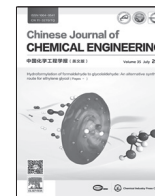




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Purification of carbazole by solvent crystallization under two forced cooling modes

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ABSTRACT

To identify the effect of solvents and anthracene on the purification of carbazole, the solvent crystallization of carbazole was investigated with xylene, chlorobenzene and tetrachloroethylene (TCE) as solvents under two forced circulation cooling (FCC) modes. The co-crystalline experimental data were obtained from runs carried out at different anthracene levels between 1% (mass) and 10% (mass). The results showed that a uniform flake carbazole crystal obtained when using xylene and chlorobenzene under the FCC-1 mode with gradual cooling rate. Nevertheless, fine flake crystals grown under shock cooling of FCC-2 mode. It is beneficial to improving the purity of carbazole with chlorobenzene as solvent under cooling mode of FCC-1. Anthracene could promote the growth of carbazole in solution, and it has a significant influence on the purification of carbazole.

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1. Introduction

Carbazole possesses large conjugated systems and desirable strong intramolecular electron transfer properties. Because of its special structure, carbazole compounds show unique properties, and wide application in photoelectrical materials [1], dyes [2], pharmaceuticals [3], etc. Carbazole and its derivatives have become new organic liquid hydrogen storage materials due to their high hydrogen storage density, and the existence of heteroatoms can effectively reduce the hydrogen storage temperature [4,5]. Carbazole based molecules have important applications in organic solar cells [6]. What's more important is that carbazole compounds are easy to modify, and various functional groups can easily introduce on the rings. With the continuous expansion of its application, higher requirements for purity of carbazole are put forward.

Moreover, carbazole itself is one of coal tar products, and raw materials are easy to get. About 90% carbazole comes from anthracene oil fraction of high temperature coal tar. In order to obtain high purity carbazole, it is necessary to separate carbazole from other components. Solvent cooling crystallization is a useful method for separation and purification of the mixture. Most

researches had devoted to produce the high purity anthracene, and carbazole is believed as a major impurity in anthracene when separate it from coal tar [7]. However, anthracene is the main impurity in carbazole in contrast. Therefore, the key to extraction and purification of carbazole is to separate it from anthracene. Nevertheless, most of the researches focus on the solvent selection and process optimization [8,9], and rarely consider the effect of solvents and role of impurities during the solvent crystallization process of carbazole.

Polyaromatic hydrocarbons with close physical properties and similar molecular structures may form solid solution. The solid solution of anthracene and carbazole would form during melting crystallization since the isomorphism of them [7]. The forms of the solid solution varies with the contents of carbazole and anthracene, from substitutional solid solution to interstitial forms, and 0.1% carbazole is the upper limit for a substitutional solid solution of carbazole in anthracene [10]. Furthermore, the solid solution also formed during solvent crystallization [11]. It is hard to obtain the carbazole with purity higher than 99% (mass) by solvent cooling crystallization. Whether the formation of solid solution between carbazole and anthracene affects the purification of carbazole during solvent crystallization is still unknown.

There are many influence factors during the solvent crystallization, such as the type of solvents used and the existing impurities or additives [12,13]. To obtain ideal crystals, the proper solubility should be in the range of 5–200 mg·ml⁻¹ at room temperature

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[12]. To realize the separation and purification of carbazole from coal tar, the solvent also need to meet the conditions that the solubility of the impurities in it are higher than that of carbazole. The impurities could lead to the change of the degree of supersaturation, the metastable zone width [14], the crystallization kinetics [15], the crystal growth [16,17], and the morphology of the final crystallization products [18,19]. Furthermore, the rates of cooling and stirring are also major factors considered for the desirable product [20], the crystal size [21,22], even the morphology could be changed [23].

The goal of this work is to obtain the detail information of separation and purification of carbazole from anthracene by solvent cooling crystallization under different cooling modes. Gas chromatography (GC), scanning electron microscope (SEM), X-ray diffractometer (XRD), differential scanning calorimeter (DSC) and fluorescence spectrometer were adopted to determine the compositions of mother liquor and crystals, and the morphology of the resulting crystals. It can provide guidance and help for the purification of carbazole during solvent cooling crystallization.

2. Materials and Methods

2.1. Materials

Anthracene and carbazole (analytical reagent, Shanghai Aladdin Biotechnology Co., Ltd) further purified by recrystallization with *N,N*-dimethylformamide and chlorobenzene, respectively. The purities are above 99% (mass). The standards of anthracene (99.4% (mass)), carbazole (99.3% (mass)) used in the experiment were purchased from West Chester, PA, and 9-fluorenone (99% (mass)), internal standard substance, Shanghai Aladdin Biotechnology Co., Ltd). The analytical reagents, chlorobenzene, xylene (a mixture of 10%–15% *o*-xylene, 45%–70% *m*-xylene, and 15%–25% *p*-xylene), and tetrachloroethylene (TCE) were used without further purification. Acetonitrile (HPLC, Dikama Technology) was used to dissolve and dilute the samples for gas chromatography and fluorescence analysis.

2.2. Solvent crystallization of carbazole

The crystallization is controlled by two forced circulation cooling (FCC) mode under the stirring rate of 450 r·min⁻¹ (controlled by a magnetic stirring apparatus). One was controlled by a temperature-controlled water bath with a forced circulating system (FCC-1), and the other was controlled by adding the tap water to the thermostatic water bath and the water flow in and flow out with a constant rate (FCC-2). Saturated solutions were prepared in a beaker by dissolving an appropriate amount of carbazole in xylene at 80 °C with agitation rate of 450 r·min⁻¹. Then, the solutions were filtered with a membrane of 0.45 μm, and transferred to a jacketed glass crystallizer (100 ml), and then cooled to 25/26 °C. The crystals were filtered from the solution quickly with a membrane of 0.45 μm, and then collected and dried at room temperature in air atmosphere more than 24 h. The crystalline behaviors of carbazole in chlorobenzene and TCE were investigated also.

Co-crystallization of carbazole with different ratios of anthracene from 1 to 10% (mass) in xylene, chlorobenzene and TCE was conducted as the same process of pure carbazole.

2.3. Analytical apparatus and procedures

2.3.1. Gas chromatography (GC)

The amounts of anthracene and carbazole were determined with an internal standard curves method by a Shimadzu GC 2014 with a DM-5 capillary column (5% (mass) diphenyl/95% (mass)

dimethyl polysiloxane, 0.32 mm × 30 m × 0.25 μl), and with a hydrogen flame ionization detector (FID). N₂ is the carrier gas with the flow rate of 1 ml·min⁻¹, and the temperature program is the same as Ref. [24]. 9-fluorenone used as the internal standard substance. A series of anthracene and carbazole standard solutions with 9-fluorenone at concentrations of 0.2, 0.4, 0.6, 1.0, and 1.2 mg·ml⁻¹ were prepared with acetonitrile (HPLC pure) as the solvent. The concentration of 9-fluorenone was fixed at 1.2 mg·ml⁻¹. Fig. 1 presents the calibration curves.

2.3.2. X-ray diffraction analysis (XRD)

A Rigaku Ultima IV (Japan) X-ray diffractometer with CuK_α radiation ($\lambda = 0.154056$ nm) was applied to determine the carbazole crystals. About 50 mg samples were tableted for XRD analysis. The voltage was set to 40 kV, and the current was set to 40 mA. The scanning speed was 4 (°)·min⁻¹ over a 2 θ range of 5°–40°. The diffraction peaks were identified by software Jade 6.0.

2.3.3. Scanning electron microscope (SEM)

Morphology of carbazole was determined by a HITACHI TM-3000 scanning electron microscope. The accelerating voltage was set to 5 kV with a vacuum of 0.01 kPa. To improve electrical conductivity, the samples were sputter-coated with a layer of gold before SEM analysis.

2.3.4. Differential scanning calorimeter (DSC)

A NETZSCH STA 449 F5 simultaneous thermal analyzer was adopted to determine the melting points of carbazole crystals. The system was purged with N₂ at a flow rate of 20 ml·min⁻¹ for 30 min. About 10 mg sample were tested from room temperature to 300 °C at a heating rate of 2 °C·min⁻¹.

2.3.5. Fluorescence analysis

A Varian Cary Eclipse (EL05033883) fluorescence spectrometer with a synchronous mode was used, and the constant energy of 1600 cm⁻¹ was adopted to analyze the crystals. The scanning range was 250–400 nm with a scanning rate of 600 nm·min⁻¹. The samples were prepared by dissolving in acetonitrile, and diluted to a concentration of 2 μg·ml⁻¹. The slits were set to 10 nm for both of excitation and emission.

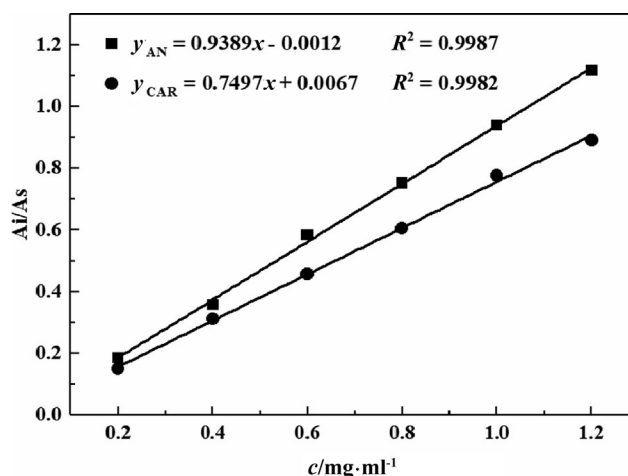


Fig. 1. Calibration curves of anthracene and carbazole with 9-fluorenone as the internal standard by GC.

3. Results and Discussion

Two forced circulation-cooling strategies were employed. Fig. 2 presents the cooling curves from 80 °C to 25 °C. For FCC-1 mode, except the initial stage, the cooling rate decreased gradually during the whole crystallization. Hence, the super-cooling degree is not very high even at the initial stage of crystallization. However, for FCC-2 mode, the cooling rate decreased sharply, especially before 40 °C. Therefore, highly super-cooling degree obtained during the crystalline process by FCC-2 mode.

3.1. Carbazole grow from different solutions

Xylene, chlorobenzene and TCE are commonly used solvents in carbazole refining process. The solubilities of carbazole and anthracene in chlorobenzene, xylene, and TCE are shown in Fig. 3. The solubilities of carbazole and anthracene in different solvents are as follows: chlorobenzene > xylene > TCE.

Fig. 4 presents the powder XRD patterns of carbazole crystals grown from the three solutions under FCC-1 mode. The crystals of carbazole belong to orthorhombic system. The prominent facets identified in the XRD patterns of carbazole are (020), (040), (211), (111), (101), and (210). Solvent is one of the main factors affecting the growth habits of polar organic crystals in solution. It was demonstrated that stereospecific adsorption of the tailor-made solvent at a particular face inhibits its growth, and affects the size and the structure of the growth unit. Nonpolar solvent, TCE could not form the strong interactions, such as hydrogen bonds, with polar solute molecules on the crystal surface, and has little influence on crystal growth. Therefore, what presented are the facets of the crystal itself. The interactions between xylene and carbazole are dominated by π - π^* interactions. Because of the strong electron-withdrawing ability of aromatic rings, the N-H groups of carbazole molecules tend to form hydrogen bonds with a strong electron donor readily. Since the strong electron donating ability of Cl, there are two major forces between chlorobenzene and carbazole, the hydrogen bonds of N-H...Cl and the π - π^* interactions. Hence, the adsorption sites of chlorobenzene on the crystal increased. As a result, the growth of some facets, such as (020), (040), and (211) slowed down further. However, the growth of the facets of (101), (111), and (210) are promoted, and the corresponding diffraction peaks are weakened, and even disappeared.

The SEM images of carbazole crystals obtained in different solvents under FCC-1 mode are shown in Fig. 5. The crystal habit of carbazole produced from different solvents is flakes, and the parti-

cle size decreased as follows: xylene > chlorobenzene > TCE. The crystals grown in xylene and chlorobenzene solution exhibited a well faceted morphology, and showed a uniform size. In addition, the crystals obtained from TCE solution are very fine, due to the low contents of carbazole in it, and it should take a long time to grow up.

The crystals grown from xylene and chlorobenzene present almost the same morphologies and sizes by using FCC-2 mode (Fig. 6), both are fine irregular flake crystals. The burst nucleation occurred since the highly cooling rate was adopted during the crystalline process, and made the resulting crystal particles fine. In solution, molecules are generally in the form of association, the growth of polar organic crystals of large size and high quality has been hindered by growth cessation phenomena [25]. There exists an upper limit for the solution concentration, above which the crystals stop growing since dimer formation adhered on the surface of the crystals. In this work, whether the crystals grown in polar solvent (chlorobenzene) or in nonpolar solvent (xylene) showed similar fine particle when using FCC-2 mode. The results demonstrate that the cooling rate plays the dominate role in the crystalline process. High cooling rate is adverse to the crystal growing up.

3.2. Effect of anthracene on the purification of carbazole

The crystallization of carbazole with different ratios of anthracene from 1% (mass) to 10% (mass) were investigated in TCE, xylene, and chlorobenzene solutions. SEM images of some co-crystals of carbazole and anthracene are also shown in Figs. 5 and 6. The impurity with bigger solubility could promote the growth of crystal [26]. The anthracene in carbazole solutions could promote the crystals growth due to the bigger solubility of anthracene in TCE, xylene, and chlorobenzene compared to carbazole (Fig. 3). For FCC-1 mode, the crystals grow up significantly with the amount of anthracene increased, and the aggregation is weakened. However, there is no difference of the morphology and size among the original carbazole crystal and the co-crystals when using FCC-2 mode.

The solubility selectivity of the solvent is one of the crucial factors during purification by solvent crystallization. Highly solubility selectivity is conducive to gaining high purity product. Table 1 lists the solubility selectivities of anthracene to carbazole ($SS_{An/CAR}$) of chlorobenzene, xylene and TCE from 30 to 80 °C. The $SS_{An/CAR}$ are from 2.44 to 3.47 for chlorobenzene, from 2.18 to 3 for xylene, and from 4.83 to 8.35 for TCE. The $SS_{An/CAR}$ of TCE is the highest among the solvents used, and it is the ideal solvent for purification of carbazole theoretically. However, TCE was not selected for purification of carbazole due to its poor solvent power. The variations of the $SS_{An/CAR}$ of chlorobenzene and xylene are not remarkable in the whole temperature range. Thus the crystalline process is relatively easy to control.

The compositions of mother liquor and the solids after crystallization are listed in Table 2. No anthracene was detected in carbazole crystals when the ratio of anthracene was less than or equal to 5% (mass) by GC. However, when the adding ratio is 6% (mass), the amounts of anthracene in the crystals are 1.65% (mass) for xylene-1, 1.79% (mass) for CB-1, 2.53% (mass) for xylene-2, and 2.18% (mass) for CB-2, respectively. Hence, it has little impact on the purity of carbazole when the ratio of anthracene is less than or equal to 5% (mass) according to the GC results. The amounts of anthracene in mother liquor increased with the adding ratios, and the contents of anthracene varied with the solvent used and the cooling mode applied. For CB-2, it is from 5.47% (mass) to 32.49% (mass), higher than that of xylene-2 (from 5.18% (mass) to 27.13% (mass)) at all anthracene ratios varied from 1% (mass) to 10% (mass). The contents of anthracene in the mother liquor

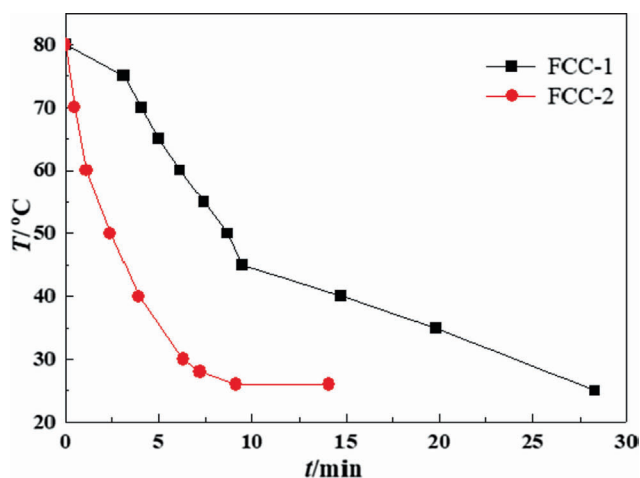


Fig. 2. Two forced circulation cooling strategies.

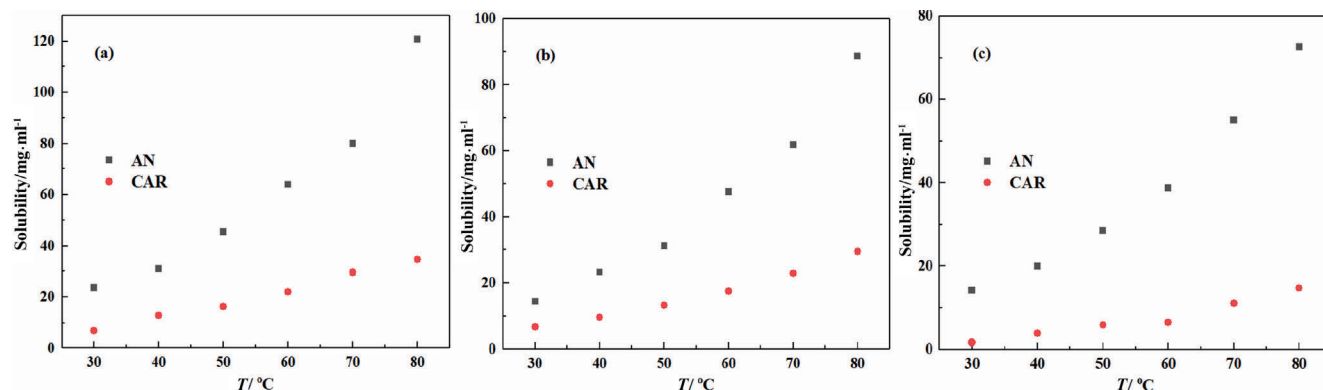


Fig. 3. Solubility data of carbazole and anthracene in different solvents, (a) CB; (b) xylene; (c) TCE.

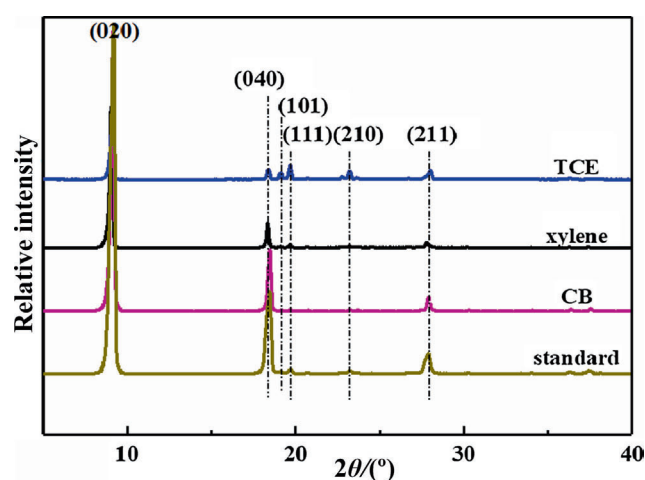


Fig. 4. The XRD patterns of carbazole crystals grown from TCE, xylene, and CB solution under FCC-1 mode.

of xylene-1 is from 6.58% (mass) to 35.28% (mass), and it is from 5.83% (mass) to 32.39% (mass) for CB-1.

The cooling rate has great influence on crystal size, morphology and purity. The burst nucleation occurred as the cooling rate increased, the crystal size decreased [27]. In addition, the crystal will wrap a large amount of impurities in it during the rapidly crystalline process [28,29], and the purity declined [30]. Therefore, when the crystal growing from the same solution, more anthracene retained in the mother liquor and little anthracene entered into the carbazole crystals under FCC-1 mode. Hence, the resultant crystals have a higher purity. Additionally, the crystals grown from chlorobenzene solution had a higher purity when the anthracene ratios surpassed 5% (mass). It ascribes to the $SS_{AN/CAR}$ of chlorobenzene are higher than that of xylene (Table 1). Hence, to separate and purify carbazole from coal tar, the FCC-1 mode with a gradual cooling rate, and with chlorobenzene as solvent is a better choice.

Synchronous fluorescence is a good method for determining multi-components, especially for PAHs [31,32]. The trace amount of anthracene might change the photoelectric properties of car-

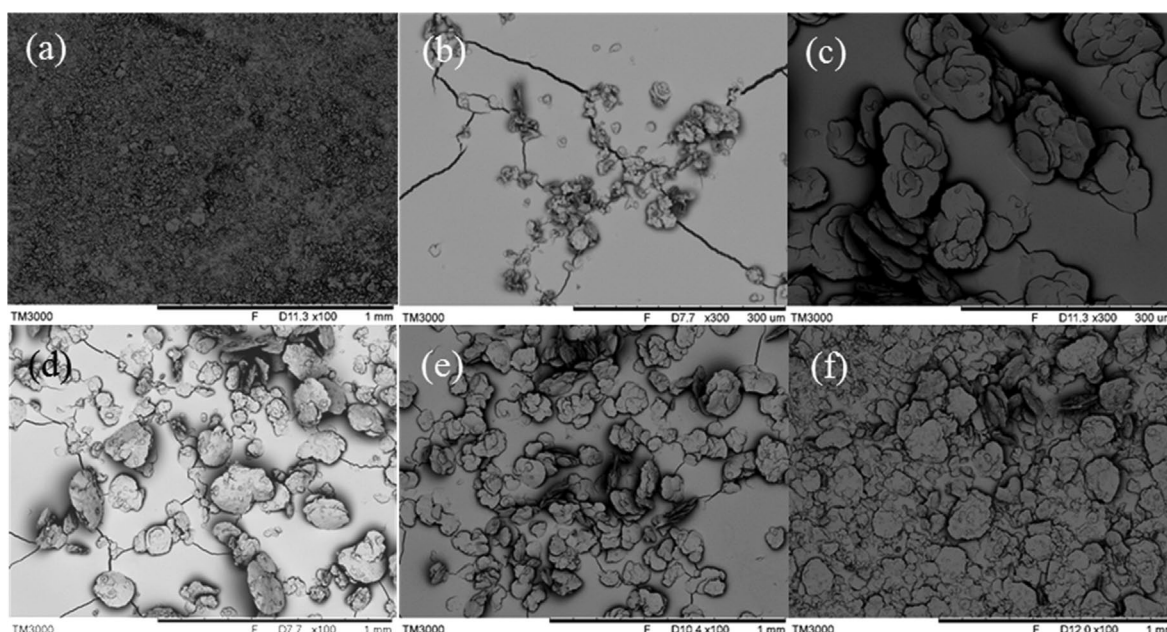


Fig. 5. The SEM images of carbazole crystals obtained in different solvents under FCC-1 mode, (a) TCE-CAR-1(100×); (b) TCE-CAR-1(300×); (c) TCE-5 % (mass) AN-1(300×); (d) xylene-CAR-1(100×); (e) CB-CAR-1(100×); (f) CB-10 % (mass) AN-1 (100×).

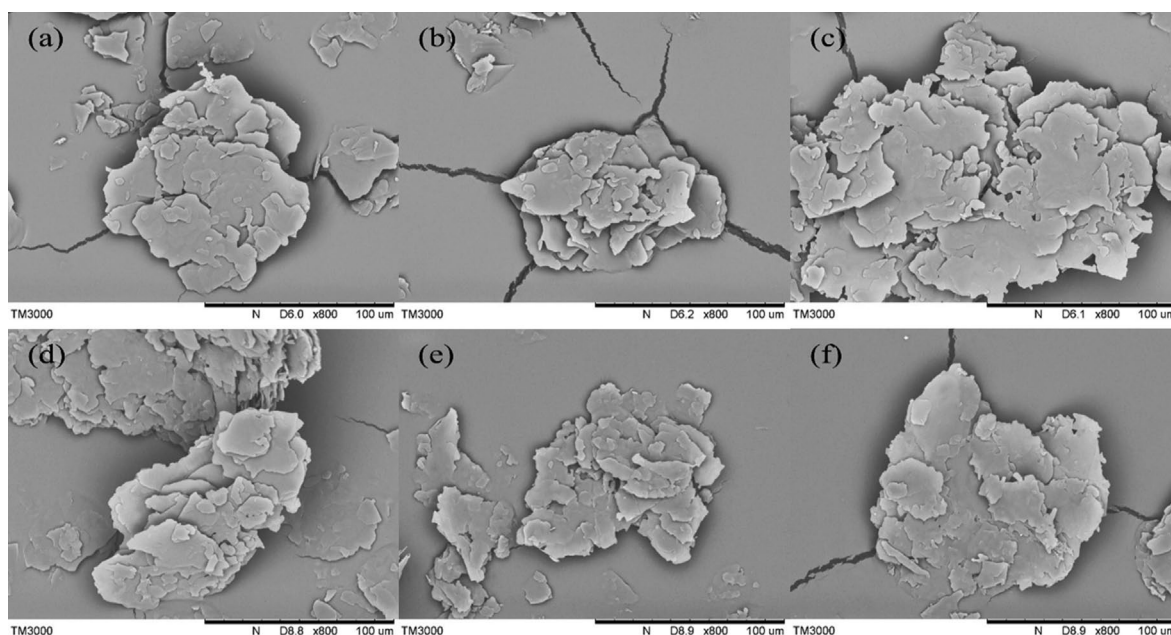


Fig. 6. The SEM images of carbazole crystals (800x) obtained in different solvents under FCC-2 mode, (a) CB-CAR-2; (b) CB-5 % (mass) AN-2; (c) CB-10% (mass) AN-2; (d) xylene-CAR-2; (e) xylene-5% (mass) AN-2; (f) xylene-10% (mass) AN-2.

Table 1

The solubility selectivity of anthracene to carbazole of chlorobenzene, xylene and TCE

Solvents	$SS_{AN/CAR}$					
	30 °C	40 °C	50 °C	60 °C	70 °C	80 °C
CB	3.47	2.44	2.81	2.9	2.96	3.46
xylene	2.18	2.43	2.36	2.7	2.69	3
TCE	8.35	5.13	4.83	5.95	4.96	4.94

Note: the solubility selectivity ($SS_{AN/CAR}$) is defined as the solubility of anthracene over the solubility of carbazole.

Table 2

Compositions of mother liquor and crystal derived from the solvent co-crystallization using chlorobenzene and xylene as the solvent

Solvent	Sample AN/% (mass)	Composition of mother liquor/% (mass)		Composition of crystal/% (mass)	
		X_{AN}	X_{CAR}	X_{AN}	X_{CAR}
xylene-1	1	6.58	93.42	0	100
	2	11.58	88.42	0	100
	5	22.96	77.04	0	100
	6	26.48	73.52	1.65	98.35
	8	33.71	66.29	2.49	97.51
	10	35.28	64.72	2.95	97.05
CB-1	1	5.83	94.17	0	100
	2	9.75	90.25	0	100
	5	21.39	78.61	0	100
	6	23.25	76.75	1.79	98.21
	8	28	72	2.26	97.74
	10	32.39	67.61	2.93	97.07
xylene-2	1	5.18	94.82	0	100
	2	8.37	91.63	0	100
	5	16.92	83.08	0	100
	6	21.97	78.03	2.53	97.47
	8	23.07	76.93	3.03	96.97
	10	27.13	72.87	3.73	96.27
CB-2	1	5.47	94.53	0	100
	2	9.70	90.30	0	100
	5	21.17	78.83	0	100
	6	22.55	77.45	2.18	97.82
	8	25.58	74.42	2.62	97.38
	10	32.49	67.51	3.27	96.73

Note: CB-1, crystallization with chlorobenzene under FCC-1 mode; CB-2, crystallization with chlorobenzene under FCC-2 mode; X_{AN} , mass fraction of anthracene; X_{CAR} , mass fraction of carbazole; xylene-1, crystallization with xylene under FCC-1 mode; xylene-2, crystallization with xylene under FCC-2 mode.

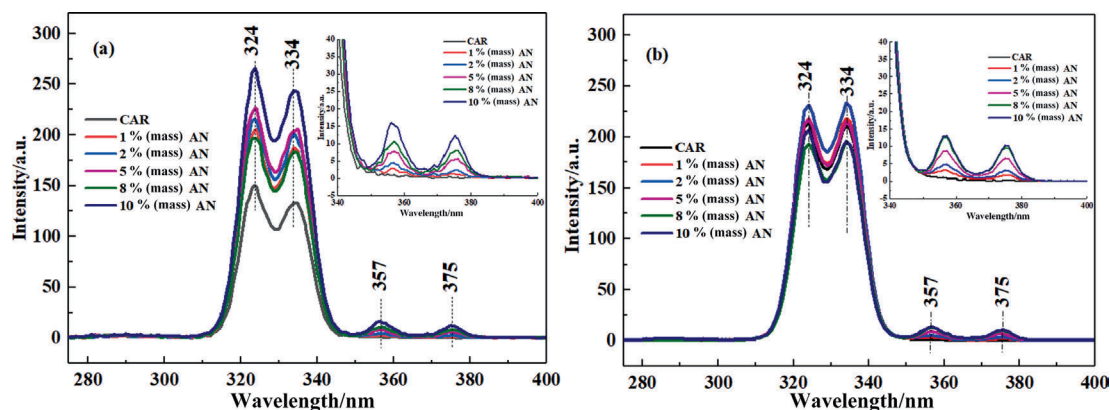


Fig. 7. The synchronous fluorescence spectra of crystals in different solvents, (a) xylene-2; (b) CB-2.

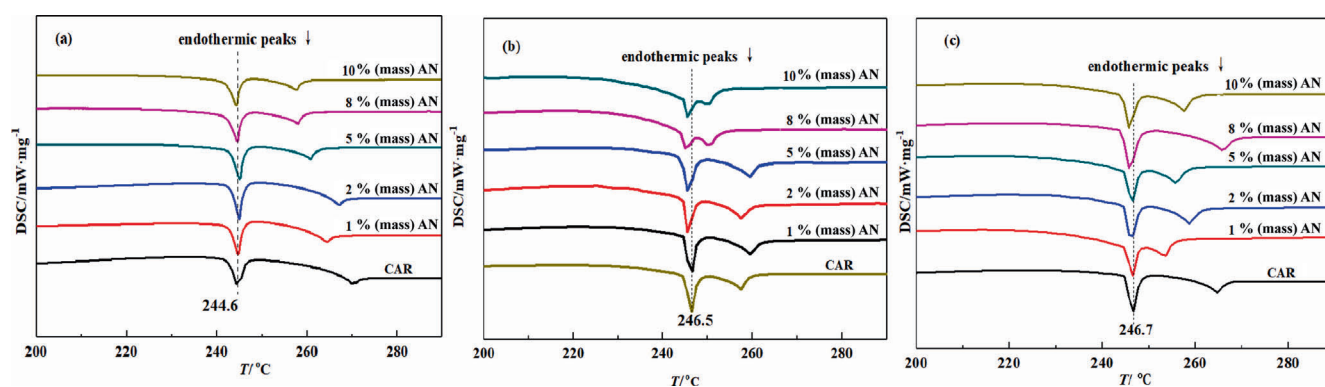


Fig. 8. The DSC curves of the co-crystals grown from different solvents, (a) xylene-1; (b) xylene-2; (c) CB-2.

bazole. Therefore, the synchronous fluorescence analysis also adopted to determine the resulting crystals. Synchronous scanning with constant energy of 1600 cm^{-1} could identify carbazole from anthracene, and no spectra overlap is found (Fig. 7). Except the pure carbazole, other co-crystals of carbazole and anthracene show both of the fluorescence peaks of carbazole (324 nm and 334 nm) and anthracene (357 nm and 375 nm), and the intensity of anthracene peaks enhanced with the ratios increased, especially when it exceeded 5% (mass). Furthermore, these peaks of anthracene are more obvious when using xylene as solvent.

Fig. 8 presents the DSC curves of the co-crystals of carbazole with different ratios of anthracene grown from xylene and chlorobenzene solutions. The DSC curves showed two endothermic peaks, one is corresponding to the melting point of carbazole, and the other is the evaporating temperature. The melting points of co-crystals grown in xylene with the FCC-1 mode are near $244.6\text{ }^{\circ}\text{C}$ (Fig. 8a) as the same as the carbazole. For xylene-2 (Fig. 8b), the peaks corresponding to the melting point of carbazole are at around $246.5\text{ }^{\circ}\text{C}$. The melting points of co-crystals show a shift to lower temperature when the ratios of anthracene are higher than 2% (mass). Therefore, it is unfavorable to the solid solution formation with shock cooling mode of FCC-2. There are notable differences between xylene-2 and CB-2 (Fig. 8c), the melting point of co-crystals grown from chlorobenzene solution have an obvious shift to lower temperature when the ratios of anthracene exceeded 5% (mass). In the case of 0.0001% (mass)–0.1% (mass) anthracene for a substitutional solid solution, an interstitial solid solution began to form when the anthracene surpassed 10% (mass) [10]. Therefore, this might attribute to the higher purity of carbazole

grown under FCC-1 mode, and the substitutional solid solution of anthracene in carbazole formed.

4. Conclusions

The effect of solvents and anthracene on the purification of carbazole were investigated by solvent crystallization under two forced circulation cooling modes. The conclusions of this study are summarized as follows.

- (1) Forced circulation-cooling mode with a gradual cooling rate is beneficial to the growth of carbazole in solution with a uniform size.
- (2) Solvent has a notable influence on the purification of carbazole. Chlorobenzene would be a better choice for the purification of carbazole and the resulting crystals have a higher crystallinity and purity.
- (3) The growth of carbazole crystal is promoted when the ratio of anthracene is below or equal to 10% (mass), but it has an adverse effect on the purity of carbazole. The influence of anthracene on the purification of carbazole became remarkable when the amounts of anthracene surpassed 5% (mass).

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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