Mutual Solubilities for Water−*o*-Nitrotoluene System and Distribution Coefficients for Furfural and Acetic Acid in Water−*o*-Nitrotoluene System

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Ten high boiling solvents were evaluated as the extraction solvent of furfural. *o*-Nitrotoluene with a high furfural distribution coefficient, high thermal stability, and having no azeotrope with furfural was chosen as a furfural extraction solvent for further study. The mutual solubilities for the water−*o*-nitrotoluene system were experimentally determined from (383.15 to 483.15) K at 25.0 bar using a static method. According to the results, it can be seen that the solubilities are temperature-dependent and can be correlated with a quadratic equation well. The distribution coefficients for furfural and acetic acid in the water−*o*-nitrotoluene system were determined from (303.15 to 483.15) K at 25.0 bar using a static method, respectively. The results show that the distribution coefficients are temperature-dependent and can be correlated with van't Hoff equation well. Considering the high selectivity for furfural, the low selectivity for acetic acid and negligible mutual solubilities with water, *o*-nitrotoluene was considered as a suitable solvent for furfural production in the solvent extraction process.

Introduction

Furfural is a key derivative, readily accessible from renewable biomass and agricultural surpluses, for the production of a wide range of important nonpetroleum-derived chemicals.[1,2] Furfural is usually produced by the dehydration of pentose (mainly xylose) in the presence of acid catalyst in the temperature range of (413 to 473) K.[1,3] Secondary reactions between furfural and its precursors are the primary cause of low yields.[4−6] Hence, high yields of furfural can only be achieved by the rapid and continuous removal of furfural from the aqueous solution. In traditional furfural production process, furfural produced is removed as quickly as possible from the reaction mixture by blowing in steam. The high consumption of steam and the release of an amount of wastewater are major drawbacks of the process based on steam-distillation. In recent years, acetic acid was suggested as a catalyst for furfural production.[7] Acetic acid was less corrosive to the equipment and more environmentally friendly than traditional mineral acid catalysts, such as sulfuric acid and phosphoric acid. Moreover, acetic acid was a byproduct of pentose production process, and it could be recovered efficiently with an extraction technology.[8] The selection of acetic acid as a catalyst will contribute to reducing industrial pollution emissions and promoting better green economy.

Solvent extraction was suggested for producing furfural with high yields and low energy consumption.[9,10] Furfural solvent extraction process is a process in which dehydration reactions of pentose (mainly xylose) are carried out in the presence of an immiscible organic solvent so that most of the furfural can be transferred from the aqueous reacting phase to the solvent phase almost as soon as it is formed to prevent furfural consecutive reactions. Then, furfural is recovered by a simple binary distillation from the solvent. Therefore, a suitable furfural extraction solvent should have a higher boiling point than that of furfural (B.P., 434.85 K) and have the property of not forming an azeotrope with furfural. Moreover, the mutual solubilities of water and solvent must not be appreciable, and it must be stable under the harsh dehydration conditions for furfural production. Also, the furfural distribution coefficient should be high as much as possible.

In this study, 10 high boiling solvents were evaluated as furfural extraction solvents. *o*-Nitrotoluene was considered as a suitable furfural extraction solvent. The mutual solubilities for the water−*o*-nitrotoluene system were experimentally determined from (383.15 to 483.15) K at 25.0 bar using a static method.[11−14] The distribution coefficients for furfural and acetic acid in the water−*o*-nitrotoluene system were determined from (303.15 to 483.15) K at 25.0 bar using a static method,[11−14] respectively.

Experimental Section

Materials. Furfural (99.5+ %), ethanol (99.5+ %), tetralin (99.5+ %), 1,2,4-trichlorobenzene (99.5+ %), *o*-nitrotoluene (99.5+ %), 2,4-dichlorotoluene (99.0+ %), decalin (98.0+ %), isophorone (98.0+ %), 1-bromonaphthalene (98.0+ %), 1-chloronaphthalene (98.0+ %), *o*-nitroanisole (99.5+ %), 1,2-dibromobenzene (98.0+ %), and acetic acid (99.5+ %) (all purchased from Sinopharm Chemical Reagent Co., Ltd.) were of analytical reagent grade and used without any treatment. Isopropanol (Merck, Germany) was of high purity; the mass fraction was better than 99.99 %, and the water mass fraction was less than 0.005 %. Deionized water was prepared in our laboratory using a four-stage reverse osmosis system.

Apparatus and Procedure. The experiments were performed using a laboratory system that was similar to the apparatus in the literature[12−14] (shown in Figure 1). The components of the system include an equilibrium cell (350 mL, 65 mm outer diameter (o.d.) × 55 mm inner diameter (i.d.) × 150 mm height, stainless steel), stainless steel tubing (1.59 mm o.d. × 0.762 mm i.d., keystone scientific), shut-off valves (HIP model 02-0120, High-Pressure Equipment Co., Eire, PA), an electric jacket, an intelligent temperature controller with a precision of ± 0.1 K, a nitrogen cylinder, a buffer vessel (the volume was about 10 L), and a magnetic stirrer (all purchased from Tianjin

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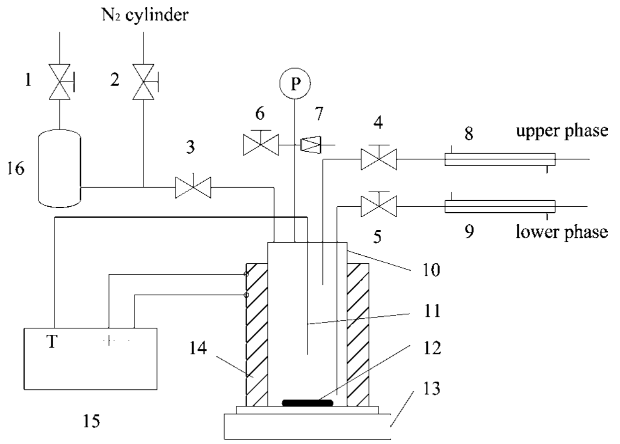

Figure 1. Schematic diagram of the mutual solubility and distribution coefficient determination system 1−6, high pressure valves; 7, safety valve; 8−9, cooler; 10, equilibrium cell; 11, thermocouple; 12, magnetic stirring rod; 13, magnetic stirrer; 14, electric jacket; 15, intelligent temperature controller; 16, buffer vessel.

University Kewei Co. of China). The lengths of sampling lines for upper and lower phases were both 80 cm with an internal volume of 0.35 mL.

The mutual solubilities and distribution coefficient were measured using the static method.[11−14] The measuring principle is the same as that described in the literature.[11−14] In the mutual solubility measuring experiments, equal volumes of water (150 mL) and *o*-nitrotoluene (150 mL) were added into the equilibrium cell. The source line inlet for upper phase and the source line inlet for lower phase were both kept a distance of about 3 cm from the phase interface to sample the representative samples. The cap of the equilibrium cell was tightened and degassed with high purity nitrogen. The shut-off valves 1, 4, 5, and 6 were closed to prevent leakage. The pressure of the equilibrium cell was maintained at 25.0 bar in the experiments by regulating valves 1 and 3 and the nitrogen cylinder. Continuous stirring of the water−*o*-nitrotoluene system was achieved with a magnetic stir bar. The equilibrium cell was heated by an electric jacket, and the temperature was controlled at the desired value by an intelligent temperature controller (fluctuates within ± 0.1 K). The water−*o*-nitrotoluene system was kept stirring for at least 8 h after reaching the desired temperature. The temperature and pressure were kept stable to establish equilibrium between water and *o*-nitrotoluene. Then the stirring was stopped, and the water−*o*-nitrotoluene system was settled down for 8 h. The aqueous phase and *o*-nitrotoluene phase separated completely because of the larger density difference between water and *o*-nitrotoluene (*o*-nitrotoluene has a density of 1.16 $\text{g}\cdot\text{cm}^{-3}$ at 303.15 K). The upper phase was water, and the lower phase was *o*-nitrotoluene.

After equilibrium was established, the saturated aqueous phase and *o*-nitrotoluene phase were sampled through the sample lines in sequence. The system pressure was kept stable during sampling processes (fluctuates within ± 0.5 bar). The sampling process did not change the equilibrium state. Each phase passes through a water-cooled heat exchanger 0.5 m in length prior to being collected. Tap water is used on the shell side to cool samples to room temperature before collection, and this aids in the prevention of sample evaporation. Approximately 4 mL of each phase was pushed out to wash the sample line to collect the representative samples. Then about 4 mL of sample was collected into a 10 mL vial. Three samples of each phase were collected for analysis at each experimental equilibrium condition. The mass of the vial and sample collected in the vial were measured using an analytical balance (Metler Toledo AL104, Shanghai, China) with an accuracy of ± 0.1 mg. Because a potential exists for phase separation to occur in the aqueous and solvent phases due to adsorption of the solute on sample line surfaces, the separated phases exit the phase separation cell through 1.59 mm o.d. × 0.762 mm i.d. stainless steel capillary tubing. This tubing minimizes dead volume, thus minimizing the effects of phase separation on sample composition.

In the furfural distribution coefficient measuring experiments, equal volumes of *o*-nitrotoluene (150 mL) and furfural aqueous solution (150 mL) were poured into the equilibrium cell, whereas the initial *o*-nitrotoluene phase furfural concentration was zero, and the initial aqueous phase furfural concentration was 5 % by mass. The furfural distribution coefficient measuring procedure was almost the same as that described above, and the only difference was that the stirring time was controlled at 2 h, and the phase separation time was controlled at 30 min. It was long enough for phase separation because of the larger density difference between water and *o*-nitrotoluene. The furfural degradation rate was very low, and the loss of furfural was negligible in such a short time.[4−6] In the acetic acid distribution coefficient measuring experiments, the initial aqueous phase acetic acid concentration was 5 % by mass, and the initial *o*-nitrotoluene phase acetic acid concentration was zero. Both the stirring time and the phase separation time were controlled at 2 h. The measuring procedure was the same as that described above.

In the solvent selection experiments, the furfural distribution coefficient was measured under atmospheric pressure at 303.15 K. The experiment was performed in a 25 mL glass test tube. The thermal stability tests of the solvents were carried out in the equipment mentioned above (shown in Figure 1). Equal volumes of solvent (30 mL) and acetic acid aqueous solution (30 mL, mass fraction concentration 5 %) were poured into the equilibrium cell. The system was maintained for 24 h at 483.15 K at 25.0 bar. Then, the system was cooled to room temperature, and the solvent−water system was poured into a 100 mL glass test tube. If there was color variance or the presence of humic-like substances was observed, the solvent was considered unstable. In the opposite case, the aqueous phase and solvent phase would be analyzed using a HP model 6890 GC with a hydrogen flame ionization detector (FID) to confirm if the degradation of the organic solvent occurred.

Analytical Methods and Procedure. The analyses of equilibrium phase samples proceeded as follows. First, a known amount (about 4 mL) of high-purity isopropanol, by weight, was added to the sample bottle. The isopropanol was used as a homogenizing cosolvent to provide a single-phase sample for analysis (the isopropanol was reagent of high purity with water mass fraction less than 0.005 %, so the amount of water contained in the isopropanol was negligible). In the mutual solubility measuring experiments, the water mass fraction of the *o*-nitrotoluene phase sample was measured by the high resolution Karl Fisher Coulomb method microwater detector (JF-5, Daqing Equipment Manufacture Co., Ltd.). The *o*-nitrotoluene mass fraction of the aqueous phase sample was analyzed using a HP model 6890 GC with an FID. Chromatographic separations were accomplished with a 30 m HP-FFAP (0.25 mm i.d., 0.25 μm film thickness, Hewlett-Packard) fused silica capillary column. Methyl benzoate was chosen as the internal standard. In the distribution coefficient measuring experiments, all analyses were performed using the HP model 6890 GC with an FID. Ethanol was chosen as the internal standard. The mass of cosolvent and internal standard substances

Table 1. Property of Solvents Investigated for the Extraction of Furfural

solvent	distribution cofficient (303.15 K)	B.P., K	M.P., K	specific gravity	azeotrope with furfural	thermal stability
tetralin	2.06	480.75	237.45	0.97	none	good
1,2,4-trichlorobenzene	2.74	486.15	290.15	1.46	none	good
o-nitrotoluene	9.16	498.15	269.05	1.16	none	good
2,4-dichlorotoluene	4.56	471.15	275.75	1.25	none	good
cis-decalin	0.96	467.75	229.85	0.89	none	good
isophorone	13.37	487.15	265.05	0.92	none	poor
1-bromonaphthalene	3.11	554.25	279.35	1.48	none	poor
1-chloronaphthalene	4.45	532.45	275.45	1.19	none	poor
o-nitroanisole	10.69	550.15	282.55	1.25	none	poor
1,2-dibromobenzene	2.78	498.65	280.28	1.98	none	poor

Table 2. Mass Fraction Mutual Solubilities for Water and *o*-Nitrotoluene from T = (383.15 to 483.15) K and p = 25.0 bar

o-nitrotoluene phase			aqueous phase		
T/K	$10^2\ x^{\mathrm{exp}}$	$[10^2(x^{\mathrm{exp}} - x^{\mathrm{cal}})]/x^{\mathrm{exp}}$	T/K	$10^2\ x^{\mathrm{exp}}$	$[10^2(x^{\mathrm{exp}} - x^{\mathrm{cal}})]/x^{\mathrm{exp}}$
383.15	0.7426	−4.18	383.15	0.2617	2.50
393.15	0.9487	3.26	393.15	0.3118	−2.63
403.15	1.1205	2.46	403.15	0.4104	1.26
413.15	1.3187	0.93	413.15	0.5133	−0.94
423.15	1.5649	−0.17	423.15	0.6507	−2.78
433.15	1.8709	−0.92	433.15	0.8651	−0.76
443.15	2.2433	−1.76	443.15	1.1562	0.79
453.15	2.7501	−0.74	453.15	1.5489	1.60
463.15	3.3672	−0.23	463.15	2.1013	2.70
473.15	4.1198	−0.18	473.15	2.8031	1.21
483.15	5.1337	1.31	483.15	3.6702	−3.18

added in the samples was measured using an analytical balance (Metler Toledo AL104, Shanghai, China) with an accuracy of ± 0.1 mg. The samples were collected after the addition of the cosolvent and internal standard substances to the sample bottles to avoid reopening the sample bottles and thus minimizing sample contact with the atmosphere.

All of the measurements reported in this article were repeated three times at each temperature, and the average value was taken. The maximum deviations of determined furfural distribution coefficient values in Table 1 were less than ± 4.2 %. The deviation of determined water solubility in *o*-nitrotoluene was less than ± 1.8 %, and the deviation of determined *o*-nitrotoluene solubility in water was less than ± 2.1 %. The deviations of determined furfural and acetic acid distribution coefficient values were within ± 3.5 % and ± 4.7 %, respectively.

Results and Discussion

Solvent Selection. Physical property data have been collected for the 10 solvents chosen in a preliminary screening of furfural extraction solvents. Experiments were conducted to determine the distribution coefficient of furfural at 303.15 K and the thermal stability of the solvents in acetic acid aqueous solutions at 483.15 K. The results of this experimental work and physical property search are summarized in Table 1. The results indicated that *o*-nitrotoluene was stable under the harshest dehydration condition for furfural production and had the property of not forming an azeotrope with furfural. Moreover, the furfural distribution coefficient for *o*-nitrotoluene was much higher than that of any other stable solvents presented in Table 1. So *o*-nitrotoluene was chosen as the furfural extraction solvent in this study.

Mutual Solubilities for the Water−o-Nitrotoluene System. The mass fraction water solubility in *o*-nitrotoluene and mass fraction of *o*-nitrotoluene solubility in water at different temperatures are presented in Table 2 and more visually given in Figure 2, where T is the absolute temperature and x^{exp} and x^{cal} are the experimental and calculated values of the solubility. From Figure 2, it can be seen that the mutual solubilities for the water−*o*-nitrotoluene system are temperature-dependent, and the mutual solubilities are very low in the experimental temperature range. The solubility of water in *o*-nitrotoluene is slightly higher than that of *o*-nitrotoluene in water under the experimental temperature range. The water solubility in *o*-nitrotoluene and *o*-nitrotoluene solubility in water were all correlated as a function of temperature by the following form of a quadratic equation:

$$\ln x = A + B(T/\mathrm{K}) + C(T/\mathrm{K})^2 \tag{1}$$

The experimental solubility values were fitted with eq 1 by the least-squares method. The values of the three parameters A, B, and C are listed in Table 3 together with the root-mean-square deviation (rmsd), namely, the standard deviation, which is defined as follows:[15]

$$\mathrm{rmsd} = \left[\frac{1}{N}\sum_{i=1}^{N}(x_i^{\mathrm{exp}} - x_i^{\mathrm{cal}})^2\right]^{1/2} \tag{2}$$

where N is the number of experimental points.

Distribution Coefficients for Furfural and Acetic Acid in the Water−o-Nitrotoluene System. The distribution coefficient of furfural is defined as the equilibrium ratio of the mass fraction of furfural in the solvent phase to the mass fraction of furfural in the aqueous phase:

$$K = \frac{x_{\mathrm{s_e}}}{x_{\mathrm{a_e}}} \tag{3}$$

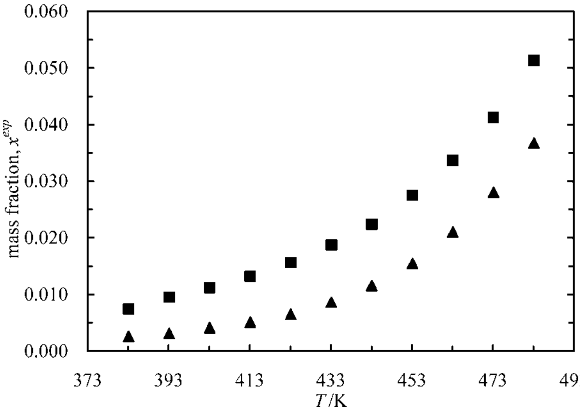

Figure 2. Mass fraction mutual solubilities for water and *o*-nitrotoluene from T = (383.15 to 483.15) K and p = 25.0 bar. ■, solubility of water in *o*-nitrotoluene; ▲, solubility of *o*-nitrotoluene in water.

Table 3. Parameters of Equation 1 for Mass Fraction Mutual Solubilities for Water and *o*-Nitrotoluene

equation parameter	A	B	C	10^4 rmsd	R^2
water solubility in *o*-nitrotoluene	−8.5484	$2.3492 \cdot 10^{-3}$	$1.8981 \cdot 10^{-5}$	2.98	0.9989
o-nitrotoluene solubility in water	−7.4134	$-1.4642 \cdot 10^{-2}$	$4.8039 \cdot 10^{-5}$	4.18	0.9994

Table 4. Distribution Coefficients for Acetic Acid and Furfural in Water−*o*-Nitrotoluene System from $T = (303.15$ to $483.15)$ K and $p = 25.0$ bar

furfural			acetic acid		
T/K	K^{exp}	$[10^2(K^{\mathrm{exp}} - K^{\mathrm{cal}})]/K^{\mathrm{exp}}$	T/K	K'^{exp}	$[10^2(K'^{\mathrm{exp}} - K'^{\mathrm{cal}})]/K'^{\mathrm{exp}}$
303.15	9.14	1.49	303.15	17.55	−1.84
323.15	8.30	−1.10	323.15	14.61	−2.46
343.15	7.82	−0.80	343.15	12.79	−0.07
363.15	7.39	−0.94	363.15	11.46	2.84
383.15	7.20	1.34	383.15	10.14	3.08
403.15	6.81	0.36	403.15	9.13	3.80
423.15	6.46	−0.95	423.15	8.02	1.09
443.15	6.33	0.66	443.15	7.26	0.40
463.15	6.00	−1.26	463.15	6.42	−3.49
483.15	5.96	1.18	483.15	5.98	−2.81

The distribution coefficient of acetic acid is defined as the equilibrium ratio of the mass fraction of acetic acid in the aqueous phase to the mass fraction of acetic acid in the solvent phase:

$$K' = \frac{x_{a_e}}{x_{s_e}} \tag{4}$$

where the subscripts *s* and *a* refer to the solvent and aqueous phases, respectively.

The distribution coefficients for furfural and acetic acid in the water−*o*-nitrotoluene system at different temperatures are presented in Table 4 and more visually given in Figures 3 and 4, respectively, where T is the absolute temperature and K^{exp} and K^{cal} are the experimental and calculated values of the distribution coefficient. From Table 4, it can be seen that the distribution coefficient is temperature-dependent. According to the liquid−liquid phase equilibrium theory, the temperature dependence of the distribution coefficient K can be correlated by the van't Hoff equation deduced from the extraction equilibrium theory:[16]

$$K = K_0 e^{-\Delta H/RT} \tag{5}$$

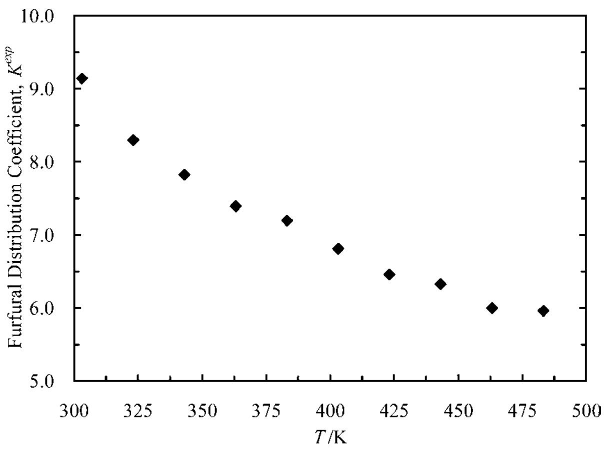

Figure 3. Distribution coefficients of furfural in the water−*o*-nitrotoluene system from $T = (303.15$ to $483.15)$ K, $p = 25.0$ bar.

where ΔH is the enthalpy change (J·mol^{-1}) for the transfer of furfural from water to *o*-nitrotoluene or the enthalpy change (J·mol^{-1}) for the transfer of acetic acid from *o*-nitrotoluene to water and K_0 is the extrapolated value for K at infinite temperature. The experimental distribution coefficient values were fitted with eq 5 by the least-squares method. The values of parameters K_0 and ΔH for furfural and acetic acid are listed in Table 5 together with the rmsd.

The results show that the furfural distribution coefficient decreases with increasing temperature. This indicates that the mass transfer of furfural from water to *o*-nitrotoluene is an exothermic process. The acetic acid distribution coefficient decreases with increasing temperature, but it is evident that the mass transfer of acetic acid from water to *o*-nitrotoluene is an endothermic process. The experimental results fit the van't Hoff equation well which indicates that the lower mutual solubilities for the water−*o*-nitrotoluene system in the experimental temperature range has little effect on the distribution coefficient. The furfural distribution coefficient for *o*-nitrotoluene was 5.96 at 483.15 K, which is still higher than that of any other stable solvents presented in Table 1 at 303.15 K. The acetic acid distribution coefficient was 5.98 at 483.15 K. This indicates that *o*-nitrotoluene has a high distribution coefficient for furfural and a very low distribution coefficient for acetic acid under the condition necessary for furfural production in solvent extraction process. Considering the high selectivity for furfural and the low selectivity for acetic acid and negligible mutual solubilities with water, *o*-nitrotoluene was considered as a suitable solvent for furfural production in the solvent extraction process. The experimental data and correlation equation in this work can be used as essential design data for the production of furfural in the solvent extraction process.

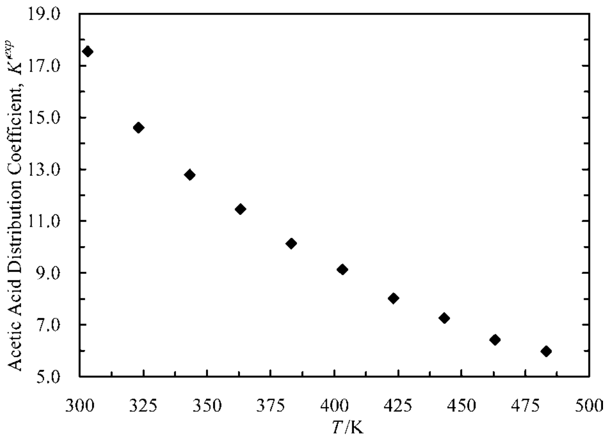

Figure 4. Distribution coefficients of acetic acid in the water−*o*-nitrotoluene system from $T = (303.15$ to $483.15)$ K, $p = 25.0$ bar.

Table 5. Parameters of Equation 5 for Distribution Coefficients of Acetic Acid and Furfural

solvent	K_0	ΔH/J·mol^{-1}	rmsd	R^2
furfural	2.8854	−2867.66	0.079	0.9938
acetic acid	1.0192	−7218.96	0.254	0.9945

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