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PREDICTION OF THE THERMAL CONDUCTIVITY OF PURE REFRIGERANTS THROUGH AN EXTENDED CORRESPONDING STATES MODEL

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ABSTRACT

An accurate evaluation of the thermophysical properties of refrigerants over wide ranges of pressure and temperature is necessary to predict the performance of possible alternatives to fully halogenated chlorofluorocarbons in refrigeration and heat pumping applications. The transport properties have, in fact, a very important part in the refrigeration plant heat exchangers design.

An Extended Corresponding States (ECS) model for the prediction of the thermodynamic properties of pure and mixed hydrocarbons was presented and more recently an implementation of ECS was proposed for the prediction of the transport properties of halocarbon refrigerants, integrating in it the former ECS thermodynamic model. In the meantime the data availability of both thermodynamic and transport properties of the alternative fluorocarbons has greatly increased.

A parallel validation of the two integrated model is here developed for five HCF fluids. Considering the predictive nature of the model and the scattering of the experimental data, the mean accuracy is good and satisfactory for the technical application requirements.

1. INTRODUCTION

The performance evaluation of the possible alternatives to fully halogenated chlorofluorocarbons in refrigeration and heat pumping applications requires an accurate prediction of the thermophysical properties of refrigerants over wide ranges of pressure and temperature. The transport properties have a very important role, for instance, in the study of the thermal behaviour of the heat exchangers of a refrigeration plant, and from the heat exchangers behaviour depends to a great extent the overall thermodynamic efficiency of the plant itself.

The utilization of the new HFC refrigerant fluids has induced a growing demand for reliable thermal conductivity and viscosity values and in the last few years a world-wide effort was undertaken for their experimental evaluation. For the new HFC fluids and their mixtures reliable thermal conductivity data are not yet sufficiently available and furthermore it has been known that the reported data are considerably scattered due also to their electric conductivity. Few experimental data for mixtures of new refrigerants are at the moment found in literature.

Because of the large interest in utilising predictive models for transport properties calculation, a validation is needed for the prediction accuracy of the proposed models and, on the other hand, an intercomparison of the experimental data from different sources is also necessary

Few years ago an Extended Corresponding States (ECS) model has been developed for the prediction of the thermodynamic properties of pure and mixed hydrocarbons and more recently an implementation of ECS was proposed for the prediction of the transport properties of halocarbon refrigerants. In this second model the ECS thermodynamic model is integrated.

The aim of this work is to test the accuracy of the ECS model in predicting the thermal conductivity of pure refrigerants of hydrofluorocarbon type and in particular of the five more often utilised HFC's. Utilising a large base of experimental data the prediction accuracy of both thermal conductivity and density for pure fluids will be verified over the pT surface, i.e. in the subcooled and saturated liquid and in the saturated and superheated vapor zones.

2. THE ECS MODEL

The thermal conductivity model utilised in this work is based on an ECS model which was originally developed in the early 1970's by Leland and co-workers [1,2] for hydrocarbons and was extended to refrigerants and their mixtures in the early 1990's by Ely and Hanley [5,6]. The ECS model is based on the idea that, with the introduction of appropriate shape factors functions of temperature and density, the properties of any fluid of a class can be derived from those of a reference fluid of the same class.

The ECS model is briefly explained with its extension to the prediction of thermal conductivity of pure refrigerants.

2.1 Thermodynamic Properties of Pure Fluids

The ECS model is an extension of the first approach of using scale factors of temperature and density to evaluate thermodynamic properties of simple spherically pure fluids. According to this approach, if two fluids of this class obey to the corresponding state principle, it is possible to lead to the conclusion developed by Leland et al.[1,2]:

$$a_j^r(\rho_j, T_j) = a_o^r(\rho_o, T_o) = a_o^r\left(\rho_j h_j, \frac{T_j}{f_j}\right) \quad (1) \quad a^r = [A(\rho, T) - A^*(\rho, T)] / RT \quad (2)$$

here a^r is the residual Helmholtz free energy referred to an ideal gas value A^* .

In eq. (2) A is the Helmholtz free energy, [J/mol]; ρ is the molar density, [mol/m³]; T is the absolute temperature [T]; R is the universal gas constant [J/mol K].

In equation (1) the subscript “ j ” indicates the fluid of interest and “ o ” a reference fluid whose thermodynamic properties are known with great accuracy. The reference fluid of this model is the refrigerant R134a, which equation of state, a modified 32-term BWR equation (MBWR), is assumed from [4]. The parameters h_j and f_j are called “*scale factors*” and are related to the critical parameters of the two fluids as follows, eq. (3):

$$h_j = \left(\frac{\rho_o^c}{\rho_j^c}\right); \quad f_j = \frac{T_j^c}{T_o^c} \quad (3) \quad h_j = \left(\frac{\rho_o^c}{\rho_j^c}\right)\varphi_j(\rho, T); \quad f_j = \frac{T_j^c}{T_o^c}\theta_j(\rho, T) \quad (4)$$

where the superscript “ c ” denotes the critical point value.

This model, developed for spherical and symmetric molecules, was extended to refrigerant fluids [3,4], which are generally nonspherical and polar. To overcome this problem, the use of molecular parameters θ and φ called *shape factors* was proposed. Introducing these parameters, the *scale factors* assume the new expressions of former eq. (4).

It is now necessary to find a definition for the parametric functions θ_j and φ_j , which are temperature and density dependent. Nevertheless, the basic thermodynamic relationship expressed by eq.(1) remains the same, but the new scale factors, eq. (4), are introduced.

All the thermodynamic properties, other than A , can be derived differentiating the fundamental equation (1) in accordance to fundamental thermodynamic relationships. For sake of brevity the analytical procedures are here omitted and reference is made to the original works [3,4].

To solve the equations so obtained for the thermodynamic properties the knowledge of component’s shape factors is required. To obtain these parameters, a broad experimental data base has to be used to develop high accuracy correlations. The fundamental problem of the ECS model is to find the analytic expression for each of the parametric functions $\varphi(\rho, T)$ and $\theta(\rho, T)$ in the eqs. (4).

Because both $\varphi(\rho, T)$ and $\theta(\rho, T)$ are functions of temperature and density they have to be regressed from experimental volumetric data presented in the same two variables. The numerical solution of this problem is in reality to much cumbersome to be obtained. The Authors of this model [4] alternatively suggest a simplified procedure: the input experimental data are limited to vapour-liquid boundary curve of the pure, so that the numerical problem reduces from two to one dimension, i.e. to the T variable.

These parameters are determined by mapping the saturation boundaries and are expressed through generalised correlations requiring the knowledge of the Pitzer acentric factor and of the fluid critical constants.

The functional forms of these correlations are:

$$\theta_j = 1 + (\omega_i - \omega_o)(\alpha_1 + \alpha_2 \ln T_{ri}) \quad (5) \quad \varphi_j = \frac{Z_o^c}{Z_i^c} (1 + (\omega_i - \omega_o)(\beta_1 + \beta_2 \ln T_{ri})) \quad (6)$$

where the subscripts “ o ” and “ i ” are referred to the reference and interest fluids respectively, T_{ri} is the reduced temperature of component and the coefficients $(\alpha_1, \alpha_2, \beta_1, \beta_2)$ are obtained by the Authors for a large number of pure refrigerant fluids [4].

2.2. Transport Properties of Pure Fluids

Ely et al [6] and Huber et al. [7] developed an extended corresponding states model for the prediction of thermal conductivity for pure refrigerants and their mixtures based on the same approach of the ECS model for thermodynamic properties. This model, whose mathematical correlations are summarised in the next section, is referred as TRAPP model.

2.2.1 - Thermal conductivity of pure fluids.

This model represents the thermal conductivity of pure fluid as the sum of two parts [7]:

$$\lambda(\rho, T) = \lambda'(\rho, T) + \lambda''(T) \quad (7)$$

where λ' represents the translation contribution due to collision effects, and λ'' represents the transfer of energy from rotational contributions.

The translation term λ' is the sum of other two terms:

$$\lambda' = \lambda^* + \tilde{\lambda} \quad (8)$$

where λ^* expresses a low-density contribution that can be calculated from standard kinetic gas theory [9] and $\tilde{\lambda}$ is the density-dependent translation contribution which can be determined using a corresponding states argument:

$$\tilde{\lambda} = \tilde{\lambda}_o(\rho_o, T_o) F_{\lambda} \quad \text{where:} \quad F_{\lambda} = \left[\frac{M_o}{M} \right]^{1/2} f^{1/2} \cdot h^{-2/3} \quad (9)$$

Here f and h are the "scale factors", described in the earlier section 2.1, while $\tilde{\lambda}_o$ is the high-density translation contribution of the reference fluid R134a, which is determined as the sum of two terms:

$$\tilde{\lambda}_o = \lambda_{ex} + \lambda_{cr} \quad (10)$$

where λ_{ex} is the excess thermal conductivity and λ_{cr} is the critical enhancement for thermal conductivity.

These terms are determined using the following equations, in which k_b is the Boltzman constant and ξ is the correlation length:

$$\lambda_{ex}(T, \rho) = \delta \left[a_1 + a_2 \delta^2 + \delta^3 (a_3 + a_4 \tau) + \delta^4 (a_5 + a_6 \tau) \right] \quad \text{with: } \delta = \frac{\rho}{\rho_c} \text{ and } \tau = \frac{T_c}{T} \quad (11)$$

$$\lambda_{cr} = \frac{R k_b T \rho C_p}{6 \pi \eta \xi} (\Omega - \Omega_o) \quad (12)$$

Reference is made to the original Authors [10].

The value of fluid density $\rho(T, P)$ is here calculated from the thermodynamic ECS model presented in the former section 2.1; this makes the transport property model strongly related to the thermodynamic one. Apart from the present development one can import the $\rho(T, P)$ function from an independent model or, moreover, from experimental data correlations.

The rotation contribution λ'' can be calculated using the modified Eucken correlation for polyatomic gases [9]:

$$\lambda'' = \frac{f_{int} \eta^*}{M} \left[c_p^o(T) - \frac{5}{2} R \right] \quad (13)$$

where η^* is the dilute gas viscosity of fluid, M is the molecular mass, c_p^o is the constant pressure heat capacity of the fluid of interest considered as ideal gas and f_{int} is a constant term equal to $1.32 \cdot 10^4$.

The whole thermodynamic and transport properties ECS model here summarised has been implemented in a computer program, referred as REFPROP, Version 4.01 [43], by the National Institute of Science and Technology (USA).

3. MODEL VALIDATION

Because the ECS model is composed of two main sections, the thermodynamic and the transport properties one, a separate validation has been developed for each of the two.

3.1. Fluid Density

In the following Figures from 1 to 5 the comparisons of experimental and calculated densities for each of the five HFC fluids are reported. The relative deviations are presented as a function of temperature and the experimental values are in general relative to both compressed and saturated liquid and saturated and superheated vapor.

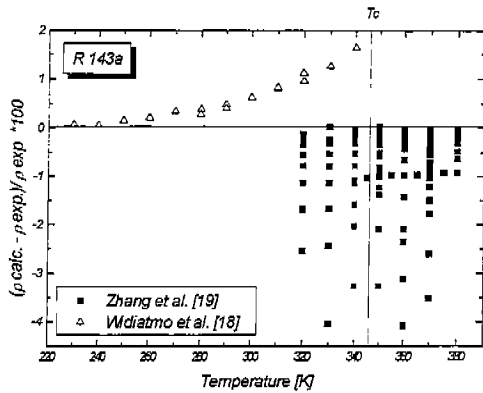


Fig. 1 - Comparison of experimental and predicted density for R143a for selected PVT data sets.

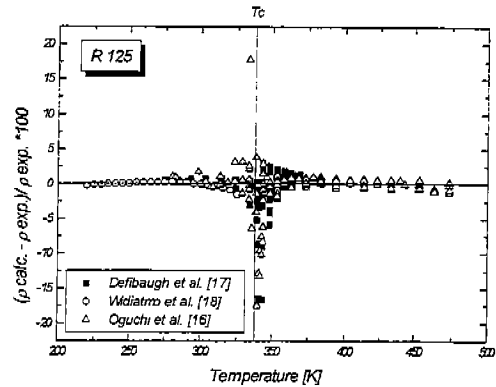


Fig. 2 - Comparison of experimental and predicted density for R125 for selected PVT data sets.

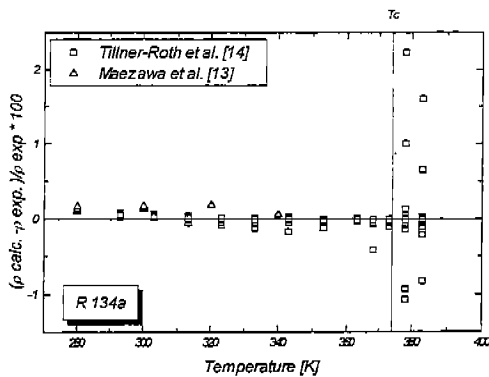


Fig. 3 - Comparison of experimental and predicted density for R134a for selected PVT data sets.

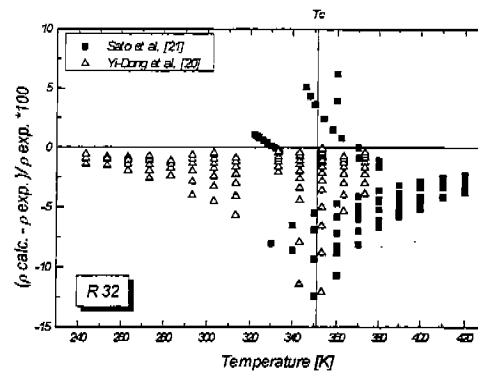


Fig. 4 - Comparison of experimental and predicted density for R32 for selected PVT data sets.

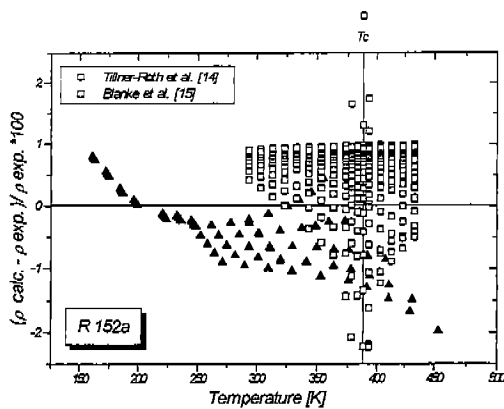


Fig. 5 - Comparison of experimental and predicted density for R152a for selected PVT data sets.

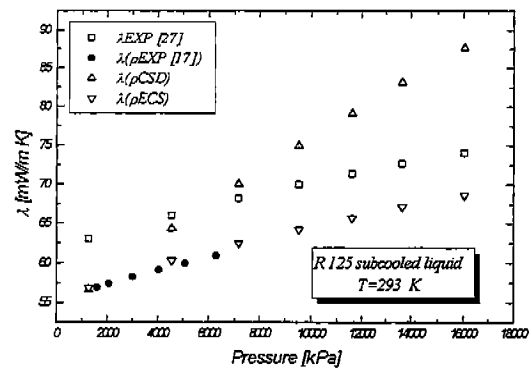


Fig. 6 - Thermal conductivity of R125, experimental and calculated calculated from experimental ρ data and from different ρ models (ECS, CSD).

3.1. Fluid Thermal Conductivity

The Figures from 7 to 11 report the relative deviations of thermal conductivity selected experimental data sets vs. the ECS predicted values as function of temperature for the five HFC fluids. In the following Tables from I to V the thermal conductivity experimental data sets, their references, ranges of pressure and temperature together with the physical state are reported. The error relative to the ECS model in terms of percent Average Absolute Deviation (AAD), BIAS, absolute

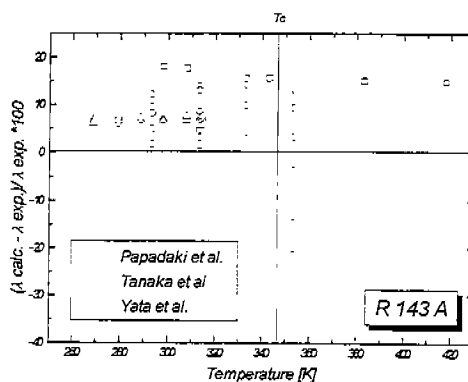
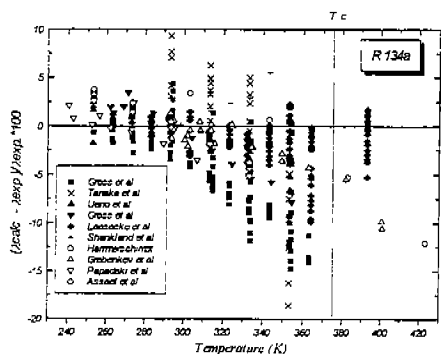


Fig. 7 - Relative deviations of thermal conductivity selected experimental data sets vs. ECS predicted values for R134a.

Fig. 8 - Relative deviations of thermal conductivity selected experimental data sets vs. ECS predicted values for R143a.

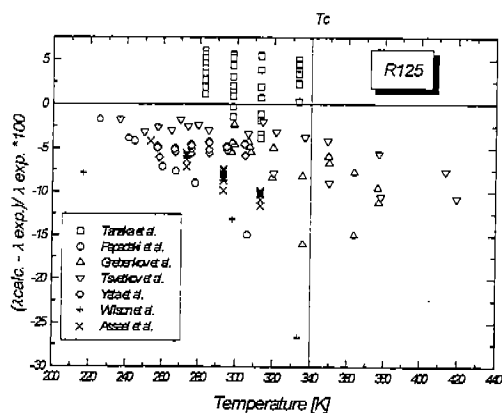


Fig. 9 - Relative deviations of thermal conductivity selected experimental data sets vs. ECS predicted values for R125.

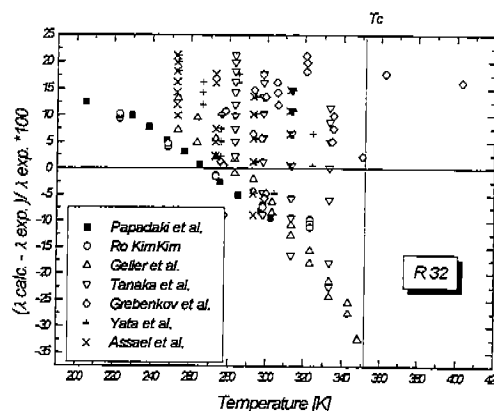


Fig. 10 - Relative deviations of thermal conductivity selected experimental data sets vs. ECS predicted values for R32.

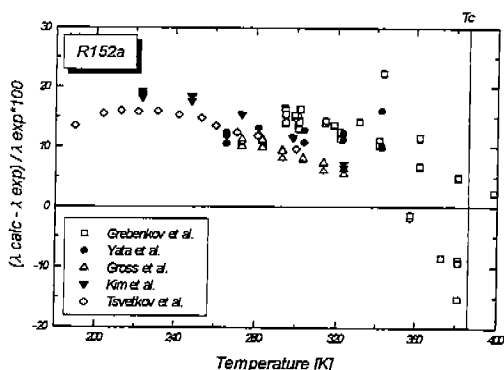


Fig. 11 - Relative deviations of thermal conductivity selected experimental data sets vs. ECS predicted values for R152a.

Table I - Thermal conductivity references for R 134a and comparison with ECS model.

Ref.	(First) Author	P (MPa)	T (K)	Phase	AAD%	BIAS%	MAX%	N
[34]	Assael	0.6 - 22.4	253 - 333	l	1.18413	0.03915	5.11	42
[23]	Grebenkov	0.7 - 19.7	293 - 400	l	2.13543	-1.92506	10.55	25
[39]	Gross	0.1 - 0.5	253 - 363	ls,v	5.19138	-1.03229	7.83	13
[33]	Gross	0.9 - 60.9	261 - 293	l,ls,v,vs	5.33836	-4.83039	14.75	88
[36]	Hammerschmidt	0.1	303 - 463	v	2.15654	-3.34652	19.16	4
[38]	Laesecke	0.1 - 68.2	263 - 393	l,v	1.78869	-1.27806	9.84	162
[35]	Papadaki		240 - 306	ls	2.18398	-0.27453	3.53	9
[37]	Shankland		298 - 343	vs	3.07714	2.15653	5.52	6
[31]	Ueno	1.0 - 30.0	193 - 353	l	5.05287	-1.4613	6.65	23
[32]	Tanaka	0.1 - 2.5	293 - 353	v	2.19587	-0.3399	18.60	33

Table II - Thermal conductivity references for R125 and comparison with ECS model.

Ref.	(First) Author	P (MPa)	T (K)	Phase	AAD%	BIAS%	MAX%	N
[24]	Assael	0.6 - 20	273 - 313	l	8.04979	-8.04979	11.55	20
[23]	Grebenkov	0.7 - 19.7	293 - 400	l	7.10001	-7.10001	16.05	19
[25]	Papadaki		205 - 303	ls	6.91825	-6.91825	14.9	7
[27]	Tanaka	0.1 - 1.0	282 - 333	v	3.07476	2.3626	5.99	34
[29]	Tsvetkov	0.1 - 1.6	236 - 419	v	4.62288	-4.62288	10.66	19
[30]	Wilson	0.00625	216 - 333	ls,v	15.52656	-15.5266	26.63	4
[28]	Yata	1.1 - 30	257 - 304	l	4.98628	-4.98628	6.05	22

Table III - Thermal conductivity references for R32 and comparison with ECS model.

Ref.	(First) Author	P (MPa)	T (K)	Phase	AAD%	BIAS%	MAX%	N
[24]	Assael	0.6 - 20	273 - 313	l	4.76091	2.04855	8.72	24
[22]	Geller		253 - 348	ls,vs	13.04625	-9.04045	32.27	22
[23]	Grebenkov	0.7 - 19.7	293 - 400	l	5.07733	5.33853	18.59	24
[25]	Papadaki		205 - 303	ls	6.42843	1.39979	12.39	10
[26]	Ro	2.0 - 20	223 - 323	l	6.39926	-0.59206	11.15	24
[27]	Tanaka	0.1 - 1.0	282 - 333	v	10.87434	2.84431	22.43	33
[28]	Yata	1.1 - 30	257 - 304	l	3.77736	-4.54305	7.49	23

Table IV - Thermal conductivity references for R143a and comparison with ECS model.

Ref.	(First) Author	P (MPa)	T (K)	Phase	AAD%	BIAS%	MAX%	N
[25]	Papadaki		205 - 303	ls	16.28147	16.28147	17.97	5
[32]	Tanaka	0.1 - 2.5	293 - 353	v	11.03276	1.17084	33.18	29
[28]	Yata	1.1 - 30	257 - 304	l	6.87999	6.87999	7.69	23

Table V - Thermal conductivity references for R152a and comparison with ECS model.

Ref.	(First) Author	P (MPa)	T (K)	Phase	AAD%	BIAS%	MAX%	N
[23]	Grebenkov	0.7 - 19.7	293 - 400	l	11.80848	9.82049	22.33	45
[33]	Gross	0.9 - 60.9	261 - 293	l,ls,v,vs	8.76461	8.76461	11.37	21
[41]	Kim	2.0 - 20	223 - 323	l	14.46784	14.46784	19.31	16
[42]	Tsvetkov	0,2 - 8	160 - 300	l	14.08792	14.08792	16.06	11
[40]	Yata	1.0 - 30	250 - 350	l	11.74541	11.74541	16.07	16

Phase: l=compressed liquid, v=superheated vapor, ls, vs=saturated liquid, vapor; N=number of points.

maximum error and the number of data of the set are furthermore reported. A total of 856 points was considered for the present study with a great difference in the total number of available data for each fluid, as the tables show.

4. DISCUSSION

4.1. Fluid Density

Considering the Figures from 1 to 5 one can see that the thermodynamic ECS model presents a volumetric prediction accuracy rather different from fluid to fluid, but with a general trend of an error increase in the near critical region. This is due to the behaviour of the MBWR32 EoS assumed for the reference fluid R134a, which is well known to loose accuracy in that zone, as any BWR EoS does. From the Figures presented the better results are for R134a, but this is misleading because this is the reference fluid for which the model has not to modulate the density coming from the MBWR32 EoS through the *shape factors*. Moreover the experimental density data utilised for the five fluids are mostly in the compressed liquid and superheated vapor zones, often far in conditions from the saturation boundaries along which the Authors have regressed the θ and ϕ functions, eqs. (5, 6). Apart from more specific considerations, subdividing for instance the P ρ T surface and considering also the polarity of the fluids, the results look to be acceptably good for the purpose, provided the residual error on density do not affect too much the second part of the model calculating the transport properties.

To test this aspect experimental density and thermal conductivity data at same conditions have to be available. In Fig. 6 for R125 in subcooled liquid state at 293K a comparison has been done between λ experimental and predicted values; the predicted values were calculated utilising density experimental data or density values generated from the present ECS model or from a Carnahan-Starling-De Santis (CSD) EoS, also reported in [43]. It was furthermore verified that CSD EoS presents for R125 in the compressed liquid zone a low prediction accuracy for density increasing with pressure and reaching, for the pressure range reported in the Figure, deviations up to 10% and more. In the same conditions the density deviation of ECS is considerably lower, as shown in Fig. 2. The λ values predicted importing the density from experimental data and from the ECS thermodynamic model are quite similar, while the λ deviations predicted importing the density from the CSD EoS are very low, and similar to the former two, near saturation (1205 kPa), but increase rapidly with pressure, in accordance with the CSD EoS error in predicting density. This confirms that the ECS λ model is sensitive to the density prediction accuracy and that an accurate thermodynamic model is required, as the ECS one is.

4.2. Fluid Thermal Conductivity

It is evident from both Figures 7÷11 and Tables I÷V the scattering of the available data; a first reading of the results can show an intercomparison of the experimental data from different sources trough the present transport properties model. In general it is not possible to state that the model systematically over- or under-predicts the conductivity, because from the relative deviations reported in the figures and, with a better precision, from the bias values reported in the tables deviations of different sign are more or less equally present trough the whole population of points.

Looking to the single fluids only for R152a an evident over-estimation is present, while for R143a the same is true only for two over three data sets. They also present the higher values of AAD with respect to the others. It is possible that these results have to be related to the high polarity of these two fluids (2.0÷2.3 debyes), although all the HFC are strongly polar. From the same point of view the more equilibrate results are obtained for R134a, which is the reference fluid of the model.

As a general trend an increase of the error toward the critical region is verified, also due to the imported error in density as discussed in the former section 4.1. For refrigeration applications the near critical region is anyway without interest. Apart from R134a it is evident that there are several great discrepancies among the data sets with furthermore excessive values of the maximum absolute errors. Assuming the AAD as the evaluation parameter and not considering the two fluids R143a and R152a, the model looks to represent sufficiently well the conductivity, apart some few more deviating data sets. But from a more general point of view with the present analysis it seems difficult to get valuable conclusions for the ECS model prediction accuracy of conductivity, mainly because of the excessive scattering, discrepancies and contradictory nature of the available data. It is then difficult to share the global error between the model and the data.

If the reported data represent the mean experimental accuracy presently achievable, as it seems evident, the model can be positively considered. Only data selected and consolidated through several sources can allow the refinement of a general model as the present one. Considering particularly the predictive nature of the model and the scattering of the experimental data, the mean prediction accuracy is good and satisfactory for the technical application requirements.

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