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TRANSPORT PROPERTIES FOR REFRIGERANT MIXTURES

A set of models to predict viscosity and thermal conductivity of refrigerant mixtures is developed. A general model for viscosity and thermal conductivity use the three contributions sum form (the dilute-gas terms, the residual terms, and the liquid terms). The corresponding states model is recommended to predict the dense gas transport properties over a range of reduced density from 0 to 2. It is shown that the RHS model provides the most reliable results for the saturated-liquid and the compressed-liquid transport properties over a range of given temperatures from 0,5 to 0,95.

Keywords: Refrigerant – Mixtures – Viscosity – Thermal conductivity – Model – Prediction.

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ВЛАСТИВОСТІ ПЕРЕНОСУ СУМІШЕЙ ХОЛОДОАГЕНТІВ

Розроблено набір моделей для прогнозування в'язкості і теплопровідності сумішей холодоагентів. Загальна модель для в'язкості і теплопровідності використовує вид суми трьох вкладів (властивості розрідженого газу, надлишкові властивості і властивості рідини). Модель відповідних станів рекомендується для прогнозування транспортних властивостей цільного газу в діапазоні наведених густин від 0 до 2. Показано, що модель RHS забезпечує найбільш надійні результати для транспортних властивостей насиченої і стиснутої рідини в діапазоні наведених температур від 0,5 до 0,95.

Ключові слова: Холодоагент – Суміші – В'язкість – Теплопровідність – Модель – Прогнозування.

I. INTRODUCTION

Refrigerant mixtures are likely replacements for several of the CFC and HCFC refrigerants. Accurate transport properties are needed for the design of equipment using refrigerant blends; the transport properties are particularly important for the design of the condenser and evaporator.

Transport property data for some of newly commercialized refrigerant mixtures are now available in the literature. But there is no comprehensive mixture model for the transport properties. A suitable comprehensive model would allow data at compositions differing from the commercial blends and even data on different, but chemically similar mixtures, to be used to supplement the data available for a particular blend, allowing extrapolation to higher or lower temperatures or pressures.

The objective of this work is to carry out a literature survey and evaluate the available and most reliable transport property (viscosity and thermal conductivity) data for refrigerant blends, in both the liquid and vapor phases, and use these data to develop a comprehensive model for the transport properties of refrigerant mixtures.

II. TRANSPORT PROPERTY MODEL AND CORRELATIONS

General model. The best known models for the viscosity and thermal conductivity calculations use the form of the sum of three contributions

$$\eta(\rho, T) = \eta_0(T) + \Delta\eta(\rho, T) + \eta_c(\rho, T), \quad (1)$$

$$\lambda(\rho, T) = \lambda_0(T) + \Delta\lambda(\rho, T) + \Delta_c\lambda(\rho, T), \quad (2)$$

where η_0 and λ_0 are the viscosity and the thermal conductivity, respectively, in the zero-density limit, $\Delta\eta = \eta - \eta_0$ and $\Delta\lambda = \lambda - \lambda_0$ are the residual terms, $\Delta_c\eta$ and $\Delta_c\lambda$ are the critical viscosity and thermal conductivity enhancement, T is the temperature, and ρ is the density.

Each contribution may be treated independently by using both theoretical and available experimental information.

The residual term for the viscosity or thermal conductivity as a function of both density and temperature describes transport property data over a wide range of parameters including the high-density region (liquid at very low temperatures, compressed liquid). As one can see from the previous survey, the

availability of the transport property data for HFC refrigerant mixtures at low temperatures is limited especially for the viscosity (the lowest temperature of the published viscosity data is 223 K at saturation and no data are available for the viscosity of compressed liquid). The critical enhancement is significant for the thermal conductivity; however, this information is currently unavailable for the refrigerant mixtures under consideration. The critical viscosity enhancement is significant only extremely close to the critical point and may be safely ignored in all practical applications. Thus, in this representation, the transport properties of

vapor and the transport properties of liquid are treated independently. The vapor viscosity and thermal conductivity are composed of two contributions: a dilute gas term, which is a function only of temperature, and a residual term, which is considered to be a function only of density.

More than 100 literature sources were analyzed and more than 2000 experimental points were chosen to develop a comprehensive model for the transport properties of refrigerant mixtures.

III. DILUTE GAS TRANSPORT PROPERTIES

Dilute gas viscosity. The dilute gas viscosity can be well represented by the Chapman-Enskog equation derived from the kinetic theory for dilute gases

$$\eta_0 = 2,6696 \cdot 10^{-2} \frac{(MT)^{1/2}}{\sigma^2 \Omega_\eta (T^*)}, \quad (3)$$

where M is molecular mass in kg/kmol; $\Omega_\eta(T^*)$ is the collision integral; $T^* = kT/\varepsilon$ is the reduced temperature; $k = 1,38066 \cdot 10^{-23}$ J/K, the Boltzmann constant; T is the absolute temperature in K; η_0 is in $\mu\text{Pa}\cdot\text{s}$; σ (in nm) and ε/k (in K) are the characteristic potential parameters.

The collision integral is presented by Kestin et al. [1], and the characteristic potential parameters σ and ε and, correspondingly, ε/k can be determined from the dilute gas viscosity data. The optimum values of the characteristic potential parameters σ and ε/k for the mixed refrigerants R404A, R407C, R410A, and R507A derived from the best fit to low-pressure viscosity measurements are given in Table 1.

Table 1 – Potential Parameters ε/k and σ for R404A, R407C, R410A, and R507C

Refrigerant	ε/k (K)	σ (nm)
R404A	279,3	0,497
R407C	339,7	0,454
R410A	317,5	0,432
R507A	294,3	0,490

The deviations of these experimental data from the values calculated by Equation 2 did not exceed $\pm 0,5\%$. For all available experimental low pressure data, the deviations did not exceed 2,2%.

The second approach for the dilute gas viscosity is based on the equation developed by Nagaoka et al. [2] for pure fluorocarbon refrigerants

$$\eta_0 \left(T_c^{1/2} M^{-1/2} P_c^{-2/3} \right) = (0,5124T_r - 0,0517)^{0,82} Z_c^{-0,81} \quad (4)$$

where T_c is the critical temperature in K, P_c is the critical pressure in MPa, $T_r = T/T_c$ is the reduced temperature, and $Z_c = P_c/\rho_c RT_c$ is the critical compressibility.

Comparisons of the experimental low-pressure viscosity data for the binary and ternary mixtures with the values calculated by Equation 4 show that in no case do the deviations exceed 3%.

We have also applied a modified corresponding states method to the dilute gas viscosity of mixed refrigerants. According to this method

$$\varphi(P_r, T_r, \rho_r, A_1, A_2 \dots) = 0, \quad (5)$$

where $P_r = P/P_c$, $T_r = T/T_c$, and $\rho_r = \rho/\rho_c$ are the reduced pressure, temperature, and density, respectively; A_i are the defining parameters (criteria) of similarity.

The main questions (which defining parameters or criteria of similarity are to be chosen and how to determine the number of those criteria) were considered by Geller et al. [3]. Based on the analysis of a large set of experimental viscosity and thermal conductivity data, they proposed to use the stretch coordinates of two different points on the saturated vapor pressure curve as the criteria of similarity for the transport properties. The first (dominant) criterion was found as $A_1 = 100P_r$ when $T_r = 0,7$. This criterion is responsible for the equality of the type of binary molecular interaction potential and for the superposition of transport property surfaces in the dense gas region. It was also noted that the contribution of the viscosity in the zero-density limit does not require any defining parameters.

Based on this approach, the following equation is obtained for the reduced dilute gas viscosity

$$\eta_0 / \eta_{0c} = -0,1069 + 1,2518T_r - 0,1439T_r^2, \quad (6)$$

where $\eta_{0,c}$ is the reference dilute gas viscosity.

The values of $\eta_{0,c}$ for R404A, R407C, R410A, and R507A are given in Table 2. Comparisons of the experimental low-pressure viscosity data for available published results with the values calculated by Equation 6 show that the deviations do not exceed 1,6%. Equation 6 has good extrapolation ability, and it may be applied if at least one experimental low-pressure viscosity point is available.

Table 2 – Reference Dilute Gas Viscosity $\eta_{0,c}$ (at $T_r = 1$) and Residual Viscosity $\Delta\eta_r$

Refrigerant	$\eta_{0,c}$ ($\mu\text{Pa}\cdot\text{s}$)	$\Delta\eta_r$ ($\mu\text{Pa}\cdot\text{s}$)
R404A	14,02	90,88
R407C	14,87	88,94
R410A	14,88	85,13
R507A	14,07	98,01

Dilute gas thermal conductivity. For the thermal conductivity, exchange of energy between translational and internal degrees of freedom plays a more prominent part. The theoretical analysis based on kinetic theory has to take into account a number of additional cross sections pertaining to the relaxation and diffusion of the internal energy and its interaction with the translational energy. Therefore, kinetic theory cannot be applied to predict the thermal conductivity, especially in the case of mixtures.

Similar to the viscosity, a corresponding states method was applied to the dilute-gas thermal conductivity. Based on this approach, the following equation is obtained for the reduced dilute gas thermal conductivity

$$\lambda_0 / \lambda_{0c} = -0,6138 + 1,7177T_r - 0,1025T_r^2, \quad (7)$$

where $\lambda_{0,c}$ is the reference dilute gas thermal conductivity at $T_r = 1$.

The values of $\lambda_{0,c}$ for R404A, R407C, R410A, and R507A are given in Table 3. Comparisons of the experimental low-pressure thermal conductivity data for available published results [4-7] with the values calculated by Equation 7 are shown in Figure 1. As one can see, the deviations do not exceed 2,1% except for the results of Perkins et al. [8]. The largest deviations are seen with R32 + propane, is such an extreme polar/non-polar blend with large size difference within range of model.

Table 3 – Reference Dilute Gas Thermal Conductivity and Residual Thermal Conductivity

Refrigerant	$\lambda_{0,c}$ (mW/m·K)	$\Delta\lambda_r$ (mW/m·K)
R404A	16,78	3,424
R407C	17,81	3,267
R410A	16,69	3,494
R507A	16,74	3,181

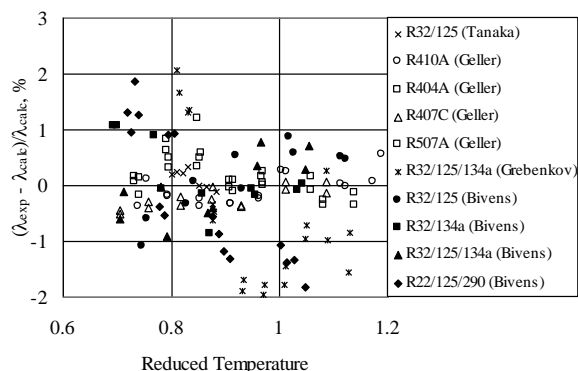


Figure 1 – Deviation of measured low-pressure thermal conductivity of mixed refrigerants from the thermal conductivity calculated by Equation 7

IV. DENSE GAS TRANSPORT PROPERTIES

The second contribution in Equations 1 and 2 (the residual term) can be considered as a function only of density or both density and temperature. The proper choice depends on the availability and accuracy of the viscosity and thermal conductivity data in the dense gas region. In this report, we consider the residual term as a function only of density.

Dense gas viscosity. For the viscosity, the reduced density limit is found to be from 2 to 2,2. Such an approach allowed us to treat the vapor viscosity and the liquid viscosity independently and to determine the viscosity in the reference point not only from the viscosity data in the vapor phase but using liquid viscosity data as well. The reduced density $\rho_r = 2$ was taken as the reference point for the residual viscosity treatment.

All mixtures under consideration as well as their pure components belong to approximately the same group of similarity defined by Geller and Paulaitis [3] with the values of the first criterion of similarity in a range from 4,8 to 5,5. For comparison, $A_1 = 9,9$ for argon and $A_1 = 1,5$ to 2 for heavy hydrocarbons. Thus, all available vapor viscosity data were treated as the reduced residual viscosity as a function of reduced density. The results are represented by equation

$$\Delta\eta/\Delta\eta_r = 0,0039\rho_r + 0,4652\rho_r^2 - 0,3016\rho_r^3 + 0,0956\rho_r^4 \quad (8)$$

where $\Delta\eta_r$ is the reference residual viscosity at $\rho_r = 2$.

The values of $\Delta\eta_r$ are given in Table 2. Comparisons of the experimental viscosity data for available published results with the values calculated by Equation 8 are shown in Figure 2. As one can see, the deviations do not exceed 4%.

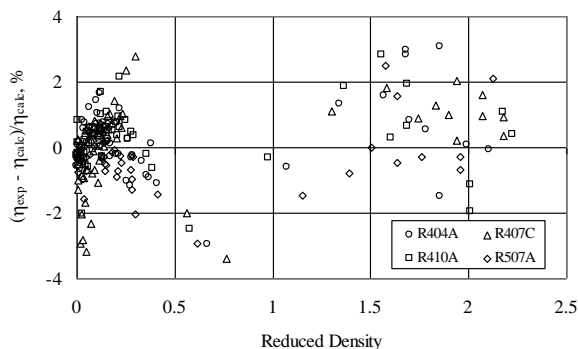


Figure 2 – Deviation of measured viscosity of R404A, R407C, R410A, and R507A from the viscosity calculated by Equation 9

Dense gas thermal conductivity. For the vapor thermal conductivity, the literature data are available only at reduced densities less than 0,4. Taking into account that the thermal conductivity shows a significant enhancement in a substantial region around the critical point, the vapor thermal conductivity and the liquid thermal conductivity were treated independently. The reference point for the residual thermal conductivity was taken at the reduced density $\rho_r = 0,2$, that is approximately in the middle of the measured range of parameters.

All available vapor thermal conductivity data were treated as the reduced residual thermal conductivity as a function of reduced density. The results are represented by equation

$$\Delta\lambda/\Delta\lambda_r = 4,5587\rho_r + 1,9318\rho_r^2 \quad (9)$$

where $\Delta\lambda_r$ is the reference residual thermal conductivity at $\rho_r = 0,2$.

The values of $\Delta\lambda_r$ are given in Table 3. Comparisons of the experimental thermal conductivity data for available published results with the values calculated by Equation 9 show that the deviations do not exceed 2%.

V. TRANSPORT PROPERTIES OF LIQUID

In recent years, a number of successful correlations of liquid viscosity and thermal conductivity have been based on the rough hard-sphere (RHS) model. The modified RHS model was applied by Assael et al. [9, 10] to the transport properties of many different fluids including hydrocarbons and alcohols, as well as pure refrigerants from the methane and ethane series. Most recently, Gao et al. [11] have used this model for prediction of the viscosity and thermal conductivity of binary and ternary refrigerant mixtures based on HFC refrigerants. Bleazard and Teja [12] presented parameter tables for the viscosity of 58 polar fluids including

refrigerants R124, R125, and R134a. Laesecke and Hafer [13] applied the RHS model for the viscosity of fluorinated propane isomers R245fa, R245ca, and R227ea. The RHS model is also used for the correlation of the liquid viscosity and thermal conductivity of mixed refrigerants in the present work.

Viscosity of liquid. The experimental viscosity data were converted to reduced viscosity according to

$$\eta^* = 6,0349 \cdot 10^8 \frac{\eta}{\rho_\mu^{2/3} \sqrt{MRT}}, \quad (10)$$

where η is the experimental viscosity in Pa·s, ρ_μ is the mole density in kmol/m³, and $R = 8,314$ J/(mol·K) is the universal gas constant.

This reduced viscosity is considered that of a rough hard-sphere fluid and fitted to the reduced viscosity of a smooth hard-sphere fluid whose density dependence is represented by the empirical and universal-for-all-fluids correlation

$$\log \left(\frac{\eta^*}{R_\eta} \right) = \sum_{i=0}^7 a_i V_r^{-i}, \quad (11)$$

$$\begin{aligned} a_0 &= 1,0945 \cdot 10^0 & a_1 &= -9,2632 \cdot 10^0 & a_2 &= 7,1039 \cdot 10^1 \\ a_3 &= -3,0190 \cdot 10^2 & a_4 &= 7,9769 \cdot 10^2 & a_5 &= -1,2220 \cdot 10^3 \\ a_6 &= 9,8756 \cdot 10^2 & a_7 &= -3,1946 \cdot 10^2. \end{aligned}$$

The reduced molar volume is defined as $V_r = V/V_0$, where V_0 is the temperature-dependent close-packed molar volume. Adjustable parameters are the roughness factor R_η and the temperature dependence of the close-packed molar volume. These parameters are determined from the available experimental data for pure fluids. R_η is usually fitted as a constant, and

$V_0(T_r)$ can be fitted as a linear function of reduced temperature (for a limited temperature range) or as a polynomial function.

Prediction of the viscosity of liquid mixtures is based on the assumption that the mixture behaves as a hypothetical pure fluid with molecular parameters given by the mole fraction average of the pure component values

$$V_{0,mix} = \sum_{i=1}^i x_i V_{0,i}, \quad (12)$$

$$R_{\eta,mix} = \sum_{i=1}^i x_i R_{\eta,i}, \quad (13)$$

where x_i are the mole fractions of the pure components, and the subscripts *mix* and *i* denote the mixture and the pure components, respectively.

Adjustable parameters in Equation 12 (R_η and V_0) were determined as follows: for R22 and R134a, these parameters are taken from the work of Gao et al. [11]; for R32, R125, R142b, R143a, R152a, and R290, these parameters were found from the most recent experimental viscosity data [13, 14]. The roughness factor R_η was fitted as a constant, and V_0 was fitted as a polynomial temperature function

$$V_0 \cdot 10^6 = \sum_{i=0}^3 a_i T_r^i \quad (14)$$

The values of R_η and the coefficients a_i of Equation 14 are listed in Table 4.

Table 4 – Roughness Factor R_η and Coefficients a_i in Equation 15

	R32	R125	R134a	R142b	R143a	R152a	R290
R_η	1,224	1,407	1,100	1,222	1,268	1,092	1,041
a_0	92,609	-34,353	49,759	89,676	-84,278	111,17	50,705
a_1	-277,90	321,10	-9,0398	-97,339	504,52	-237,97	-9,9289
a_2	378,77	-407,53	7,1778	54,912	-656,80	257,28	0
a_3	-177,13	159,24	-7,2956	0	271,30	-93,345	0

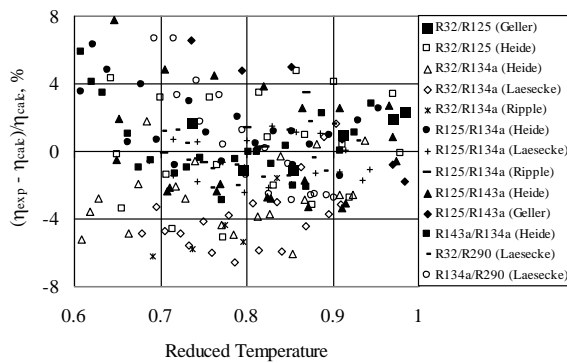


Figure 3 – Deviation of measured viscosity of binary mixtures from the viscosity calculated by Equation 11

For fluorinated propane isomers R245fa, R245ca, and R227ea, R236ea, and R236fa, R_η and V_0 can be found in the paper [13]. Comparisons of the experimental viscosity data of binary mixtures with the values calculated by Equation 12 are shown in Figure 3. As one can see, the deviations are mostly in

a range of $\pm 5\%$ except for a few experimental points where the deviations are up to 8%.

Thermal conductivity of liquid. The experimental thermal conductivity data were converted to the reduced thermal conductivity according to

$$\lambda^* = 1,936 \cdot 10^7 \frac{\lambda}{\rho_\mu^{2/3} \sqrt{RT}}, \quad (15)$$

where λ is the experimental thermal conductivity in W/(m·K).

The density function of the reduced thermal conductivity is found to be universal for all fluids and represented by the empirical correlation

$$\log \left(\frac{\lambda^*}{R_\lambda} \right) = \sum_{i=0}^4 a_i V_r^{-i} \quad (16)$$

$$a_0 = 1,0655; \quad a_1 = -3,538; \quad a_2 = 12,120; \quad a_3 = -12,469; \quad a_4 = 4,562.$$

The roughness factor R_λ for the thermal conductivity of pure refrigerants was found from the most

reliable experimental data and was fitted as a polynomial temperature function

$$R_{\lambda} = \sum_{i=0}^3 a_i T_r^i. \quad (17)$$

The values of R_{λ} are given in Table 5.

For the mixture thermal conductivity, R_{λ} is given by the mole fraction average of the pure component values

$$R_{\lambda,mix} = \sum_{i=1}^i x_i R_{\lambda,i}, \quad (18)$$

where x_i are the mole fractions of the pure components.

Table 5 – Coefficients a_i in Equation 18

	R32	R125	R134a	R142b	R143a	R152a	R290
a_0	0,98590	10,281	1,1659	1,7701	0,42061	-0,22460	-0,85450
a_1	2,4422	-32,688	1,7780	-1,7994	2,3532	3,4732	10,814
a_2	-3,5072	40,005	-3,4044	1,8385	-2,1513	-1,6137	-17,225
a_3	2,3633	-15,218	2,3646	0	1,6135	0	9,1347

Comparisons of the experimental thermal conductivity data with the values calculated by Equation 16 show that the deviations for all mixtures are mostly in a range of ± 6 to 8%.

VI. CONCLUSIONS

A literature survey to identify the available viscosity and thermal conductivity data for refrigerant mixtures containing HFC refrigerant in both vapor and liquid phases is conducted. The most accurate data on which to base the viscosity and thermal conductivity models development is evaluated.

A set of models can be recommended to predict viscosity and thermal conductivity of refrigerant mixtures. The dilute-gas viscosity is calculated with the Chapman-Enskog method, Nagaoka method, and the corresponding states method with approximately the same accuracy. The saturated liquid viscosity may be predicted with the ECS model over a range of reduced temperature from 0,6 to 0,95 or the RHS model without temperature limitation. The corresponding states model is recommended to predict the dense gas and liquid viscosity over a range of reduced density from 0 to 2. This model may be applied if at least one experimental dense-vapor or liquid viscosity point is available in the mentioned density range. If no experimental data is available the reference state viscosity may be found from the RHS model.

The dilute-gas and the dense-gas thermal conductivity is calculated with the corresponding states method. Calculations of the dense-gas thermal conductivity are valid in the range of reduced density from 0 to 0,7. The saturated-liquid thermal conductivity and the compressed-liquid thermal conductivity may be predicted with the RHS model or with the ECS model over a range of reduced temperature from 0,5 to 0,95. The RHS model provides the more reliable results.

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СВОЙСТВА ПЕРЕНОСА СМЕСЕЙ ХЛАДАГЕНТОВ

Проведен обзор литературы для определения доступных данных о вязкости и теплопроводности смесей хладагентов, содержащих альтернативные хладоны в обеих паровой и жидкой фазах. Оценены наиболее точные данные, на основе которых базируется разработка моделей вязкости и теплопроводности. Разработан набор моделей для прогнозирования вязкости и теплопроводности смесей хладагентов.

Общая модель для вязкости и теплопроводности использует вид суммы трех вкладов (свойства разреженного газа, избыточные свойства и свойства жидкости). Показано, что вязкость разреженного газа рассчитывается с помощью метода Челмена-Энскога, метод Нагаоки и метода соответственных состояний с примерно одинаковой точностью. Вязкость насыщенной жидкости может быть предсказана с помощью модели расширенного закона соответствующих состояний (ЗСС) в диапазоне приведенных температур от 0,6 до 0,95 или с помощью модифицированной модели твердых сфер (МТС) без ограничения температуры. Модель соответствующих состояний рекомендуется для предсказания вязкости плотного газа и жидкости в диапазоне приведенных плотностей от 0 до 2. Эта модель может быть применена, если имеется хотя бы одна экспериментальная точка по вязкости плотного газа или жидкости в указанном диапазоне плотностей. Если такие экспериментальные данные отсутствуют, приведенная вязкость можно найти из модели МТС.

Теплопроводность разреженного и плотного газа рассчитывается по модели соответственных состояний. Расчеты теплопроводности плотного газа правомерны в диапазоне приведенных плотностей от 0 до 0,7. Теплопроводность насыщенной и сжатой жидкости может быть предсказана по модели ЗСС или по модели МТС в диапазоне приведенных температур от 0,5 до 0,95. Модель МТС обеспечивает более надежные результаты.

Сравнения наиболее надежных экспериментальных данных с расчетами по предлагаемым моделям показывают, что отклонения для всех смесей, в основном, лежат в диапазоне экспериментальных погрешностей.

Ключевые слова: Хладагент – Смесей – Вязкость – Теплопроводность – Модель – Прогнозирование.

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