N_{N2} = 0$ and $N_{H2} = 0$. For water, however, it is a flow N_{H2O} which is constant.

Then if water is component 1 and the gas in the mixture is components 2 to n, then one gets:

$$N_i = 0, i = 2..n \tag{25}$$

Putting Eq(25) into Eq(17) one gets:

$$J_1 = (1 - x_1)N_1, J_i = -x_i N_1, i = 2..n \tag{26}$$

Defines a new variable $D_{1,mix}$ with the following definition:

$$J_1 = -\rho D_{1,mix}\frac{dx_1}{dz} \Rightarrow \frac{dx_1}{dz} = -\frac{J_1}{\rho D_{1,mix}} = -\frac{(1-x_1)N_1}{\rho D_{1,mix}} \tag{27}$$

Putting this into Eq(24) the following will be generated:

$$\frac{dx_1}{dz} = \frac{1}{\rho}\sum_{j=2}^{n}\frac{x_1 x_j}{D_{1,j}}\left(\frac{J_j}{x_j}-\frac{J_1}{x_1}\right) = -\frac{N_1}{\rho}\sum_{j=2}^{n}\frac{x_j}{D_{1,j}} = -\frac{N_1(1-x_1)}{cD_{1,mix}} \tag{28}$$

Then for component *i* we can calculate the diffusion coefficient from component *i* to the mixture in the following way:

$$D_{i,mix} = (1-x_i)\left(\sum_{j=1,j\neq i}^{n}\frac{x_j}{D_{i,j}}\right)^{-1} \tag{29}$$

4.4 Diffusivity in liquids

For liquids it is better to speak about gradient in chemical potential $(d\mu_i/dz)$ instead of concentration. Also the diffusivity is much lower than for gases, and relatively much lower than for viscosity and thermal conductivity. This is the case since the molecules must pass between each other. Consequently, we cannot expect the formulas for diffusivity to work at the liquid phase.

However the Figure G6 in Appendix G, provides an example of calculation and measured diffusivity for CO_2 in liquid water.

5 COMPARING VISCOSITY WITH EXPERIMENTAL DATA

5.1 About the comparison

The comparison is done in three steps. First, we studied pure components at low pressure. Then we looked at mixtures at low pressure and finally mixtures at high pressure. The reason is related to the fact that the models are built on each other. The experimental data is taken from the the review work on available transport properties by Li et al [6, 7]. All the data has been organized and, if necessary, converted to a common set of input and output units before comparison. The collected data and the results from the model evaluations are shown in appendix A to G.

We have not discussed how the measurements were done or their experimental accuracy for each reference. However, the accuracy was estimated by comparing the series.

To see the difference the AAD was calculated. AAD (Absolute Average Deviation) is defined as:

$$AAD = \frac{1}{n}\sum_{i=1}^{n}\frac{\mathrm{abs}\left(y_{measure,i} - y_{calc,i}\right)}{y_{measure,i}} \qquad (30)$$

in which y refers to viscosities, thermal conductivities or diffusivities.

5.2 Plotting of results

All the plots are 2 dimensional plots with variation in one variable. For binary mixture at low pressure one usually varies the composition. In that way we are able to see where the inaccuracy comes from, whether it is from the pure component estimation or from the calculation of the mixture effects. In addition the plots show how non-linear a particular system is and how this non-linearity is captured by the different models.

5.3 Reconciliation of the experimental data

A binary the system is defined by three variables: The mole fraction of one of the components, pressure and temperature. In the 2D—plots generally, the composition varies while the temperature and pressure are kept constant.

It is a benefit to compare the data from two sources even if they are done at some different temperatures (Typical 10K difference) or pressure. Also some references do their measurements at slightly different temperatures.

To get to the same temperature and pressure (or two other variable as composition and pressure) one correct the measured value that are plotted by calculating the difference after the following formula:

$$\eta\, \eta_c(T_c, P_c, x_c) = \eta_m - \Delta\eta, \qquad \Delta\eta = \eta_{calc}(T_m, P_m, x_m) - \eta_{calc}(T_c, P_c, x_c) \qquad (31)$$

The same is done for the conductivity (λ) and diffusivity(D). Subscript *m* is measured value η_m measured at T_m, P_m, x_m and subscript *c* is corrected value or value where one does the comparison. Here it is assumed that $\Delta\eta \ll \eta_m$ and also that error in $\Delta\eta$ is much smaller than the error in the measured value.

For viscosity REFPROP[3] is used to do the correction and for conductivity the Mason and Saxena model[20] with regressed binary interaction coefficient ($\varepsilon_{i,j}$) is used.

Example: One set of experimental values are at temperatures between 20 and 22 °C and the second set is at 25°C, both at 1 bar pressure. The composition is varied in both experiments. To be able to compare, all are adjusted to a chosen temperature of 22°C. This gives:

T_c=22°C, P_c=1Bar=P_m, x_c=x_m

5.4 Viscosity for pure components at low pressure

The results from the comparison of low pressure pure component viscosities are shown in Appendix A. The viscosity for pure components at low pressure will only vary with temperature, so if measurements exist, it is possible to regress a chosen model to the measurements. One challenge is to decide on the choice of model. The regressed models are compared with the model from Chung and with REFPROP. The coefficients for a regressed model are listed in Table A1 and the comparison between the models is shown in Table A2. The default model refers to Equation A1, A2 or A3 in these two tables

For the default method, the maximum AAD is 0.2%.
For substances used not listed in Table A1, Chung's model has been used to calculate the low pressure pure component viscosities. n Chung's model the molecular weight, acentric factor, dipole moment, critical temperature, critical specific volume are the component specific properties and the model uses no empirical coefficients that need to be regressed. One exception of is for polar components where a parameter, κ, that is used which can be regressed from measurements. In this evaluation it is only H_2O that use the parameter κ. Maximum error (AAD) is 2.3% for components that neither are polar nor quantum gases, but the AAD is 11% for H_2 and 7% for H_2O over the full temperature range (T_{Min} to T_{Max}).

5.5 Viscosities for mixtures at low pressure

Appendix B provides details on the prediction of viscosity for binary mixtures at low pressure. Low pressure means 1 bar, except for the data from Maltsev[33] for H_2 where the pressure was 3.0 bar. For the pure component viscosities are calculated with the regressed (or Chung's) model as explained in 5.4.

A summary of the error (AAD) for the Chung's, Reichenberg's calculations with REFPROP compared to experimental data is shown in Table B1. Reichenberg's model shows lower error than Chung's model for all mixtures. This is expected, since Reichenberg uses the pure component viscosity as part of the model, which can be improved by regression from the experimental viscosities (Section 5.4)

For most mixtures REFPROP does better than the Reichenberg's model, but the average value is slightly better for Reichenberg[11]. For some mixtures like Ar-CO_2, REFPROP fails at higher temperatures. More importantly is the fact that the REFPROP approach is not able to simulate all components, neither can it be extended. The Reichenberg's method can be used for all mixtures where dipole moment and viscosity for pure components are known.

The results from the various models are shown graphically in Figures B1 to B14.

Figure B14 shows measurements and simulation with Reichenberg's model for CO_2-SO_2 when the mixture is in liquid phase. Here the Chung-TRAPP and Reichenberg-TRAPP models are both able to calculate viscosity in liquid phase.

5.6 Viscosity for mixtures at high pressure

Appendix C shows tables and curves for viscosity at high pressure. Gas mixtures at high pressure are generally modelled using the TRAPP methodology in this evaluation as already explained earlier. In the implementation of the TRAPP model, propane has been used as the reference fluid and the correlation from Younglove and Ely [18] has been used to calculated the propane viscosity. For the pure component low pressure viscosity contribution, either the regressed model from Table A1 or the Chung's model has been used. For the mixture effect, either Chung's or Reichenberg's model has been applied. The combinations are shown in Table C2. These models s are also compared to the predictions from REFPROP v9.0

Here REFPROP has the smallest deviation and shows an AAD of 0.2% or lower for 3 of 6 cases. As expected for the same reason as for mixture at lower pressure, the Chung-TRAPP model shows higher deviation than the Reichenberg-TRAPP model.

For the mixture CO_2-CH_4 a total of 132 measurements are conducted at various pressures (34-690 bar), concentrations and temperatures (50-200°C). All results are shown in the Figures C2-C6. The AAD is slightly better using the Reichenberg-TRAPP model compared to REFPROP. It looks like the Reichenberg-TRAPP method performs better for pressures below 300 bar while REFPROP gives better predictions for pressures above 300 bar and at low temperatures. However, the experimental data is limited.

The TRAPP model with Vogel[19] correlation for propane is also evaluated and included in Table C1 (in Appendix C). It provides a different viscosity compared to the other correlations, but the average deviation becomes the same. This method is not shown in any of the curves.

5.7 Preferred methods:

The methods of Reichenberg-TRAPP and possibly REFPROP are preferred for assessments of mixtures at low and high pressure. For pure components at high pressure, the models used by REFPROP show the lowest deviation compared to the measurements

These are the preferred methods:

Pure components at low pressure:	REFPROP and the regressed models
Mixture at low pressure:	Reichenberg
Pure components at high pressure	Not checked: Expected to be REFPROP.
Mixture at high pressure:	Reichenberg-TRAPP or REFPROP

	Preferred model	Average AAD	Max AAD	Components max AAD
Pure component low pressure	REFPROP or curve adapted (Default method)			
Mixtures low pressure	Reichenberg	1.3%	3.2%	CO_2-Ar
Mixture high pressure	Reichenberg-TRAPP	1.2%	2.2%	CO_2-CH_4
	REFPROP	0.8%	2.6%	CO_2-CH_4

6 COMPARISION OF THERMAL CONDUCTIVITY

6.1 Thermal conductivity for pure component at low pressure

The thermal conductivity for pure components at low pressure is needed as input with calculating the mixture conductivity at both low and high pressures for the models that are evaluated. For that reason a regression to polynomial expression has been done. REFPROP v9.0 was used to estimation and the coefficients and the polynomial expression is shown in Appendix D. The simplified model is regressed over the same temperature range as provided as the acceptable by the models in REFPROP. For Ne and SO_2 the model by Chung was used. In Table D2 (in Appendix D) the calculated thermal conductivities from the regressed polynomial model and the Chung model are compared to the REFPROP models. The expected accuracies of the REFPROP models are also listed in Table D2. For further details, see section 2.1.2 and 3.1.2 for treatment outside the regressed temperature region. As described in section 1.1, the range from 218 K(=-55°C) to 1623 K (=1350 °C) will be of interest.

As described in previous sections, the model by Chung is not based on measured data on pure components but critical and molecular properties. The deviation is high for polar component as H_2O (24%) and for quantum gases as H_2 (17%), but for the other gases max deviation is 6.7%.

6.2 Thermal conductivity for mixtures at low pressure.

In Appendix E a summary of the calculated thermal conductivities for mixtures at low pressure compared to available experimental data are shown. The different models that are evaluated are: the mixture model by Chung, three variants of the Mason and Saxena model and the models available in REFPROP All the experimental data are performed at 1 bar except the data on CH_4 mixtures by Christensen and Fredenslund [34]

The three variants of the Mason and Saxena's model that are used are:
1: Where viscosity is used to calculate the reduced conductivity ratio according to Eq(14).
2: Where viscosity is not used but with the reduced conductivity ration estimated from Eq(15) .
3: With regressed binary interaction coefficient $\varepsilon_{i,j}$ (Eps) as described in section 3.2.3. For series where one can compare more than one reference, $\varepsilon_{i,j}$ is regressed.. The regressed values for $\varepsilon_{i,j}$ can be found in Table E2. For other binary mixtures than the ones listed in Table E2 the available experimental data was either inconsistent or too few, and the value of 1.0 was used. With $\varepsilon_{i,j}$ equal to 1.0, variant 2 and 3 of the Mason and Saxena model becomes the same.

As expected the calculation where $\varepsilon_{i,j}$ is adapted provides better results compared to calculation in which $\varepsilon_{i,j}$ is not adapted. Also the method including the use of viscosity gives slightly better results.

As expected the Chung's method does it worse than the Mason and Saxena's method since it does not uses pure component parameters.

Table E1 (in Appendix E) provides further details on the different series
REFPROP predicts very high conductivity for the mixture CO_2-Ar so in Table E1 the overall average AAD for all the experimental data, the comparison is shown with and without this mixture.

6.3 Thermal conductivity for higher pressure

The results from the comparison between models and experimental data for thermal conductivity at higher pressure are shown in Appendix F.

Three models are evaluated in this assessment, Chung's method and TRAPP with two variants of Mason and Saxena model for the mixture thermal conductivity at lower pressure. In addition the models available in REFPROP v9.0 are included in the comparison. The two variants of the Mason and Saxena model are with and without regressed binary interaction factor, $\varepsilon_{i,j}$, for the reduced thermal conductivity ratio of the two gases. Chung's model is used both for the low pressure and the high pressure contribution.
The summary of the results from the comparisons are listed in Table F1. Experimental data are few and there are only data for binary mixtures with CO_2 for Argon (Ar), Methane, Hydrogen and N_2O at higher pressure.

The model using TRAPP with Mason and Saxena's model for low pressure mixture contribution with regressed binary interaction coefficient shows the lowest average AAD when compared to available experimental data. It also has the lowest maximum AAD of the models. In the TRAPP implementation Propane is used as reference fluid with the reference equation from Younglove [18].

Regressed $\varepsilon_{i,j}$ improve the predictions for all series except one, the measurement of Christensen for CH_4 [34]. From figure E4 it is shown that there seem to be a discrepancy between the sources for experimental data.

6.4 Preferred methods

This is the preferred method for prediction of thermal conductivity:

Pure components at low pressure:	REFPROP or the regressed polynomial model
Mixture at low pressure:	Mason and Saxena model with regressed $\varepsilon_{i,j}$
Pure components at high pressure	Not evaluated:
Mixture at high pressure:	The TRAPP model using the Mason and Saxena model with regressed $\varepsilon_{i,j}$ for the low pressure mixture contribution

Table 13 Preferred methods with deviation

	Preferred model	Average AAD	Max AAD	Components max AAD
Pure component low pressure	REFPROP or polynomial model			
Mixtures low pressure	Mason and Saxena with regressed $\varepsilon_{i,j}$	3.5%	6.6%	CO_2-N_2O
Mixture high pressure	TRAPP using Mason and Saxena model with adjusted $\varepsilon_{i,j}$ for low pressure contribution	4.7%	8.9%	CO_2-CH_4 100 to 700 bar

7 COMPARISION OF DIFFUSIVITY

The results from the comparison of the diffusion coefficient models are shown in Appendix G.

Table G1 in Appendix G shows the available experimental data for diffusion coefficients for relevant binary gas mixtures used for this study. . In addition experimental data is also available for CO_2 mixed with air and MEA but not included in this study. All the experimental data were from low pressure measurements. However, the high pressure/high density term were checked with liquid water.

In table G1 the error from the predictions using the model by Wilke and Lee [27] and from Fuller et.al [28] is also listed with reference to the graphical results.

The Wilke and Lee model performed best for all cases done at low temperatures. For two gases measured at high temperature as the CO_2-CO_2 and CO_2 -O_2, the Fuller model was slightly better. Polling[1] have done a comparison of the predicted gas diffusion coefficient using the two models on various binary mixtures and they found that average deviation was 9.6% with the Wilke and Lee model and 5.4% with the Fuller model. This was expected since the Fuller method uses an atomic parameter for each component that has been regressed from experiments-However, those results were compared at one temperature only, so for the binary mixtures in this study, the Wilke model showed a better overall performance with an average AAD of 8.5% (for the gas phase) compared to 15.3% with the Fuller model.

The Wilke and Lee model does well for non-polar components at low temperature. It is not as good for polar components as for water. At higher temperatures the deviations from the Wilke and Lee model are higher than Fuller model

No measurements were done at higher pressure/higher densities from the available experimental data, but a few are done for liquid water. For calculating at higher pressure/higher densities, the Riazi and Whitson[31] model is used, although this model is not intended to use in the liquid region. This model uses the viscosity as parameter in the calculations. Using correct viscosity for liquid water the AAD becomes 131% with Wilke and Lee's model. This implies an estimated diffusivity about two times too high. Using the TRAPP method for estimating the water viscosity for the result are 6 times too high and the diffusion coefficient is calculated 3 times to high.

Table 14 Preferred methods with deviation

	Preferred model	Average AAD	Max AAD[1]	Components max AAD
Binaries low pressure	Wilke and Lee	8.5%	23%	CO_2-CO_2 at 900 to 1400 K

[1]: For gas phase, liquid water is higher

8 FUTURE WORK

In this study the transport properties of mixture of gases at low and high pressure are evaluated and compared with available experimental data from literature. However, data was not found for all binaries, and only a few experiments were done at higher pressure. No literature data was found for diffusivity at higher pressure. More measurements at higher pressure would have been a benefit.

To make the analysis complete, it would also have been a benefit to collect more data for pure components at higher pressure. Then one could generate better knowledge on how much error that comes from the mixture effects and how much that comes from the pure component contribution.

9 CONCLUSION

Models for prediction of the transport properties for fluid mixtures of CO_2 with impurities coming from various capture sources has been evaluated and compared. The models that are evaluated depend on using the pure component predictions that has added the non-ideal effects from mixing at low pressure and finally added the effect of higher pressure. In addition models for conductivity and diffusivity need prediction for the viscosity as a parameter.
First the different models were described. Polling [1] has described all models in details. Then the available models were compared with measurements.

Here is a summary of the preferred models for the given components:

Table 15 Preferred methods for calculation for mixtures with CO_2

	Viscosity	Conductivity	Diffusivity
Single component low pressure	REFPROP or Regressed [1]	REFPROP or Regressed[1]	
Mixtures at low pressure	Reichenberg	Mason and Saxena with adjusted $\varepsilon_{i,j}$	Wilke[2]
Mixture at high pressure	Reichenberg-TRAPP or REFPROP	Mason and Saxena with adjusted $\varepsilon_{i,j}$ – TRAPP	Riazi[3]

[1]: Regressed are regressed models adapted from experiments or calculated with REFPROP
[2]: Fuller gives usually better results for other mixtures
[3]: No tests are done and Riazi[31] is the only one that is programmed

No evaluation of models has been done for pure components at higher temperatures.

APPENDIX A: MODELLING OF VISCOSITY FOR PURE COMPONENTS

For the default model the viscosity is calculated after the following formulas, T given in Kelvin.

$$\eta = a_1 T^{\frac{1}{2}} + a_2 T^{a_3} \tag{A1}$$

$$\eta = a_1 T^{\frac{1}{2}} + a_2 T + \frac{a_3 T^{\frac{3}{2}}}{1000} \tag{A2}$$

$$\eta = a_1 x^{\frac{1}{2}} + a_2 x^1 + a_3 x^{\frac{3}{2}} + a_4 x^2 + a_5 x^{\frac{5}{2}}, \qquad x = T/100 \tag{A3}$$

For other components the Chung's model is used

Table A1: Coefficients used for calculating pure components

Co-mp-one-nt	Eq.	a_1	a_2	a_3	a_4	a_5	T_{Min} (K)	T_{Max} (K)	Based on REF-PRO-P[2]	Reference
CO_2	A2	-2.645	0.845	-11.061			220	1000	Yes	Fenghour[35]
Ar	A1	28.36	-80.50	0.207			160	2000	No	Zeberg-Mikkelsen[8]
CH_4	A1	13.39	-47.94	0.325			195	1050	No	Zeberg-Mikkelsen
CO	A3	-29.9	139.45	-46.487	5.781	0	100	800	Yes	NIST 14, 9.08[2]
H_2	--	[2]					15	200	Yes	McCarty[10]
H_2	A1	-1.552	2.92788	0.6457			200	2000	No	Zeberg-Mikkelsen[8]
H_2O	A2	-2.746	0.488	-0.739			300	1000	Yes	Hubert[36]
Ne	A1	36.69	-49.52	0.325			50	2000	No	Zeberg-Mikkelsen
N_2	A3	-27.57	138.45	-49.912	8.87	-0.603	100	1625	Yes	Lemmon[37]
N_2O	A3	-6.15	8.566	6.993	-0.622	0	200	500	Yes	[1]
O_2	A3	-36.94	159.69	-54.27	9.16	-0.598	100	1625	Yes	Lemmon

[1]: Extended Corresponding States model. Fitted to extremely limited data, estimated uncertainty 10%

[2]: Hydrogen uses between 15 and 200 K:

$$\eta = 0.9972884 * \frac{(-2.3413534 + 70.451242x)}{1.0 + 0.7321488x - 0.1055658x^2}, x = \frac{T}{100}$$

Table A2: Comparison between the models, average absolute deviation (AAD)

Component	T_{Min} T_{Max} (°C)	T_{Min} – T_{Max}		4 – 150 °C[1]		Use REFPROP
		Chung – REFPROP	Default –REFPROP	Chung – REFPROP	Default – REFPROP	
CO_2	-55 – 1350	1.7%	0.2%	1.0%	0.1%	Yes
Ar	-55 – 1350	2.3%	0.2%	0.6%	0.2%	No
CH_4	-55 – 660	0.6 %	0.2 %	1.0 %	0.5%	No
CO	-55 – 1225	1.9%	0.1%	2.8%	0.1%	Yes
H_2	-55 – 1225	11.5%	0.7%	9.5%	0.2%	No
H_2O	25 – 1350	8.2%	0.1%	10.5%	0.2%	Yes
N_2	-55 – 1350	1.4%	0.02%	0.8%	0.02%	Yes
N_2O	-55 – 500	1.1%	0.1%	1.5%	0.01%	Yes[2]
O_2	-55 – 1350	0.8%	0.01%	0.3%	0.02%	Yes
SO_2	See [3]					No

[1]; Except H_2O, where from 25 °C

[2]: REFPROP reports an estimated uncertainty of 10% for N_2O

[3]: REFPROP V90 was not able to calculate viscosity of SO_2, and no regression is done for SO_2

Default model = Chung's model. Figure B13 gives some comparison for SO_2

APPENDIX B: COMPARISION OF VISCOSITY FOR BINARIES AT LOW TEMPERATURE

Table B1: AAD (Average Absolute Deviation) for collected binary mixtures at low pressure.

Second component	T(°C)	AAD			Fig	Comment
		Chung	Reichen-berg	REF-PROP		
Ar	25 – 600	2.5%	1.1%	3.2%	B1	REFPROP Overestimate the viscosity at high pressure
CH_4	25 – 200	3.5%	1.2%	0.7%	B2	REPPROP best, especially at low temperature. At 200 °C is Reichenberg as good as REFPROP
CO	25-300	1.4%	0.9%	0.3%	B3	REFPROP best at all temperatures
H_2	25-3000	5.2%	1.1%	2.3%	B4,B5	Reichenberg best at all temperatures. Chung also fails at viscosity for pure H_2
H_2O	30	2.0%	1.3%	0.3%	B6	Measurements to 4% H_2O (Saturation concentration 4.2%), REFPROP calculate only to 2% H_2O.
N_2	25-600	0.8%	0.6%	0.4%	B7-B8	Little difference between pure CO_2 and N_2. Reichenberg provides pure component properties from curve fitting of REFPROP
N_2O	26 – 275	2.0%	1.9%	1.8%	B9-B10	Little difference between N_2O and CO_2 and low curvature. Large difference between different experimental series
O_2	25 – 400	2.5%	1.9%	1.7%	B11	Little difference between REFPROP and Reichenberg
SO_2	-35 to 80	1.5%	1.2%	-	B11-B13	One serie closest to Chung and one closest to Reichenberg. REFPROP was not able to calculate SO_2
AVERAGE		2.5%	1.25%	1.34%		Not including SO_2

Table B2: Simulation model used

Model in figure	Model used for		Colour in the figures
	Single component	Mixture	
Chung	Chung	Chung	Blue
Reichenberg	Default	Reichenberg	Green
REFPROP	REFPROP	REFPROP	Red

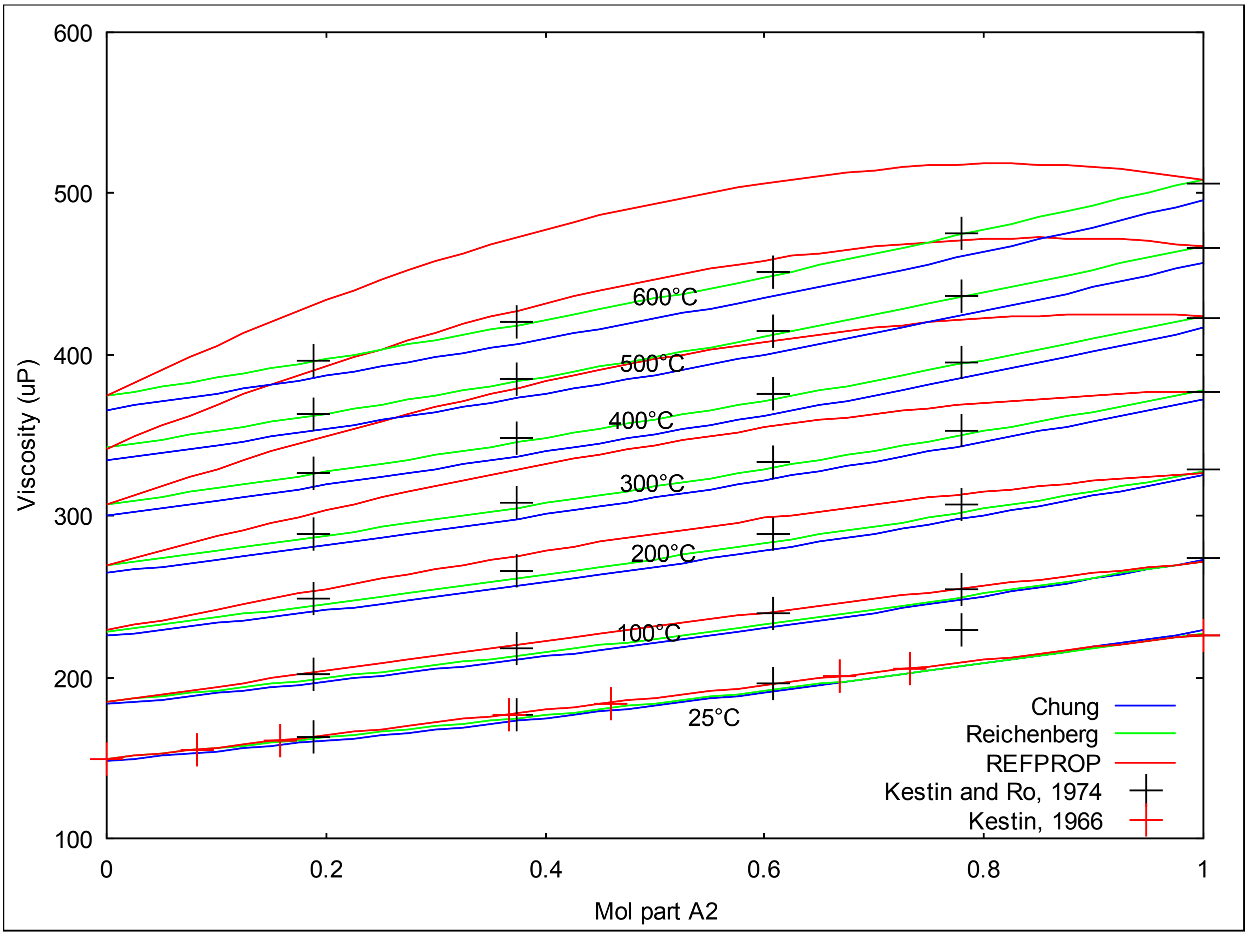

Figure B1: Viscosity for mixture CO_2-Ar [38, 39]

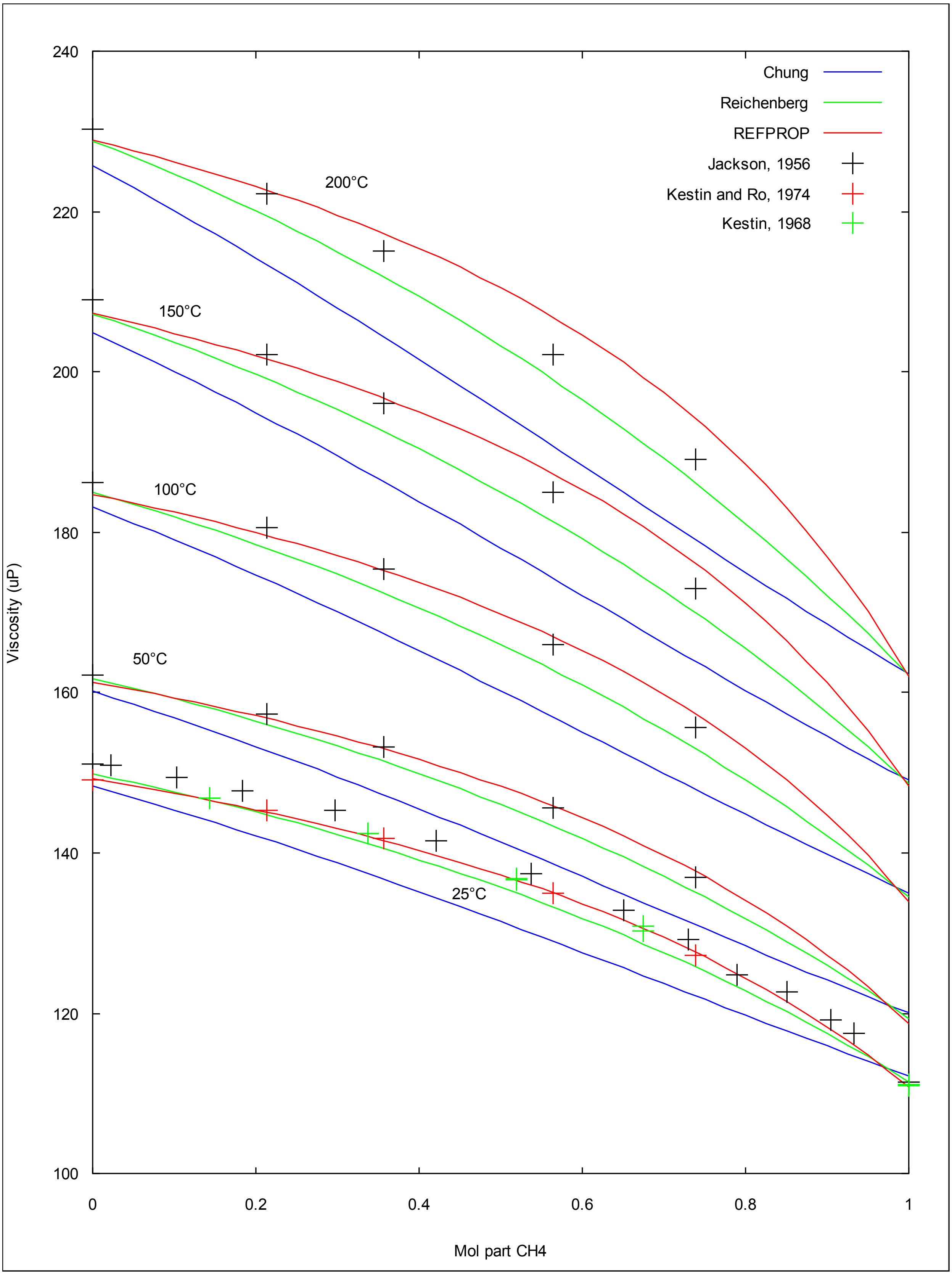

Figure B2: Viscosity for mixture CO_2-CH_4 [38, 40, 41]

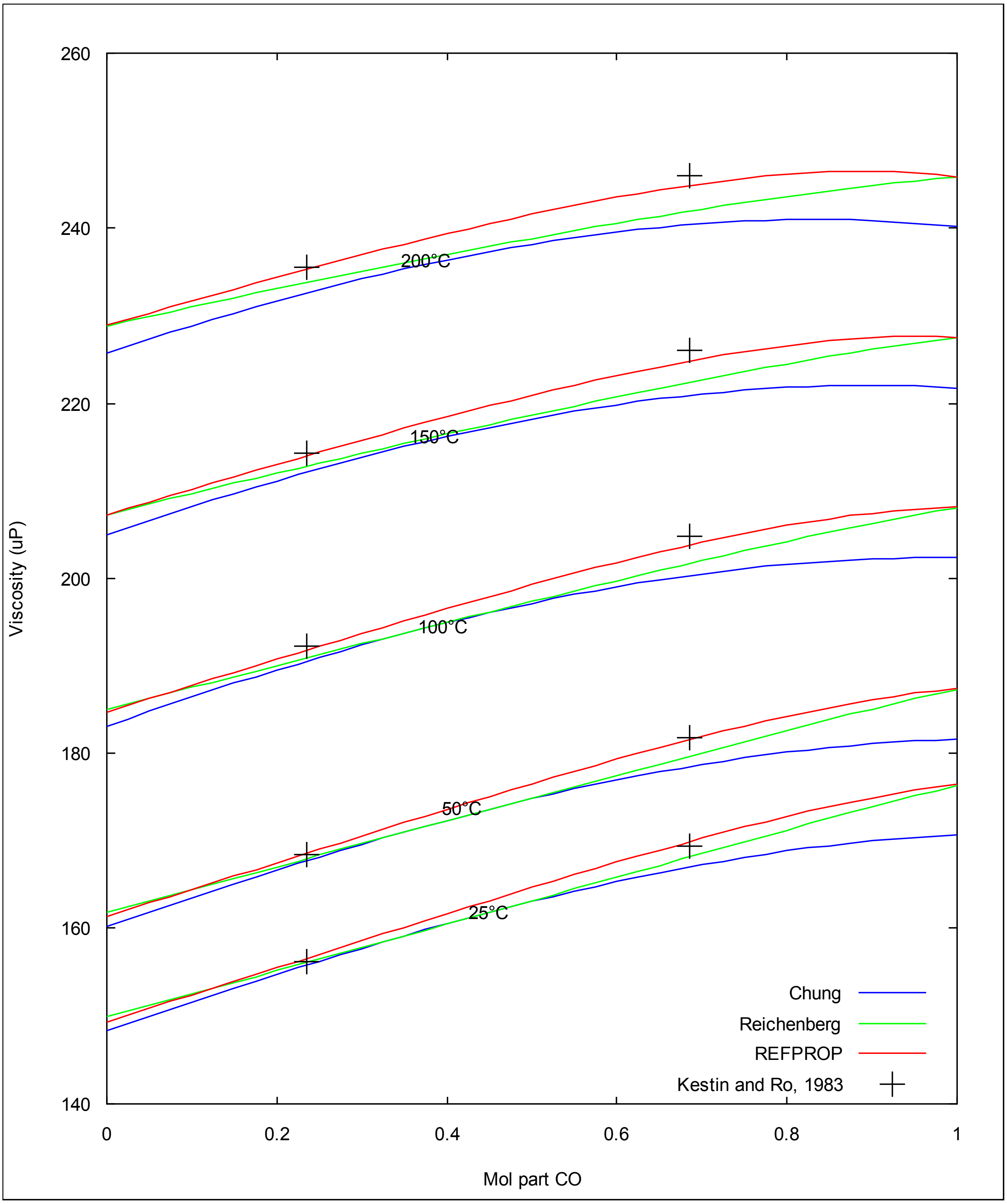

Figure B3: Viscosity for mixture CO_2-CO [42]

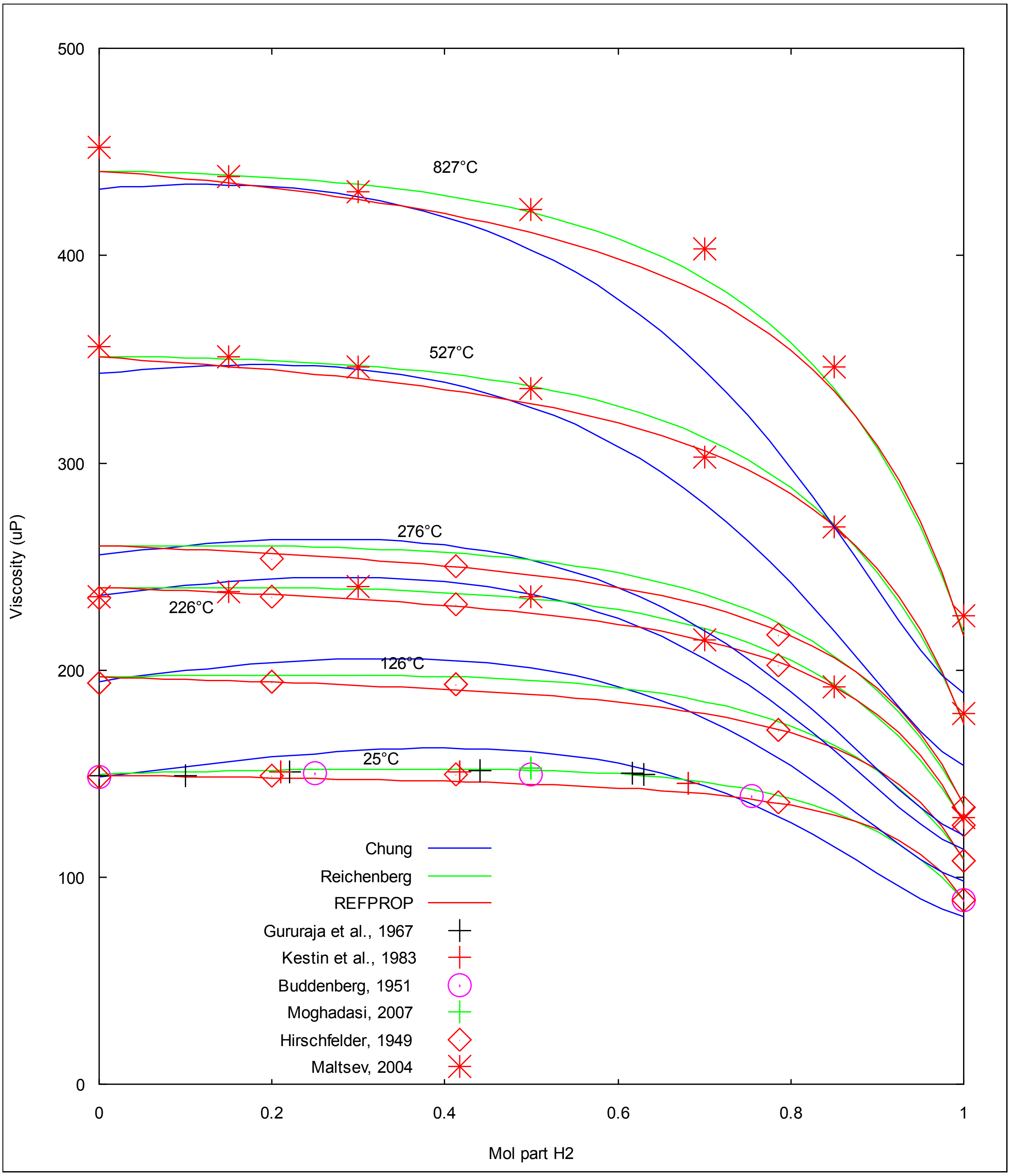

Figure B4: Viscosity for mixture CO_2-H_2 [33, 42-46]. Maltsev measured at 3 bar, the rest at 1 bar..

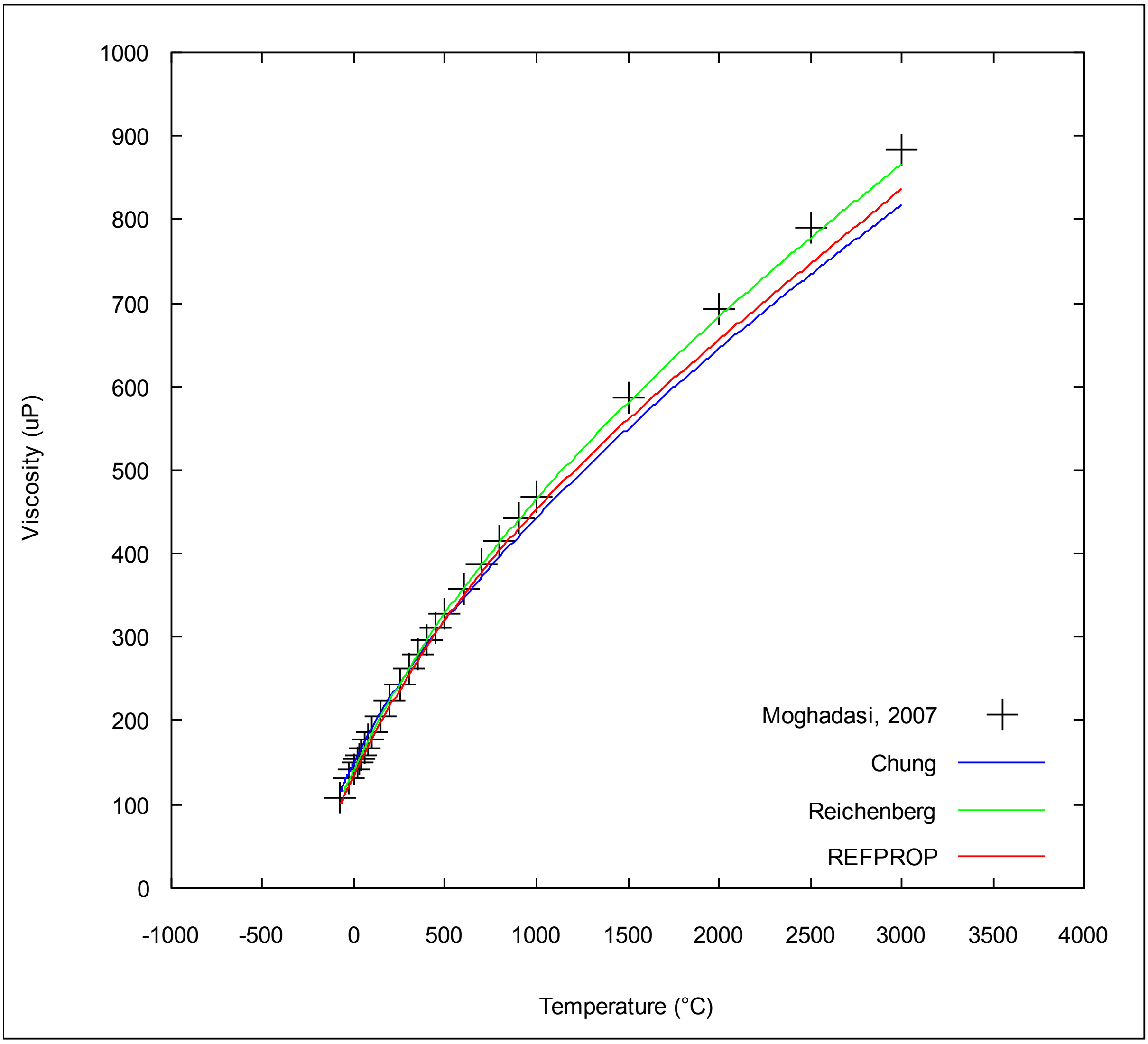

Figure B5: Viscosity as function of temperature for 50%H_2, 50% CO_2 at 1 atm [45]

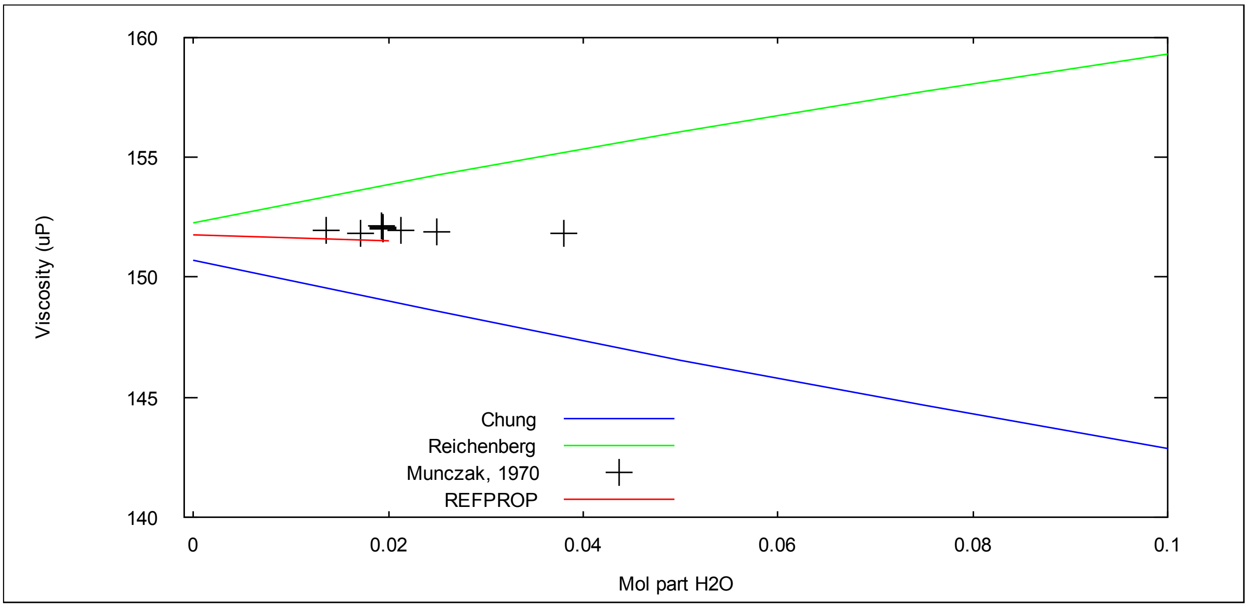

Figure B6: Viscosity for mixture CO_2-H_2O at 30°C. REFPROP was only able to calculate to 2% H_2O [47]

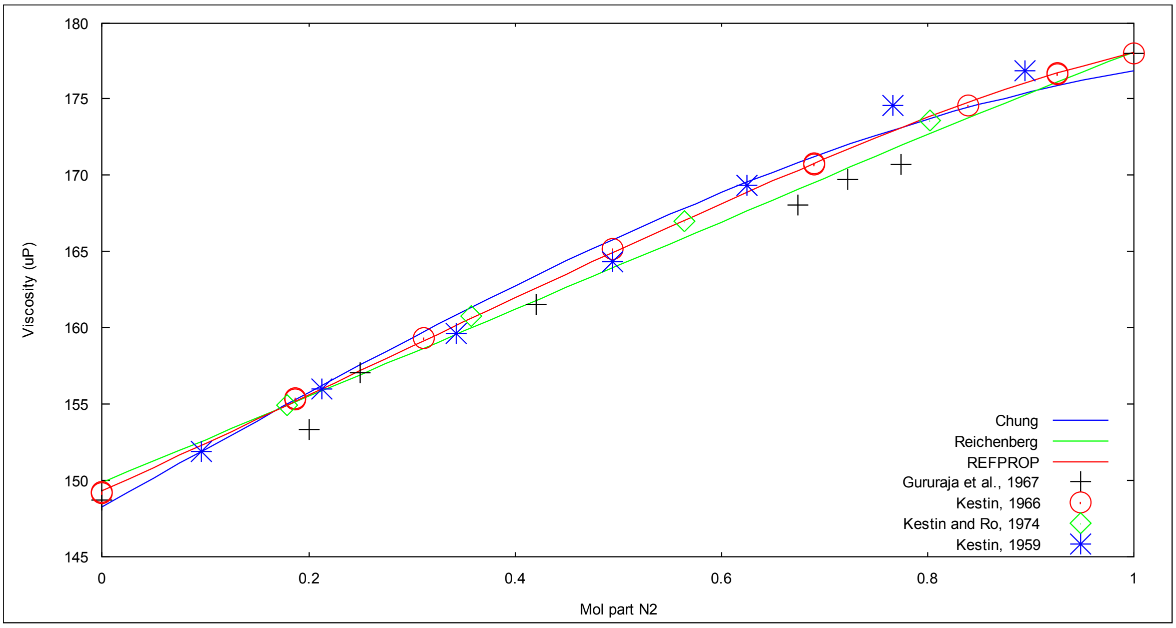

Figure B7: Viscosity for mixture CO_2-N_2 at 25°C [38, 39, 43, 48]

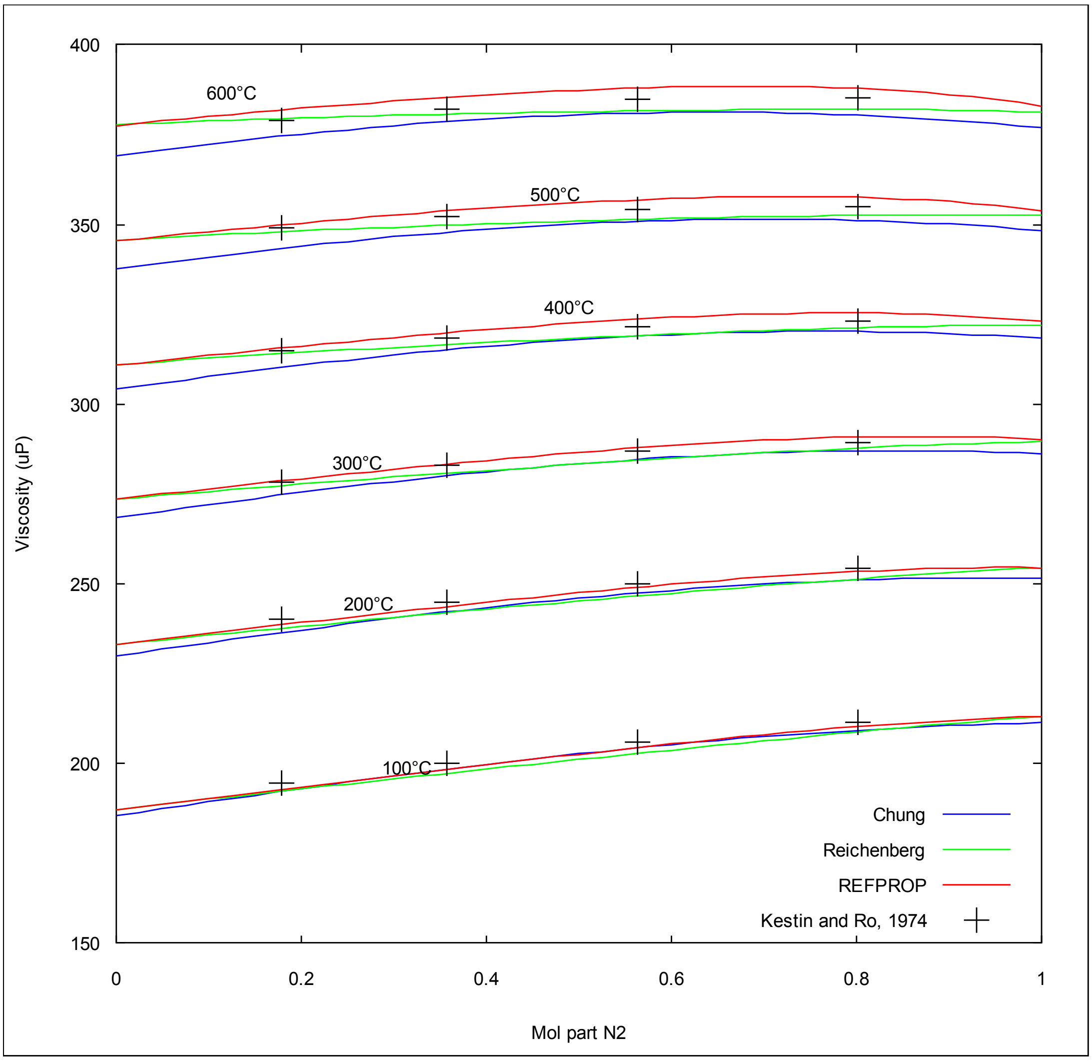

Figure B8: Viscosity for mixture CO_2-N_2 at higher temperatures [38]

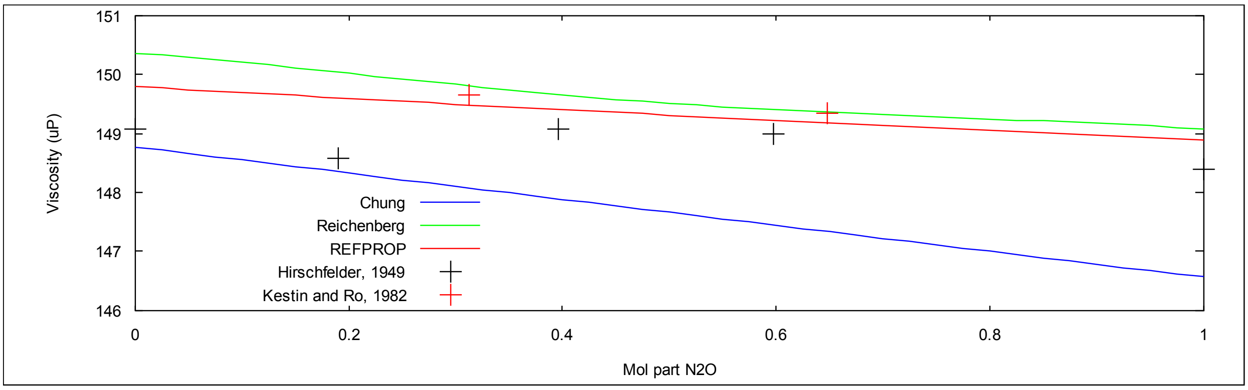

Figure B9: Viscosity for mixture N_2O-CO_2 at 26°C [46, 49]

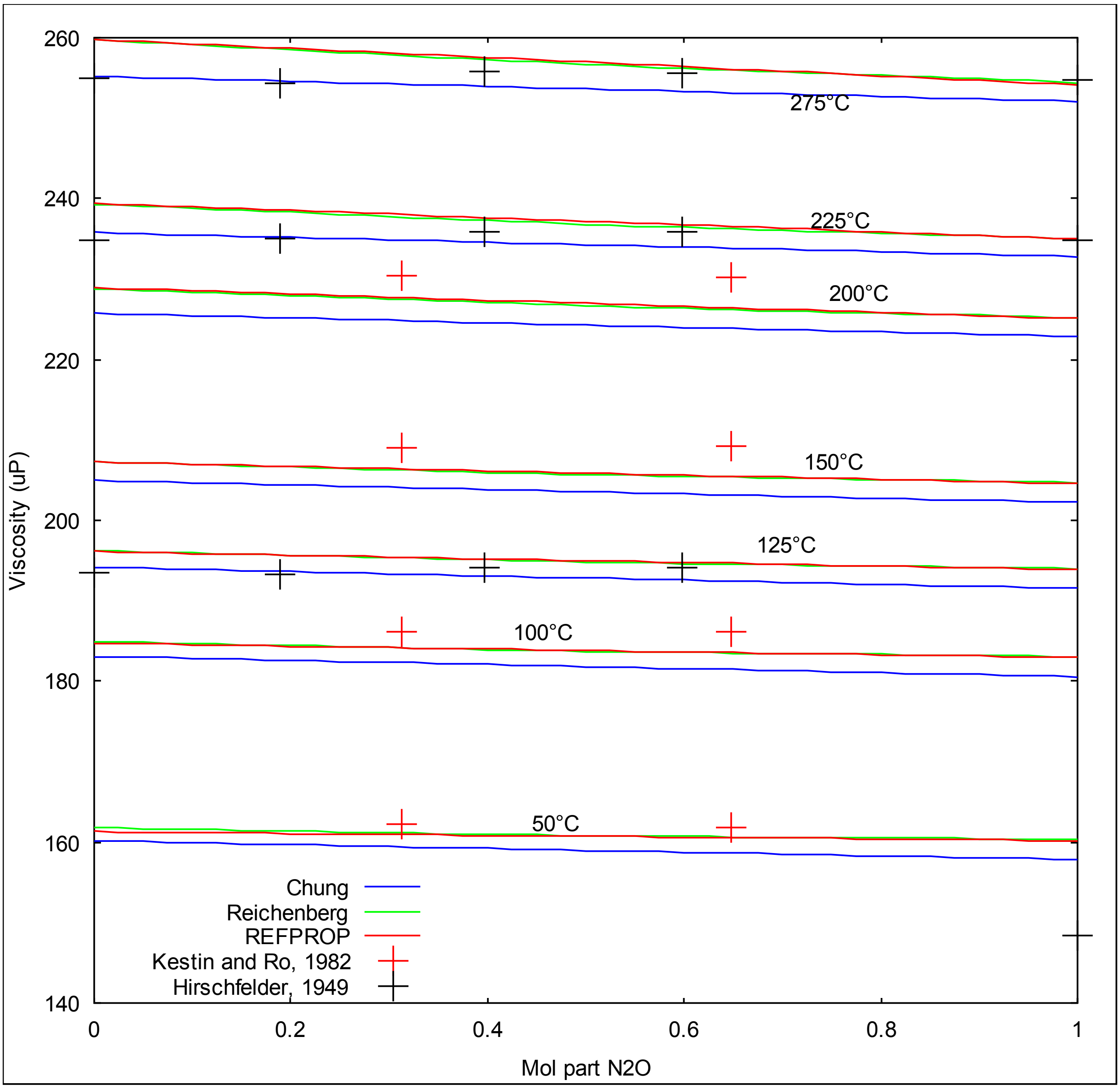

Figure B10: Viscosity for mixture N_2O-CO_2 at higher temperatures [46, 49]

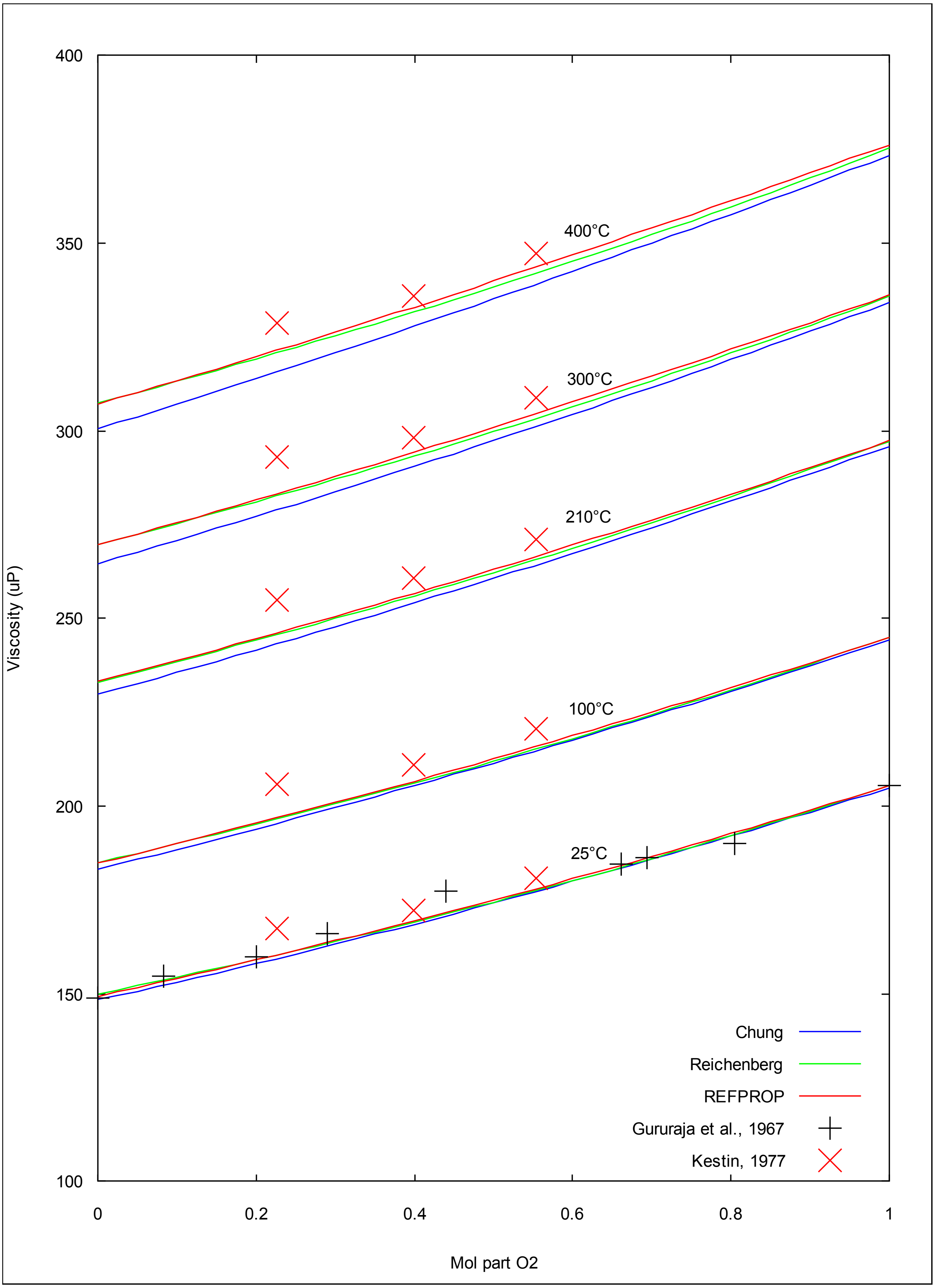

Figure B11: Viscosity for mixture CO_2-O_2 [43, 50]

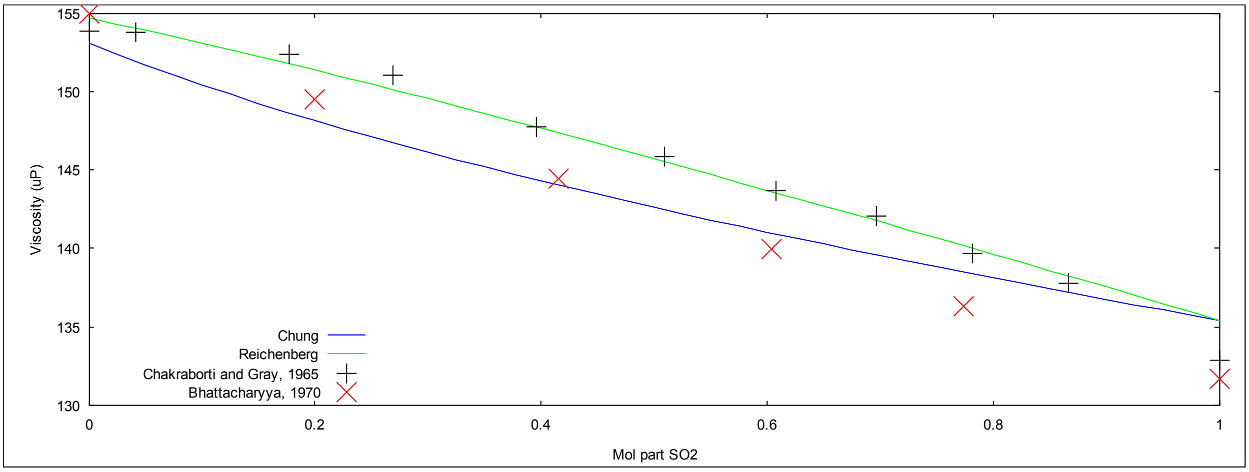

Figure B12: Viscosity mixture SO_2-CO_2 at 35 °C [51, 52]

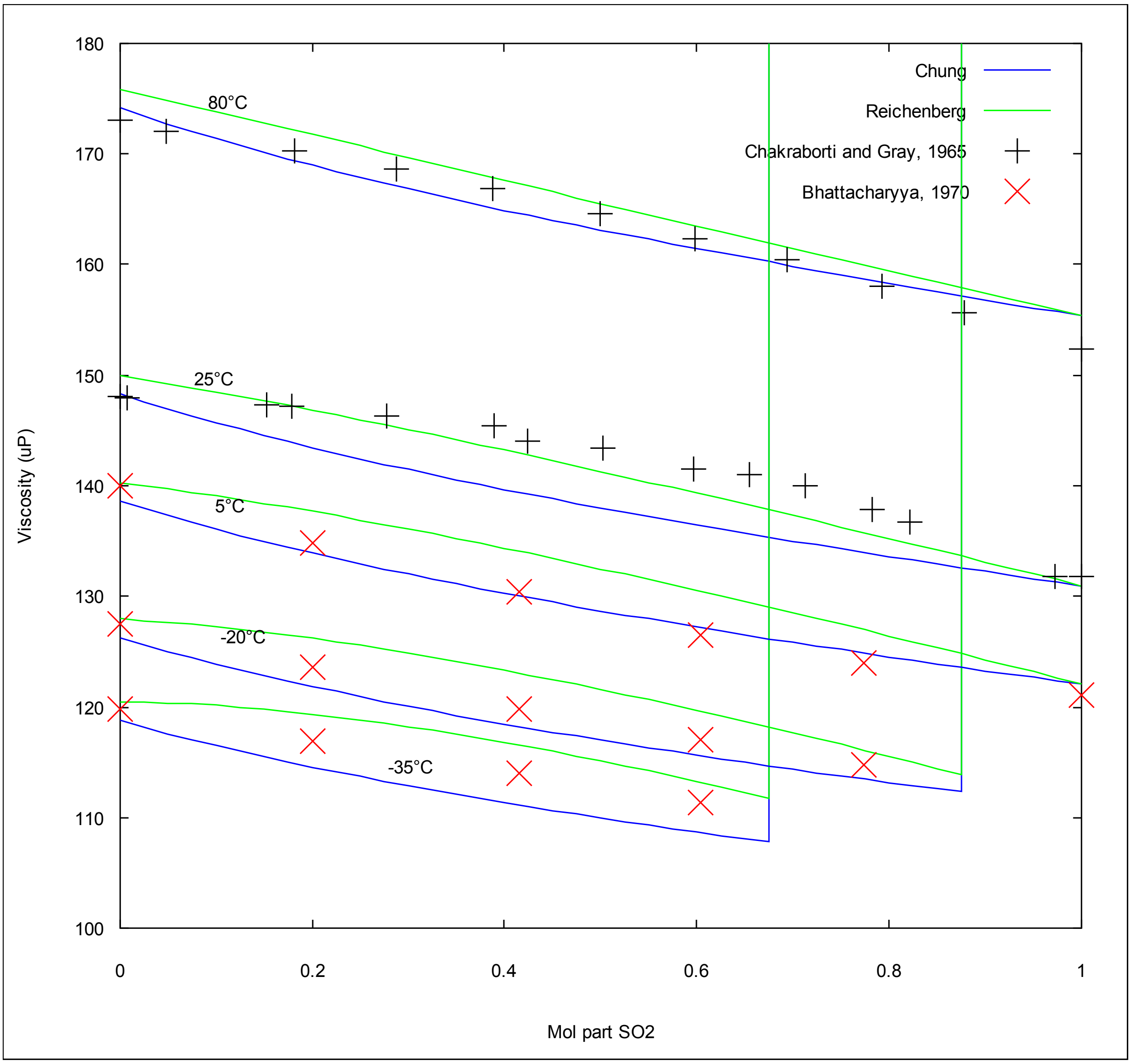

Figure B13: Viscosity for mixture SO_2-CO_2 at gas phase at different temperatures [51, 52]

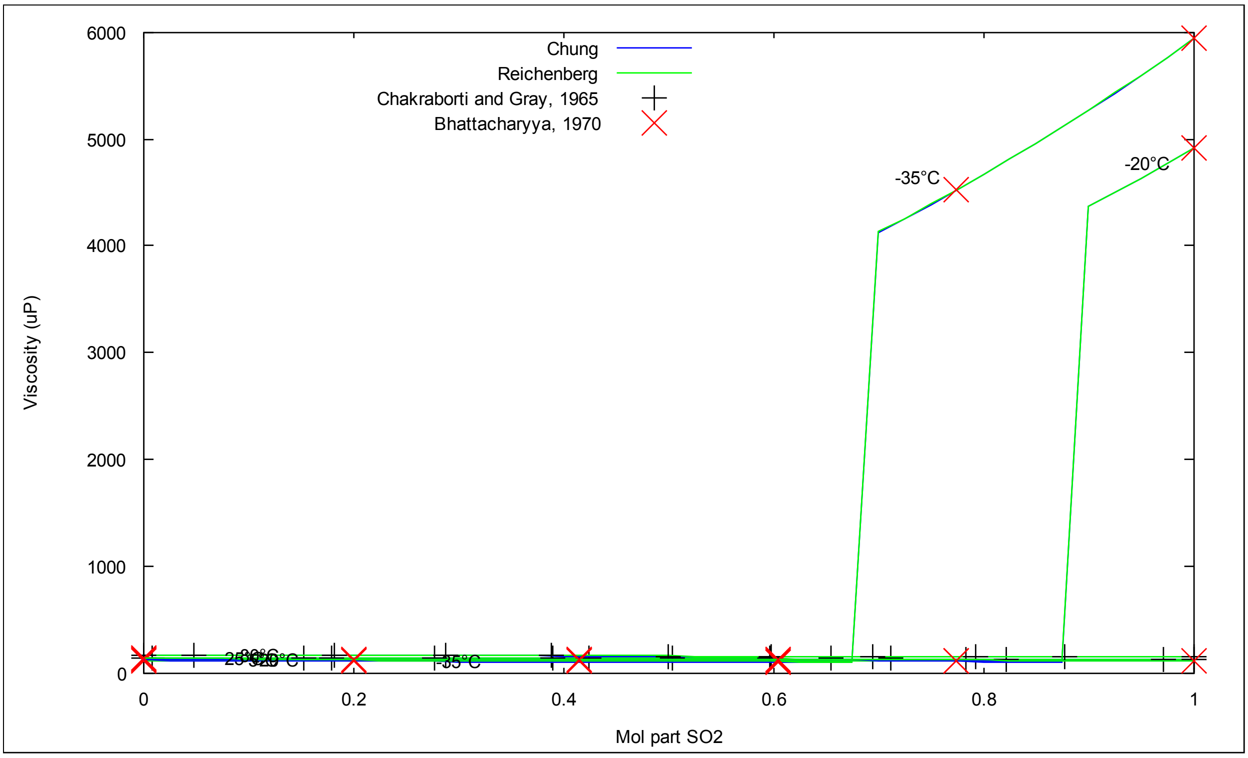

Figure B14: Viscosity for mixture CO_2- SO_2 at liquid phase [51, 52]

APPENDIX C: COMPARISION OF VISCOSITY AT HIGHER PRESSURE

Table C1: Area and deviations for measurements at high pressure

Figure	Mixture CO_2 -	P_{Max}	X_2 (%)	T(°C)	Deviation AAD				Reference
					TRAPP		TR-APP Vogel[1]	REF-PROP	
					Chung	Reichenberg			
C1	Ar	25	0-100	25	1.4%	1.0%	0.8%	0.15%	Kestin 1966[39]
C2-C5	CH_4	683	24% - 76%	50 – 200	3.6%	2.2%	3.4%	2.6%	Dewitt 1966[53]
C6	CH_4	27	14% - 100%	25	2.3%	0.8%	0.8%	0.2%	Kestin 1968[41]
C7	N_2	120	38%	19	2.1%	1.1%	1.5%	1.3%	Hanley 1976[54]
C8	N_2	21	9% - 89%	20	1.0%	0.9%	0.5%	0.2%	Kestin 1959[48]
C9	N_2	25	0% - 100%	25	1.0%	1.3%	0.4%	0.14%	Kestin 1966[39]
Average					1.9%	1.2%	1.2%	0.8%	

[1]: TRAPP with Vogel[19] correlation for Propane and Reichenberg[13] with low pressure

Table C2: Models used

Model on graph	Component	Mixture low pressure	Mixture high pressure	Colour on graph
Chung	Chung	Chung	TRAPP	Blue
TRAPP	Default	Reichenberg	TRAPP	Green
REFPROP	REFPROP	REFPROP	REFPROP	Red

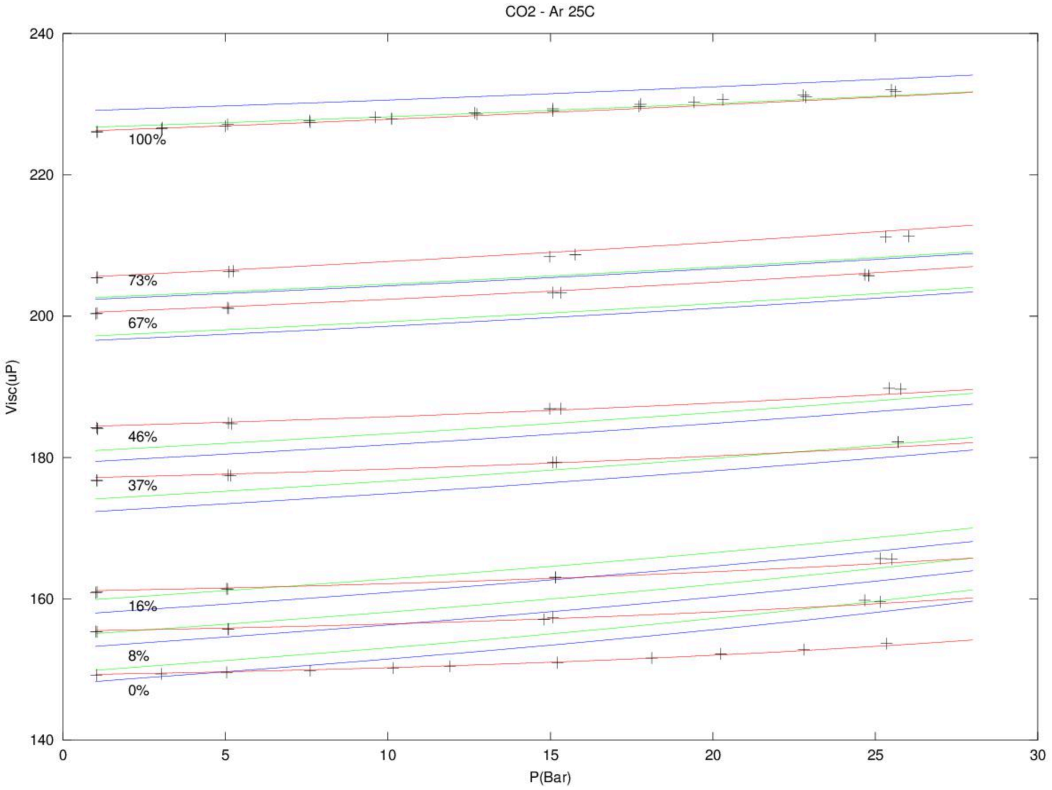

Figure C1: Mixture CO_2-Ar at 25°C, where part Ar is shown. Simulation with measurements from Kestin 1966 [39]

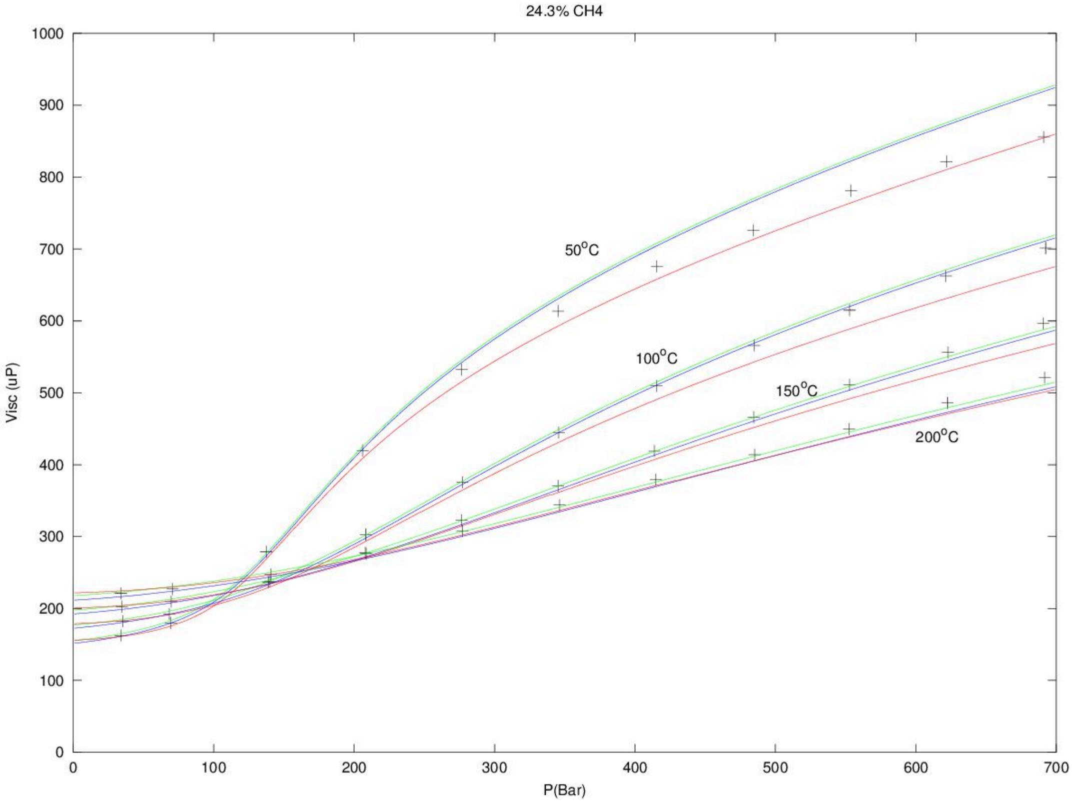

Figure C2: Mixture CO_2-CH_4 at 24.3% CH_4, simulations with measurements from Dewitt 1966[53]

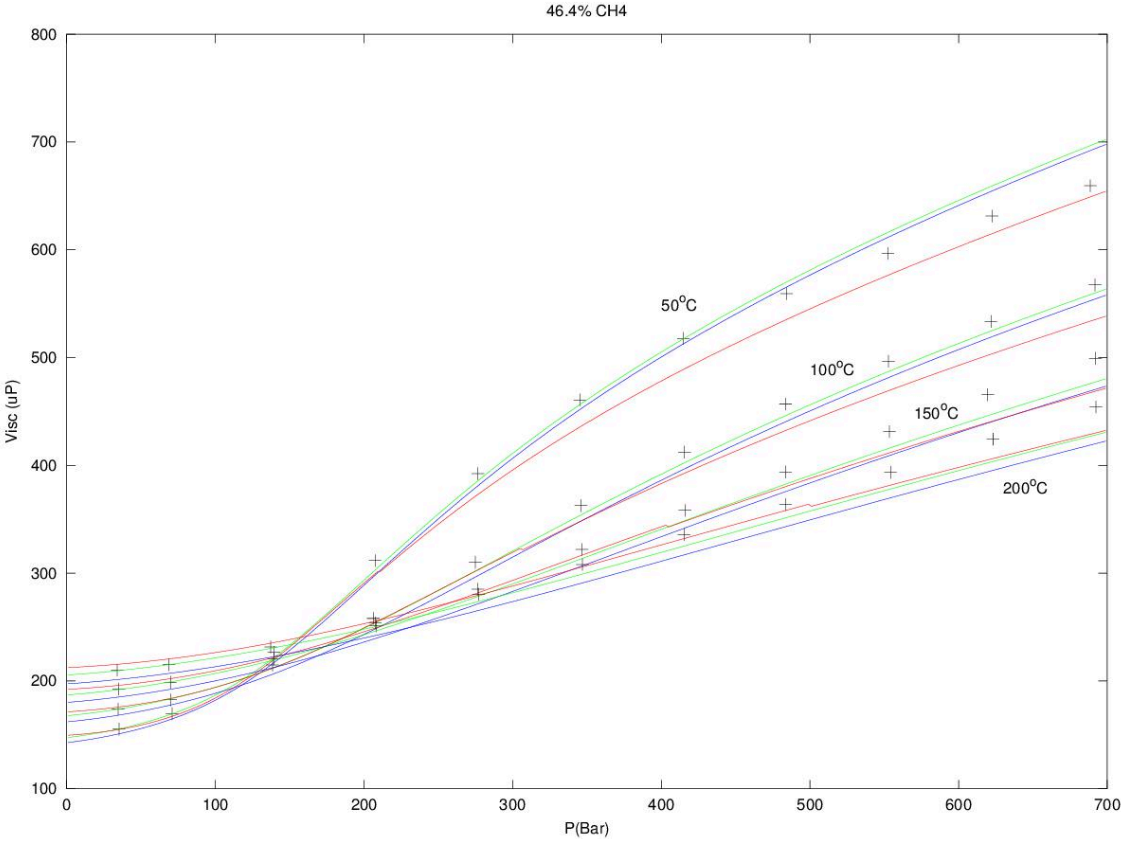

Figure C3: Mixture CO_2-CH_4 at 46.4% CH_4, simulations with measurements from Dewitt 1966[53]

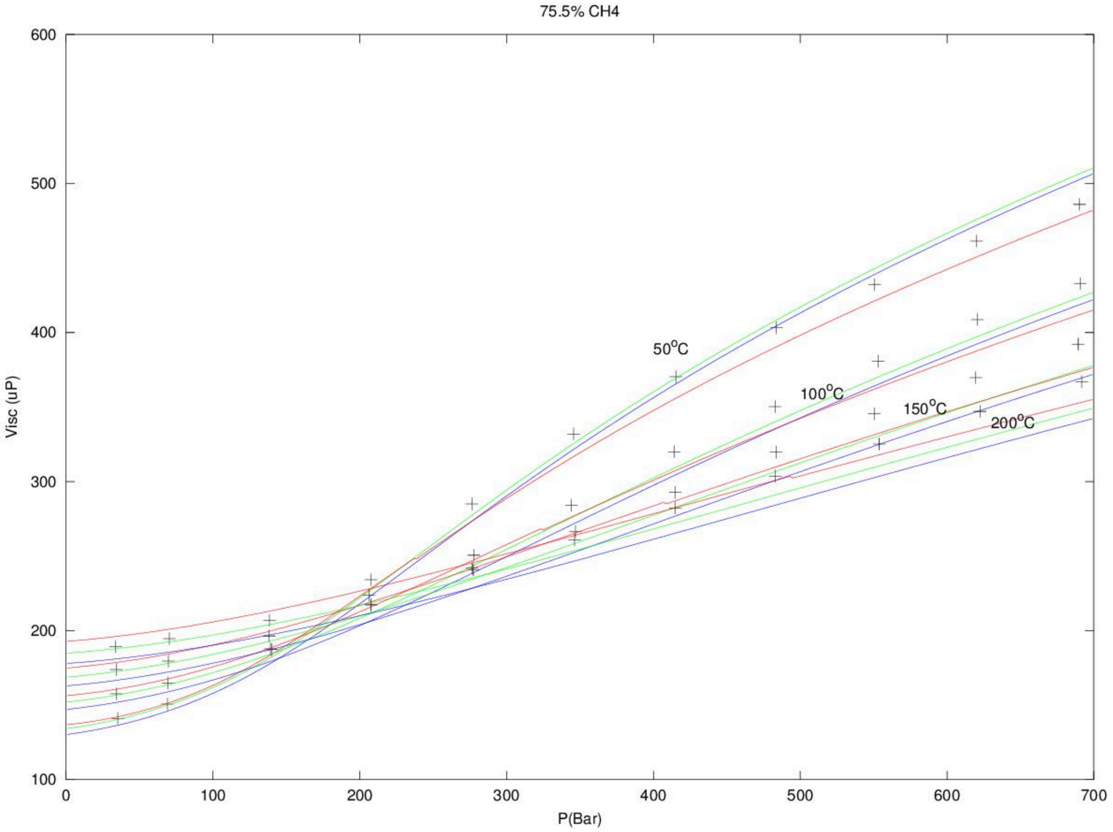

Figure C4: Mixture CO_2-CH_4 at 75.5% CH_4, simulations with measurements from Dewitt 1966[53]

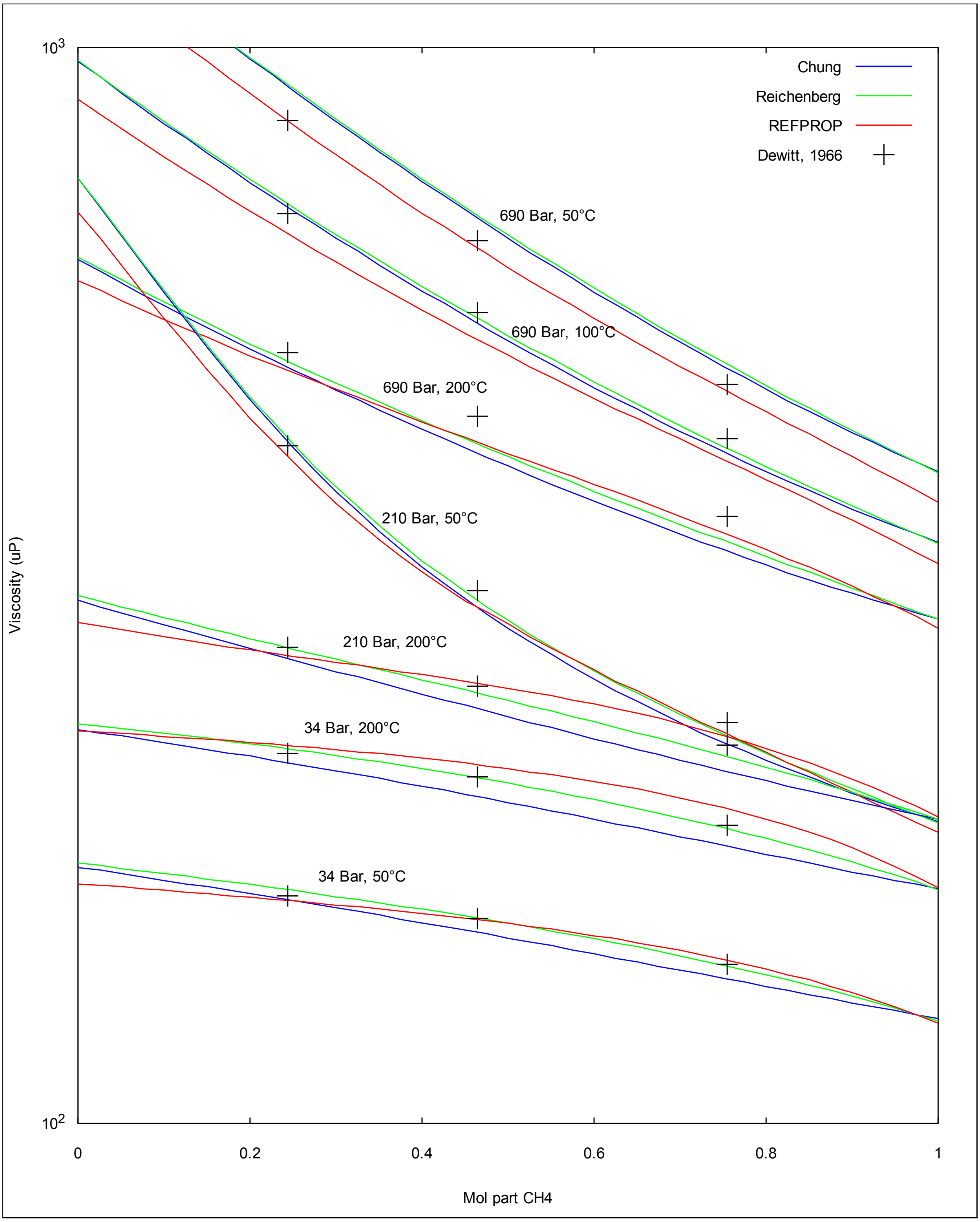

Figure C5: Viscosity for mixture CO_2 - CH_4 as a function of composition for some temperatures and pressures. The models Chung and Reichenberg use TRAPP to calculate at higher pressure

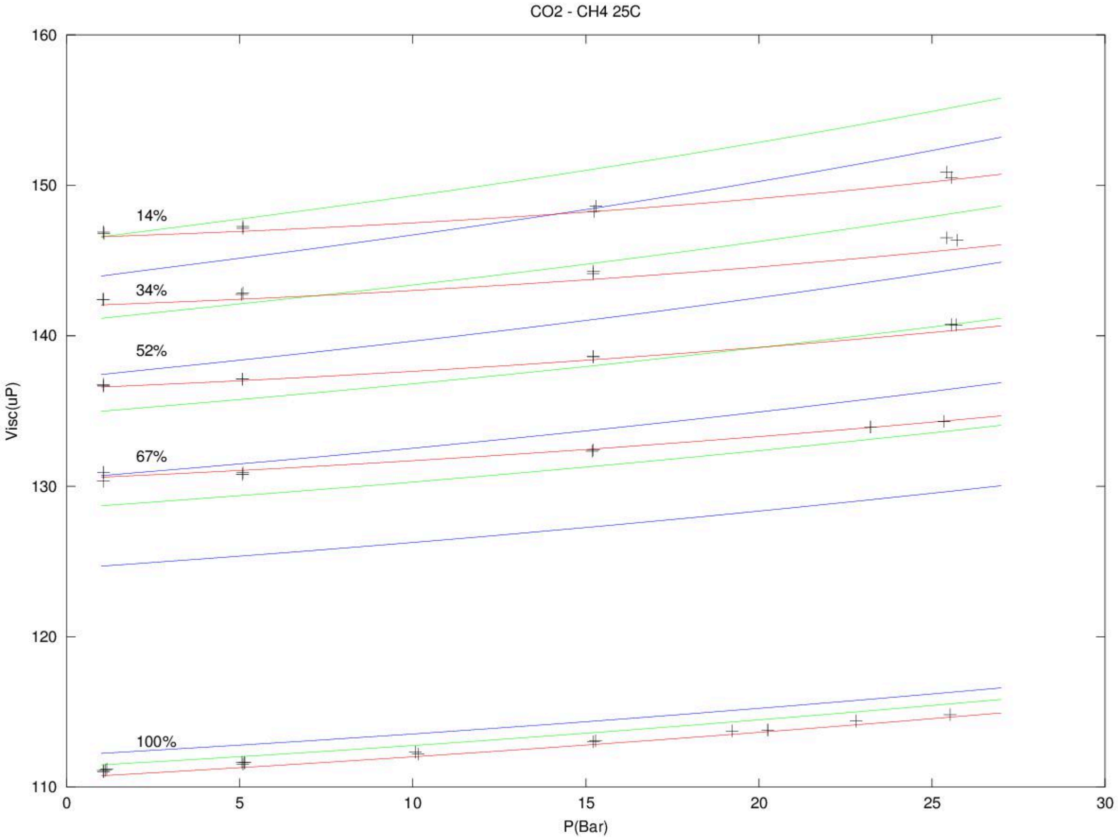

Figure C6: Methane at 25C, with measurements from Kestin 1968[41]

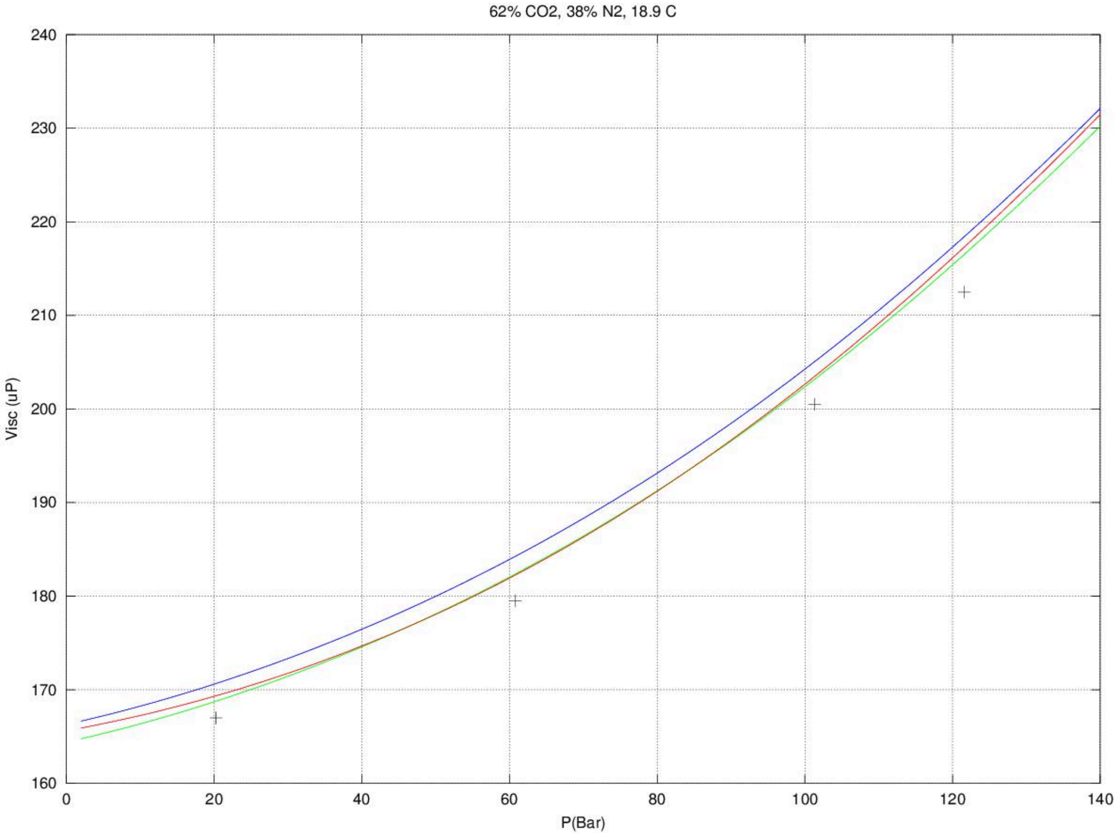

Figure C7: CO_2-N_2 at 18.9°C and 38% N_2 with measurements from Hanley 1976[54]

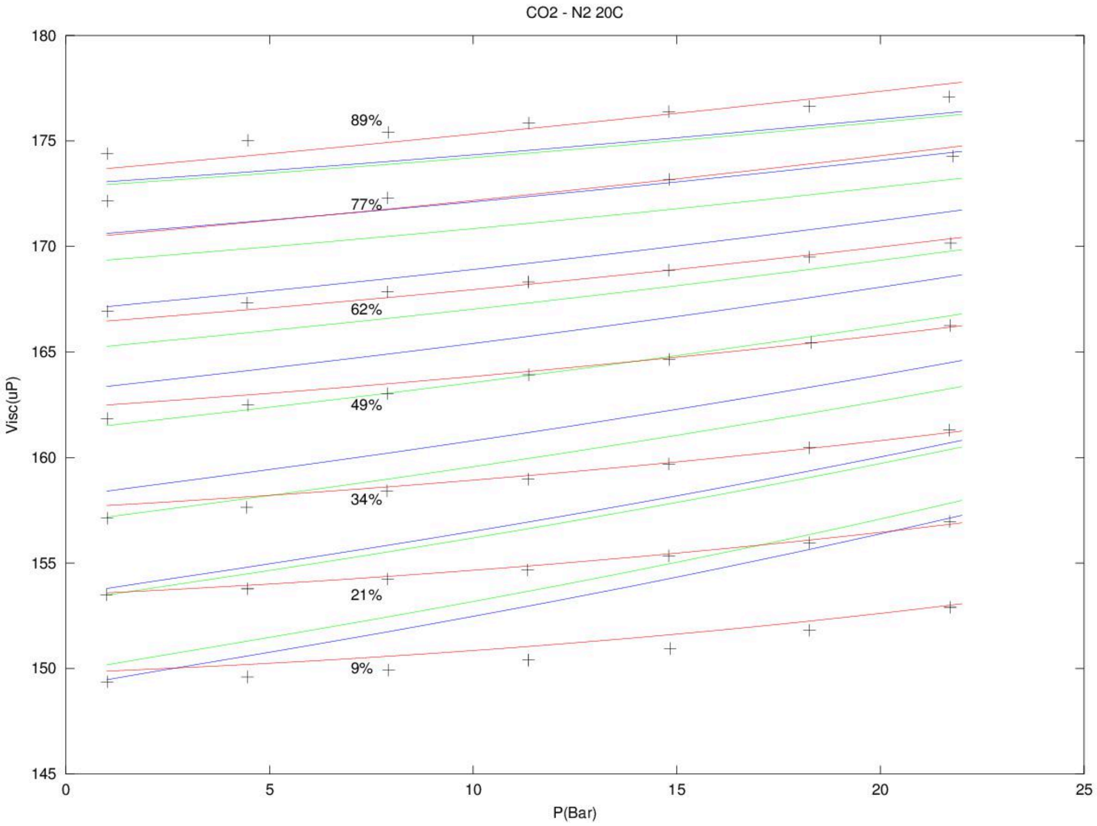

Figure C8: CO_2-N_2 at 20°C with measurements from Kestin 1959[48]

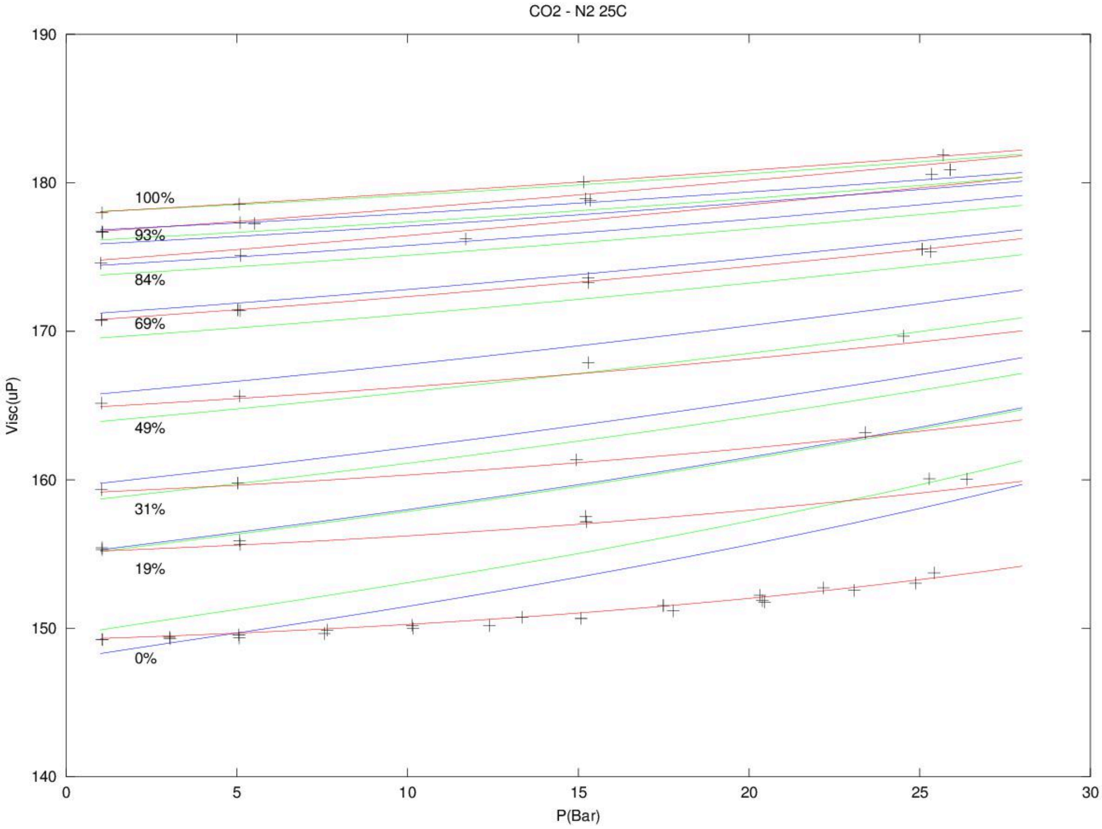

Figure C9: CO_2-N_2 at 25°C with mole part N_2 on the curves with exp. from Kestin 1966[39]

APPENDIX D: CONDUCTIVITIES FOR PURE FLUIDS

Thermal conductivity at low pressure, parameters to the expressions below are found from regression of data from REFPROP for temperatures between T_{Min} and T_{Max}

$$\lambda = \lambda_p(T) = \frac{(a_0 + a_1x + a_2x^2 + a_3x^3 + a_4x^4)}{1000}, x = \sqrt{T/100}$$

For temperatures below T_{Min} and above T_{Max} Chung's model (Section 3.1.1) is used with $\lambda_{Chung}(T)$ for $T < T_{Min}$

$$\lambda = \lambda_{Chung}(T)\ \frac{\lambda_p(T_{min})}{\lambda_{Chung}(T_{min})}$$

similar formula is used for $T > T_{max.}$

Table D1: Model parameters used

Component	a_0	a_1	a_2	a_3	a_4	T_{Min}	T_{Max}
CO_2	30.0779	-61.93371	43.81387	-8.18559	0.57067	220	1623
Ar	-5.55709	7.29499	5.29926	-1.16584	0.09937	218	1623
CH_4	-27.22552	93.26383	-88.33826	39.70462	-4.61273	218	900
CO	-22.74269	46.46195	-20.05345	6.76485	-0.6881	173	1500
H_2	-146.88591	296.90815	-118.86594	39.0262	-3.12797	200	1400
H_2O	48.596057	-56.844026	22.81191	0	0	273	1300
Ne	Uses Chung's model						
N_2	-13.67953	21.02439	0.85356	0.1272	0	218	1400
N_2O	10.09885	-24.80272	21.20986	-2.35084	0	200	700
O_2	-6.27055	7.52293	8.68347	-1.45036	0.13044	200	1400

Table D2: Average absolute deviation

	T_{Min}[1]	Chung – REFPROP		Default-REFPROP[2]		Reference REFPROP
		Whole area	4 to 150 °C	Whole range	Accuracy given by REFPROP	
CO_2	220	3.7%	2.9%	0.1%	<5%	Vesovic 1990 [25]
Ar	218	0.3%	0.15%	0.01%	2%	Lemmon 2004 [37]
CH_4	218	3.0%	1.1%	0.06%	2.5% up to 625 K	Friend 1989 [55]
CO	218	6.7%	0.9%	0.08%	4% to 6%	NIST 14
H_2	218	17%	7.3%	0.07%	about 10%	McCarty 1972 [10]
H_2O	273	24%	24%	0.08%		Kestin 1984 [56]
N_2	218	4.6%	1.9%	0.03%	2%	Lemmon 2004 [37]
N_2O	218	2.9%	4.2%	0.02%	10%	Huber 2003 [57]
O_2	218	4.2%	1.2%	0.01%	2%	Lemmon 2004 [37]

[1]: $T_{Max,}$ Same as in Table D1

[2]: Shows the accuracy of the regression, but does not tell anything about accuracy of the predicted value

APPENDIX E: COMPARISION OF CONDUCTIVITY FOR BINARY MIXTURES AT LOW TEMPERATURE

Table E1: Comparison of binary mixtures at low temperatures

Second component	T(°C)	ADD					
		Chung	Mason S. w visc[1]	Mason S.	Mason S. w $\varepsilon_{i,i}$	REFPROP	Fig
Ar	0 – 200	6.0%	2.9%	2.9%	2.2%	82%	
	REFPROP calculate very high conductivity for mixture. Measurements from Barua[58] gives lower value than both models (Mason S. becomes equal REFPROP at pure components)						
CH_4	-50 – 35	4.9%	4.9%	4.9%	4.9%	4.5%	E3-E4[2]
	25 and 35	2.9%	4.3%	4.5%	1.5%	3.7%	E3-E4[3]
	Results from Christensen are different from all simulations and other experiments.						
CO	No data						
H_2	-15 – 200	26%	10%	12%	5%	22%	E5-E11
	Good consistency between different measurement series						
H_2O	25 – 330	8.4%	2.6%	2.5%	1.8%	-	E12-E14
	Most references gave low values even at points where water is liquid. Dijkema[59] gave some high values, not shown. (25 m//mK at 25°C)						
Ne	No data						
N_2	27 – 774	2.7%	2.4%	2.5%	2.5%[4]	2.6%	E15-E16
	Experiments from Barua (1968) were different from other experiments and not included in the comparison. About same conductivity for CO_2 and N_2. In addition, the effect of $\varepsilon_{i,j}$ on the model becomes relatively low. Also not enough consistency in measurement to calculate $\varepsilon_{i,j}$						
N_2O	15-400	6.0%	6.6%	6.6%	6.6%[4]	6.5%	E17
	Only one reference, Saxena[60], but the results are not good since it gives wrong conductivity towards pure CO_2 where measurements fit the model.						
SO_2	50-100	5.7%	6.1%	6.0%	6.0%[4]	-	E18
	Neither REFPROP nor literature was found for pure SO_2 so calculated conductivity for pure SO_2 is not expected to be right. Also only one reference. Not able to judge the quality of that measurements. However, it seems to fit well towards pure CO_2 where the simulation is expected to be right. Not able to find $\varepsilon_{i,i}$						
O_2	96	3.6%	1.1%	1.1%	1.1%[4]	2.3%	E19
	Only one reference and low deviation for the Mason and Saxena's method, and then not able to find $\varepsilon_{i,j}$						
Average all		7.9%	4.6%	4.8%	3.5%	20%	
Average without Ar		8.2%	4.8%	5.1%	3.9%	7.6%	

[1]: Uses Default model for components and Reichenberg for mixtures for calculating viscosity

[2]: All measurements

[3]: Without Christensen

[4]: Not adjusted $\varepsilon_{i,j}$ and then it becomes the same as Mason and Saxena's method.

Table E2: Adjusting with Mason an Saxena's model

Second component	Ref used in adaption	ε_{ij}	AAD	Comment
Ar	Yorizane	1.054	2.2%	
CH_4	Yorizane	1.110	4.5%	
H_2	All at 0 °C and about 1 bar	1.268	5.0%	$\varepsilon_{i,j}$ = 1.268 on figures
	All at about 1 bar	1.408	5.0%	
H_2O	All, at 330°C	0.911	1.8%	

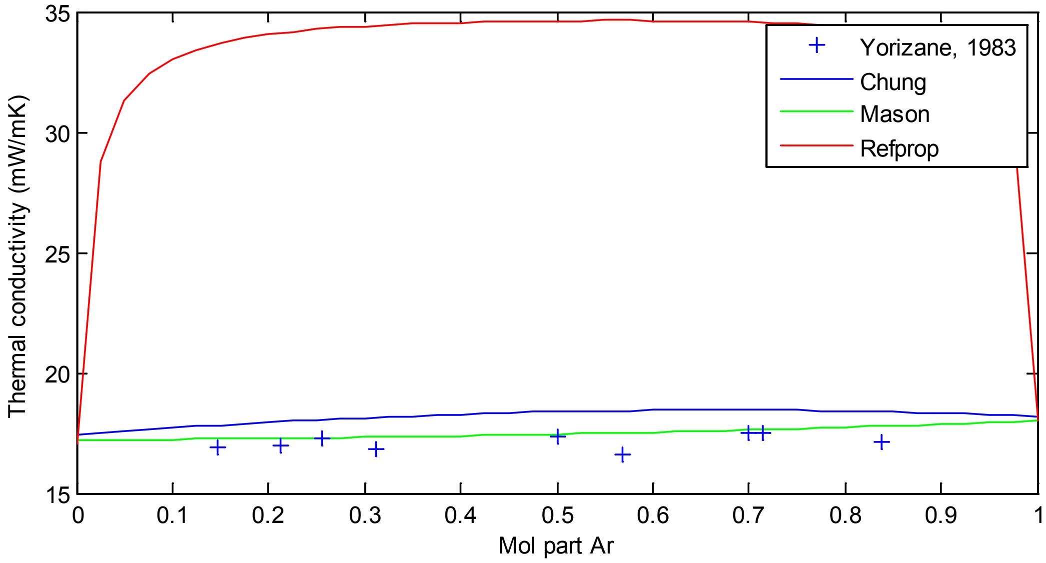

Figure E1: Thermal conductivity for mixture CO_2-Ar from Yorizane[61]

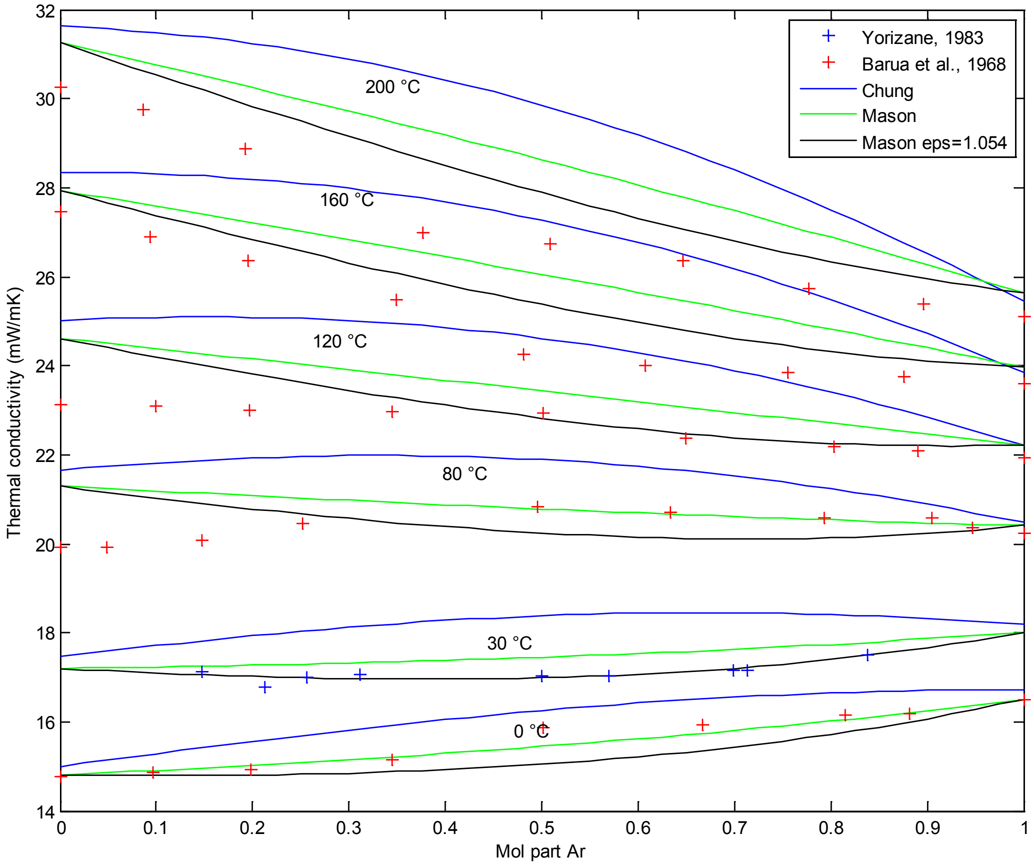

Figure E2: Thermal conductivity for mixture CO_2-Ar from Yorizane[61] and Barua[58]

2

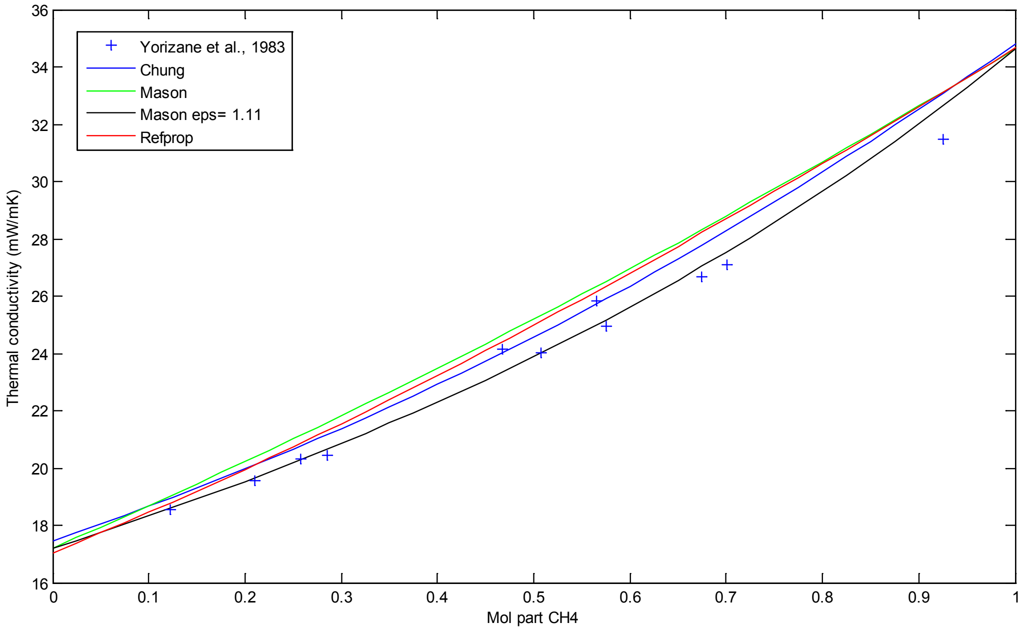

Figure E3: Thermal conductivity for mixture CO_2-CH_4 from Yorizane[61]

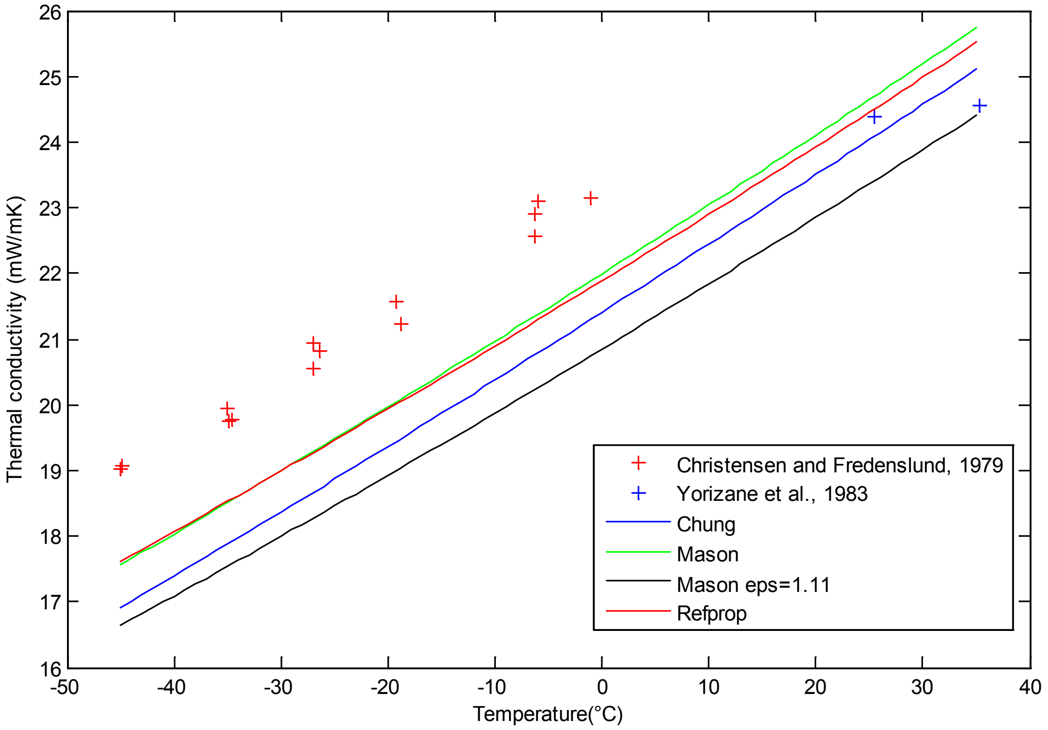

Figure E4: Thermal conductivity for mixture CO_2-CH_4 at 50% CO_2 and 50% CH_4, 2.8 Bar pressure from Christensen[34] and 1.0 bar from Yorizane[61]. Experiments from Yorizane are corrected to 2.8 bar as described in section 5.3

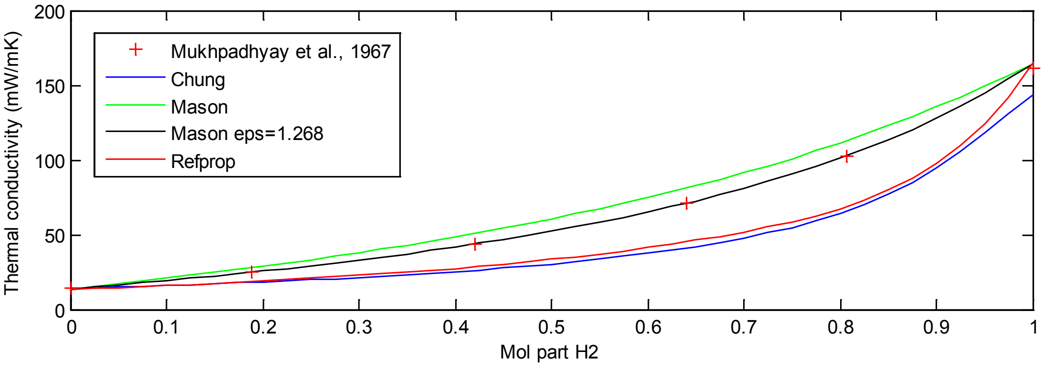

Figure E5: Conductivity for mixture CO_2-H_2 at -15°C from Mukhpadhyay [62]

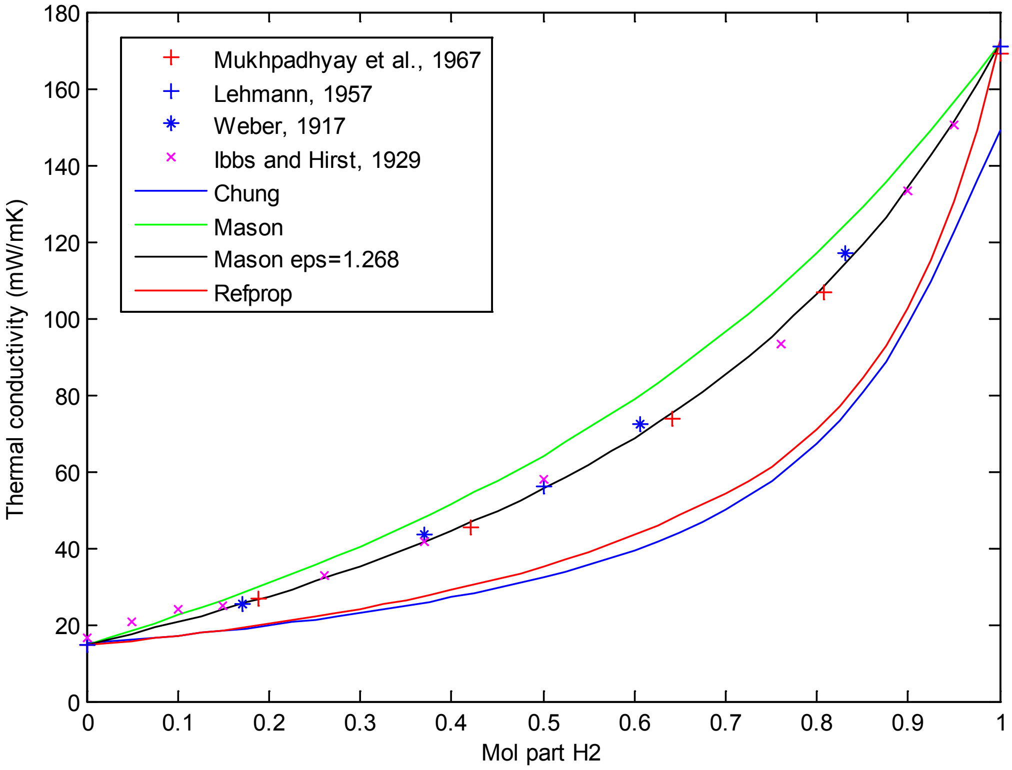

Figure E6: Conductivity for mixture CO_2-H_2 at 0°C from Mukhpadhyay [62], Lehmann [63], Weber [64] and Ibbs [65]

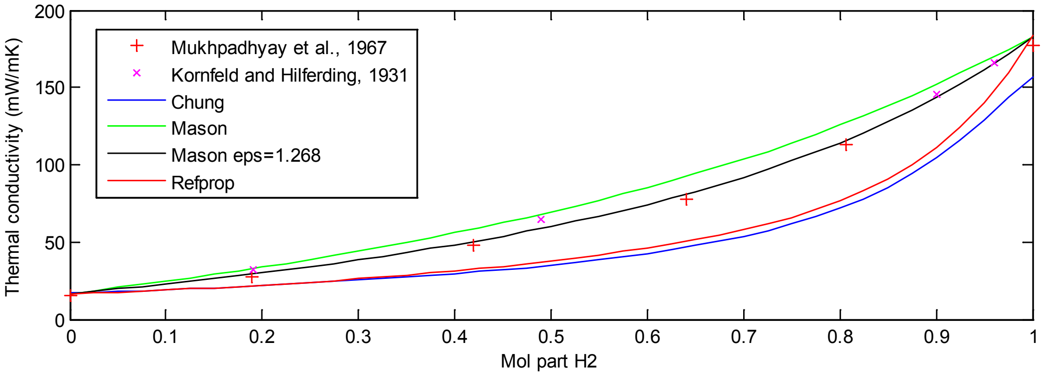

Figure E7: Conductivity for mixture CO_2-H_2 at 22°C from Mukhpadhyay [62] and Kornfeld [66]

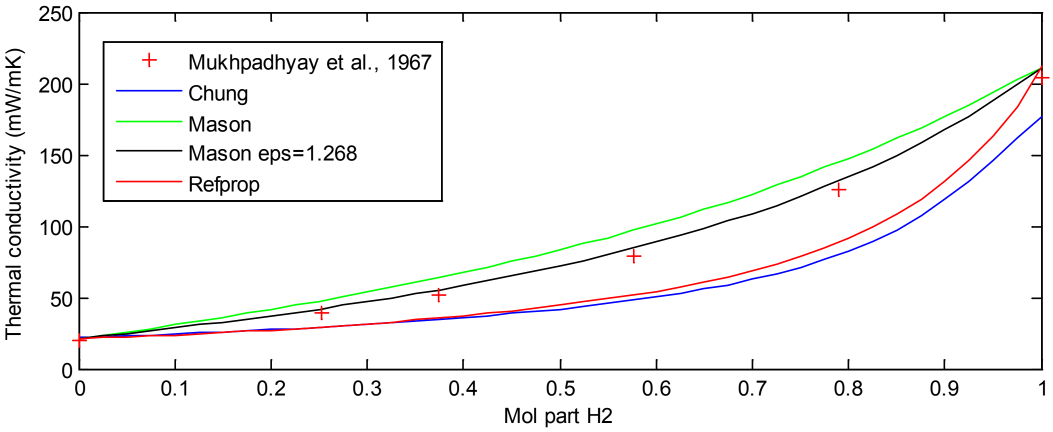

Figure E8: Conductivity for mixture CO_2-H_2 at 80°C from Mukhpadhyay [62]

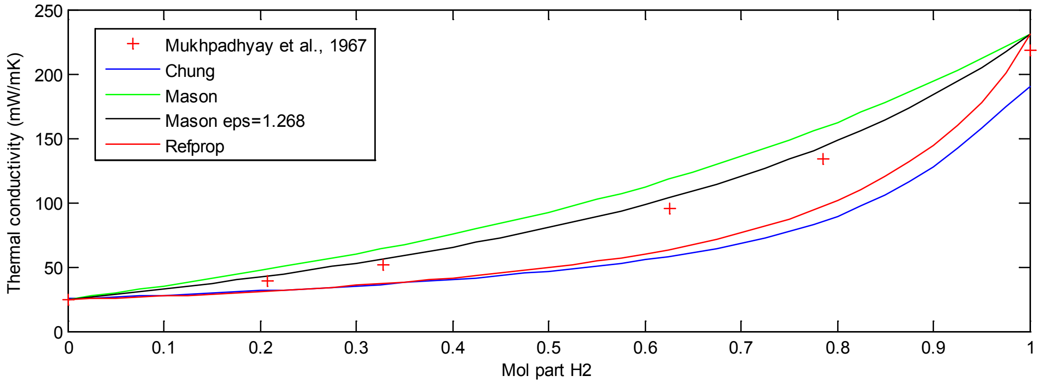

Figure E9: Conductivity for mixture CO_2-H_2 at 120°C from Mukhpadhyay [62]

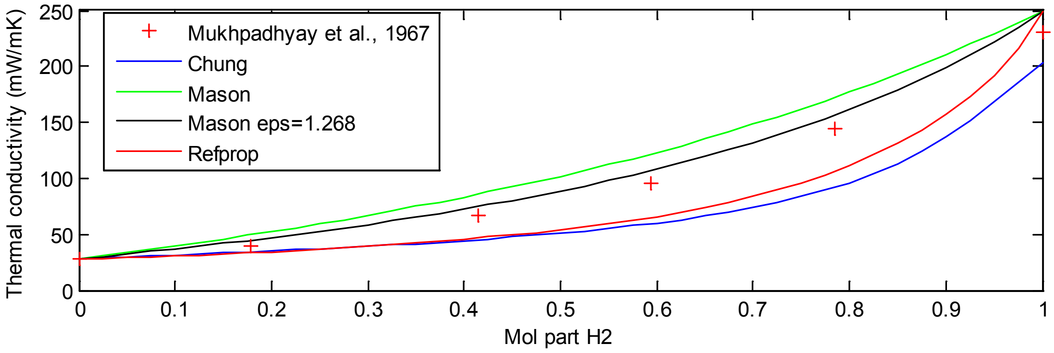

Figure E10: Conductivity for mixture CO_2-H_2 at 160°C from Mukhpadhyay [62]

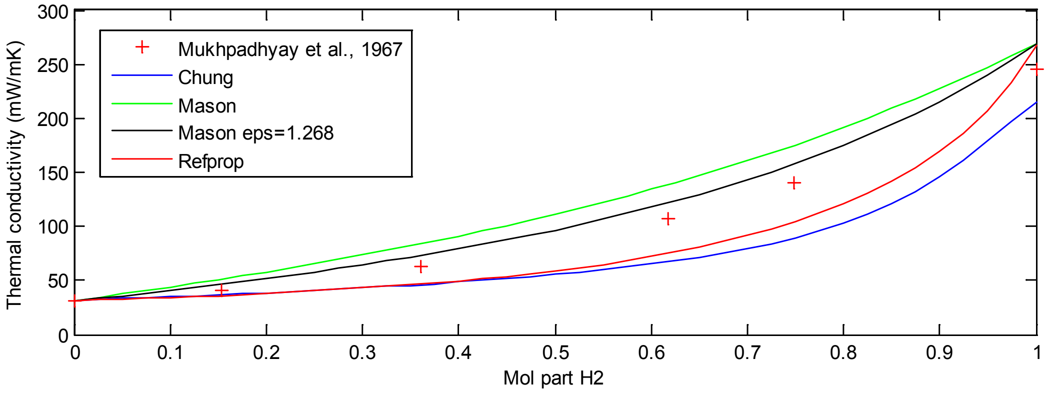

Figure E11: Conductivity for mixture CO_2-H_2 at 200°C from Mukhpadhyay [62]

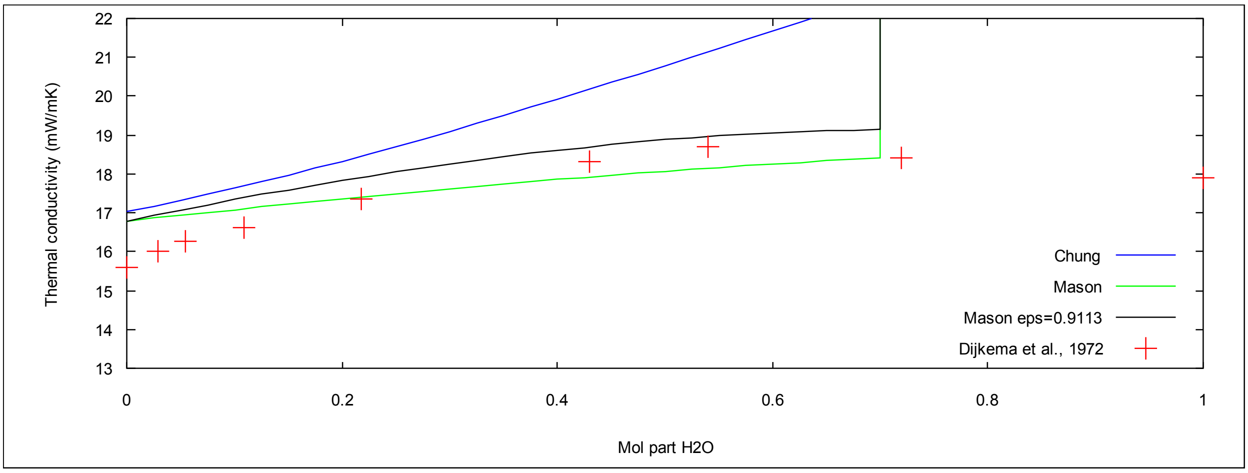

Figure E12: Conductivity for mixture CO_2-H_2O at 25 °C from Dijkema[59]

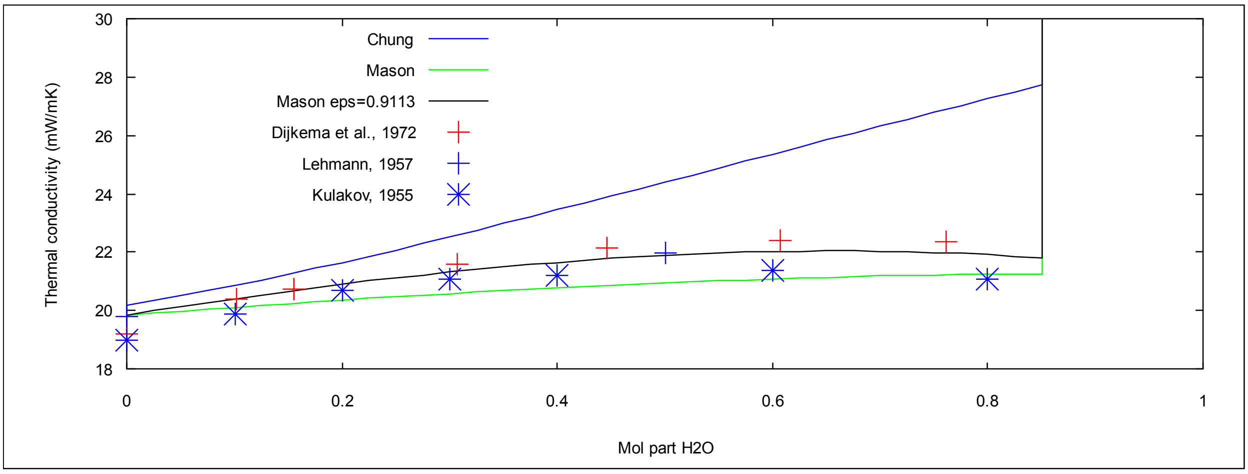

Figure E13: Conductivity for mixture CO_2-H_2O at 62.5°C from Dijkema[59], Lehmann[63] and Kulakov[67]

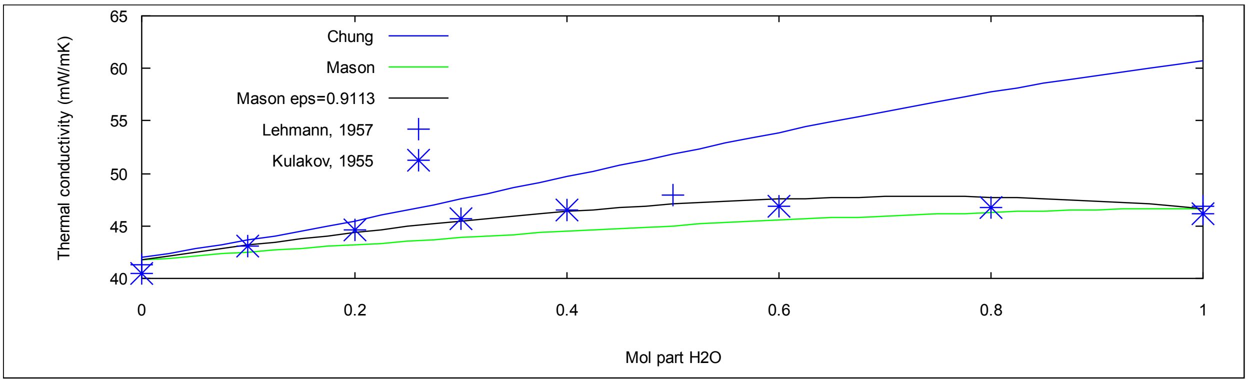

Figure E14: Conductivity for mixture CO_2-H_2O at 330 °C from Lehmann[63] and Kulakov[67]

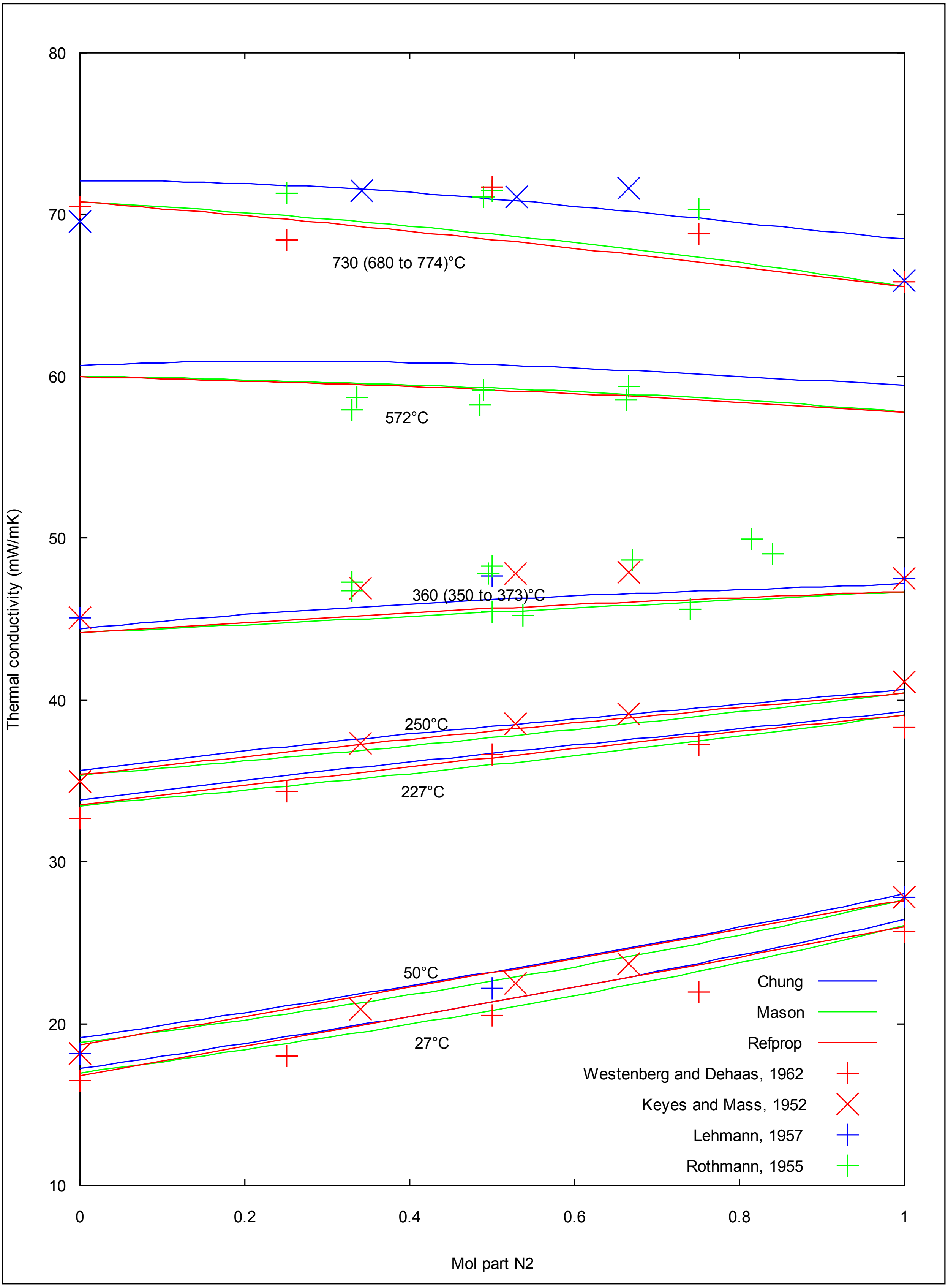

Figure E15: Conductivity for mixture CO_2-N_2 at different temperatures from Westenberg [68], Keyes[69], Lehmann[63] and Rothmann[70]

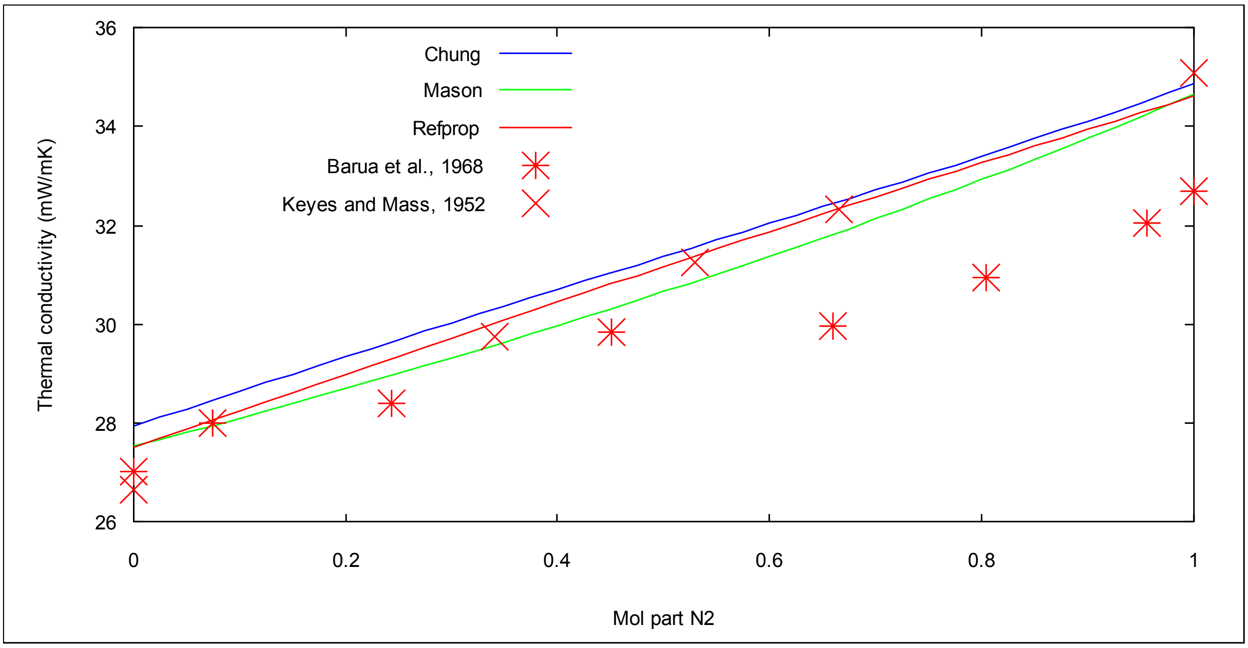

Figure E16: Conductivity for mixture CO_2-N_2 at 155 °C. Showing that Barua (1968)[58] is different from the others as Keyes[69]

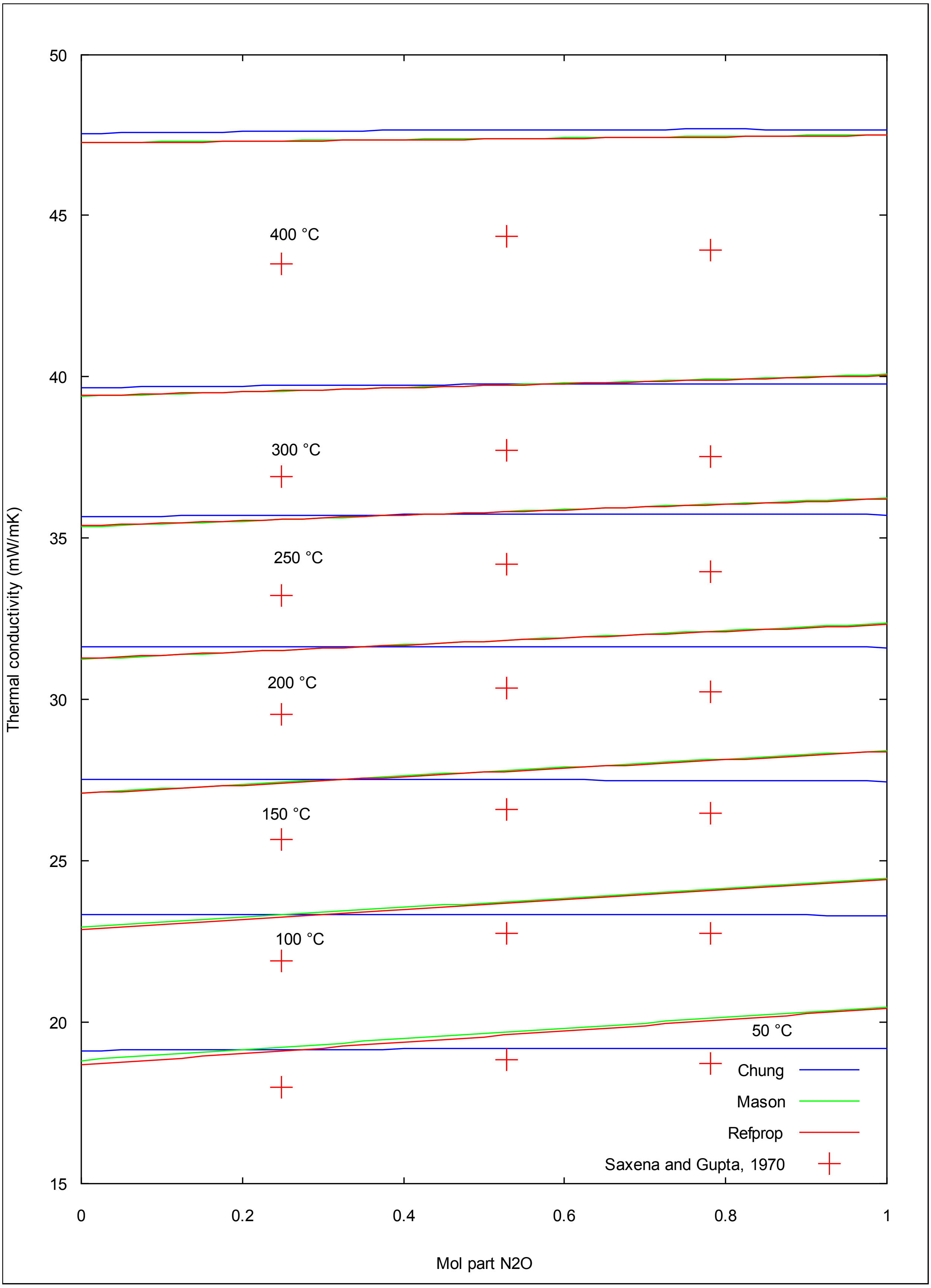

Figure E17: Conductivity for mixture CO_2-N_2O from Saxena [60] Did also measure at 350°C and 450°C not shown, from Saxena[60]

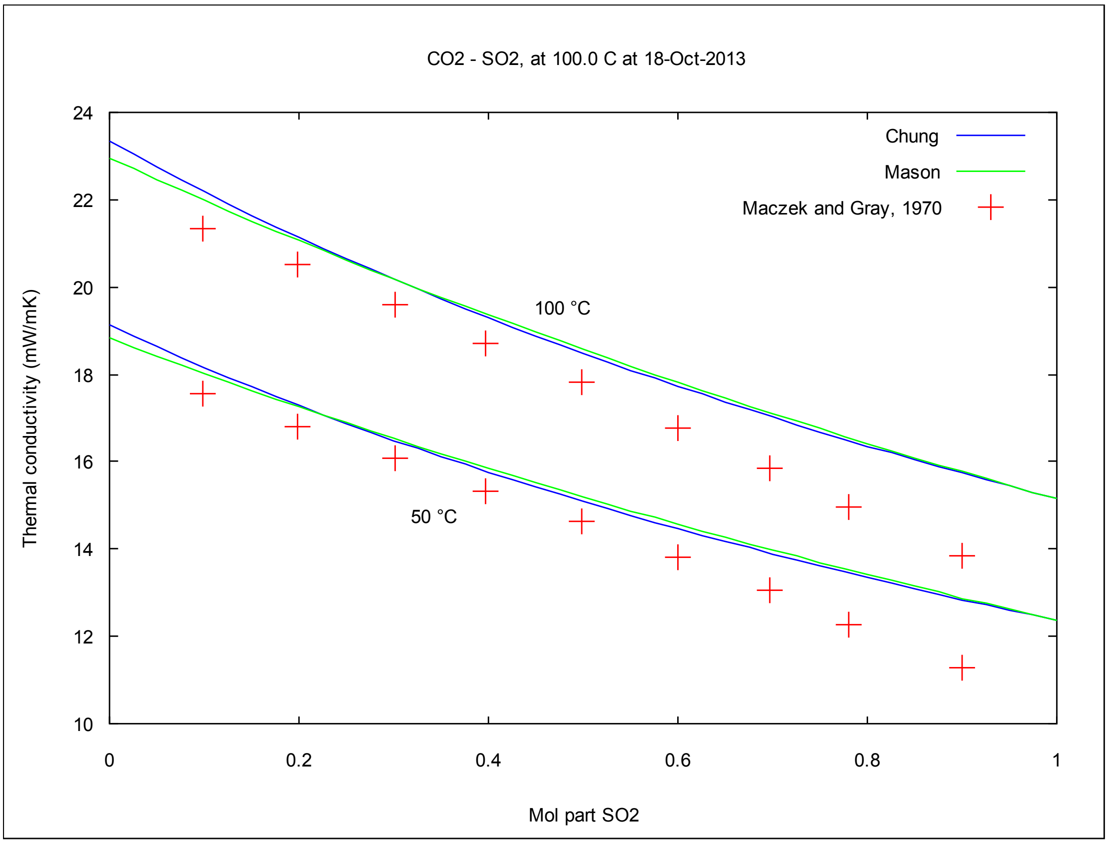

Figure E18: Conductivity for mixture CO_2-SO_2 from Maczek[71]

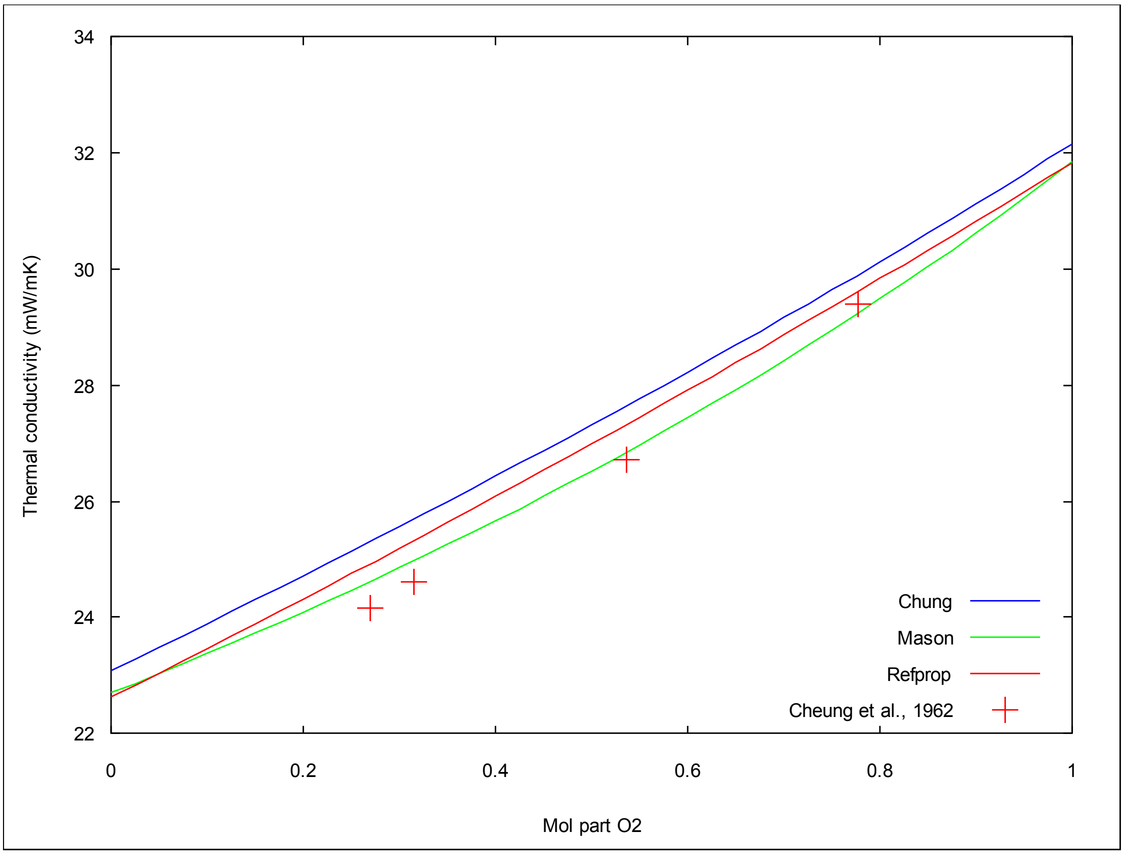

Figure E19: Conductivity for mixture CO_2-O_2 at 97 °C from Cheung[72]

APPENDIX F: COMPARISION OF THERMAL CONDUCTIVITY AT HIGHER PRESSURE

Table F1: Area and deviation for measurements at higher pressure

Figure	Mix-ture CO_2-	P_{Max} (bar)	X_2(%)	T(°C)	Deviation AAD in %				Reference
					Chung	TRAPP, Mason and Saxena[1]		REF-PROP	
						$\varepsilon_{i,j}$=1	$\varepsilon_{i,j}$ adapted		
F1-F3	Ar	120	17%, 48%, 74%	26	1.6%	2.9%	2.0%	86%	Kestin 1982, [73]
	Compare with Yorizane (1983) on same figure shows good quality of measurement. REFPROP off as in figure E1. Chung below and the TRAPP above the curve. Chung good.								
F1-F5	Ar	91	0 – 100%	25, 35	2.5%	4.3%	2.5%	90%	Yorizane 1983 [61]
	Show good comparison with Chung's model except at pressures of 11 bar and below.								
F6-F7	CH_4	20	51%	-45 to 0	12%	4.2%	8.9%	5.9%	Christensen [34]
	From figure E4 it looks as Christensen is inaccurate.								
F8-F10	CH_4	700	24%, 46%, 76%	63, 98, 131	12%	11%	8.6%	5.2%	Rosenbraum, 1969 [74]
	Here all models give higher conductivity than this reference above 200 bars. REFPROP does well at below 100 bars and it gives same results as Yorizane, 1983.								
F11-F12	CH_4	90	0-100	25, 31	4.2%	4.3%	1.5%	2.1%	Yorizane 1983 [61]
	Here both REFPROP and adapted Mason and Saxena do well. High consistence in measurement								
F13	H_2	75	21%, 48%, 74%	27	41%	10%	3.1%	36%	Kestin 1983 [42]
	As shown in Figure E7 both REFPROP and Chung's model are not able to calculate this mixture with high difference in conductivity between H_2 and CO_2. To make the graph readable, this models is only included at 21% H_2								
F14-F16	N_2O	45	20%, 41%, 64%	28	5.0%	6.0%	6.0%[2]	1.8%	Imaishi, 1984 [75]
	Here does REFPROP very well, Mason S. and Chung are one each side of the measurements								
Average					11%	6.1%	4.7%	32%	

[1]: TRAPP used for higher pressure, Mason S. for mixture

[2]: No $\varepsilon_{i,j}$ adapted in Mason S. method

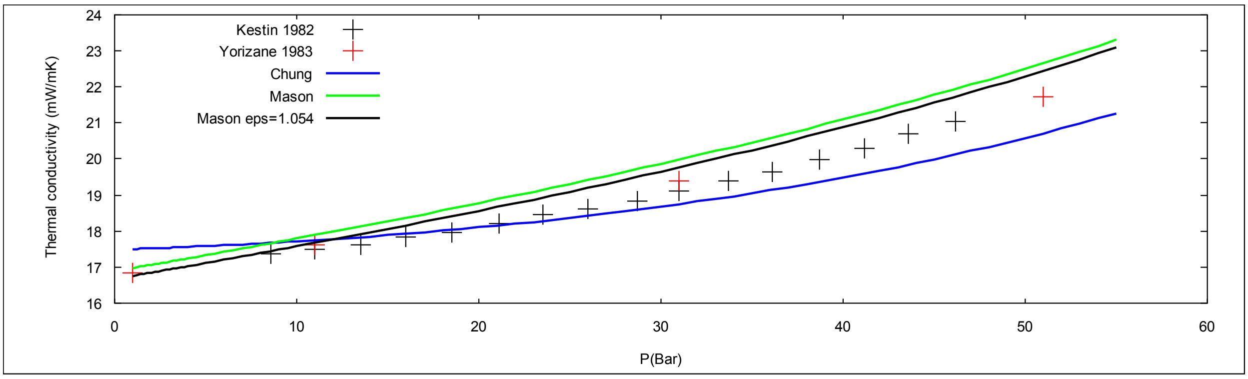

Figure F1: Conductivity for mixture with 17% Ar, 83% CO_2 at 26.3°C. REFPROP is not included wince it is considerable off, see Figure E1. From Kestin[73] and Yorizane[61].

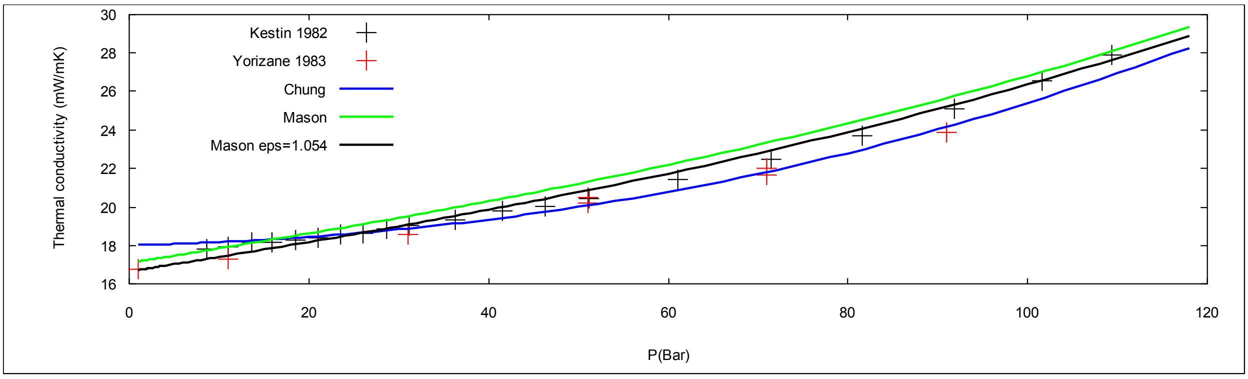

Figure F2: Conductivity for mixture with 48% Ar, 52% CO_2 at 26.3°C. REFPROP is not included wince it is considerable off, see Figure E1. From Kestin[73] and Yorizane[61].

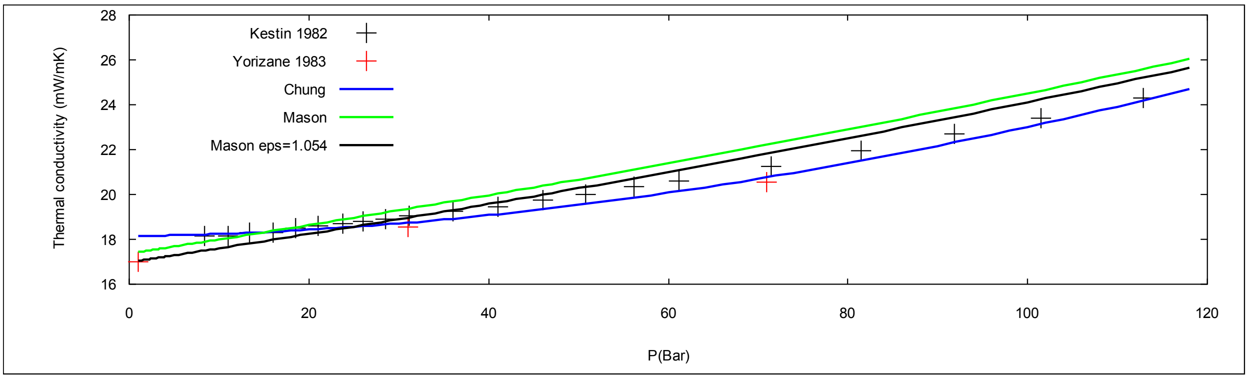

Figure F3: Conductivity for mixture with 73.6% Ar, 26.4% CO_2 at 26.3°C. REFPROP is not included wince it is considerable off, see Figure E1. From Kestin[73] and Yorizane[61].

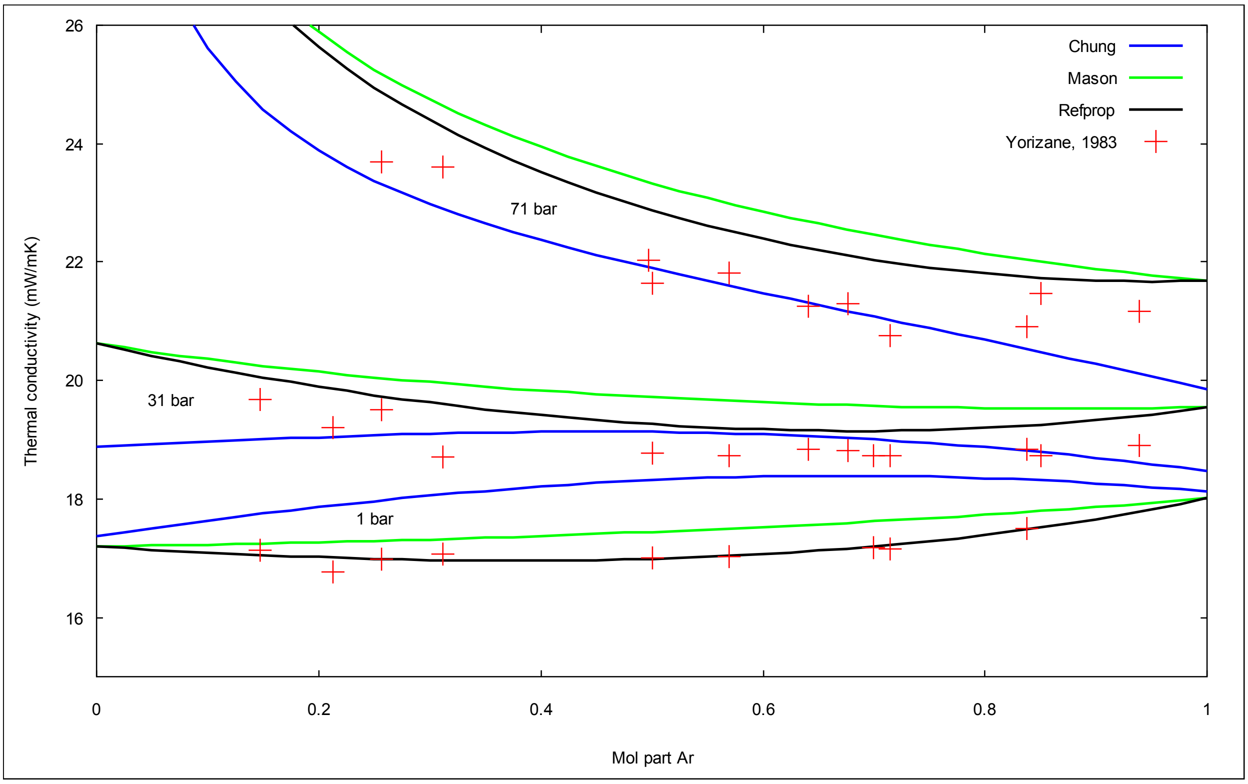

Figure F4: Conductivities at different pressures at 30°C for mixture Ar-CO_2, from Yorizane[61].

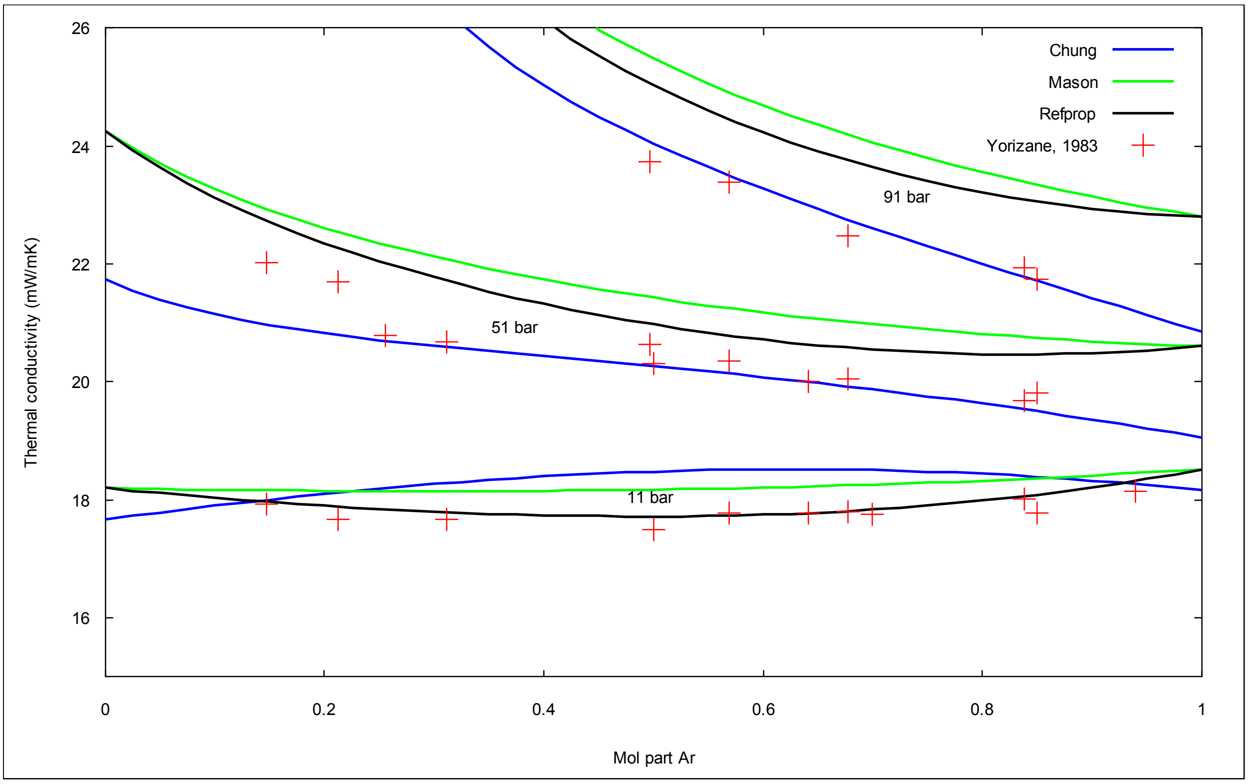

Figure F5: Conductivities at different pressures at 30°C for mixture Ar-CO_2, from Yorizane[61].

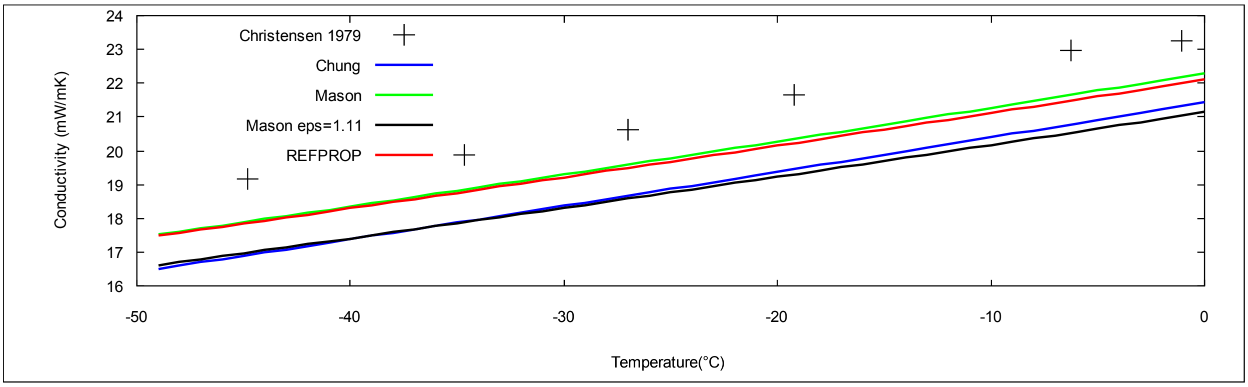

Figure F6: Thermal conductivity for mixture with 49.4% CO_2 and 50.6 % CH_4 at 2.8 Bars compared with Christensen [34]

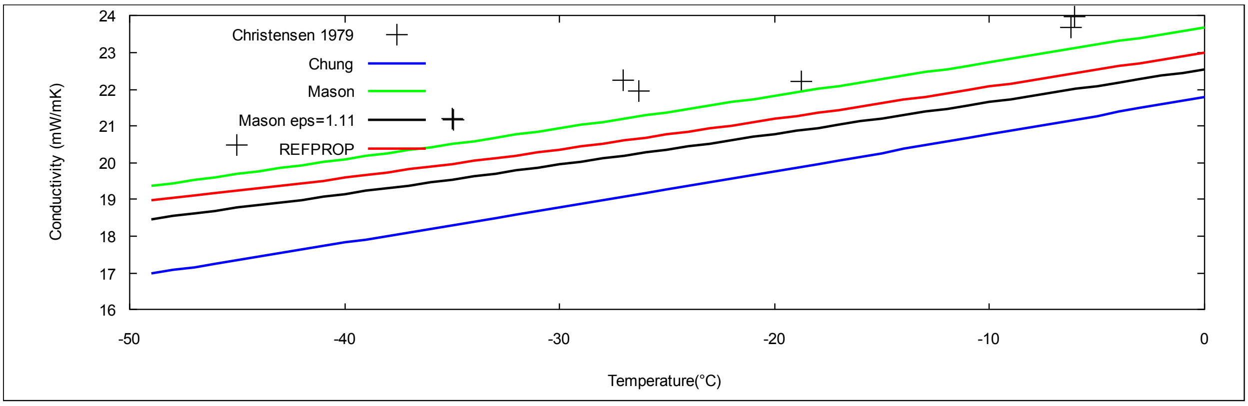

Figure F7: Thermal conductivity for mixture with 49.4% CO_2 and 50.6 % CH_4 at 14.5 Bars compared with Christensen [34]

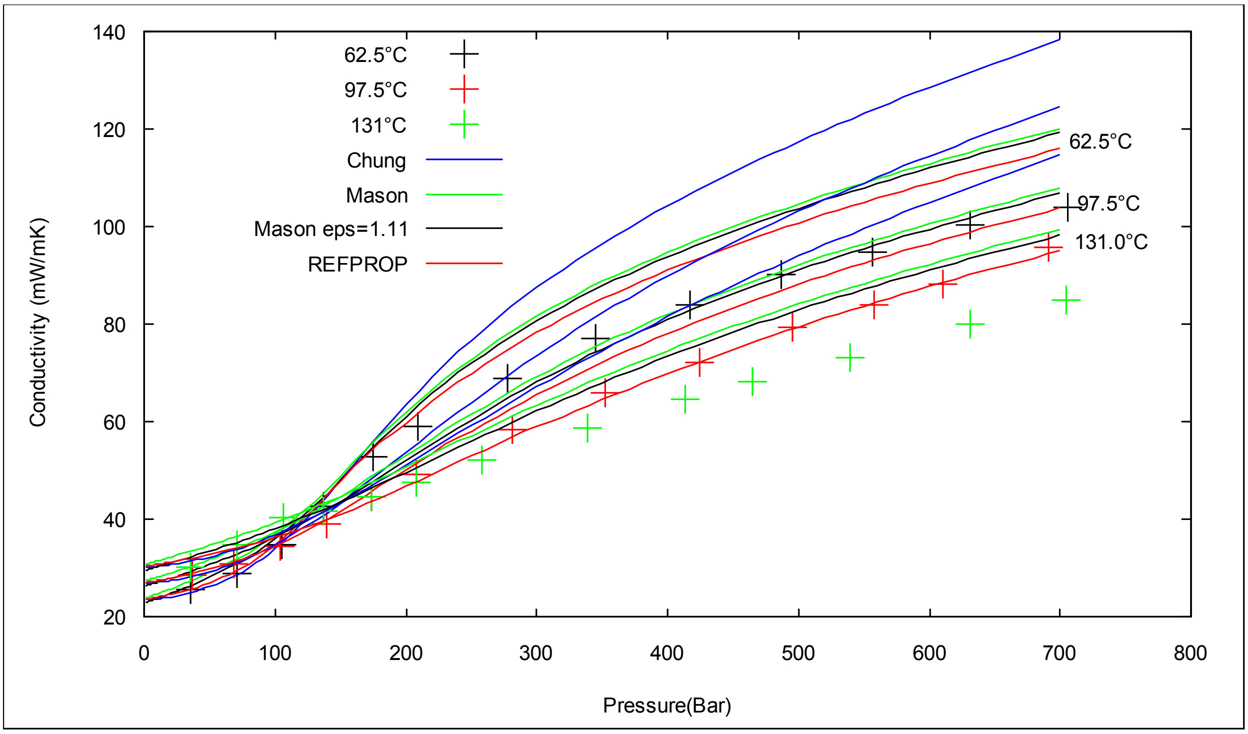

Figure F8: Thermal conductivity for mixture with 75.7% CO_2 and 24.3% CH_4 compared with Rosenbraum (1969) [74]

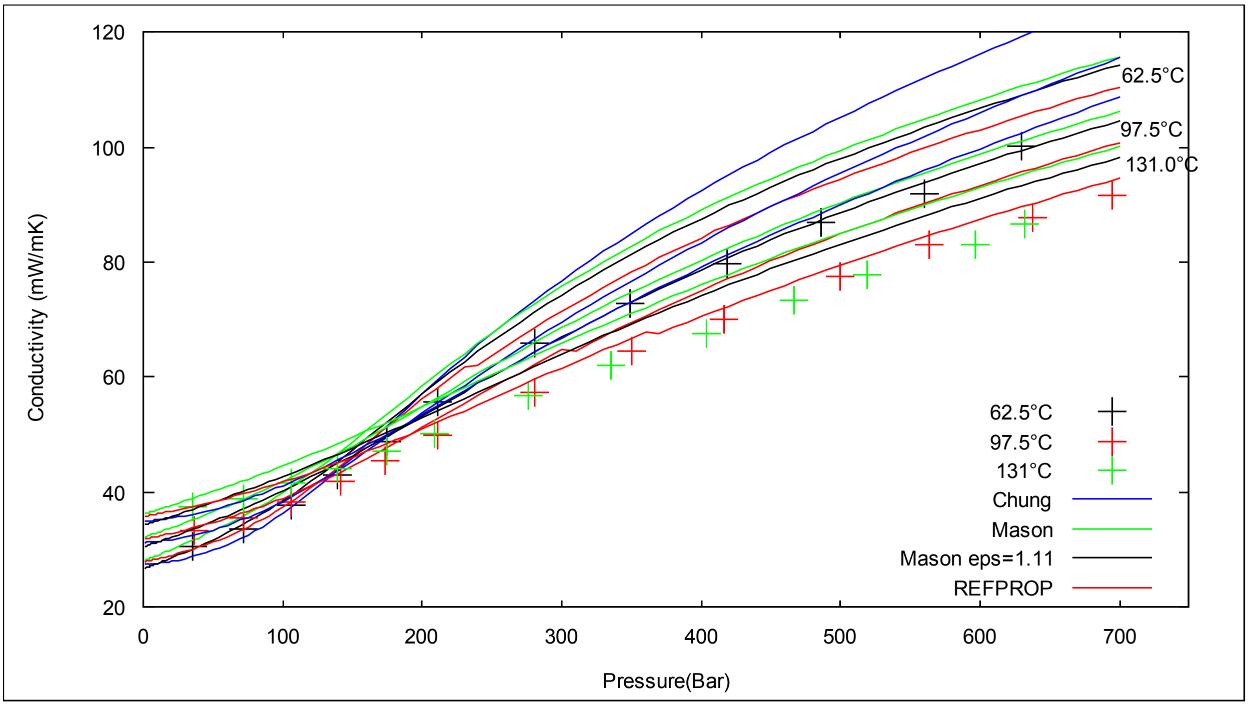

Figure F9: Thermal conductivity for mixture with 53.6% CO_2 and 46.4% CH_4 compared with Rosenbraum (1969) [74]

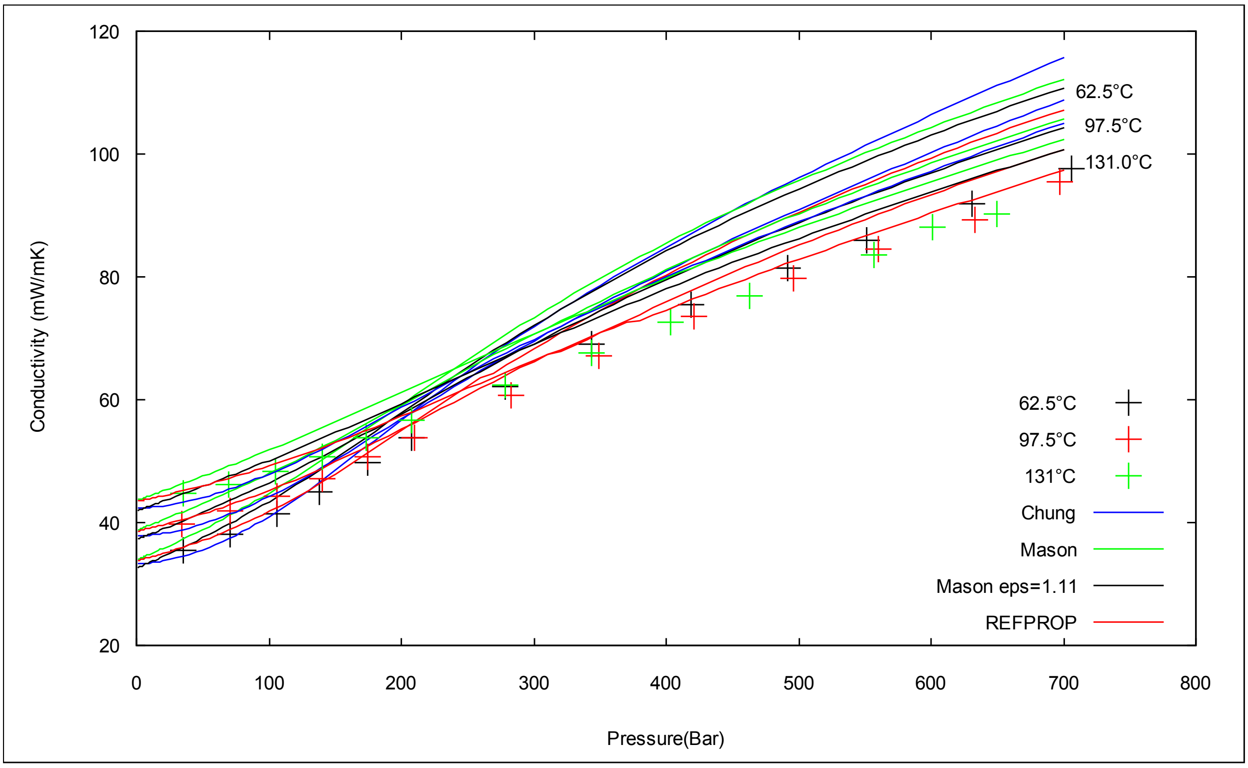

Figure F10: Thermal conductivity for mixture with 24.5% CO_2 and 75.5% CH_4 compared with Rosenbraum (1969) [74]

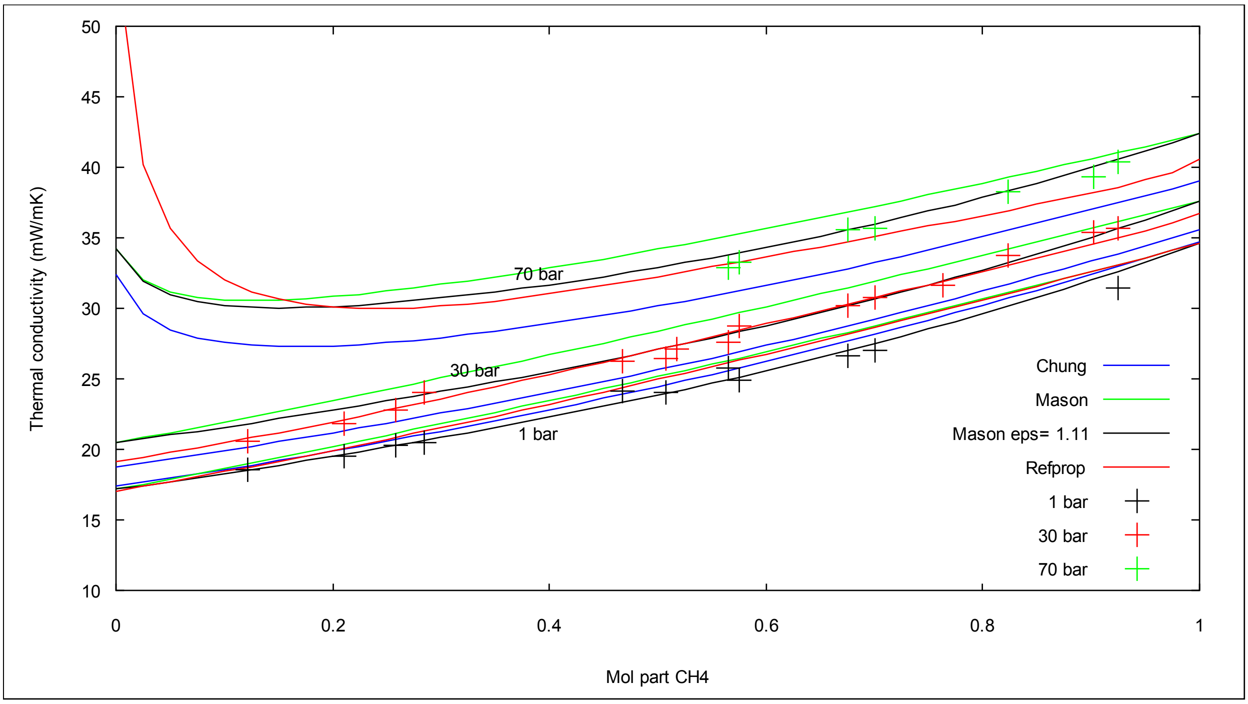

Figure F11: Thermal conductivity for mixture CO_2-CH_4 at 30 °C, measurement at 25.1°C and 35.5°C (But not possible to see difference when changed to 30°C) from Yorizane 1983 [61]

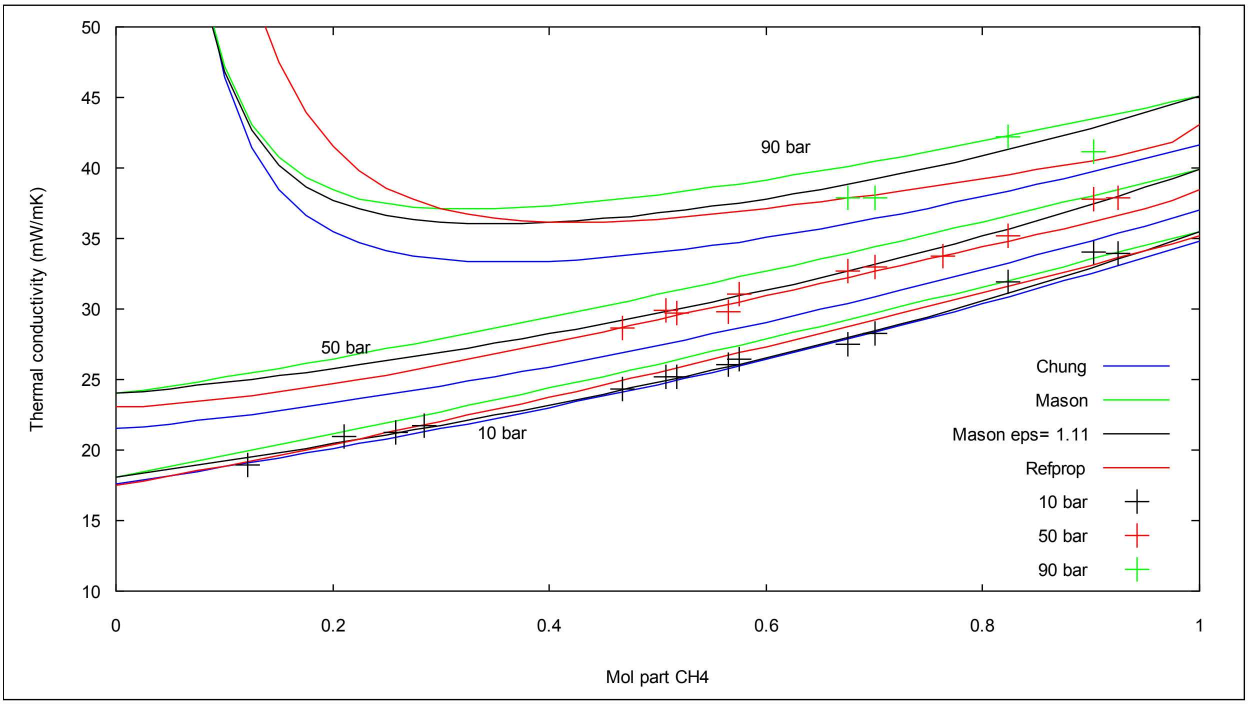

Figure F12: Thermal conductivity for mixture CO_2-CH_4 at 30 °C. Measurement done at 25.1°C and 35.5°C from Yorizane 1983 [61]. This is at different pressures than figure D9. Measurements is from Yorizane 1983[61].

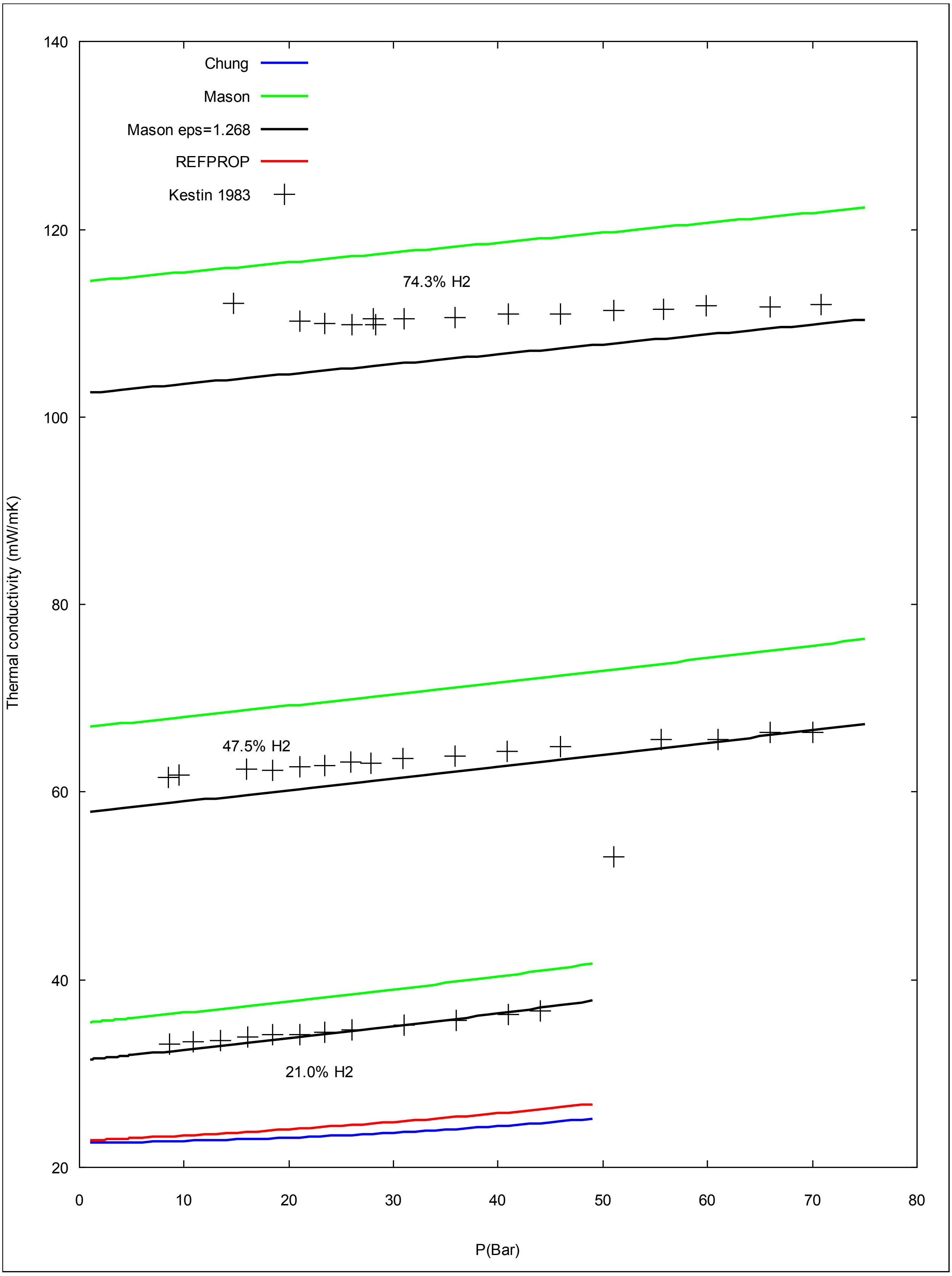

Figure F13: Conductivity for mixture CO_2-H_2 as function of pressure at 27°C and different concentrations of H_2. REFPROP and Chung are off and will hit the curve below and are of that reason only shown for 21% H_2. Also see figure E6. From Kestin[42]

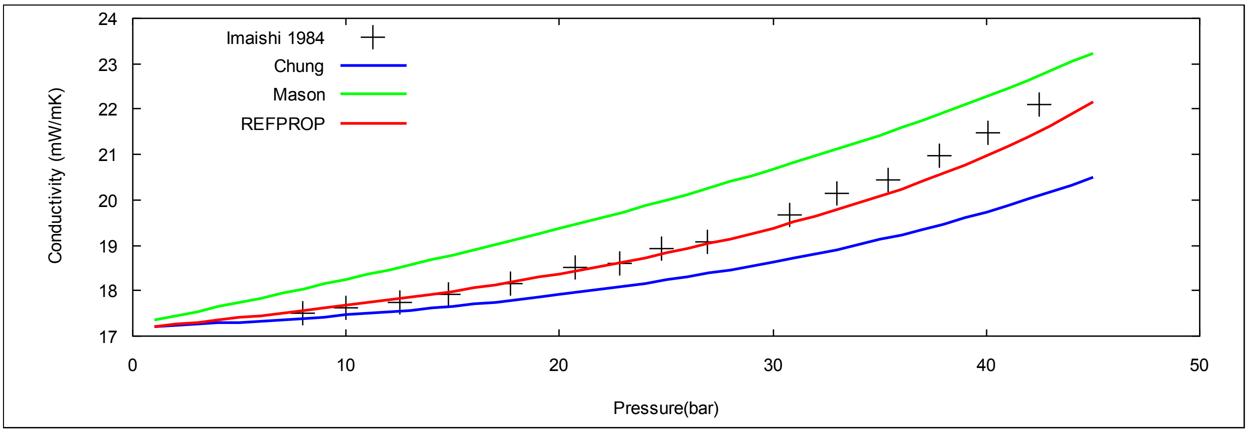

Figure F14: Conductivity for mixture with 79.6% CO_2 and 20.4% N_2O at 27.6°C from Imaishi, 1984 [75]

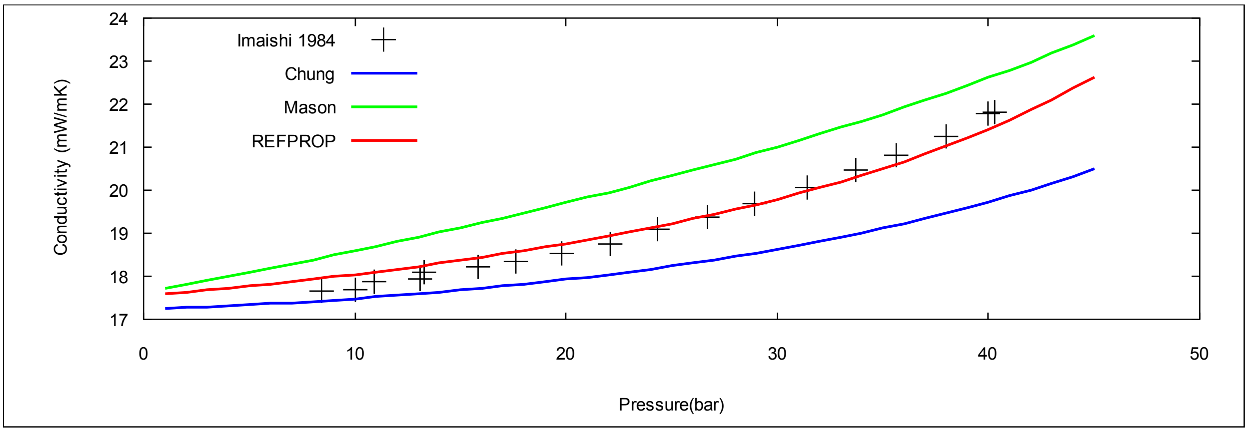

Figure F15: Conductivity for mixture with 58.9% CO_2 and 41.1% N_2O at 27.6°C from Imaishi, 1984 [75]

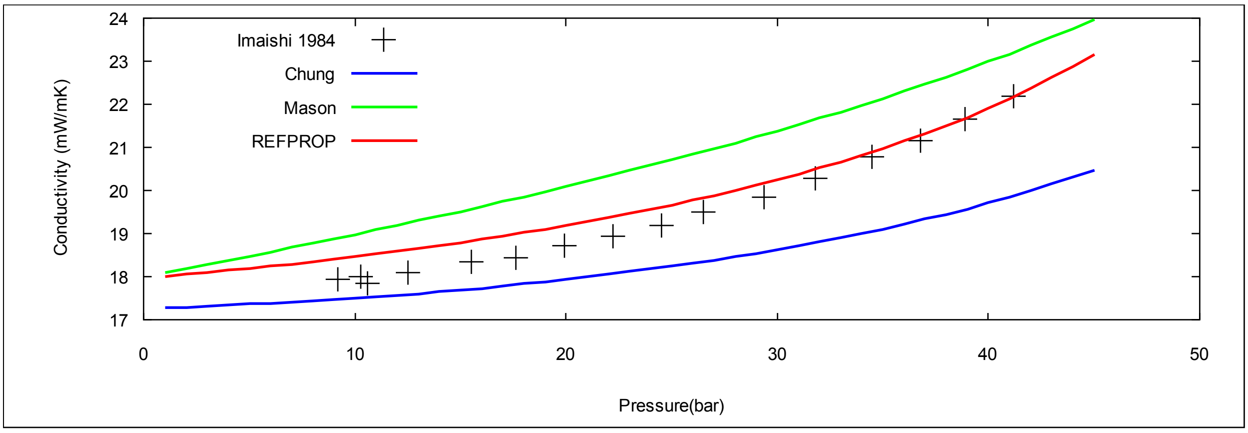

Figure F16: Conductivity for mixture with 35.6% CO_2 and 64.3% N_2O at 27.6°C from Imaishi, 1984 [75]

APPENDIX G: COMPARING OF DIFFUSIVITIES AT LOW PRESSURE

Table G1: Comparison of binaries

Binary	T(°C)	ADD		Figure
		Fuller	Wilke	
CO_2-CO_2	-80 to 90	20%	7%	G1
	900 to 1400	20%	23%	G2
	The second component uses C^{14}.			
CO_2-Ar	0 to 200	5.1%	4.1%	G3
CO_2-CO	20 to 200	4.5%	2%	G4
CO_2-H_2O gas	35 to 80	21%	7.8%	G5
CO_2-H_2O liquid	5 to 75	314%	209%	G6
CO_2-H_2O liquid, correct viscosity	5 to 75	210%	131%	
CO_2-N_2	17 to 200	4.4%	2.6%	G7
	300 to 850	6%	4.0%	G8
CO_2-N_2O	-80 to 90	38%	5.4%	G9
CO_2-O_2	0 to 2700	6.9%	9.2%	G10
CO_2-SO_2	-10 to 200	27%	20%	G11
Average for gas phase		15.3%	8.5%	

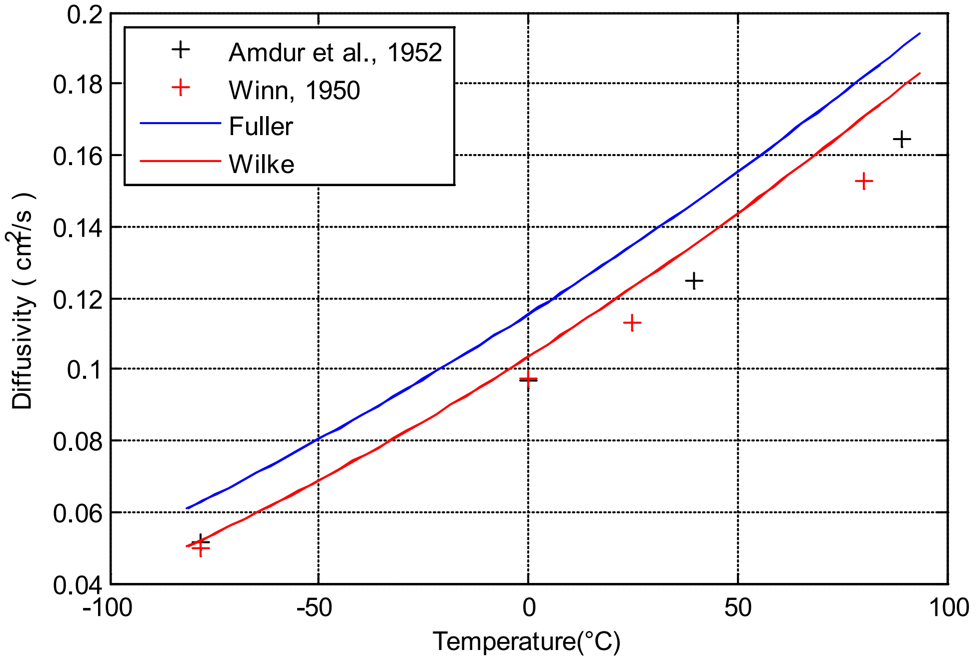

Figure G1: Diffusivity CO_2-CO_2 with data from Amdur[76] and Winn[77]

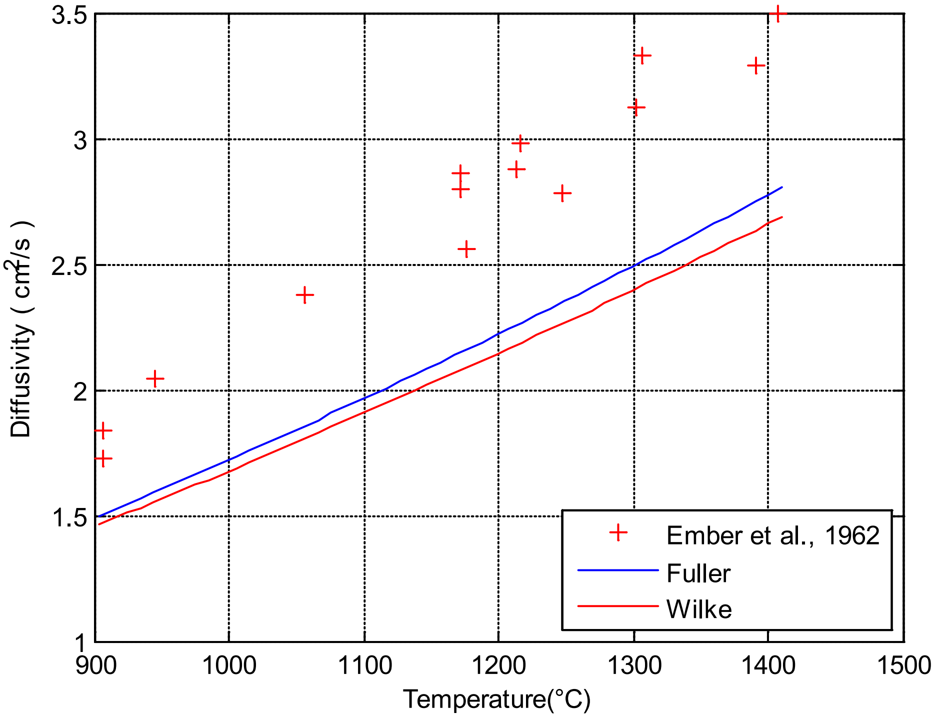

Figure G2: Diffusivity CO_2-CO_2 at higher temperatures with data from Ember[78]

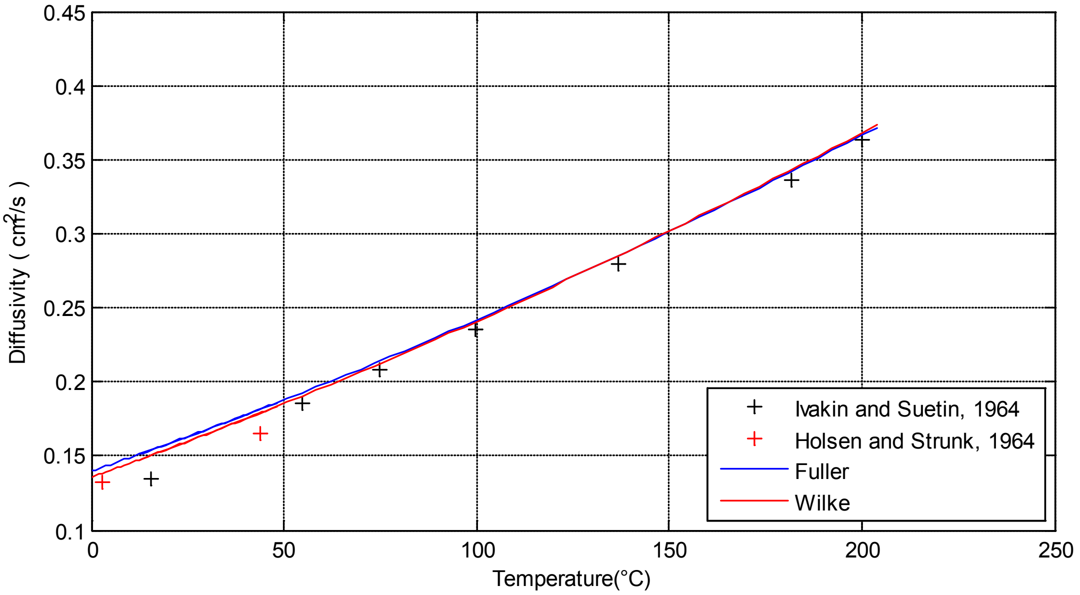

Figure G3: Diffusivity for CO_2-Ar with data from Ivakin[79] and Holsen[80]

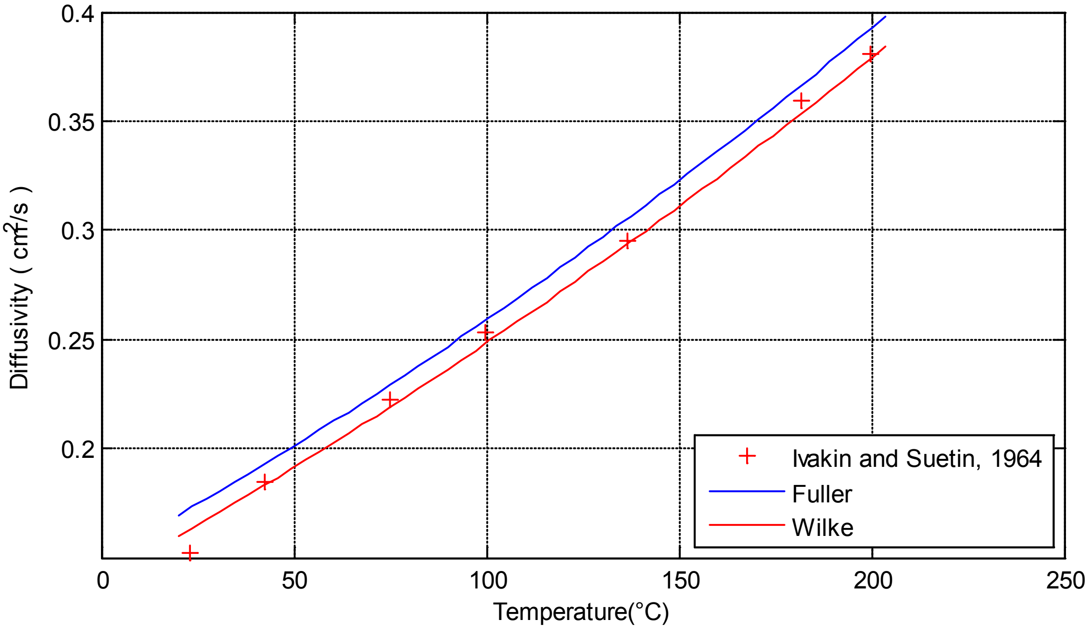

Figure G4: Diffusivity for CO_2-COwith data from Ivakin[79]

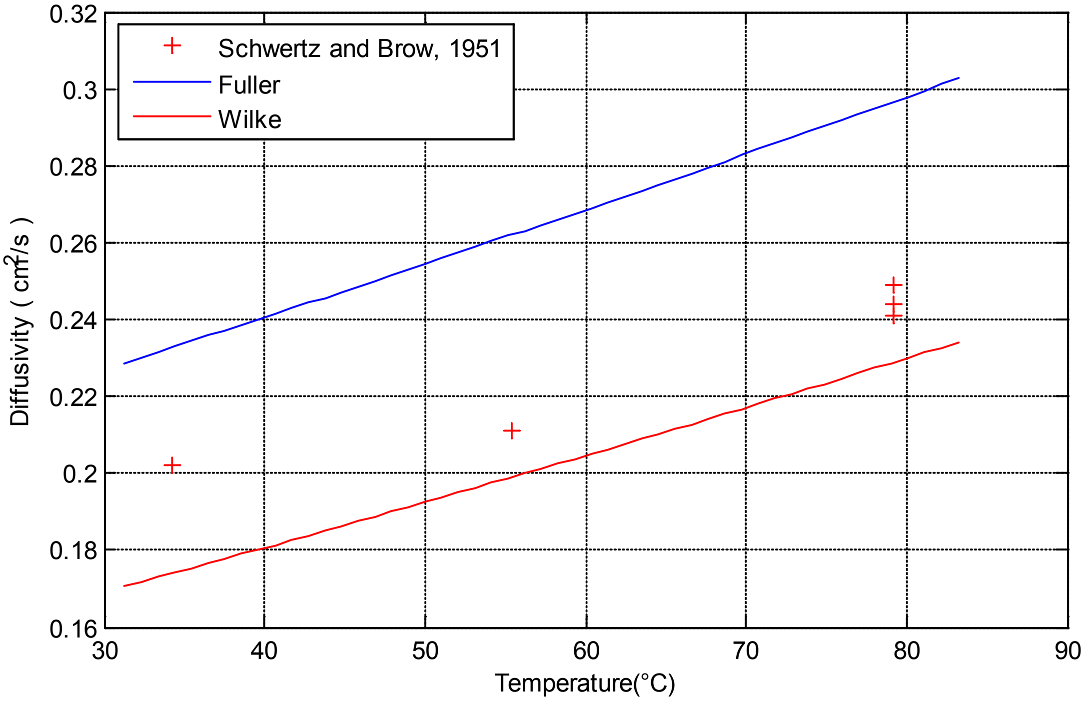

Figure G5: Diffusivity CO_2-H_2O: Normalized to 1 atm pressure with data from Schwerts [81]

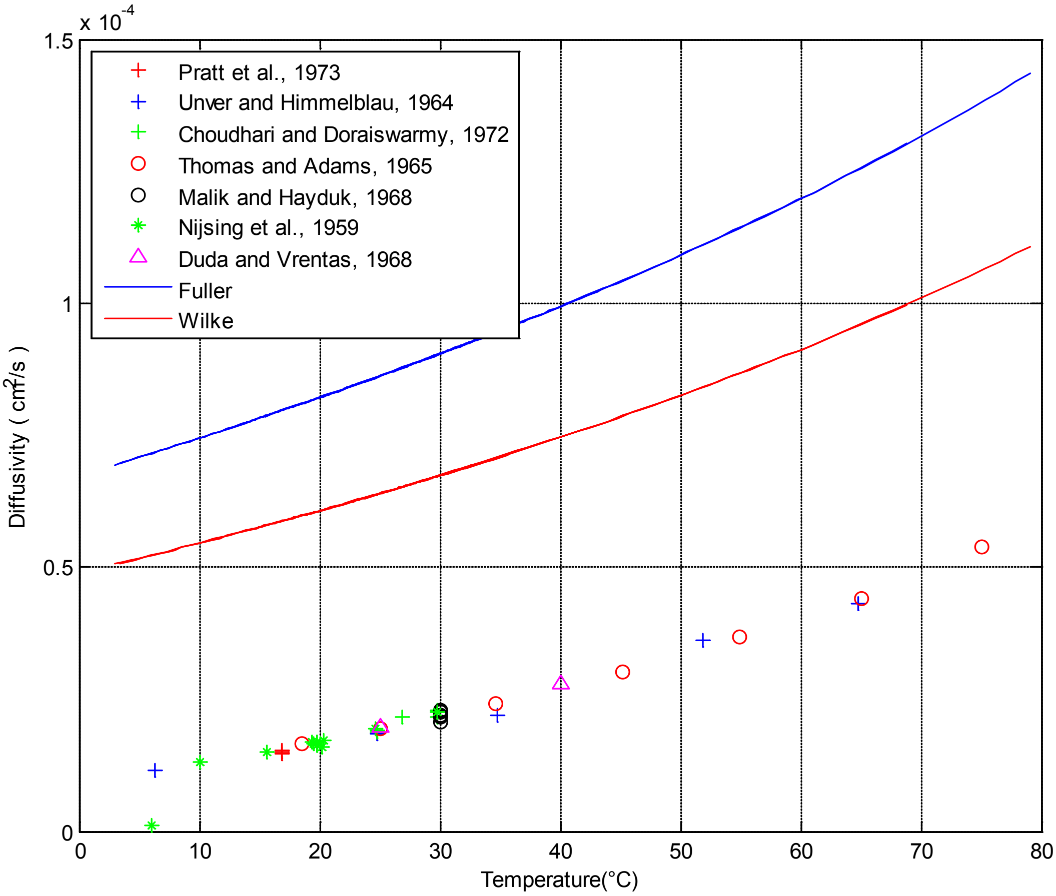

Figure G6: Diffusivity CO_2-H_2O at liquid phase with data from ref[82-88]

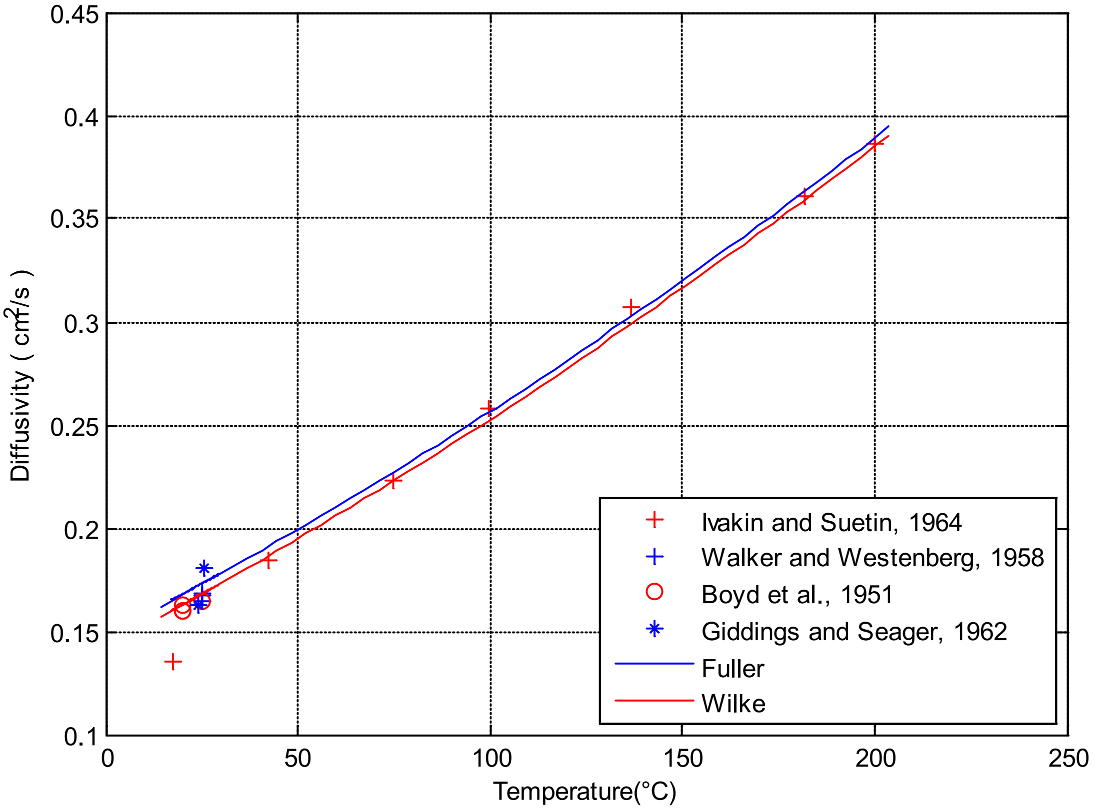

Figure G7: Diffusivity CO_2-N_2 with data from ref [79, 89-91]

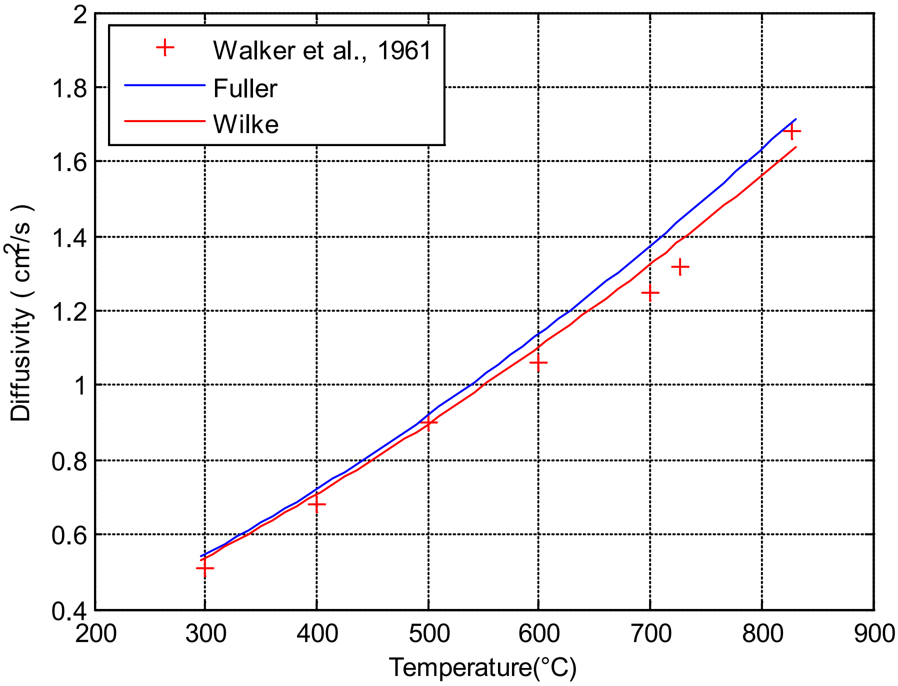

Figure G8: Diffusivity CO_2-N_2 at higher temperatures with data from Walker [92]

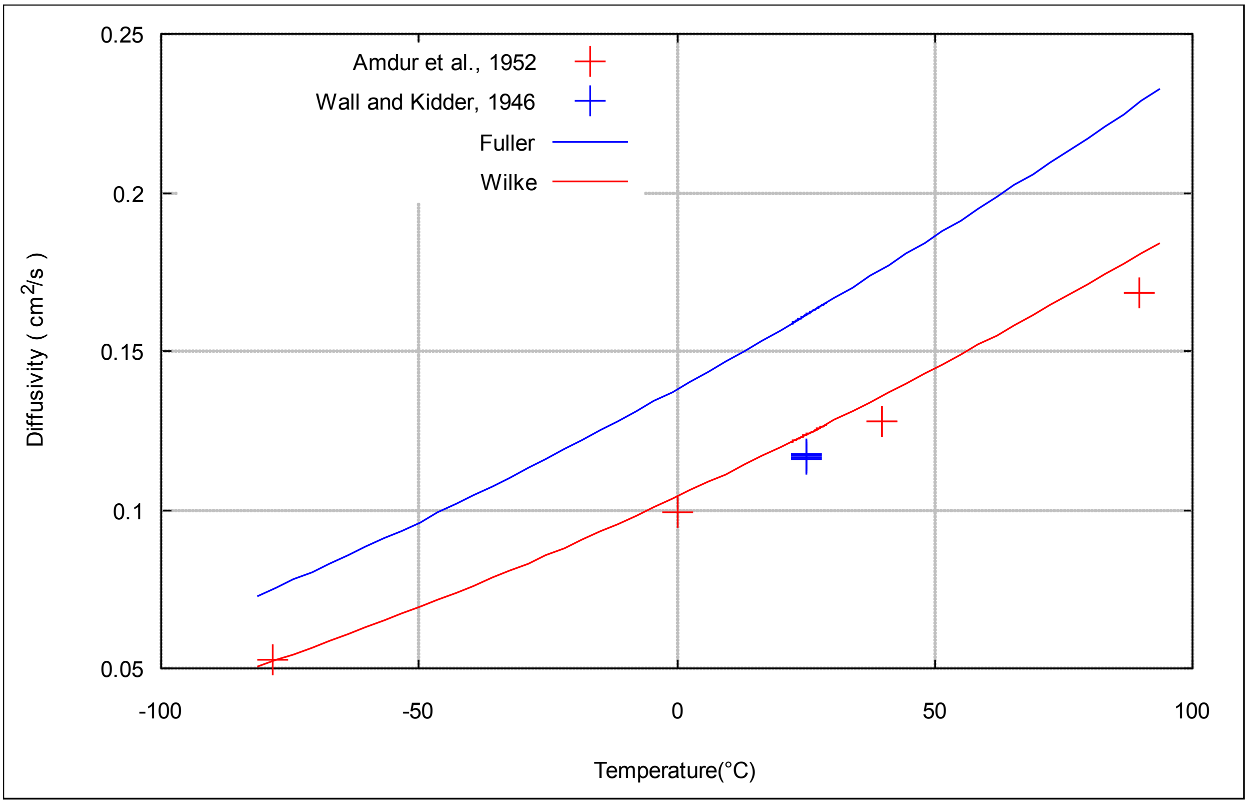

Figure G9: Diffusivity CO_2-N_2O with data from Amdur [76] and Wall [93]

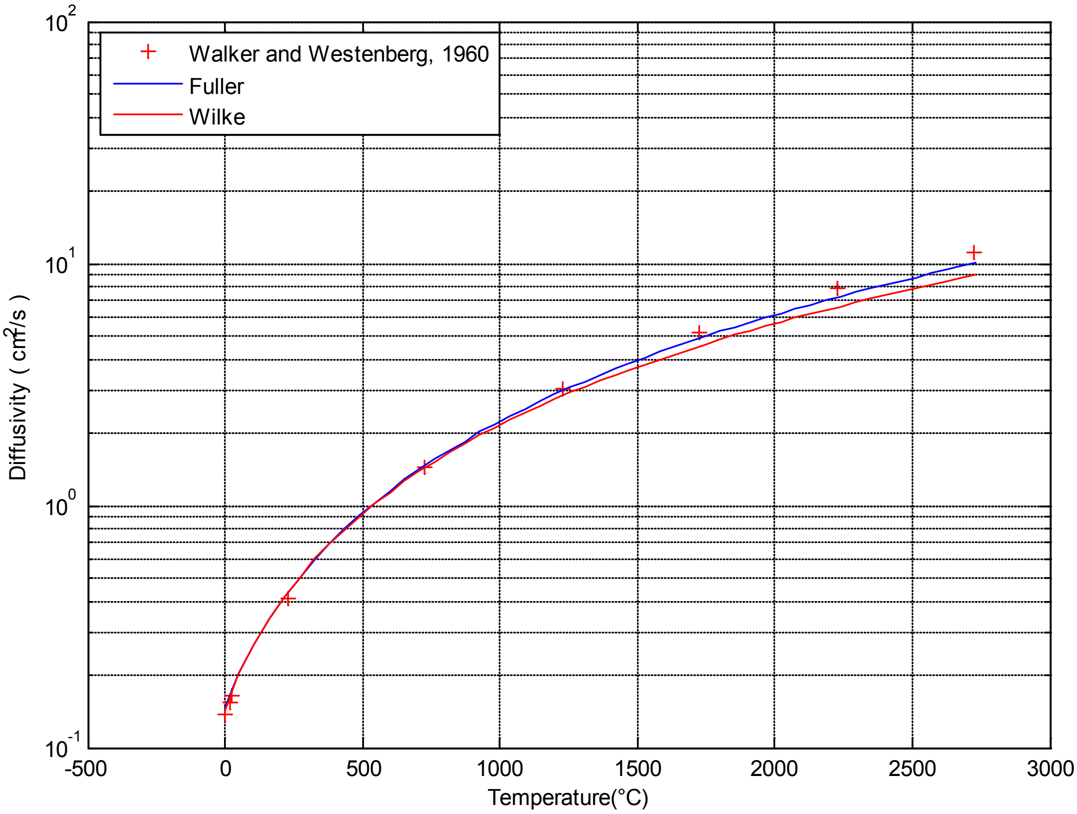

Figure G10: Diffusivity CO_2-$O_2$with data from Walker [94]

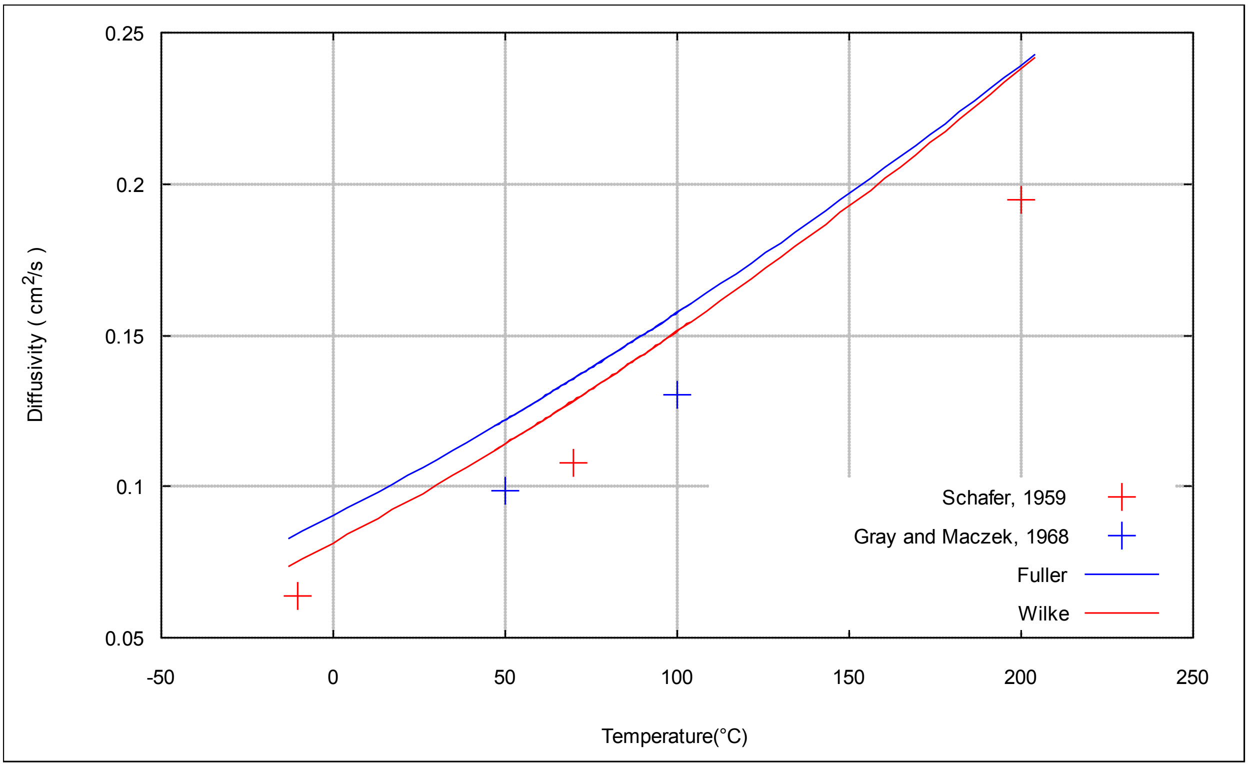

Figure G11: Diffusivity CO_2-SO_2 with data from Schafer[95] and Gray[96]

SYMBOLS USED

Latin letters

A	Area (m^2)
A,B,C,D,E,F,..:	Constants where numeric value is given.
$A_1, A_{2,..}$	Curve adapted constants, any dimension
C_v	Heat capacity at constant volume (J/mol K)
c:	Concentration (mol/m^3)
f_i, g_i	Factor in the TRAPP method, see Eq(7)
G	Gibbs energy (J/mol)
J_i	Mol flux in mixture of component *i*. (mol/m^2s)
k	Boltzmann's constant, $\approx 1.380622 \times 10^{-23}\, J/K$
L	Any length (m)
M	Mole weight (g/mol)
M'	Mole weight M' = M/1000, (kg/mol): Should not be used
m_m	Molecular weight (kg)
Q	Heat flux (W/m^2)
N_i	Total mole flux of component *i*, diffusion + convection (mol/m^2s)
N_A	Avagardo constant (=6.022139×10^{23} Molecules/Mol)
n	Number of mol (mol)
T	Temperature (K)
T^*	$= T/(\varepsilon/k)$, dimensional temperature for calculation of collision integral
v	Velocity, m/s
V	Volume (m^3/mol)
V'	Volume (cm^3/mol) $V' = V * 10^{-6}$
x_i	Mol fraction of component *i*
dz	Infinitesimal distance
Z	Compressibility factor, PV=ZRT no dimension

Greek

λ	Thermal conductivity W/mK
Ω_v	Collision integral, effective collision diameter become$\Omega_v \times \sigma$, no dimensions
τ	Shear force, N/m^2
η	Viscosity, Ns/m^2
σ	Molecule diameter, Å (Ångstrøm)
ρ'	Density, kg/m^3
ρ	Density mol/m^3 ($\rho' = \rho M/1000$)
ε	Minimum potential energy between two molecules (J). Uses usually ε/k.
$\varepsilon_{i,j}$	Binary parameter in Mason and Saxena's formula. See section 0
μ_i	Chemical potential,

$$\mu_i = \frac{\partial G}{\partial n_i} \;\; at\ constant\ T, P, n_{j \neq i}$$

κ	Special parameter for polar components in Chung's model
ω	Acentric factor (No dimension)
μ	Dipole momentum (Debye)
μ_r	Non dimensional dipole momentum

$$\mu_r = 131.3 \frac{\mu}{(V_c' T_c)^{1/2}}$$

Subscript

b Boiling at 1 atm.
c Critical state T_c = critical temperature
c Corrected value in Eq(31)
r Reduced size $T_r = T / T_c$: No dimension
max Maximum value
min Minimum value

Superscript

ABRIVATIONS

AAD: Average absolute deviation.

$$AAD = \frac{1}{n}\sum_{i=1}^{n}\frac{\mathrm{abs}\left(y_{measure,i} - y_{calc,i}\right)}{y_{measure,i}}$$

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