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Prediction of Thermal Conductivity of Liquid and Vapor Refrigerants for Pure and their Binary, Ternary Mixtures using Artificial Neural Network

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Abstract

The determination of thermophysical properties of hydrofluorocarbons (HFCs) is very important, especially the thermal conductivity. The present work, investigated the potential of an artificial neural network (ANN) model to correlate the thermal conductivity of (HFCs) at (169.87-533.02) K, (0.047-68.201) MPa and (0.0089- 0.1984) W.m.⁻¹K⁻¹ temperature, pressure and thermal conductivity ranges respectively, of 11 systems from 3 different categories including five pure systems (R32, R125, R134a, R152a, R143a), four binary mixtures systems (R32+R125, R32+R134a, R125+ R134a, R125+R143a), and two ternary mixtures systems (R32+R125+ R134a, R125+ R134a+ R143a). Each one received 1817,794 and 616 data points, respectively. The application of this model for these 3227 data points of liquid and vapor at several temperatures and pressures allowed to train, validate and test the model. This

study showed that ANN models represent a good alternative to estimate the thermal conductivity of different refrigerant systems with a good accuracy. The squared correlation coefficients of thermal conductivity predicted by ANN were $R^2 = 0.998$ with an acceptable level of accuracy of RMSE = 0.0035 and AAD= 0.002 %. The results of applying the trained neural network model to the test data indicate that the method has a highly significant prediction capability.

Key words: Refrigerant, Pure system, Mixtures, Thermal conductivity, Artificial Neural Network, Predictive model.

Introduction

In recent years, It has been highlighted the need to establish a balance between "energy consumption" and "environmental protection". The introduction of new refrigerants to replace Chlorofluorocarbons (CFCs) and Hydrochlorofluorocarbons (HCFCs) and the adaptation of new techniques should allow a reduction of the overall environmental impacts. To reduce security risks, another line of research has been developed towards the use of a secondary cooling loop (indirect cooling). The multiple processes used in industry are often based on heat exchanges.

Refrigerants cause global warming and ozone depletion bring on the environmental problems. So, UN drew up the Montreal Protocol (MP) and its London and Copenhagen Amendments [1]. Prior to the 1980s, the principal classes of chemicals used as refrigerants in the refrigeration industry were CFCs and HCFCs. Based on growing evidence of ozone depleting potential (ODP) [2] of CFCs based on the Montreal Protocol, different industries turned to HCFCs as substitutes for CFCs. While HCFCs have a lower ODP than CFCs, they still damage the ozone layer and have become topic to scheduled period out by 2030,

Hydrofluorocarbons (HFCs) were used as an acceptable alternative to CFCs and HCFCs because they possess several characteristics, including near-zero ODP and low Global warming potential (GWP), similarity to CFCs and HCFCs in physical properties, short atmospheric lifetimes, less or non-flammable and not expensive [3].

During the previous periods, owing to the understanding of the impact of refrigerants in the destruction of the ozone layer, the concepts of ozone depletion potentials (ODPs)[4] was designed to determine the relative capacity of a chemical to destroy ozone, [5,6,7,8]. Consequently, GWP is the purpose of several studies leading to the publication of a wide range of literature data [9].

Many researchers focused their efforts on the measurement of the thermophysical properties of these compounds (Refrigerants), aiming to find proper substitutes. Thermal conductivity is one of the major thermophysical properties, as defined by Fourier's equation, it is the ability of a material to transmit heat by means of conduction. Thermal conductivity, K ($\text{W. m}^{-1}\text{K}^{-1}$), is essential to the knowledge of heat transfer, in particular for refrigerants. In fact, the higher the liquid and vapor thermal conductivity, the higher the heat transfer coefficient.

The context of this work is related to the problem of the substitution of CFC and HCFCs. Experimental aspect (technical and measurements) and modeling aspects of the data obtained were first considered. The products studied are mixtures (pure, binary and ternary) compounds of R32, R125, R134a, R152a and R143a, which are HFCs of high interest to the industries of air conditioning and refrigeration.

Different authors have used diverse equations and methods to predict and reproduce the thermodynamic properties of refrigerant systems. Most of these attempts have been restricted to limited systems and to the best of the knowledge of no systematic work devoted to test the ability of these methods to predict the thermodynamic properties of different categories of systems is available. A number of theories are available providing equations to predict the

thermal conductivity of liquids, these equations include theoretical calculations which consider intermolecular distances [10]; there are equations based on the theory of group contribution [11] or molecular descriptors [12,13]; equations involving some fixed parameters in order to describe the thermal conductivity [12,14]. An empirical approach based on a limited number of fixed physical parameters [15] was considered; a new equation to describe the thermal conductivity of R-32, R-125, R134a, and R-143a for practical use, applicable over a wide range of temperature and pressure has also been proposed [16]. Semi-empirical approaches for correlating thermal conductivity data for multicomponent liquid mixtures [17,18] were studied, a method to estimate the thermal conductivity and the viscosity similar to cubic equations of state of halogenated hydrocarbon of pure substances in vapor and liquid regions [19,20] was proposed; thermal conductivity modeling of refrigerant mixtures in a three-parameter corresponding states format [21] was carried out. There are an extensive studies have been published in the literature on the thermal conductivity of mixed refrigerants [22,23,24] which suggested a correlation to calculate the thermal conductivity and viscosity of some alternative refrigerant mixtures such as R-507, R-404A, R-407C, and R-410A.

Recently, some researchers used artificial neural networks (ANNs) to correlate thermal conductivity property [25]:and obtained much better results than the traditional approach. To minimize the difficulties of experimental measurements and also their time-consuming and costly nature, it is desirable to develop predictive methods for estimating the phase behavior of these kinds of systems.

The purpose of this study was therefore the application of artificial neural network for representing/predicting the liquid and vapor thermal conductivity of pure refrigerants including R32, R125, R134a, R152a, R143a and their binary, ternary mixtures at different temperatures and pressures.

Methodology and modeling

Neural network

Artificial neural networks (ANNs) consist of a number of neurons working in unity to solve various scientific and engineering problems such as estimation of physical and chemical properties [26]. ANN can operate like a black box model, which requires no detailed information about the system or equipment. The ANN can learn the relationship between input and output based on the training data.

ANNs have been used in many engineering applications such as control systems, in classification and in modeling complex process transformations. Detailed information about artificial neural networks can be found in the following References [27,28]. ANNs is a widely used numerical method which is able to model any kind of data set even complex ones.

Selection of network parameters

The artificial neurons are arranged in layers where in the input layer receive inputs (u_i), namely experimental data and each succeeding layer receives weighted outputs (w_{ij} , u_i) from the preceding layer as its input resulting therefore a feedforward ANN, in which each input is feed forward to its succeeding layer where it is treated. The outputs of the last layer constitute the outputs to the real world [29]. In such a feed forward ANN a neuron in a hidden or an output layer has two tasks:

(a) It sums the weighted inputs from several connections plus a bias value and then applies a transfer function to the sum as given by (for neuron j of the hidden layer):

$$z_j = f_h\left(\sum_{i=1}^n w_{ji}^I u_i + b_{hj}\right); j = 1, 2, \dots, m \quad (1)$$

(b) It propagates the resulting value through outgoing connections to the neurons of the succeeding layer where it undergoes the same process as given by (for instance outputs Z_j of the hidden layer fed to neuron k of the output layer gives the output V_k):

$$V_k = f_o\left(\sum_{j=1}^m w_{kj}^h z_j + b_{ok}\right); k = 1, 2, \dots, i \quad (2)$$

Combining equations 1 and 2 the relation between the output V_k and the inputs u_i of the NN is obtained:

$$V_k = f_0\left(\sum_{j=1}^m w_{kj}^h f_h\left(\sum_{i=1}^n w_{ji}^l u_i + b_{hj}\right) + b_{ok}\right); k=1, 2 \dots, i \quad (3)$$

The output is computed by means of a transfer function, also called activation function. It is desirable that the activation function has a sort of step behavior. Furthermore, because continuity and derivability at all points are required features of the current optimization algorithms[30,31,29] three kinds of transfer function are considered in this study:

Hyperbolic tangent sigmoid transfer function:

$$f(a) = \frac{e^a - e^{-a}}{e^a + e^{-a}} \quad (4)$$

Logarithmic sigmoid transfer function:

$$f(a) = \frac{1}{1+e^{-a}} \quad (5)$$

Pure linear transfer function:

$$f(a) = a \quad (6)$$

Training of network

A Feed forwardback propagation (FFBP) which is very effective in representing nonlinear relationships among variables was used in this work BP algorithm (BPA) is the most widely used in ANN, and has different variants. FFBP with Levenberg-Marquardt (BP) training algorithm and one hidden layer was chosen because it is suitable for modeling the relationship between input data and output variable[32].

Testing of network

The criteria used for measuring the performance of the network are the absolute fraction of variation (R^2), and the root mean square error (RMSE), which can be calculated using the following equations.

The fraction of absolute variance is given by:

$$R^2 = 1 - \frac{\sum_{i=1}^N (K_{cal,i} - K_{exp,i})^2}{\sum_{i=1}^N (K_{exp,i})^2} \quad (7)$$

The root mean square error value is calculated by

$$RMSE = \sqrt{\sum_{i=1}^N (K_{cal,i} - K_{exp,i})^2 / N} \quad (8)$$

Here, N is the number of data patterns in the independent data set, $K_{cal,i}$ indicates the values predicted by ANN, $K_{exp,i}$ is the measured value of one data point i [33,34].

The accuracy of the model to reproduce and predict the thermal conductivity of refrigerant systems at different temperatures and pressures may be evaluated using the statistical parameters [35,36]: namely, the absolute average deviation (AAD), and the mean square error (MSE) which are defined as follows:

The mean square error value is calculated according to:

$$MSE = \frac{1}{N} \sum_{i=1}^N (K_{exp,i} - K_{cal,i})^2 \quad (9)$$

The absolute average deviation value is calculated by [29]:

$$AAD = \frac{1}{N} \sum_{i=1}^N |K_{exp,i} - K_{cal,i}| \quad (10)$$

The Average Absolute Relative Deviation:

$$AARD\% = \frac{100}{N} \sum_{i=1}^N \left| \frac{K_{exp,i} - K_{cal,i}}{K_{exp,i}} \right| \quad (11)$$

Optimization of the network architecture

Optimization of the network architecture is the major task in ANN. The parameters that affect the performance of the network are the number of neurons in the hidden layer, the number of hidden layers, the transfer function and the training algorithm. The network architecture can be optimized by varying the above parameters (using trial and error method) to achieve the results with good accuracy [33,34].

Results and Discussion

In the present study, a total of 3227 data points of thermal conductivity data were considered. The collected experimental data points were divided into three different sub- datasets five pure systems, (R32, R125, R134a, R152a, R143a), four binary mixtures systems(R32+R125, R32+R134a, R125+ R134a, R125+R143a), and two ternary mixtures systems (R32+R125+ R134a, R125+ R134a+ R143a) at several temperatures and pressures. Each one received 1817, 794, 616 data points respectively.

For developing the neural network model, inputs and outputs should be first defined. The input parameters included, Temperature (T (k)) and pressure (P (MPa)), namely two macroscopic inputs which characterize the physical conditions of the system. Molecular weight (M (g. mol⁻¹)), critical temperature (T_c (k)), critical pressure (P_c (MPa)), critical density (D_c (Kg. m⁻³)), and mass fraction of liquids or vapor phase (X) were the other selected inputs for pure systems they were specified as pseudo critical mixtures property for binary and ternary systems(9 inputs), and Thermal conductivity was the network output (1 output). The network is simple in structure and easily analyzed mathematically is schematically illustrated in Fig. 1 and Table 1 shows the physical properties values of these compounds accompanied with related references. This study employs experimental data from the open literature for each (pure, binary, ternary) systems, the details listed in Tables 2 to 4 respectively.

The available correlations for prediction can be described as follows:

$$K = f(T, P, M, T_c, P_c, D_c, X_i, X_j, X_k) \quad (12)$$

Calculation of the pseudo critical mixtures properties

The different equations of state were developed from the knowledge of the properties P , V , T of pure substances. In the case of mixtures of known composition, it is necessary to make use of mixing rules for calculating the average properties of the mixture. Pseudo properties in the case of critical mixtures, are generally obtained from the rule of Kays (1936):

$$T_{PC} = \sum_{i=0}^n X_i * T_{Ci} \quad (13)$$

$$P_{PC} = \sum_{i=0}^n X_i * P_{Ci} \quad (14)$$

$$D_{PC} = \sum_{i=0}^n X_i * D_{Ci} \quad (15)$$

$$M_{PC} = \sum_{i=0}^n X_i * M_{Ci} \quad (16)$$

$$K = f(T, P, M_{PC}, T_{PC}, P_{PC}, D_{PC}, X_i, X_j, X_k) \quad (17)$$

n=1 for pure refrigerant, $X_i = 1, X_j = 0, X_k = 0$, n=1, 2 for binary mixtures, $X_i + X_j = 1, X_k = 0$ and n=1, 2, 3 for ternary mixtures, $X_i + X_j + X_k = 1$.

201 Procedure for neural network modeling of the thermal conductivity

202 ANN modeling is described consists in the following four steps: (a) extract the results from
203 experiments or theoretical calculations (b) train the network using experimentally or
204 theoretically predicted values (c) testing of network with data, which are not used for
205 training, (d) identify the best network architecture based on statistical performance values
206 [33,34].

207 (i) phase of collection of experimental data as complete as possible. From this, the following
208 procedure was considered:

209 (ii) division of data base to three parts (DB of pure systems, BD of binary systems, BD of
210 ternary systems).

211 (iii) phase of pre-treatment and analysis of the data.

212 (iv) dividing data into three subsets-train, validation and test.

213 (v) phase of choice of the parameters of the neural network (neural network (FFBP), neurons
214 number in hidden layer, activation function in hidden layer (tan-sig), activation function in
215 output layer (Tang-sig)).

216 (vi) saving the parameters of the optimal ANN.

Table 5 shows the structure of the optimized ANN model for each pure systems, binary systems, ternary systems and global systems *Tan-sig* is a sigmoid transfer function used in the hidden layer and used in the output layer for all systems, in this table, R² values and RMSE of the trained networks have been shown for different number of hidden layer neurons. The best configuration with minimum error measure (RMSE) and appropriate R² value. The parameters (weight and biases values) of the best selected NN model for each system (pure, binary, ternary, global) are listed in Tables 6 to 9 respectively, where W^1 is the input-hidden layer connection, W^h is the hidden layer output connection, b_{hj} and b_{ok} are biases of the neuron j of the hidden layer and the neuron of the output layer respectively.

Table 10 shows the regression line of the network model equation during training, validation, and prediction sets of pure systems, binary systems, ternary systems and global system RMSE, MSE, AAD, AARD, and SSE, are collected in Table 11 showing that there was a significant correlation between experimental and predicted values. For each plot, the ANN method provide good results with high correlation coefficients (R^2_{exp}, R^2_{cal}). Comparison between the output data and calculated values of the thermal conductivity of different refrigerant systems with ($R^2_{pure}=0.997$, $R^2_{binary}=0.998$, $R^2_{ternary}=0.998$, $R^2_{global}=0.997$), shows slopes close to unit and an intercept values close to zero, confirming the agreement between experimental and predicted values (Fig. 2).

Reduction technique

The reduced data refers to the analysis and transformation of the input and target variables having different physical units with different range values in order to minimize the input parameters (9 inputs to 7 inputs) and to improve the learning quickness, the present study use this method to calculate the minimum and the maximum of each vector variable and scaling the data with respect the upper limits. Finally, the values of the outputs from the neural network reduced are converting before being presented.

242 For data reduce, the following equations are considered:

$$G_r = \frac{G - G_{min}}{G_{max} - G_{min}} \quad (17)$$

$$X_i = X_i + 1 \quad (18)$$

$$T_r = \frac{T}{T_c}, P_r = \frac{P}{P_c} \quad (19)$$

245 For converting (to real value) of data estimated by ANN:

$$G = G_r * (G_{max} - G_{min}) + G_{min} \quad (20)$$

$$K = f(T_r, P_r, M_r, D_r, X_i, X_j, X_k) \quad (21)$$

247 Where G is the original data value, G_r is the corresponding reduced variable; min, max are the
248 minimal and the maximal values of each vector respectively.

249 The validation agreement, coefficients of determination (R^2_{exp}) and (R^2_{cal}), and different
250 errors values RMSE, MSE, AAD, AARD, and SSE, are collected in Table 12. The
251 performance of the present developed ANN (RN2, RN3) models were compared to the
252 previously developed ANN models (RN1) to estimate the thermal conductivity for original
253 (real) data and also the reduced data. The three models provided high values of the correlation
254 coefficient R^2 , as illustrated in Fig. 3 otherwise the RN1 model obtained was similar to RN3;
255 while the RN2 model led to a better fit of experimental data than the RN1, RN3 models.

256 Artificial neural network prediction

257 The assembled of new experimental data points, namely 660 data points, for harmful
258 refrigerants covered thermal conductivity range 0.0106-0.15 W/m. K, temperature range of
259 164.77–463K, and pressure range of 0.059–69.58 MPa. These collected data were distributed
260 into 363 data points for three pure systems (R22, R124, R142b), 240 data points for two
261 binary systems (R22+R142b, R22+R152a) and 57 data points for one ternary system
262 (R142b+R124+R22). In this part the range of experimental data parameters and the number of

data points accompanied with their references in Tables 13, 14 and 15 for different systems (pure, binary, ternary), respectively.

The prediction of the new data base (DB2) of the harmful refrigerants by no harmful refrigerants using (DB1) are given in Fig. 4 this indicates that the new approach (RN3) improved the prediction of thermal conductivity, producing the following RMSE, MSE, AAD, AARD, SSE and R^2 values, 0.0123, 0.0002, 0.0094%, 17.92%, 0.10 and 0.974, respectively (Table 16).

These results show the good predictive ability of the ANN model. In general, in Fig. 5 the residual did not exceed -0.025 to 0.04 for new data base (DB2) and ± 0.02 for data base (DB1), (except for a few experimental points). The illustration of harmful refrigerants for all (pure, binary, ternary) systems by different colors that shows an appropriate R^2 value, 0.95, are shown in Fig. 6 this confirms that the method outperforms prediction ability. Fig. 7 shows the prediction of DB2 by DB1 in the range $[0.01, 0.03] \text{ W.m}^{-1} \cdot \text{K}^{-1}$.

All experimental data points (DB1+DB2), namely the whole 3887 data points, were divided into three different sub-datasets: eight pure systems, (R32, R125, R134a, R152a, R143a, R22, R124, R142b), six binary mixtures (R32+R125, R32+R134a, R125+R134a, R125+R143a, R22+R142b, R22+R152a), and three ternary mixtures (R32+R125+R134a, R125+R134a+R143a, R142b+R124+R22), namely 2180, 1034, 673 data points respectively.

Figs. 8, 9 and Table 17 shows a significant correlation with percentage residual error varying in the range of ± 0.02 , a high R^2 value, 0.998, and fairly accurate prediction the absolute average deviation error (AARD), estimated to be 5.8%, as well as a high robustness.

Conclusion

In this paper, a methodology for choosing an ANN model to predict thermal conductivity was presented. The methodology starts with a wide search in order to select the model with

minimum complexity, optimal performance and choice of the respective parameters (inputs and outputs, activation functions, training algorithm, reduction technique, hidden layers).

An artificial neural network model was used to predict the thermal conductivity of pure refrigerants and their binary, ternary mixtures for liquid and vapor phase of 11 systems from 3 different categories containing five pure systems (R32, R125, R134a, R152a, R143a), four binary mixtures (R32+R125, R32+R134a, R125+ R134a, R125+R143a) and two ternary mixtures (R32+R125+ R134a, R125+ R134a+ R143a). This study involved 3227 data points including the temperature, the pressure, the molecular weight, the critical temperature, the critical pressure, the critical density and the mass fraction of liquid and vapor phases of refrigerants.

In this work, the feed forward Backpropagation (BP) algorithm with Levenberg–Marquardt (LM) training was applied to estimate the thermal conductivity. One relevant model (RN1) we attempted to improve their performance by using the reduction technique to attain (RN3). The best network model obtained consisted of the best fit of the validation agreement plot, and high R^2 value, 0.997 was obtained. Furthermore, the statistical analysis showed that the model (RN3) was able to yield quite satisfactorily estimates for pure, binary and, ternary refrigerant systems, as well as the global system, the model was shown to be robust.

The performance of the proposed ANN model was also examined through its application to test a data set consisting of 660 experimental points of thermal conductivity for various systems for diverse pure, binary and ternary mixtures of refrigerants over a wide range of temperatures and pressure. The results of this evaluation indicate that the developed ANN model was capable to predict thermal conductivity with a high coefficient of determination.

The results of applying the trained neural network model to the test data indicate that the method has a very good prediction capability with respect to not only the temperature and pressure ranges but also the refrigerant types.

313 Lastly the gathering of all experimental data points DB1 and DB2 (RN4) gave a high R^2 value,
314 0.998, and a low average absolute relative deviation error (5.8%).

315 **Nomenclature**

316	AAD	Average absolute deviation
317	AARD	Average absolute relative deviation
318	ANN	Artificial neural network
319	CFCs	Chlorofluorocarbons
320	FFBP	Feed forward back propagation
321	GWP	Global warming potential
322	HCFCs	Hydrochlorofluorocarbons
323	HFCs	Hydrofluorocarbons
324	M	Molecular mass (g.mol^{-1})
325	M_c	Critical molecular mass (g.mol^{-1})
326	MSE	Mean square error
327	N	Number of data points
328	ODP	Ozone depleting potential
329	P	Pressure (MPa)
330	P_c	Critical pressure (MPa)
331	R^2	Coefficient of determination
332	regression equation	
333	RMSE	Root mean square error
334	T	Temperature (K)
335	T_c	Critical temperature (K)
336	W	Weights
337	X	Mass fraction of liquid or vapor phase
338	α	Slope of the linear regression
339	equation	
340	β	y Intercept of the linear regression
341	equation	
342	<i>Subscripts/superscripts</i>	
343	Cal	Calculated
344	exp	experimental
345	max	maximum
346	min	minimum
347	n	component
348	*	harmful refrigerant
349	<i>Greek letters</i>	
350	K	Thermal conductivity ($\text{W.m}^{-1} \cdot \text{K}^{-1}$)
351	D	Density (Kg.m^{-3})
352	D_c	Critical density (Kg.m^{-3})
353	<i>Acronyms</i>	
354	ASHRAE	American society of heating, refrigerating and air conditioning engineers

355 MP Montreal Protocol
356

357

358 **References**

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Table 1
The critical properties of compounds used in this work

Component	Molecular weight (g/mol)	T_c (K)	P_c (Mpa)	D_c (Kg/m ³)	Reference
R32	50.020	351.55	5.83000	430.0	[56]
R125	120.02	339.45	3.59000	571.0	[56]
R134a	102.00	374.18	4.05629	508.0	[49]
R152a	66.050	386.44	4.52000	365.0	[44]
R143a	84.040	346.25	3.81100	434.0	[44]
R22*	86.470	369.35	4.99000	513.0	[56]
R124*	136.48	395.65	3.63400	560.0	[44]
R142b*	100.50	410.25	4.24600	459.0	[44]
R152a*	66.050	386.44	4.52000	365.0	[44]

Table 2
Sources and ranges of data base used in this work for pure systems

Component	T (K)	P(Mpa)	X _i	(W. m. ⁻¹ K ⁻¹)		Reference
R134a	293.15-353.15	0.10-2.540	1	0.0123-0.0256	32	[37]
	272.46-400.47	0.1005-8.943	1	0.1165-0.0948	34	[38]
	202.83-303.05	1.2919-5.177	1	0.08445-0.1272	28	[39]
	253.25-363.15	0.096-6.097	1	0.01205-0.10605	76	[40]
	203.00-393.00	0.09-68.201	1	0.01036-0.14495	207	[41]
	273.15-363.15	0.10-2.8	1	0.01096-0.02036	38	[42]
	252.97-333.20	1.71-22.43	1	0.0707-0.1113	32	[43]
	169.87-290.06	10.00	1	0.0862-0.1430	19	[44]
	303.00-463.00	0.10	1	0.01376-0.02758	05	[45]
	223.15-323.15	2-25	1	0.07448-0.12087	24	[46]
	213.01-292.88	1.00-21.34	1	0.087297-0.13048	51	[47]
	232.75-323.25	2.00-20.00	1	0.0751-0.1184	23	[48]
	295.85-532.94	0.10-50.00	1	0.01276-0.10494	521	[49]
	295.85-354.95	1.379-4.147	1	0.05715-0.08401	102	[50]
R125	172.74-290.02	10.00	1	0.0679-0.1116	16	[44]
	283.15-333.15	0.100-2.010	1	0.01229-0.01777	51	[51]
	253.04-313.46	1.24-16.03	1	0.0578-0.0793	19	[52]
	187.43-413.61	0.18-6.040	1	0.011-0.1075	28	[53]
	254.45-354.35	0.104-7.033	1	0.01212-0.08426	93	[54]
	231.25-324.05	2.00 -20.00	1	0.0524-0.0984	24	[55]
	231.25-324.05	2.00 -20.00	1	0.0524-0.0984	24	[48]
R32	283.15-333.15	0.10 -3.00	1	0.01084-0.02025	51	[51]
	252.67-312.83	3.68-17.63	1	0.1148-0.1704	21	[52]
	223.15-323.15	2.00-20.00	1	0.11035-0.1984	22	[46]
	233.45-334.95	0.098-6.194	1	0.01081-0.17762	55	[54]
	232.55-322.95	2.00-20.00	1	0.1141-0.1927	22	[55]
R152a	263.15-363.15	0.082-6.22	1	0.01215-0.1263	67	[40]
	223.15-323.15	2.1-20.1	1	0.0928-0.145	25	[56]
	303-423	0.1	1	0.01491-0.02592	4	[45]
	189.61-299.33	7.71-8.55	1	0.1076-0.159	11	[57]
	211.69-294.29	0.79-18.5	1	0.10259-0.14752	38	[58]
R143a	293.15-353.15	0.1-3.98	1	0.0126-0.0375	29	[37]
	298-383	0.1	1	0.0123-0.01873	4	[45]
	233.15-323.45	2-20	1	0.0576-0.1121	21	[59]

Table 3

Sources and ranges of data base used in this work for binary systems

Binary Systems	T (K)	P(Mpa)	$X_i+X_j=1$	$K(W. m.^{-1}K^{-1})$	N	Reference
R32+R125	283.15-298.15	0.10-1.20	1876-0.8222	0.0117-0.01502	69	[51]
	232.65-323.95	2-20	0.2522-0.7595	0.0639-0.1667	120	[55]
R410A	213-293	2-30	0.249-0.75	0.0814-0.1785	60	[60]
	255.04-409.8	0.101-3.69	0.5+0.5=1	0.02276-0.00998	50	[61]
R32+R134a	223.15-323.15	2-25	0.3057-0.7496	0.08165-0.17378	72	[46]
	193.2-316.1	2-30	0.249-0.75	0.0846-0.1953	84	[60]
R125+R134a	232.75-323.55	2-20	0.191-0.785	0.0597-0.1143	98	[48]
R507A	254.71-372.17	0.101-2.647	0.5+0.5=1	0.01007-0.02138	34	[61]
	297.95-332.55	1.465-3.775		0.05-0.0637	128	[62]
	312.59-424.24	0.105-1.902		0.01468-0.02528	79	[63]

Table 4

Sources and ranges of data base used in this work for ternary systems

Ternary systems	T (K)	P (Mpa)	$X_j+X_k=1$	$K(W. m.^{-1}K^{-1})$	N	Reference
R32+R125+R134a 407C 407C	193.1-293	2-30	0.19-0.23	0.0797-0.144	44	[64]
	232.55-324.15	2-20	0.18-0.61	0.0641-0.1505	168	[65]
	253.27-389.83	0.101-2.447	0.23+0.25+0.52=1	0.00968-0.02012	38	[61]
	303.9-424.25	0.1055-2.038	0.23+0.25+0.52=1	0.0133-0.02454	97	[66]
404A	252.8-393.09	0.101-2.763	0.44+0.04+0.52=1	0.0099-0.02234	46	[61]
	233.55-322.95	2-20	0.44+0.04+0.52=1	0.0563-0.1056	24	[59]
	311.32-428.94	0.13-1.841	0.44+0.04+0.52=1	0.02584-0.01451	91	[67]
	297.85-332.65	1.277-3.818	0.44+0.04+0.52=1	0.05073-0.06532	108	[70]

N: Number of experimental data; **R410A** (R32, R125: 50%, 50%); **R507A** (R125, R143a: 50%, 50%), **407C** (R32, R125, R134a: 23%, 25%, 52%); **404A** (R125, R134, R143: 44%, 4%, 52%).

Table 5

The values of coefficient of determination, R^2 and RMSE of different transfer function with the optimal number of neurons in each case

Type of Network		Input layer	Hidden layer	Output layer	R^2	RMSE
		Neurons Number	Activation function	Activation function		
Pure systems	FFBP	13	Logsig	Tansig	0.99312	0.0039
	FFBP	16	Logsig	Purelin	0.99411	0.0033
	FFBP	13	Tansig	Tansig	0.99615	0.0027
	FFBP	19	Tansig	Purelin	0.98434	0.0054
	FFBP	25	Purelin	Tansig	0.75703	0.0200

Binary systems	FFBP	16	Logsig	Tansig	0.9971	0.0037
	FFBP	15	Logsig	Purelin	0.9956	0.0045
	FFBP	13	Tansig	Tansig	0.9980	0.0030
	FFBP	19	Tansig	Purelin	0.9911	0.0064
	FFBP	25	Purelin	Tansig	0.9909	0.0065
Ternary systems	FFBP	15	Logsig	Tansig	0.9934	0.0046
	FFBP	16	Logsig	Purelin	0.9928	0.0048
	FFBP	12	Tansig	Tansig	0.9983	0.0023
	FFBP	19	Tansig	Purelin	0.9933	0.0046
	FFBP	25	Purelin	Tansig	0.9080	0.0157
Global system(RN1)	FFBP	09	Tansig	Tansig	0.9925	0.0063
	FFBP	16	Logsig	Tansig	0.9941	0.0057
	FFBP	15	Logsig	Purelin	0.9825	0.0081
	FFBP	13	Tansig	Tansig	0.99667	0.0036
	FFBP	19	Tansig	Purelin	0.99279	0.0052
	FFBP	25	Purelin	Tansig	0.81528	0.0251

Table 6

Weights and biases of the optimal ANN architecture (Pure systems)

Input-Hiddenlayer							Output layer	
Weights(w_j^I)						Bias	Weights (w_j^h)	Bias
T	P	P_{PC}	T_{PC}	P_{PC}	D_{PC}	b_{hj}	λ	b_{OK}
4.1835	-26.0332	3.548	-0.73562	1.9594	-2.3406	-24.0384	8.1261	-1.078
3.1419	3.6054	-0.36381	-1.9651	-2.152	-4.9293	5.0308	-2.9587	
1.0815	-1.6325	0.4712	-0.27291	-0.90651	-1.3208	-2.0767	-0.78916	
-1.0815	0.8861	1.4178	1.1202	-1.3863	0.31068	0.21321	-1.852	
4.186	-27.8174	3.0948	-0.53317	1.8773	0.31068	-25.6167	-12.7112	
0.018849	-10.7969	-2.5628	-4.3041	4.3532	-5.1926	-3.7738	3.0238	
-2.758	-3.7144	-0.13042	-0.16113	-3.4486	-3.4213	-3.7011	-1.4468	
3.643	3.5253	1.06	1.6562	-1.7157	3.1487	-0.10328	1.4675	
0.20958	21.8944	7.2547	7.4236	-5.9335	7.6625	9.693	9.5999	
0.67147	-0.19387	1.7556	0.184331	0.37153	-0.39813	0.68139	-2.7786	
-3.7149	1.3188	-2.8947	-1.3753	-4.3276	-6.2882	-4.335	3.7792	
-4.7261	33.3708	9.3397	11.3213	10.045	11.4176	10.549	-4.4501	
1.7355	-0.72955	-0.66524	-0.53499	-1.8261	-1.2245	2.8832	1.4051	

Table 7

Weights and biases of the optimal ANN architecture (Binary systems)

Input-Hidden layer connections								
Weights(w_j^I)								Bias
T	P	M_{PC}	T_{PC}	P_{PC}	D_{PC}	X_I	X_J	b_{hj}

-1.1368	0.35087	0.061351	-0.092349	0.19155	0.015487	0.9552	0.93462	1.9505
-0.52848	-0.27209	-0.34413	1.5926	-1.8845	-2.2342	-0.89647	0.091077	0.72922
-9.2944	15.2025	-1.3931	-5.7894	-0.69764	-3.0922	-1.2185	0.63763	10.2253
0.74509	-0.046277	-2.1833	1.0658	1.6071	-1.7698	2.1854	-0.63659	-2.6666
-5.463	1.3739	0.35375	-3.044	-0.13265	3.0506	-0.70419	1.0513	0.063846
9.2908	-15.1812	0.67375	5.4099	0.5982	3.5809	0.51316	-0.9871	-9.9995
0.5417	-0.049875	-1.4994	1.3517	2.4231	-1.1869	2.38	-1.1708	-2.6427
-0.52533	-0.28921	1.232	2.1296	-1.0579	-2.9281	-0.47692	0.60561	0.72659
-5.2406	1.3486	1.0374	-3.6427	-1.5027	0.75957	-1.1621	0.60392	-0.0015349
-0.22325	-0.55471	-0.67886	-3.2055	0.02742	0.096161	1.0705	1.0084	-2.6841
-0.69205	0.062437	-0.88158	0.7745	0.60294	1.371	-0.51057	-0.9332	-1.8227
3.4761	21.2807	0.9883	1.1499	-0.0055736	-0.49002	0.76189	0.62307	21.9264
3.4725	12.7136	0.97232	1.2258	0.24431	-0.20199	0.64987	0.51264	14.4721
Hidden layer -Output connections								
Neuron	1	2	3	4	5	6	7	8
Weights (w_j^h)	2.1988	2.5544	-3.6827	1.0359	0.43907	-3.6939	-1.1012	-2.4532
Neuron	9	10	11	12	13	Bias (b_{OK})		
Weights (w_j^h)	-0.44522	-0.52743	3.8007	8.5022	-8.5843	0.80292		

Table 8
Weights and biases of the optimal ANN architecture (Ternary systems)

Input-Hidden layer connections									
Weights(w_j^I)									Bias
T	P	M_{PC}	T_{PC}	P_{PC}	D_{PC}	X_I	X_J	X_K	b_{hj}
-6.7739	31.4165	3.2577	0.39732	1.036	-2.527	-3.2365	-4.0231	-2.7244	25.5898
-0.29425	-0.9485	-0.21473	0.21213	1.2451	-0.7214	0.11886	-0.74649	-2.0618	3.0249
-2.47	0.18387	0.75536	1.8232	0.99324	-0.27279	0.11747	1.3872	-0.52704	-1.2649
6.7677	-0.902	0.55508	-0.22995	-0.18338	1.1255	-0.24203	-0.49338	0.82683	-1.0511
12.3596	-35.956	-3.9554	-1.2256	-0.82614	2.2185	3.2681	5.1516	3.3689	-28.9329
-8.6059	30.2842	3.1171	0.62656	0.4441	-3.11	-3.9329	-4.3703	-3.1823	24.2517
5.3871	-0.45338	0.17102	0.88106	1.5367	0.51125	-0.22673	0.45733	-0.37837	0.89032
-4.024	0.6629	-0.072903	0.8168	1.6132	0.075941	-0.90256	0.61739	-0.52554	0.30976
-0.55489	0.97025	0.81368	-1.2384	-1.1274	0.27533	-0.68395	2.6689	-0.7989	-0.52271
-1.9379	0.18981	-0.039593	-0.46625	-1.1118	-0.55954	1.1551	0.28289	0.45203	-1.8192
-0.93241	0.1198	-1.3877	-0.25618	-0.12932	0.32091	0.38547	0.91658	-0.77965	-2.7939
-1.7399	0.087514	0.91754	0.57086	-0.14689	0.19638	0.45431	-0.51917	-0.50043	-0.42572
Hidden layer -Output connections									
Neuron	1	2	3	4	5	6	7		
Weights(w_j^h)	5.5039	0.16279	2.6773	0.11532	-5.1089	-9.9932			0.33928
Neuron	8	9	10	11	12	Bias (b_{OK})			
Weights (w_j^h)	0.45079	0.36474	3.0944	-1.4005	-2.6727	1.1341			

Table 9

Parameters (weight and bias) of the ANN of global systems (pure systems +Binary systems +Ternary systems)

Input-Hidden layer connections									
Weights(w_j^1)									Bias
T	P	c	c	c	c			X_K	
6.957	-12.2166	2.9853	-1.3155	2.0151	-2.1339	3.3188	2.9528	2.015	-9.5758
1.4863	0.65639	-3.5468	-3.212	-0.67515	-5.9821	5.0425	-6.9349	3.4699	-3.1199
10.335	-81.7504	6.0135	-1.6097	4.2269	-3.718	17.1393	16.9112	12.3423	-63.1976
11.5506	-19.1889	1.0367	-2.2234	-4.1378	-10.7707	0.96966	6.8634	4.6281	-15.8314
-5.1279	1.2623	7.8443	5.3259	-5.9467	5.445	-8.7607	11.7747	-4.9305	-1.0918
1.4196	0.70906	-5.2684	-3.4587	-1.2712	-5.6834	6.5972	-7.3254	4.5641	-3.5073
-10.6208	23.0268	-10.2401	2.4067	-6.6956	6.7064	-4.5555	-4.6063	-3.5096	14.769
1.1016	-1.1453	-9.782	0.29611	-5.9066	6.0638	0.4596	0.41237	0.30055	-1.9767
10.1973	-21.9597	9.7397	-2.2774	6.3447	-6.3376	4.2893	4.3408	3.3243	-14.2148
-0.81496	0.045279	-1.263	0.050917	-0.43153	0.67735	-0.233	-0.21751	-0.17218	-2.5694
-5.9358	41.3513	-5.2625	1.0139	-2.699	3.4824	-8.3715	-8.3472	-6.1807	31.8419
-5.8418	41.3729	-4.9489	0.9846	-2.5971	3.2643	-7.8934	-7.8601	-5.8206	32.3549
-7.2882	13.0275	-2.485	3.8655	-2.1155	8.6409	0.18179	-2.7696	-1.5401	10.0868
Hidden layer -Output connections									
Neuron	1	2	3	4	5	6	7		
Weights(w_j^h)	0.5244	-1.9121	-2.2127	0.4186	-0.034145	1.8947	-5.637		
Neuron	8	9	10	11	12	13	Bias (b_{OK})		
Weights (w_j^h)	0.21922	-6.1822	11.9478	7.0574	-9.0366	0.49239	11.0953		

Table 10

Linear regression vectors (linear equation: $y^{cal} = \alpha y^{exp} + \beta$, with, α = slope β = y intercept, R^2 correlation coefficient)

Systems	N		α	β	R^2
Pure systems	1817	Training phase	1.000	0.00120	0.99667
		Validation phase	1.000	0.00120	0.99539
		Test phase	0.990	0.00030	0.9944
		Total	1.000	0.00096	0.99615
Binary systems	794	Training phase	1.000	0.00120	0.99810
		Validation phase	1.000	0.00094	0.99790
		Test phase	1.000	0.00130	0.99780
		Total	1.000	- 0.00120	0.99800
Ternary systems	616	Training phase	1.000	0.0001100	0.99810
		Validation phase	1.000	0.0001700	0.99910
		Test phase	1.000	0.0002900	0.99880
		Total	1.000	0.0000064	0.99830
Global systems (RN1)	3227	Training phase	1.000	0.00054	0.99668
		Validation phase	1.000	0.00067	0.99641
		Test phase	1.000	0.00071	0.99690
		Total	1.000	0.00058	0.99667

Table 11
The values of the different errors of this study

Systems	RMSE	MSE	AAD	AARD%	SSE	R^2_{Cal}
Pure systems	0.0027	0.0000	0.0024	07.6812	0.0244	0.9977
Binary systems	0.0030	0.0000	0.0020	07.4144	0.0074	0.9988
Ternary systems	0.0023	0.0000	0.0013	04.8599	0.0033	0.9989
Global system(RN1)	0.0036	0.0000	0.0023	08.1295	0.0411	0.9980

Table 12
Linear regression vectors and error performance between models obtained by RN1, RN2, RN3

	α	β	R^2_{exp}	RMSE	MSE	AAD	AARD%	SSE	R^2_{cal}
RN1	01.000	0.00058	0.99667	0.0036	0.0000	0.0023	08.1295	0.0411	0.9980
RN2	01.000	-0.00082	0.99805	0.0027	0.0000	0.0018	05.2531	0.0237	0.9988
RN3	01.000	-0.00330	0.99683	0.0035	0.0000	0.0023	07.7516	0.0393	0.9981

Table 13
Sources and ranges of new data base for pure systems.

Pure Systems	T (K)	P(Mpa)	$X_i = 1$	K(W. m. ⁻¹ K ⁻¹)	N	Reference
R22	298.15-393.15	0.1-5.76	1	0.0106-0.0682	130	[69]
	252.48-333.32	0.1-26.58	1	0.0667-0.1141	37	[43]
	223.15-323.15	2.1-20.1	1	0.0713-0.1262	50	[57]
	303-463	0.1	1	0.01089-0.02064	05	[45]
	208.56-289.6	0.9	1	0.0875-0.1222	16	[58]
R142b	293.15-353.15	0.1-1.35	1	0.0109-0.0165	21	[37]
	302.201-304.346	1.6425-69.5827	1	0.08064-0.11163	32	[39]
	223.15-323.15	2.1-20.1	1	0.0734-0.1183	25	[57]
	298-418	0.1	1	0.01249-0.0214	05	[45]
	210.4-289.55	4.31-7.59	1	0.0872-0.1164	07	[58]
R124	252.4-333.1	0.62-18.67	1	0.0584-0.0909	35	[52]

Table 14
Sources and ranges of new data base for binary systems

Binary Systems	T (K)	P(Mpa)	$X_i + X_j = 1$	K(W.m. ⁻¹ K ⁻¹)	N	Reference
R22+R142b	223.15-323.15	2.1-20.1	0.2796-0.729	0.01664-0.1207	75	[57]
	164.77-295.76	2.7-8.14	0.377-0.765	0.0854-0.1422	16	[58]
R22+R152a	223.15-323.15	2.1-20.1	0.2488-0.750	0.0759-0.1391	75	[57]
R22+R152a	176.6-297.45	2.44-8.02	0.269-0.765	0.0916-0.1512	13	[58]
R415	308.22-415.55	0.12-1.684	0.5+0.5=1	0.0139-0.0225	61	[70]

Table 15
Sources and ranges of new data base for ternary systems

Ternary Systems	T (K)	P(Mpa)	$X_i+X_j+X_k=1$	K(W. m. ⁻¹ K ⁻¹)	N	Reference
R409	305.67-427.13	0.0591-1.364	0.15+0.25+0.6=1	0.0118-0.0196	5 7	[71]

N: Number of experimental data; **R415** (R22, R152a: 50%, 50%); **R409** (R142b, R124, R22: 15%, 25%, 60%).

Table 16
Comparison between the prediction model of different results of new data base (DB2) obtained by each model RN1, RN2, RN3

Model	Systems	N	RMSE	MSE	AAD	ARD%	SSE	R^2_{Cal}
RN1	Pure	363	0.0284	0.0008	0.0212	48.7426	0.2925	0.8446
	Binary	240	0.0116	0.0002	0.0094	21.046	0.0486	0.9754
	Ternary	57	0.0304	0.0009	0.0202	115.911	0.0525	-2.7634
	Total	660	0.0244	0.0006	0.0169	44.7115	0.3936	0.8982
RN2	Pure	363	0.0218	0.0005	0.0147	23.0868	0.1723	0.9084
	Binary	240	0.0167	0.0004	0.0121	23.2145	0.1017	0.9484
	Ternary	57	0.0021	0.0000	0.0017	10.8684	0.0002	0.9827
	Total	660	0.0204	0.0004	0.0126	22.0780	0.2743	0.9291
RN3	Pure	363	0.0140	0.0002	0.0108	21.1563	0.0710	0.9623
	Binary	240	0.0090	0.0001	0.0092	14.6979	0.0292	0.9853
	Ternary	57	0.0020	0.0000	0.0017	10.8850	0.0002	0.9832
	Total	660	0.0123	0.0002	0.0094	17.9208	0.1004	0.9742

Table 17
Statistical analyses of the error of the predicted results (RN4).

System	N	MSE	MSE	AAD	AARD%	SSE	R^2_{Cal}
DB1+DB2	3887	0.0028	0.0000	0.0018	5.78	0.0303	0.9988

Fig.1. Feed forward neural network for the prediction of thermal conductivity of refrigerants.

(a)

(b)

(c)

(d)

Fig. 2. Comparison of experimental and calculated values for the whole data set (a) Pure systems, (b) Binary systems, (c) Ternary systems, (d) Global system.

(e)

(f)

Fig. 3. Comparison of reduced and calculated values for the whole data set: (e) RN2 (number of neurons=17), (f) RN3 (number of neurons=13) for global system.

(a)

(c)

Fig.4. Comparison between data base DB1 and DB2 for pure, binary and ternary systems: (a) RN1, (b) RN2, (c), RN3, for the prediction model.

Fig.5. Plot of the residuals for calculated values of thermal conductivity from the ANN model versus their experimental values for DB2, [$Residual = K_{exp,i} - K_{cal,i}$].

Fig.6. Prediction of the new data base DB2.

Fig. 7.The prediction between the DB2 and the actual data base DB1 in the range [0.01, 0.03].

Fig. 8.Regression analysis plot for the optimal model between output and target of thermal conductivity(RN4).

Fig. 9. Plot of the residuals for calculated values of thermal conductivity from the ANN model and their experimental data points (RN4).