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Solid–liquid phase equilibrium study of *n*-octadecane + lauryl alcohol binary mixtures

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1. Introduction

The storage of energy in suitable forms, which can conventionally be converted into the required form, is a present day challenge to the technologists. Energy storage not only reduces the mismatch between supply and demand but also improves the performance and reliability of energy systems and plays an important role in conserving the energy. One of the prospective techniques of storing thermal energy is the application of phase change materials (PCMs) [1,2]. A phase change material (PCM) is capable of absorbing or releasing great amount of energy in a form of latent heat ($\triangle H$) during phase transitions between solid–solid or solid–liquid phases over a narrow temperature range. This action is typically transient, *i.e.*, it will occur until a latent heat of the PCM is absorbed or released [3–5].

Most of the investigations on developing PCMs were focused on organic or inorganic compounds [6]. Apart from these compounds, a considerable number of binary and ternary eutectics of organic or inorganic compounds may be tailored with almost any desired melting point. However, the number of studies on the latent heat storage by a eutectic PCM is limited in the literature. Thus, at first, we focused our attention on the binary mixtures composed of *n*octadecane and lauryl alcohol. The solid–liquid phase behavior of the mixtures was investigated by means of differential scanning

ABSTRACT

Differential scanning calorimetry (DSC) is used to investigate the thermal properties of *n*-octadecane and lauryl alcohol and their binary system. The experimental results show that the phase diagram of the *n*-octadecane + lauryl alcohol binary system presents a eutectic behavior for the solid–liquid equilibrium line, and the corresponding mole fraction of *n*-octadecane in eutectic point is 0.257. The eutectic melting temperature (T_E) is 292 K, and the latent heat of melting of eutectic mixture ($\triangle_{fus}H_E$) is 245 J · g⁻¹. Furthermore, the activity coefficients of components in mixtures of *n*-octadecane + lauryl alcohol have been correlated by the Wilson, NRTL and UNIQUAC models, respectively. The best correlation of the solid–liquid equilibrium data has been obtained by the NRTL equation, in which the average root-mean-square deviation of temperature is 0.15 K.

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calorimetry. Furthermore, the activity coefficients of components in (*n*-octadecane + lauryl alcohol) mixtures have been correlated by the Wilson, NRTL and UNIQUAC models to investigate the performance of these expressions for predicting SLE of *n*-octadecane with lauryl alcohol, respectively.

2. Experimental section

2.1. Materials

Highly pure samples of *n*-octadecane, and lauryl alcohol were obtained from J&K Scientific Company. (Beijing, China), all of them were guaranteed reagent grade with purity higher than 99.5%. Their melting temperatures and enthalpies of fusion were measured using DSC (Metter DSC30). The melting points of the pure samples were in close agreement with literature data and no further purification was made. Properties of pure compounds, $T_{\rm m}$, the melting temperature; and $\triangle_{\rm fus}H$, the molar enthalpy of fusion were listed in table 1 together with those reported in literature. These results agree well with the references.

2.2. Equipment

The calorimetry experiments were performed on a Metter DSC30 differential scanning calorimeter equipped with sample and reference standard aluminium crucibles, each with a capacity of 40 μ L. During the scanning operation, high purity nitrogen gas



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TABLE 1

Physical constants of pure compounds: $\Delta_{fus}H$, and T_m , denote the molar enthalpy of fusion, the melting temperature, respectively.

Component	$\Delta_{\rm fus} H/(\rm kJ\ mol^{-1})$	$T_{\rm m}/{\rm K}$
n-Octadecane	61.65 ^a	300.95 ^a
	61.706 ^b	301.15 ^b
	61.7 ^c	301.35 ^c
	60.760^d	301.0 ^d
Lauryl alcohol	39.05 ^a	297.15 ^a
	31.380 ^e	297.50 ^f
	40.2 ^c	297.05 ^c
	40.17 ^g	300.2 ^g

^a This work.

^c Haynes [8].

^d Kolesnikov and Syunyaev [9]. Domanska [10].

^f Bylicki and Plesnar [11].

^g Acree Jr. [12].

was purged through the DSC chamber at 50 ml \cdot min⁻¹. The measurements were carried under fixed conditions of constant heating rate of $1 \text{ K} \cdot \text{min}^{-1}$. This scanning rate is low enough to approach the equilibrium measurement conditions. Before analysing, the heat flow was calibrated by using the heat of fusion of an exactly known quantity of indium: temperature measurement with the Pt100 sensor was calibrated by using a pan containing an exactly known quantity of indium as well as lead and zinc in separate compartments. The uncertainties of the measurements are estimated to be ± 0.2 K for the temperature and $\pm 1\%$ for the enthalpy.

2.3. Sample preparation

The samples used in this work were prepared on an analytical balance (Sartorius A200S) with a ±0.2 mg accuracy. The weighed compounds were placed in a glass tube, then heated and stirred under a nitrogen atmosphere until 10 K above the higher melting point of the components. The mixtures were then allowed to cool down to room temperature and kept in a freezer at 273 K. Then a small amount of solid about 5 mg was taken and sealed in a Mettler sample crucible for the analysis.

2.4. SLE measurement

As described in detail previously [13,14], we adopted the method in which a solidus and a liquidus curves were estimated from the onset and peak temperatures obtained from the measured DSC curves. The onset temperatures at each first peak almost show constant value for the eutectic temperature. The next peak temperatures were liquidus temperature.

Before each measurement, the sample pans were heated in the DSC until a temperature 15 K above the melting point of the heavier compound of the mixture. At this temperature the pans were kept for additional 20 min. Afterwards, the pans were cooled down until a temperature 25 K below the melting point of the lighter compound and kept in this state for 30 min. This pre-treatment was performed in order to avoid thermal memory and polymorphic effects. Only after such treatment the experimental run for measuring, the equilibrium data was carried out.

3. Results and discussion

3.1. Determination of solid-liquid phase diagrams

The equilibrium temperatures for varying mole fractions of noctadecane, x_1 , with lauryl alcohol were listed in table 2 and shown

TABLE 2

Measured solid-liquid equilibrium data for the system {n-octadecane (1) + lauryl alcohol (2): x_1 denotes the mole fraction of *n*-octadecane, the component 1 in this work; T, the temperature of solid-liquid phase transformation, ΔH , the enthalpy of the eutectic reaction; $T_{\rm E}$, the eutectic temperature; $\gamma_1^{\rm exp}$ and $\gamma_2^{\rm exp}$, the experimental activity coefficients of component 1 or 2, respectively.

<i>x</i> ₁	T/K	$\Delta H/(J \cdot g mix^{-1})$	$T_{\rm E}/{\rm K}$	γ_1^{exp}	γ_2^{exp}
1.0000	300.95	0		1.000	
0.9512	300.35	19.3	292.05	1.001	
0.8980	299.85	33.2	291.95	1.017	
0.7984	298.85	72.2	292.00	1.053	
0.7015	298.30	105.4	292.15	1.145	
0.6019	297.85	128.9	292.25	1.286	
0.5513	297.45	154.7	292.30	1.357	
0.5008	296.95	163.0	292.10	1.433	
0.4728	296.60	167.4	292.15	1.474	
0.4138	295.55	187.9	292.15	1.541	
0.3744	294.85	198.9	292.05	1.604	
0.3142	293.80	211.8	292.15	1.747	
0.2890	293.05	221.0	292.00	1.781	
0.2090	293.15	193.4	292.00		1.019
0.1724	293.85	168.6	291.95		1.012
0.08414	295.55	82.0	292.15		1.002
0.0000	297.15	0			1.000

^a Estimated uncertainties in temperature, T, and mole fraction compositions, $x_1: \delta T = \pm 0.28 \text{ K}; \ \delta x_1 = \pm 0.0005.$

graphically in figure 1. The phase diagram of the (n-octadecane + lauryl alcohol) system was found to be of the simple eutectic type.

Tammann's plot [15] was shown in figure 2. From the characteristic Tammann's triangle shape obtained after a linear regression of the enthalpy of each eutectic reaction versus the sample composition, the eutectic composition and the enthalpy of fusion at the eutectic point of the (n-octadecane + lauryl alcohol) system, were accurately determined: $x_1 = 0.257$, and $\triangle_{fus}H_E = 245 \text{ J} \cdot \text{g}^{-1}$, respectively. The eutectic temperature determined from all appropriate DSC curves is $T_{\rm F}$ = 292 K.

The eutectic mixture had large latent heat and suitable transition temperature in the range of indoor comfortable temperature. It could be applied for thermal energy storage as PCM.

Experimental solid-liquid equilibria investigated in this work are characterized mainly by the following: positive deviations from ideality were found, thus the activity coefficient, γ , is greater than 1 (see the values of activity coefficients in table 2 and figure 1; ideal solution, $\gamma = 1$).



FIGURE 1. (Solid + liquid) phase diagram for {n-octadecane(1) + lauryl alcohol(2)}. \bigcirc , fusion temperature; \Box , eutectic temperature; ideal solution $(\ln \gamma_i = 0,$ equation (2)); -, description given by the NRTL model.

^b Domanska [7].

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