

# Crystallization Behavior of Polyvinyl Alcohol With Inorganic Nucleating Agent Talc and Regulation Mechanism Analysis

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## Research Article

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## Abstract

Polyvinyl alcohol (PVA) decompose before melting, making it difficult for melt processing. This phenomenon limits the applications of PVA. Therefore, to widen the application of PVA, in the present work, the PVA films have been prepared by the flow casting method. And talc added as an inorganic nucleating agent to improve the crystallinity of PVA. The effects of talc on the crystallization behavior of PVA are tested by differential scanning calorimetry (DSC), Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and successive self-nucleation annealing (SSA). The crystallization kinetics behavior of talc was further studied by Mo Zhishen equation and Avrami equation. The results show that the addition of talc to the PVA film promotes its crystallization, and as the content of the nucleating agent increases, the crystallinity tends to increase, and the thickness of the wafer becomes uniform due to the increase of nucleation points. These results show that talc regulates the crystallization of PVA, improves the crystallinity and crystallization rate of PVA, and has a heterogeneous nucleation effect. PVA itself exhibits hydrophilicity because it contains large number of hydroxyl groups, and has good physical and chemical properties. In addition, the mechanical properties and water resistance have been further improved by improving the crystallinity of PVA, and the practicability of PVA film in actual production was improved.

## 1. Introduction

Polyvinyl alcohol (PVA) is a linear synthetic polymer produced by partial or complete hydrolysis of polyvinyl acetate to remove the acetate groups. Because the molecular chain contains a large number of hydroxyl functional groups, it has high hydrophilicity and biocompatibility. It is a kind of polyhydroxy polymer with excellent comprehensive performance[1, 2]. In recent times, due to the attention paid to environmental issues, biodegradable and environmentally friendly materials have attracted the attention of researchers[3-7]. PVA has been extensively studied as a material with excellent properties and is widely used in industrial, commercial, medical and food applications, including paper sheets, surgical threads, resins and food packaging[8-11]. However, pure PVA film has poor water resistance and mechanical properties[12]. The crystallization behavior and crystal structure of polymers affect their physical and mechanical properties, which is a challenge for studying the properties of polymers[13, 14].

Crystallization rate of crystalline polymers varies greatly due to their different molecular structures and crystallization conditions[15, 16]. Polymer segments are easily affected by small molecular compounds or other impurities during crystallization, resulting in heterogeneous nucleation. The crystallinity can be easily improved by adjusting the nucleating agent used. Effective nucleating agent provides an active surface for polymer adsorption, with higher crystallinity and more uniform crystal structure[17-19]. The physical and mechanical properties of polymer materials are affected by crystallinity to a certain extent, such as hardness, transparency, tensile strength and solvent resistance. Therefore, crystallinity is an important factor for polymer materials[20, 21].

There are many hydroxyl groups in the PVA chain, leading to the formation of hydrogen bonds between molecules and within molecules, which affects the motion of the molecular chain and further affects the crystallization of PVA[22]. On the other hand, due to its multi-hydroxyl and strong hydrogen bonds, the melting temperature is higher than its decomposition temperature, that is, PVA decomposes before melting, and the thermoplastic processing window is narrow[23]. In order to solve these problems, a large number of studies have been conducted on the structural properties and crystallization behavior of PVA and PVA composites in the past decades[24-28]. -OH as PVA side gene is smaller and similar to hydrogen, so other substances can enter the crystallization point without causing stress[24]. Various plasticizers, inorganic nucleating agents and nanofillers are often added to improve the crystallinity of PVA and affect its physical and chemical properties[29]. Such as graphene[19, 30], carbon nanotubes[31-33], MMT[34, 35], SiO<sub>2</sub>[36], talc[37] and CaCO<sub>3</sub>[38]. Qiu et al. found that the size of talc affected the mechanical properties of PVA. They found that the smaller the size of talc was, the better were the mechanical properties of the composites were[37]. However, few people studied the effect of talc content on the crystallinity, crystallization kinetics and wafer thickness of PVA.

In this study, the effect of talc on the crystallization behavior of PVA and the regulation of the crystallization properties of PVA films were tested by controlling the talc content in PVA films. The results showed that talc was added to PVA film to change the crystallization behavior of molecules, which had a heterogeneous nucleation effect and promoted the crystallization of PVA. The crystallinity increased with the increase of nucleating agent content. And due to the increase of nucleation point, the wafer thickness becomes uniform.

## 2. Experiments

### 2.1. Materials

PVA (117) with a molecular weight of 83 000 g/mol and the alcoholysis degree of 98%- 99% was purchased from Kuraray Co., Ltd. (Japan). Talc powder (3000 mesh) was obtained from Sigma-Aldrich company. All of these materials were directly used without further purification.

### 2.2 Preparation of PVA films with different talc contents

A certain proportion of talc powder, PVA and deionized water were mixed in three flasks, and the solid content of talc was 0 %, 3 %, 6 % and 9 %. After stirring at 90 °C for 2 h in a water bath, the obtained mixed solution was poured into a beaker and placed in a numerical control ultrasonic cleaner (TYD-TG4 - 90 KW, Taiyuanda) for defoaming. Mixed solution after defoaming was spread on smooth glass. The film was dried at room temperature for 48 h, and then removed on the glass plate and sealed for preservation. The experimental process is shown in Fig 1.

### 2.3. Characterization

To determine the relative crystallinity, the Q2500 differential scanning calorimeter (TA Company of the United States) was used. The sample was heated to 235 °C at a rate of 10 °C / min and kept at constant temperature for 3 min to eliminate the thermal history under N<sub>2</sub> atmosphere, and then cooled to 30 °C at a rate of 10 °C / min to obtain the crystallization curve. The sample weight was 3-5 mg. The relative crystallinity was calculated using formula (1):

$$X_C = \frac{\Delta H_f}{\Delta H_f^0} \quad (1)$$

X<sub>C</sub> represents the relative crystallinity. ΔH<sub>f</sub> represents the melting enthalpy of the sample, J/g. ΔH<sub>f</sub><sup>0</sup> represents the standard melting enthalpy of PVA.

SSA technology was used to characterize semi-crystalline polymers. Semicrystalline polymer can cause molecular segregation during cooling crystallization from melt using during DSC measurements. After thermal adjustment, the final DSC heating curve reveals the melting point distribution induced by SSA treatment. This is due to the heterogeneous nature of the chain structure of the analyzed polymer. Under N<sub>2</sub> atmosphere, a certain amount of the samples was heated to above 235 °C for 3 min to eliminate the thermal history, and then cooled to 40 °C. Then heated up again to 225 °C for 5 min, cool down to 40 °C. Heated up to 215 °C for 5 min, cooled down to 40 °C. The above steps were repeated until the temperature reaches the end of the crystallization temperature, the temperature rises again to above the melting point, and the obtained curve is the SSA curve. X-ray diffraction (XRD) measurements were performed using a Bruker D8 Advance device (40KV, 30 mA, Bruker Co., Germany) with Cu irradiation at a scanning rate of 10°/min in the 2θ range of 5° - 40°.

### 3. Results And Discussion

#### 3.1 DSC analysis

Fig 2 ( a ) and ( b ) shows the SEM images of composite films with different talc content. The images indicate good dispersion and uniform distribution of the talc in the composites. The DSC crystallization curves of PVA films with different talc contents are shown in Fig 2 ( c ). With the increase of talc content, the initial crystallization temperature and crystallization peak temperature move to high temperature direction, and the crystallization peak enthalpy of samples with talc content more than 3 % is significantly higher than that of pure PVA samples. This indicates that talc has exhibited heterogeneous nucleation effect in PVA film and increased its crystallization temperature. With the increase of talc content, more nucleation sites for PVA crystallization are provided, which promotes the increase of crystallinity and crystallization temperature. Fig 2 ( d ) shows the crystallization data of PVA films with different talc content. It can be seen that the crystallinity and crystallization peak temperature of PVA film with 3 % talc has decreased slightly, while the crystallinity and crystallization peak temperature increased after adding 6 % and 9 % talc. The initial crystallization temperature T<sub>c-onset</sub> increases with talc content. This is because the addition of talc forms the nucleation point for PVA crystallization, reduces the driving force of nucleation, and the spherulites are easier to form, so the crystallinity increases.

#### 3.2 Non-isothermal crystallization analysis

The DSC crystallization curves at different cooling rates are shown in Fig 3. It can be seen that the non-isothermal peak of PVA films with different talc content moves to lower temperature with the increase of cooling rate, and the peak width increases gradually. This is because when the cooling rate is relatively low, the activity of the PVA molecular chain is relatively strong, and there is enough time for the accumulation and self-arrangement of the molecular chains. If the cooling rate is high, the mobility of the polymer molecular chain will decrease, the accumulation and self-arrangement of the molecular chain cannot keep up with the temperature change, and the molecular chain does not have enough time to form a crystal structure, which will broaden the crystallization peak[39]. As shown for the composite with 9% talc sample in Fig 3(d), due to the crystallization and cooling process there is a lag period, so when the cooling rate increases, the lag period will increase. Therefore, the crystallization peak shifts to low temperature on the DSC curves.

The relationship between crystallinity and temperature of PVA films with different talc content under non-isothermal crystallization and the relationship between lgΦ and lgt are shown in Fig 4. When the relative crystallinity is the same, the larger the cooling rate is, the smaller the corresponding temperature is, and the more time is needed. It further shows that the higher the cooling rate, the lower the temperature is needed to reach the same crystallinity, and the more imperfect the crystallization is. Longer the time required to reach the same crystallinity, the smaller the crystallization rate. Through longitudinal comparison, it is found that under the conditions of the same relative crystallinity and cooling rate, the larger the amount of talc, the higher the temperature corresponding to the PVA film, and the shorter the time required. At this time, the activity of the PVA molecular chain is higher, and the crystallization starts at a higher temperature, and the crystallization rate is higher.

The crystallization parameters obtained by fitting the Mozhishen equation are listed in Table 1. Combined with Fig 4, it can be found that lgΦ vs lgt curves have a good linear relationship, indicating that Mo Zhishen equation can better describe the non-isothermal crystallization process of PVA films with different talc content. The value of α is less than 1 in pure PVA film. With the increase of talc content, α increases and becomes greater than 1, indicating that the addition of talc makes the Avrami index greater than the Ozawa index. When PVA films with different talc content reaches reached the same crystallization percentage, F(t) has increased and the crystallization rate decreased with the increase of talc content. This may be because talc acts as a nucleation point to promote crystallization during the crystallization process, and it accelerates the nucleation rate. However, with the increase of talc content, due to the increase of nucleation point, crystallinity and the number of spherulites, the growth rate of spherulites is affected. The resistance of macromolecules to migrate to the surface of PVA crystal increases, so the crystallization rate decreases.

Table 1. Non-isothermal crystallization parameters of PVA films with different talc content

Talc content	X(t)	$\alpha$	F(t)	$R^2$
0%	10%	0.97	14.45	0.99
	40%	0.99	16.59	0.99
	70%	1.00	18.62	0.99
3%	10%	0.91	14.79	0.99
	40%	0.99	18.19	0.99
	70%	1.04	21.87	0.98
6%	10%	1.14	14.79	0.97
	40%	1.17	19.05	0.99
	70%	1.11	21.87	0.98
9%	10%	1.14	15.13	0.99
	40%	1.20	19.05	0.99
	70%	1.19	21.87	0.99

### 3.3 Isothermal crystallization analysis

Fig 5 shows the isothermal crystallization DSC curves of PVA film with different talc content. With the increase of the crystallization temperature ( $T_c$ ), the crystallization peak of the isothermal crystallization curve broadens, and the enthalpy of crystallization fusion decreases. It shows that the time to complete the crystallization becomes longer, and the crystallinity decreases at the same time. This is because when  $T_c$  is low in heterogeneous nucleation, the supercooling degree increases, the driving force of crystallization increases, the crystallization occurs quickly, and the crystallinity increases. When the  $T_c$  is higher, the supercooling degree is lower, and the mobility of the PVA molecular chain segment increases, which is easy to be discharged into the crystal lattice and easy to escape, so the crystallization time is longer. It can be seen from Fig 5 that with the increase of talc content in PVA, the crystallization rate and  $T_c$  increase. It indicates that the addition of talc has enhanced the heterogeneous nucleation of PVA, and the molecular chain could be crystallized at a higher temperature, thus enhancing its crystallization ability.

Fig 6 shows the curves of relative crystallinity of PVA films with different talc content and Avrami equation fitting curves. As the crystallization temperature increases, the crystallization time of the PVA film increases. There is a critical  $T_{c\text{-iso}}$  with the fastest crystallization rate. Crystallization rate is dominated by crystal nucleation below this critical temperature. Above this critical temperature, the crystal growth rate is the dominant mechanism due to the increase in molecular mobility. The lines in the Avrami equation fitting curve are almost parallel, indicating that the fitting effect is good.

Table 2 presents the isothermal crystallization parameters of pure PVA film and PVA/talc film. With the increase of  $T_c$ , the  $t_{1/2}$  of PVA films with different talc content increases, and the K value decreases with the increase of  $T_c$ . It shows that the crystallization rate becomes slower at higher  $T_c$ . The Avrami index n of pure PVA film varied from 2.21 to 3.05. When the content of talc in PVA increases, the value of  $t_{1/2}$  becomes smaller at a similar  $T_c$ , indicating that the crystallization rate of the sample is accelerated due to the heterogeneous nucleation of talc. The K value represents the crystallization rate. The larger the K value is, the faster the crystallization rate is. In the vicinity of the similar  $T_c$ , the K value increases with the increase of talc content, indicating that the crystallization rate is accelerated, and the addition of talc promotes the nucleation of PVA. With the increase of talc content in PVA, the n value has followed a decreasing trend and is less than 3. It shows that the nucleation method of the sample has changed from a biased spherical three-dimensional growth to a two-dimensional and three-dimensional mixed growth, and the degree of crystal perfection has decreased. However, the variation range of n value is not large, indicating that the addition of talc has little effect on the growth mode of PVA crystal.

Table 2. Isothermal crystallization parameters of pure PVA film and PVA/talc

Talc content	Tc	n	K	R <sup>2</sup>	t <sub>1/2</sub>
0%	196	2.21	40.73	0.99	0.25
	199	2.47	13.8	0.99	0.3
	204	3.05	1.34	0.99	0.8
3%	204	2.4	8.709	0.99	0.35
	206	2.32	6.025	0.99	0.39
	210	2.95	5.754	0.99	1.59
6%	203	1.77	3.467	0.99	0.4
	207	2.07	1.548	0.99	0.53
	209	2.25	4.073	0.99	1.6
9%	203	2.67	30.9	0.99	0.24
	205	2.41	2.39	0.99	0.6
	209	2.51	2.63	0.99	1.28

### 3.4 SSA analysis

Fig 7 shows the SSA fitting curves of pure PVA and PVA/talc films and the percentages of different wafer thicknesses with different amounts of talc. It can be seen from Fig 7 ( a-d ) that the number of endothermic peaks have not changed with the increase of talc content, but the height difference between the peaks decreased. Table 3 shows the wafer thickness data of pure PVA film and PVA/talc film at each melting temperature. According to the comprehensive analysis, compared with PVA with talc, the wafer thickness of pure PVA is distributed between 3 nm and 8 nm. When the addition amount of talc has exceeded 3%, the maximum wafer thickness decreased. It can be seen from Fig 7 ( a-d ) that with the addition of talc, the proportion of each melting peak becomes uniform, which proves that due to the heterogeneous nucleation effect of talc and the increase of crystallization rate, the nucleation growth of each wafer thickness increases at the same rate. After adding talc, the PVA film has a smaller wafer thickness change range, and as the talc content in PVA increases, the proportion of thinner wafers gradually increases, and the maximum wafer thickness also gradually decreases. This shows that the thickness of PVA film becomes uniform at different melting temperatures after adding talc, and the trend becomes more and more obvious with the increase of talc content. The percentage of PVA film with different content of talc with different wafer thickness is shown in Fig 7(e). This phenomenon is because when talc is not added, the crystallization of PVA film is mainly homogeneous nucleation. Nucleation point formation is closely related to temperature. Crystal growth is more complete at higher melting temperature, so the content of thicker wafer is higher. The addition of talc has played the role of heterogeneous nucleation, which in turn has increased the nucleation point of PVA film crystallization and has not changed with temperature. As a result, the PVA molecular chain has more nucleation sites to attach when it is orderly arranged into the lattice, and all nucleation sites have grown together. The crystals interact with each other and do not have enough time to improve the crystal, so the proportion of thinner wafer thickness increases.

Table 3. Wafer thickness of PVA film with different talc content at each melting temperature

Sample	Temperature (°C)	Wafer thickness (nm)	Sample	Temperature (°C)	Wafer thickness (nm)	Sample	Temperature (°C)	Wafer thickness (nm)	Sample	Temperature (°C)	Wafer thickness (nm)
Talc	197.00	2.90	3%	201.51	3.23	6%	197.40	2.93	9%	198.73	3.02
	206.20	3.66		206.29	3.67		202.11	3.28		203.53	3.40
	208.65	3.93		210.95	4.23		206.86	3.73		208.38	3.90
	216.51	5.18		215.79	5.03		211.78	4.35		213.52	4.62
	218.91	5.73		220.81	6.26		217.02	5.29		218.87	5.72
	225.25	7.98		225.71	8.21		222.40	6.78		224.40	1.

### 4. Conclusions

In this study, the crystallization behavior of PVA films with different talc content have been studied. The results indicate that the addition of talc has increased the nucleation point of PVA, and the heterogeneous nucleation has increased the crystallinity. Non-isothermal crystallization analysis has shown that with the increase of talc content in PVA, the nucleation point in PVA has increased, the number of spherulites increased, and the total crystallization rate slowed down. Isothermal crystallization analysis has shown that with the increase of talc content in PVA, the crystal growth mode of the sample has tended to be two-dimensional from three-dimensional. SSA analysis has indicated that with the increase of talc content in PVA, the proportion of thinner wafers has increased, and the difference of wafer thickness was also smaller. In summary, the addition of Talc regulated the crystallization behavior of PVA and promoted its crystallization due to heterogeneous nucleation. The crystallinity increased with the increase of nucleating agent content.

## Declarations

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### Conflict of Interest

The authors declare no conflict of interest.

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## Figures

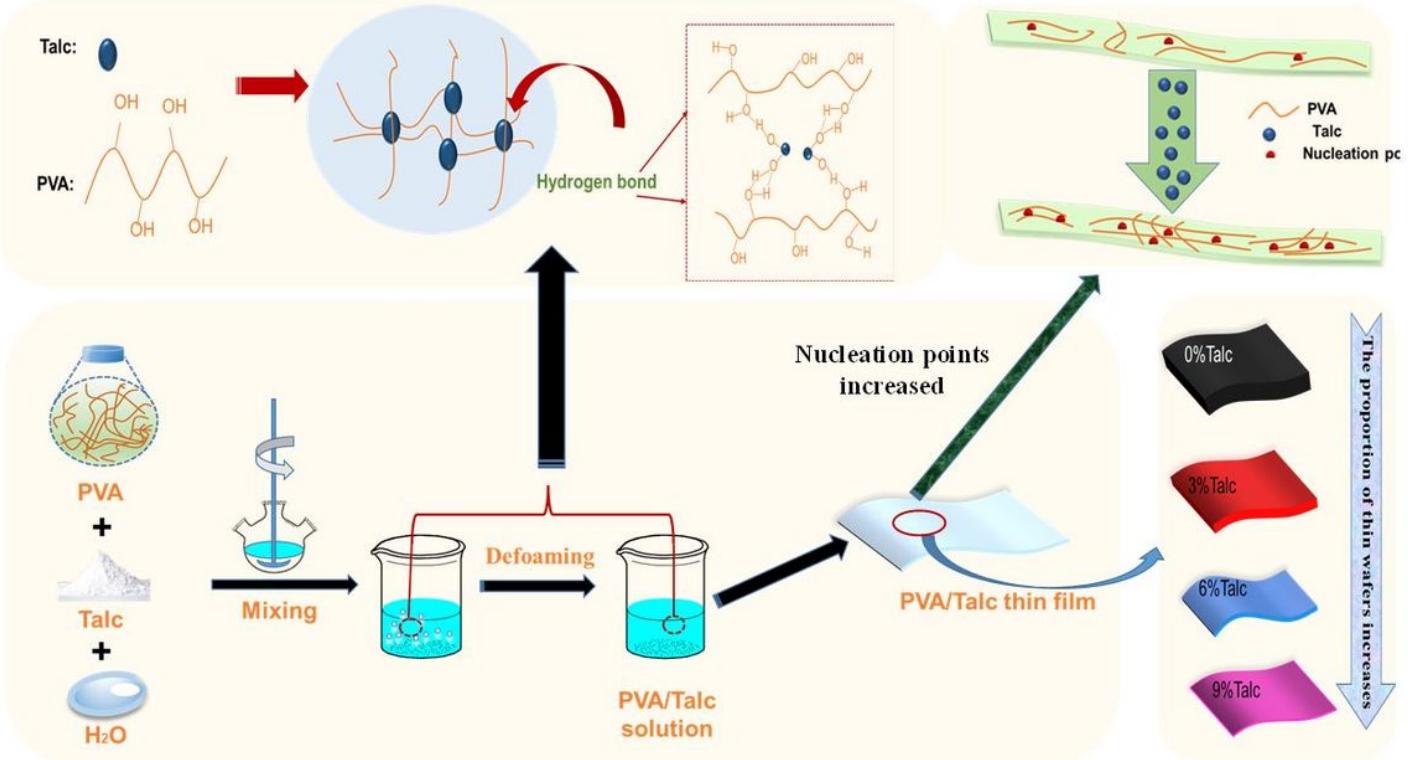


Figure 1

Diagram of preparation process of PVA films with different talc contents

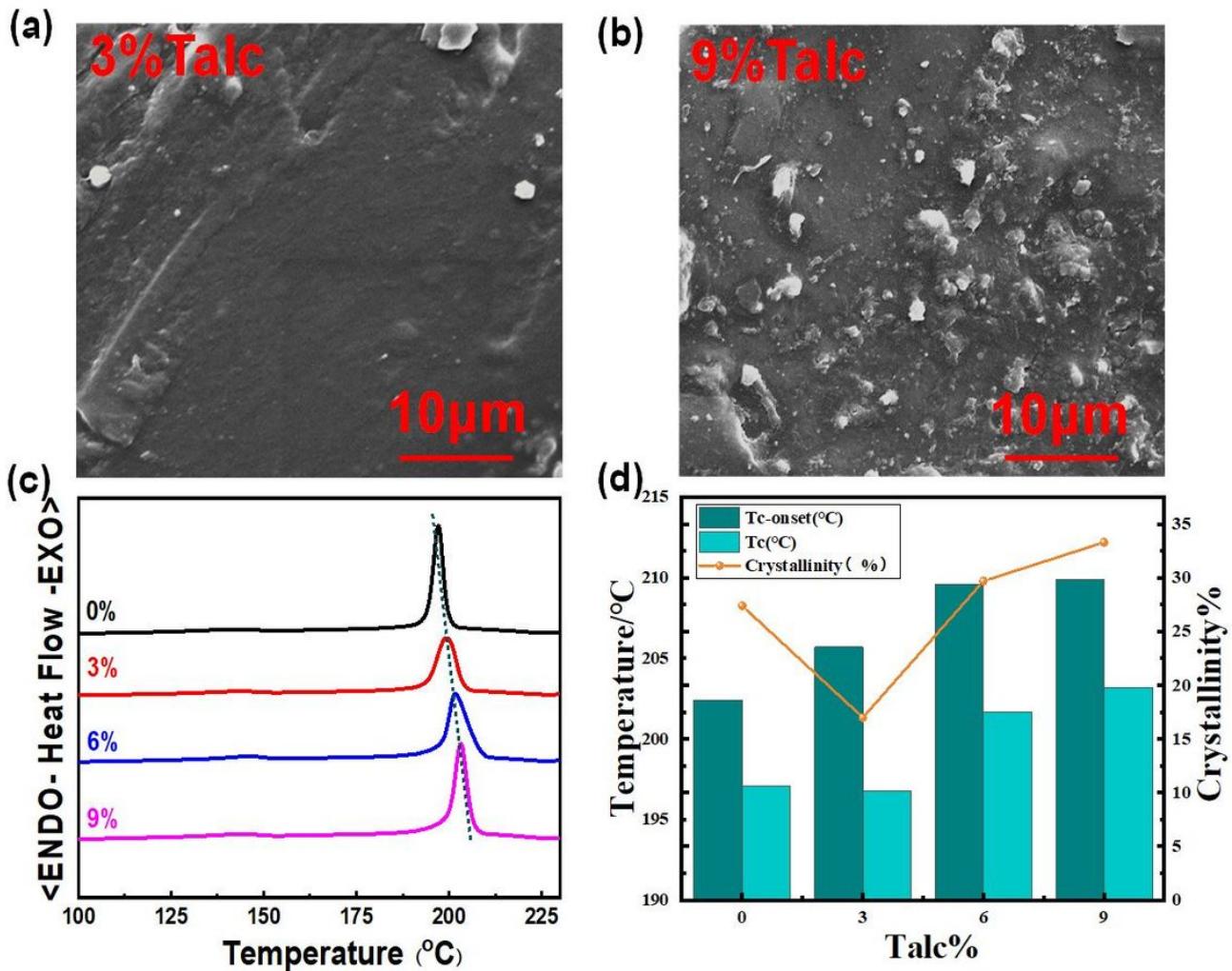


Figure 2

Microstructure and crystallization behaviors of PVA films with different talc contents. (a-b) SEM images of 3 % and 9 % talc films. (c) DSC curves of PVA films with different talc contents. (d) DSC curves of PVA films with different talc contents.

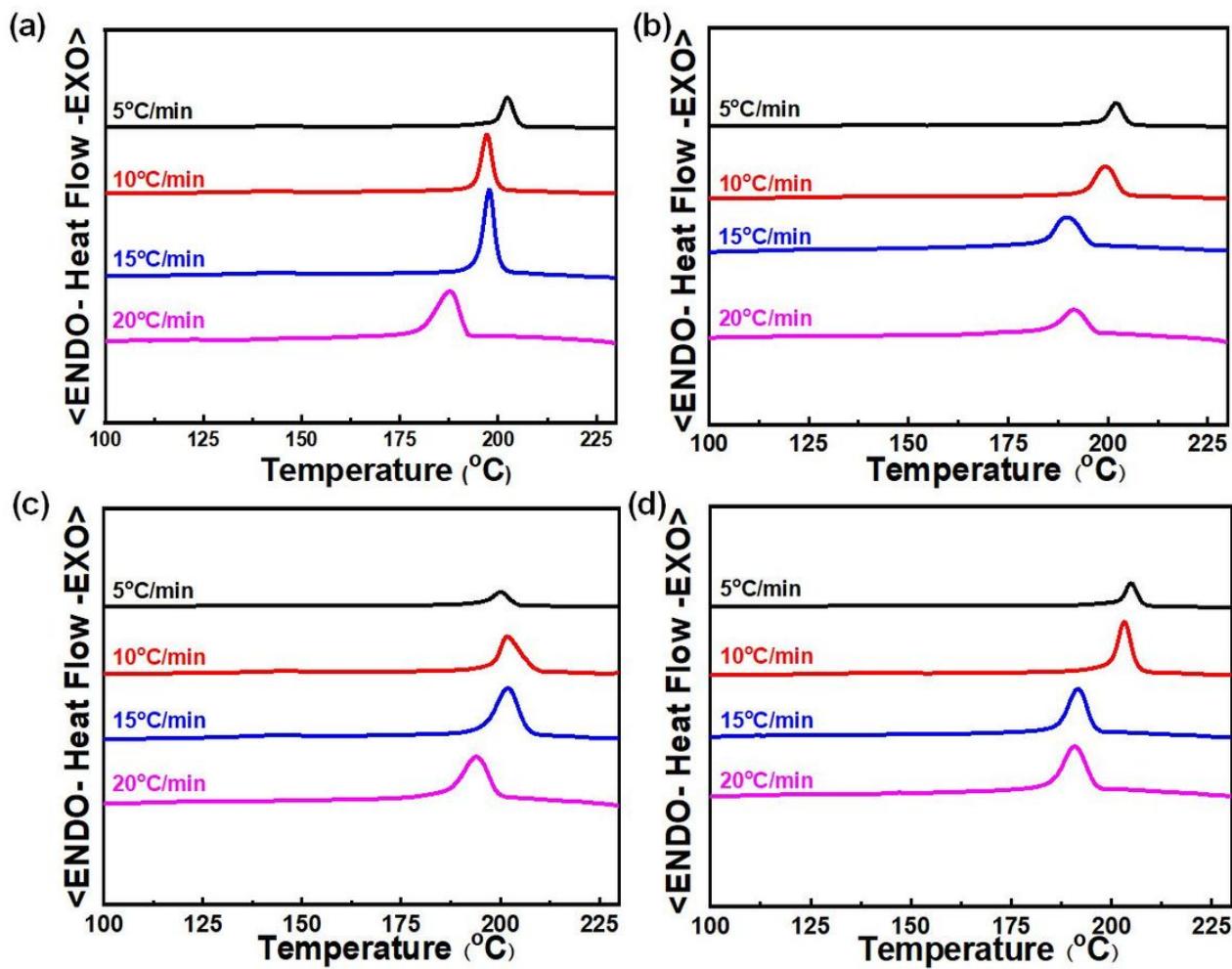


Figure 3

DSC curves of PVA films with different talc contents. ( a ) 0 %, ( b ) 3 %, ( c ) 6 %, ( d ) 9 %.

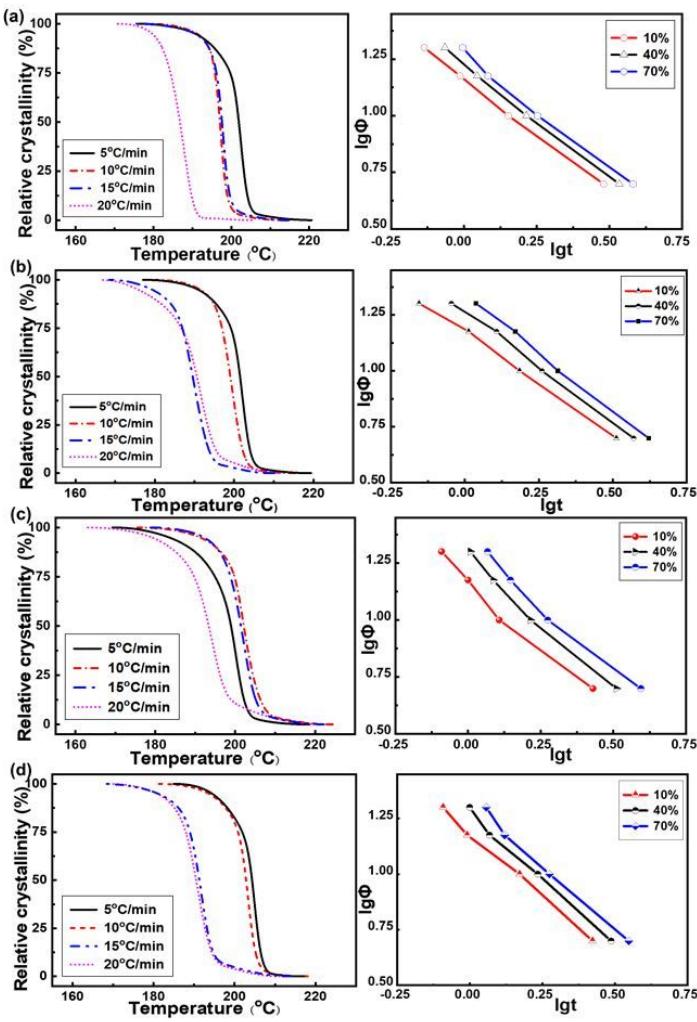


Figure 4

The curves of crystallinity versus temperature and the plots of  $\lg\Phi$  and  $\lg t$  of PVA films with different talc contents under non-isothermal crystallization. (a) 0%, (b) 3%, (c) 6%, (d) 9%.

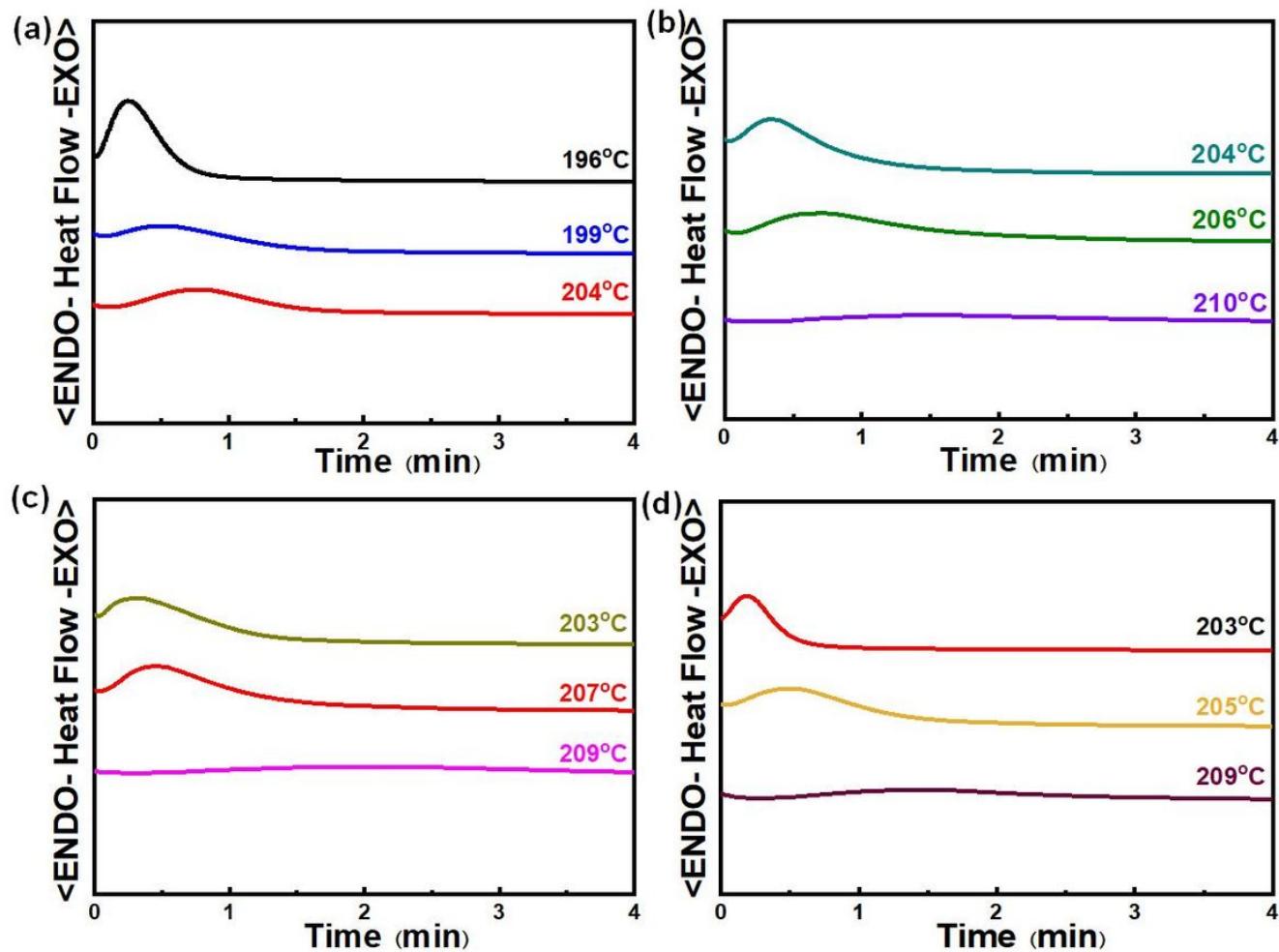


Figure 5

DSC curves of PVA films with different talc contents. (a)0%, (b)3%, (c)6%, (d)9%.

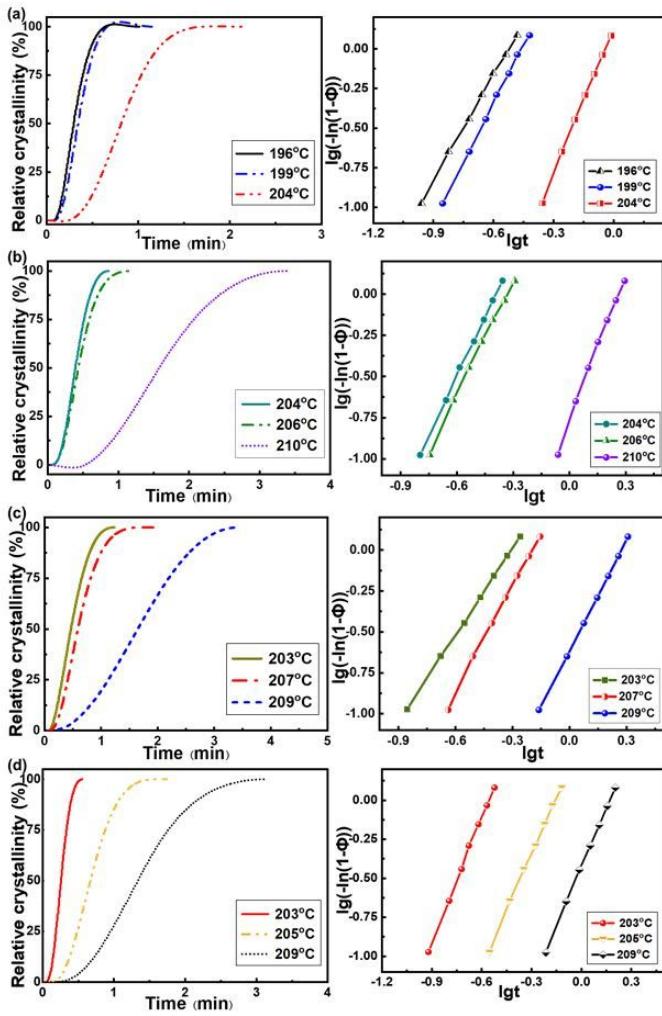


Figure 6

The curves of relative crystallinity of PVA films with different talc content with time and Avrami equation fitting curves. (a)0%, (b)3%, (c)6%, (d)9%.

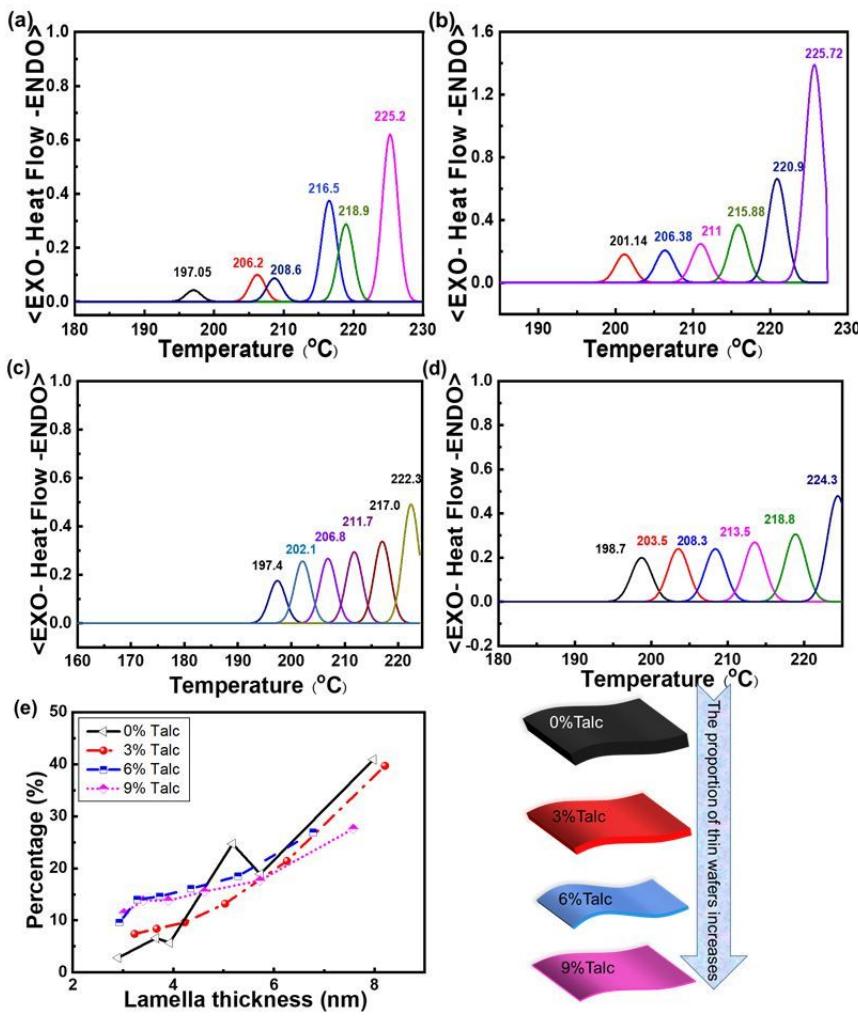


Figure 7

(a-d) SSA fitting curves of PVA/talc films with different amounts of talc. (e) The percentage of different wafer thicknesses of PVA films with different talc contents (a)0%, (b)3%, (c)6%, (d)9%.