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PII: S0141-8130(18)35261-9
DOI: https://doi.org/10.1016/j.ijbiomac.2018.12.180
Reference: BIOMAC 11339

To appear in: *International Journal of Biological Macromolecules*

Received date: 3 October 2018
Revised date: 7 December 2018
Accepted date: 19 December 2018

Please cite this article as: Wei Wu, Jinxuan Tao, Peitao Zhu, Hongsheng Liu, Qiliang Du, Jie Xiao, Wutong Zhang, Shaobo Zhang , A new characterization methodology for starch gelatinization. Biomac (2018), https://doi.org/10.1016/j.ijbiomac.2018.12.180

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A new characterization methodology for starch gelatinization

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Abstract:

A gelatinization degree control system, with a combination of Artificial Neural Networks (ANNs) and computer vision, was successfully developed. An intelligent measurement framework was purposely designed to achieve a precise investigation on phase transition and morphology change of starch in real time, as well as a process control during gelatinization. Base on a variation of birefringence number, the degree of gelatinization (DG) control system provided a direct and fast methodology without subjective uncertainty in studying starch gelatinization. In the course, the whole system was a cascade structure with the hot-stage temperature chosen as the inner-loop parameter, thus the granule morphology and birefringence at different DG could be easily observed and compared in real time, and the relative transition temperature was simultaneously calculated.

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1. Introduction:

Starch is one of the most abundant natural polymers and usually consists of two components: mainly linear amylose and highly branched amylopectin. In the presence of water, starch gelatinization could be triggered by heating [1, 2]. In the course, starch granules gradually swell and finally lose their crystallinity and molecular organization, allowing the amorphous regions to become more accessible to water and greatly swell [3, 4].

Because of its industrial significance, various methods, including differential scanning calorimetry (DSC), rheological measurements, microscopy with hot-stage and viscosity measurement, for the study of starch gelatinization and the associated properties have been developed [5-9]. Evidently, a researcher has an array of methods available and the choice would naturally depend on the facilities available as well as the objective to be achieved in the experiment. DSC is the most widely used method to investigate phase transitions through measuring thermal behavior changes during the process. However, the method has a limitation for system moisture content and sometimes induced inconsistent conclusions arose from different experimental operations [10, 11]. In addition, numerous rheological analysis technologies were also employed to record the torque required to balance the viscosity when starch slurry is subjected to a programmed heating and cooling cycle [12-15]. But such methods could not provide online information of granules and are hard to explore the detailed progress of starch phase transitions, such as the behavior of granule swelling and changes in crystalline structure.

Microscopy with a hot-stage is another popular and relatively simple method to evaluate gelatinization feature, basing on a series morphology change of starch granules during heating process [2]. Microscopy observation under normal and polarized light provides a direct way to inspect a variation of granule and birefringence. In the past years, some imaging processing softwares have been developed to study

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such changes. Chen et al [1] applied Gun Image Manipulation Program to study a change of average diameter of corn starches with different amylose content at some certain temperature and reported that the sequence of diameter increase rate and final accretion ratio was waxy > maize > G50 > G80. Since birefringence is an indicative of starch granules with ordered molecular orientation, a few researches were also performed to investigate the brightness changes of optical density of birefringence [16, 17]. However, these visual observations were usually marked by subjective uncertainties and time-consuming. Considering a real-time control of starch gelatinization with a desired degree is of great significance in some scientific research and industrial production [18-20]. Recently, our group developed a new methodology combining microscopy observation with Artificial Neural Networks (ANNs) for a further study of starch gelatinization [21]. In the case, birefringences were automatically identified by computer vision and then the relative birefringence number of the image was quickly calculated, which provides a unified standard for microscopy observation without subjective uncertainty. Resulted from the number change, temperature of phase transition was detected and consequently the degree of gelatinization (DG) corresponded to the temperature was quickly provided. In addition, the all evaluation process could be completed in a few seconds.

In the present study, the artificial intelligence method was further developed with a DG control heating system, which derived from a combination of image recognition by computer vision and a cascade control system, to achieve a precise investigation in situ on the changes of morphology and structure of starch in real time during gelatinization process. This study would be useful for a deep understanding of starch gelatinization and potentially provides a valuable way for the precise observation and control of starch-based reaction system.

2. Material and methods2.1 Material

Native corn starch with about 24% amylose employed in this experimental work

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was obtained from Hebei Derui Starch Co., Ltd (China). The initially moisture content of starch is approximately 13.5%, which was measured by an infrared heating balance (Model DHS-20, China) through heating samples to 110 °C for 20 min.

2.2Methods

2.2.1 Optical microscopy

A polarization microscope (BHS-2, Olympus Vanox, Japan) equipped with a Micropublisher 3.3 RTV camera and a hot-stage (THMS600, Linkam, UK) thermos system was applied.

Initially 0.5 wt% starch suspensions were prepared in glass vials, and then one drop was placed on a glass cover-slip and sealed with silicon adhesive to prevent moisture loss. Each specimen was heated from room temperature to 90 °C at 1 °C/min. The camera interval timer was set as 30s so that an image was captured at each 0.5 °C temperature increase. The magnification was set at 500×.

2.2.2 An image-based heating system through DG controlling

A new workstation with image recognition server and controller was designed and developed, which was applied to replace the traditional heating control system of hot-stage through the new function of DG recognition. As shown in Fig-1, the image-based control system mainly consists of two parts: image acquisition and DG recognition, and a modified heating system with DG controller.

1) Image acquisition and DG recognition

The original images were firstly acquired by the Micropublisher RTV system and then transferred by internal bus to the workstation where the birefringences were automatically identified by computer vision and subsequently the relative birefringence number of the image was calculated in real time. Starch-SSD, developed from Artificial Neural Network was performed on the image recognition server to monitor the change in DG. In fact, the workstation acted as a DG controller as well as an image recognition

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server.

2) A modified heating system with a DG controller

The function of DG controller was implemented on the workstation. A fuzzy logic controller (FLC), was employed for DG controller due to the lack of accurate dynamic model of the gelatinization process. Inputs of the DG controller were the difference of the expected number of starch granules and the actual number, and its variation, while the DG controller output the desired temperature of the hot-stage. The relative calculated result of DG controller worked as a proper temperature set-point to the T95 Heating Controller, which was used to precisely maintain the temperature of THMS 600 hot-stage under the microscope. A serial bus was used for data communication between the workstation and the T95 Heating Controller.

It is worth to note that, before this study, tens of thousands of native and modified starch granules images, which were of various types, regions and even from different processing stage, etc., were collected and kept in the data center of cloud for data storage, management and processing.

2.2.3 Differential scanning calorimetry (DSC)

A Perkin-Elmer DSC Diamond-8000 with a refrigerated cooling system and nitrogen purge gas was used in the experimental work. The melting point and enthalpies of indium were used to calibrate the temperature and heat capacity. DSC measurements of each specimen were performed in triplicate, and the results were presented as the mean.

DSC samples containing approximately 30 wt% starch and 70 wt % water were prepared initially and kept at room temperature for 2-3 h. The samples were heated from 30 ℃ to 90 ℃ at a scanning rate of 1 ℃/min, which was same as the program of hot stage.

2.2.4 Samples analysis with other traditional methods

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2.2.4.1 Sample preparation

A comparison between a new observation method with in situ artificial intelligence evaluation and a traditional offline observation method was provided. In the traditional system, partially gelatinized starch samples were prepared by thermal treatment using a rheometer (HR-2 Discovery Hybrid Rheometer, American), and the processing conditions was designed the same as the hot-stage heating program. In the rheometer system, 3 wt% starch suspension as heated from room temperature to the specific temperature at a rate of 1 °C/min. After that, the prepared samples were poured into undefiled petri dishes and dried in an oven (Shanghai Hengyi Scientific Instrument Co., Ltd，China) at 45 °C for 15h. Subsequently, the dried samples were kept in a desiccator before further measurements.

2.2.4.2 Scanning electron microscope (SEM)

The morphologies of starch samples were studied using a SEM (EVO18, Zeiss, Germany). The granules were affixed to a specimen holder using an aluminum plate and were coated with gold in a vacuum evaporator. The samples were then measured by SEM operated at an accelerating voltage of 10.00 kV.

2.2.4.3 X-ray diffraction analysis (XRD)

XRD analysis was performed using an X-ray Polycrystalline Diffractometer (D8 Advance, Bruker AXS, Germany) operating at 40 kV and 40 mA with Cu Kα radiation. Starch samples were equilibrated in a desiccator with saturated $BaCl_2$ solution at room temperature for two weeks before measurement to decrease the influence of water contents on the crystallinity of starch. X-ray diffraction patterns were obtained range from 4° to 50° with a scanning step size of 0.013°. The relative crystallinity (RC) was quantitatively estimated as a ratio of the crystalline area to the total area as described by Cheetham and Tao [22].

3. Result and discussion

3.1 Study gelatinization offline

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In the past years, to investigate the detailed information of starch gelatinization, a number of studies have been carried out to observe the morphology and phase transitions at specific temperature [13,23-25]. However, these works were mainly completed offline and usually a conversion from temperature to DG was required, which provided an indirect and time-consuming method. In the process, at least 46s was required to measure the relative transitions through an observation of birefringence change from the first picture to the last one of the gelatinization process. As shown in Fig-2, an illustration of the relationship between DG and transition temperature, as well as the corresponding morphology of starch granules observed under normal light (NL) and polarized light (PL) at specific DG, was provided. Obviously, in the course of gelatinization, DG value presented a non-linear increase with a rise in temperature. Based on DG change, it is distinctly observed that gelatinization took place at approximately 62 °C, accompanying with significant granules swelling phenomenon. At the end of gelatinization (about 73 °C), although all starch granules completely lost birefringences, a few still kept their granule shapes. Apparently, such model is effective in the detailed investigation of starch gelatinization. However, it is worth noting that, such offline studies were mainly performed by manual operation, which are usually marked by subjective uncertainties, long and costly litigation over discovery.

In this work, starch samples with the specific DG were also prepared by a rheometer using the same processing method as hot-stage. After the treatment, the morphology and appearance of granule were characterized by optical microscope and SEM, respectively. As shown in Fig-3, natural corn starch consisted of homogeneous polygonal granules and kept shapes with well-defined edges, exhibiting bright birefringences under polar light. After a slight gelatinization with 5% DG, most granules were swelled and a few started to lose their birefringences. However, such changes did not result in an evident change in granule appearance. With an increased in DG, granules greatly swelled and some lost granule shapes with an absence of

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birefringences. At the same time, rougher surface, more serious deformations, degradation and aggregation of starch granules were observed by SEM observation. As DG was increased to 60%, an extensive destruction in appearance was distinctly observed. The starch samples with different DGs were also studied using XRD, the corresponding changes of crystal pattern and RC are showed in Fig-4. It can be seen that native starch presented typical characteristics of A-type crystalline form, exhibiting strong diffraction peaks at 2θ values of 15°and 23°and a doublet at 17°and 18°. Apparently, although the pattern intensity gradually became weak, starch samples with 5%, 15% and 60% DG still presented their characteristic peaks and kept unchanged crystal type. However, a reduced RC value was clearly observed with the increase in DG, which is consistent with the study of microcopy and SEM observation.

3.2 New observation system with DG controller

Usually, a traditional microscopy observation of gelatinization is performed through a variation of temperature with a combination of hot-stage. In the measurement, a desired temperature was provided by heating control system and thus the change in granule with increasing temperature was observed in situ and in real time. After it, the relative detailed information at specific temperature, including morphological change and transition temperature, was achieved by offline visual observation and DG was subsequently calculated. However, such traditional methods were usually time-consuming and subjected to subjective uncertainties. In addition, most of these studies were simply carried out with the variable of temperature, which is hard to achieve a desired starch gelatinization degree in real time during the modification and synthesis of starch based system. In our recent work, a variation of birefringence was automatically recognized through a combination of microscopy with Artificial Neural Networks ANNs, and then DG observation was realized by machine learning algorithms [21]. The architecture of the method is provided in Fig-5. Starch-SSD, as a core of the object detection method, was developed from ANNs and mainly consists of

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three parts: data collection, feature learning, and results evaluation. In this work, the birefringences of starch were automatically identified and calculated. Accompanying with such number change, the temperature of phase transition was simultaneously provided and consequently the DG at specific temperature was achieved, which is much faster and provides a unified standard for microscopy observation without subjective uncertainty.

In this study, based on the newly designed DG control method, the artificial intelligence methodology was further developed to control gelatinization process in real-time, together with an online study of granule morphology at specific DG. The block diagram of the DG control system was presented in Fig-6. The whole system was a cascade structure with the hot-stage temperature chosen as the inner-loop parameter. In fact, the system input was the expected number of starch granules while the system output was the actual number of starch granules recognized by the method of Starch-SSD. Since temperature significantly affected gelatinization and tended to vary easily, it was chosen as the controlled variable of the inner-loop. Therefore a fluctuation of the process temperature could be reduced greatly before the gelatinization process was affected, which contributed to stabilize the final DG.

3.2.1 A design of temperature controller

A basic PID control law [26], as shown in Eq（1）, where K_P, K_I, K_D is the proportional gain, integral gain and differential gain respectively, was employed for hot-stage temperature control e_T, and the difference of the expected temperature and the actual temperature, was used to calculate the corresponding output u_T, according to a linear combination of a proportional, an integral and a differential calculation.

$$u_T(t) = K_P e_T(t) + K_P K_I \int_0^t e_T(t)dt + K_P K_D \frac{de_T(t)}{dt} \qquad (1)$$

3.2.2 A design of DG controller

An accurate and complete mathematical model of gelatinization process was

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difficult to generate because of the uncertainties in the knowledge of complicated gelatinization behavior. In this study, an FLC was applied for DG control since fuzzy rules were an intuitive carrier of the field knowledge of manual operations [27]. The frame of an FLC was shown in Fig-7, which showed the input information was processed through fuzzification, inference mechanism and defuzzification successively, and a rule base was used as the foundation of inference process. The inputs of the FLC were the DG error (e_{DG}), and its derivative de_{DG}. Fuzzification translated these two numeric values into linguistic values. The fuzzifications of e_{DG} and de_{DG} were performed using triangular and trapezoidal shape membership functions, and each signal was eventually partitioned into seven fuzzy sets entitled as Large Negative (LN), Medium Negative (MN), Small Negative (SN), Zero (Z), Small Positive (SP), Medium Positive (MP) and Large Positive (LP), as shown in Fig-8. It presented similar fuzzy sets of output signal variation Δu_{DG}, i.e., the increased temperature commanded to the inner loop.

A logical framework was developed to design reasoning system based on fuzzy control laws and represented as fuzzy rules in the rule base. Table-1 showed fuzzy rules for controlling DG, where the left column and the top row contained the linguistic values of the antecedent's variables, and each square represented the linguistic value of the consequent of a rule. Evaluation of each fuzzy rule and the combination of all the results of individual rules were performed according to Mamdani's inference method. Denote $A_i, B_i,$ and C_i the fuzzy sets related to e_{DG}, de_{DG} and u_{DG}, and the corresponding membership values were given as $A_i(e_{DG})$, $B_i(de_{DG})$, and $C_i(u_{DG})$. Each rule was evaluated by "and" operation, i.e., to find the minimum value of the membership values of the two inputs, to obtain a final result. The fuzzy set $\tilde{C}_k$ was calculated as

$$\tilde{C}_k(u_{DG}) = min\{A_i(e_{DG}), B_j(de_{DG})\} \quad (2)$$

If an output fuzzy set has more than one membership value resulted from several

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rules, choose the maximum membership value as the unique result.

$$C_k(u_{DG}) = \max\{\tilde{C}_m(u_{DG})\} \quad (3)$$

To calculate the final crisp output value, output membership functions were truncated at their corresponding membership values, and then consolidated into one area. The center of gravity (COG) of the area was used as the crisp output value. Suppose a function f(x) depicted the corresponding curve of the area, with maximum value x_{max} and minimum value x_{min}, the COG of the area could be calculated as

$$x_{COG} = \frac{\int_{x_{min}}^{x_{max}} x f(x) dx}{\int_{x_{min}}^{x_{max}} f(x) dx} \quad (4)$$

3.2.3 A comparative study using DSC, microscopy with temperature-control and DG-control system

Starch gelatinization at different degree was also investigated by DSC through an analysis of the change of thermal enthalpy. As shown in Fig-9, corn starch with about 70% water content presented a typical peak of gelatinization, and the area under the peak is directly proportional to the enthalpic change, which provides a quantitative measure of the energy transformation. According to the area change, transition temperature at a specific DG was calculated after the measurement. It is seen that, as DG value was 5%, 15% and 60%, the corresponding transition temperature was 63.9 ℃, 65.8 ℃ and 69.2 ℃ respectively. Apparently, such indirect method required at least a few minutes to achieve the relative results and it is impossible to monitor the DG change during gelatinization process in real time.

Comparatively, in the traditional microscopy observation system, based on a variation of birefringences of starch granules in gelatinization process, the morphology changes were initially observed by human sight after measurement and subsequently the relative results were evaluated by manual operation. In this course, DG was calculated through the number change and consequently the corresponding temperature

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was achieved. However, the DG-control system provided a direct and intelligent way to study the detailed information of gelatinization online and in real time through a combination of an image recognition block and a number controller of birefringence.

A variation of birefringence during gelatinization studied by the new system was presented in Fig-10. Apparently, the granule morphology and birefringence at different DG could be easily observed and compared. For example, at 60% DG, meaning 60% granules had lost their birefringences, it can be seen that most granules kept their contact shapes but they were slightly swelled. At the same time, the relative transition temperature was also automatically provided through measuring the number change in birefringence, which was approximately 69.4 ℃. Similarly, such information at any specific DG point could be simultaneously provided during the process of online observation. It is worthy to note, based on birefringence number change from the image, the traditional offline method with manual operation is time-consuming, and usually more than 5 min was required to observed the change and achieve the relative results. Comparatively, the new proposed method provided a direct and faster way to investigate and evaluate the detailed information of starch system at specific degree, and the whole process could be completed in 3-4 s. Moreover, a combination of ANNs and computer vision also provided a uniform standard to distinguish the slight difference between the birefringences, which was hard for human vision and usually induced some insistent and confused results from subjective difference of individual. However, it is worth to note, every method has its limitations and it is unlikely that any single method would be able to give a complete picture of the gelatinization properties. In further study, the new methodology will be purposely designed with other methods for a deep understanding of starch phase transitions.

4. Conclusions

In this study a new intelligence methodology was further developed via a DG control system, which provides a new way to directly study the detailed information of

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granule morphology and phase transition at specific DG in real time. Compared to the traditional observation method performed by human operation, which is time-consuming and subjective, the proposed DG-control observation and study effectively provide a fast way and a unified standard without subjective uncertainty. Such process control method positively gives a promising approach to real-time design and control physical (or chemical) reactions at specific condition for starch-based system in industrial applications.

Acknowledgments

The authors from SCUT, China would like to acknowledge research funds National Key R & D Program of China (2018YFD0400702), NSFC (31571789), the 111 project (B17018) and the Fundamental Research Funds for the Central Universities （2018KZ10）.

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Tables

Table-1 Rule base for DG controlling

$e_{DG}\backslash de_{DG}$	LN	MN	SN	Z	SP	MP	LP
LN	LP	LP	LP	LP	SP	SP	Z
MN	LP	LP	LP	MP	SP	Z	SN
SN	LP	LP	MP	SP	Z	SN	SN
Z	LP	MP	SP	Z	SN	SN	MN
SP	MP	SP	Z	SN	SN	MN	MN
MP	SP	Z	SN	MN	MN	MN	LN
LP	Z	SN	MN	LN	MN	LN	LN

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Figures and captions

Fig-1 Diagram of the device with DG controller (A: image acquisition and DG recognition; B: a modified heating system with DG controller)

Fig-2 The relationship between temperature and DG in the process of gelatinization and the relative morphology of starch granules at specific DG.

Fig-3 Morphology of starch granules at specific DG by microscopy observation under normal light (NL) and polar light (PL), and SEM observation, respectively.

Fig-4 XRD traces of corn starches with different DG and the relative RC (from bottom to top: DG is 0%, 5%, 15% and 60% respectively).

Fig-5 Schematic representation of the artificial intelligence methodology.

Fig-6 Block diagram of the DG control system.

Fig-7 Structure of fuzzy logic controller.

Fig-8 Fuzzy sets of (a) error, (b) rate of change of error, and (c) output variation.

Fig-9 DSC curve of corn starch and the corresponding transition temperature at specific DGs.

Fig-10 Birefringence of starch granules at specific DG observed by DG control method.

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Fig-1

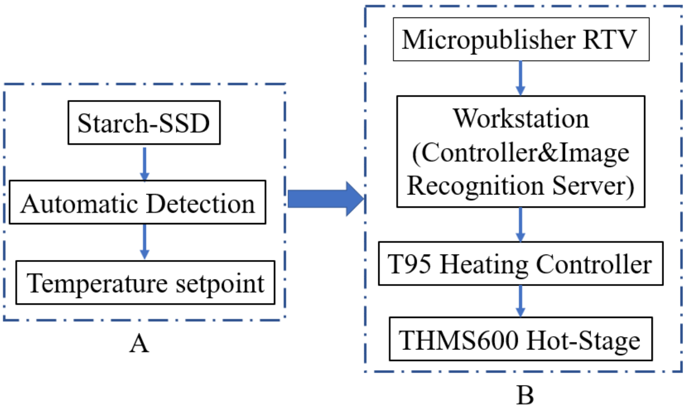

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Fig-2

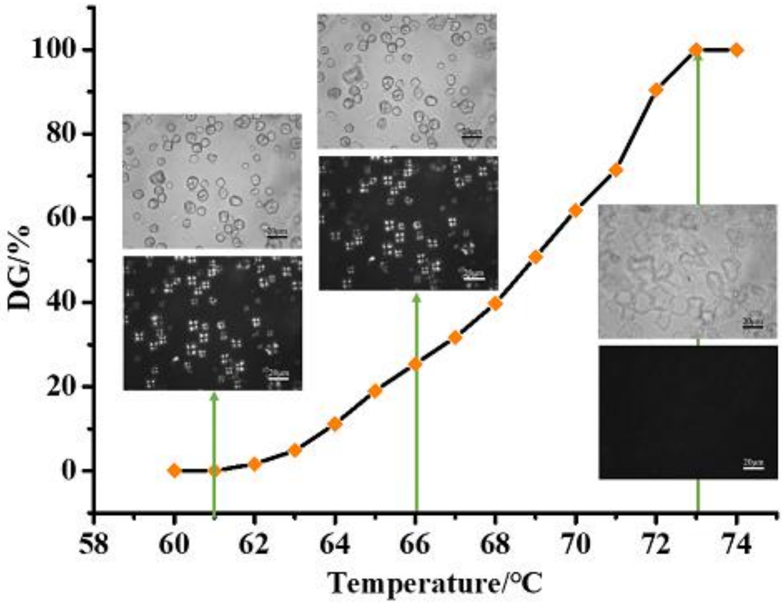

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Fig-3

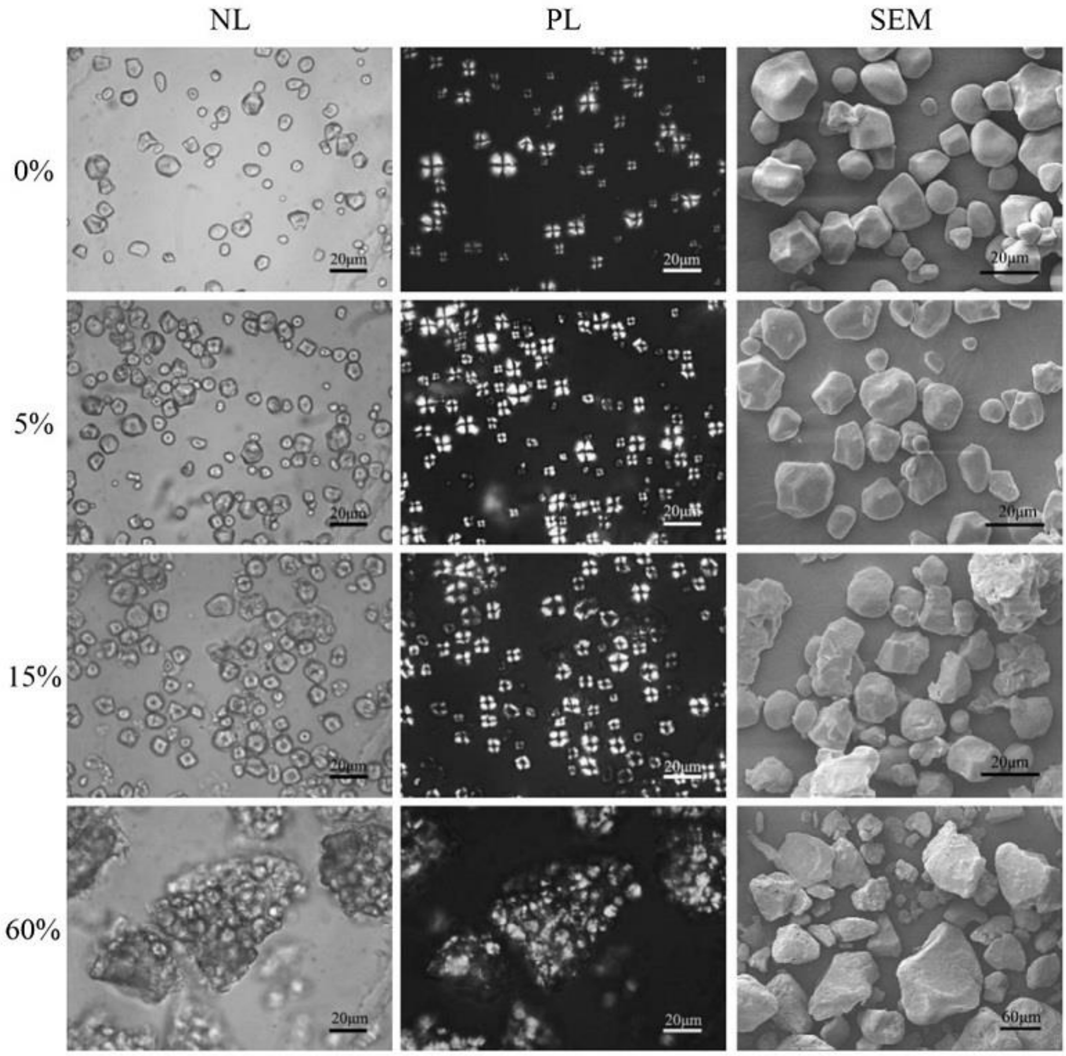

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Fig-4

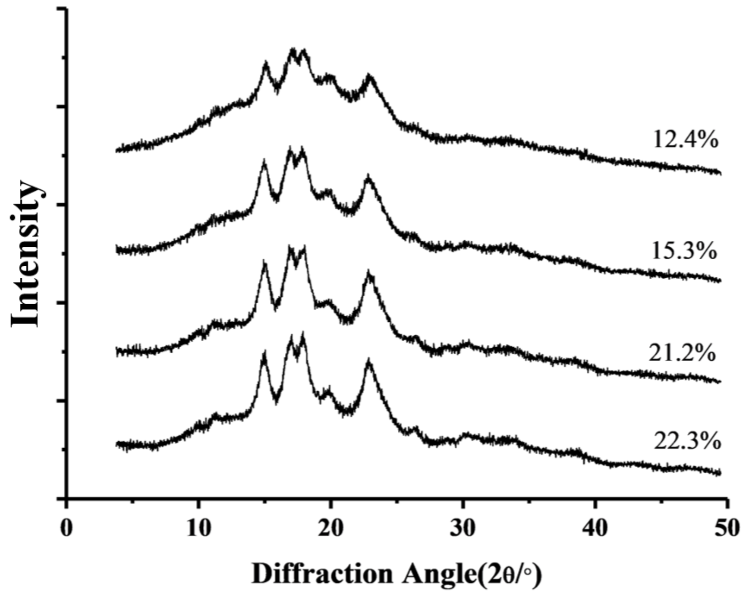

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Fig-5

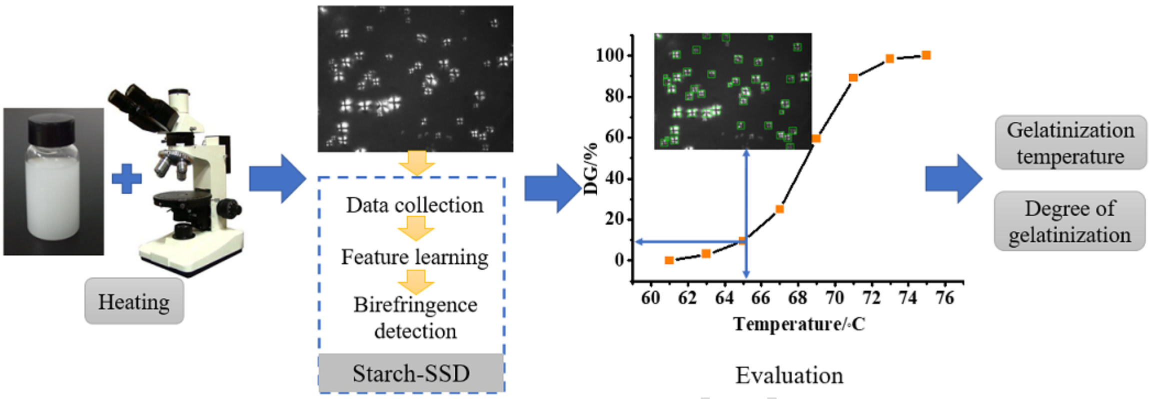

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Fig-6

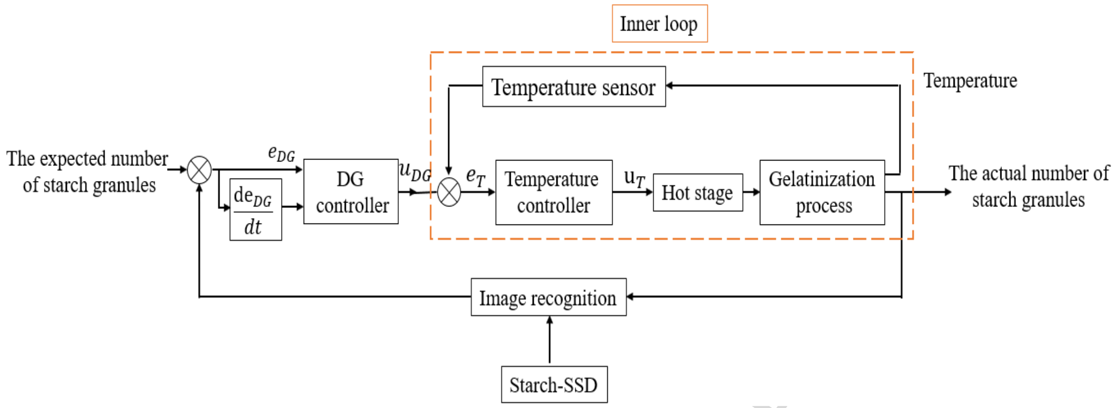

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Fig-7

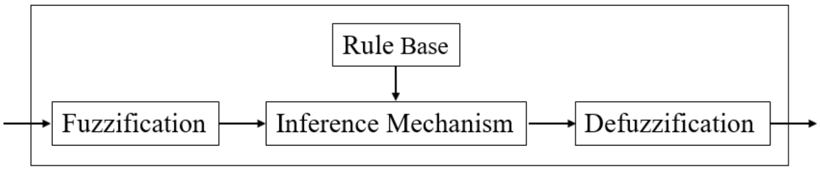

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Fig-8

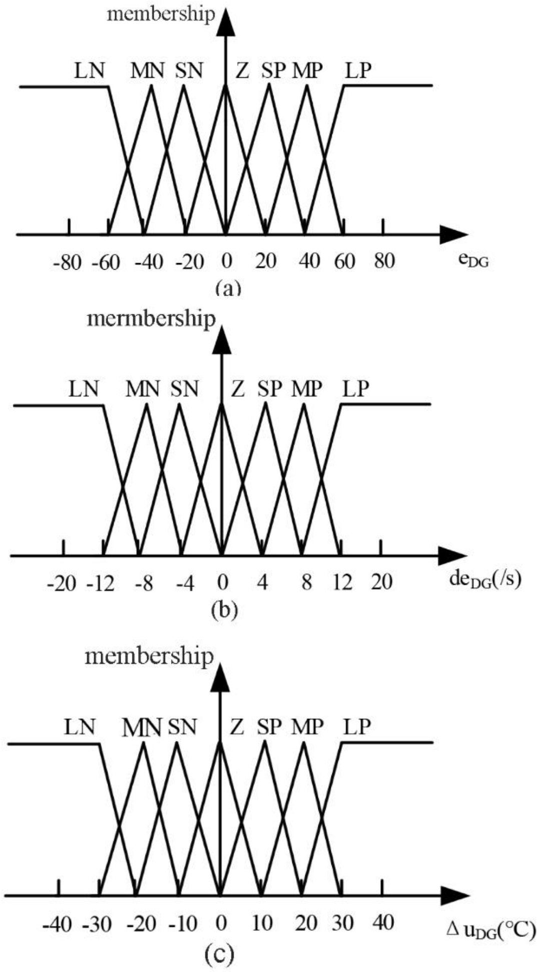

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Fig-9

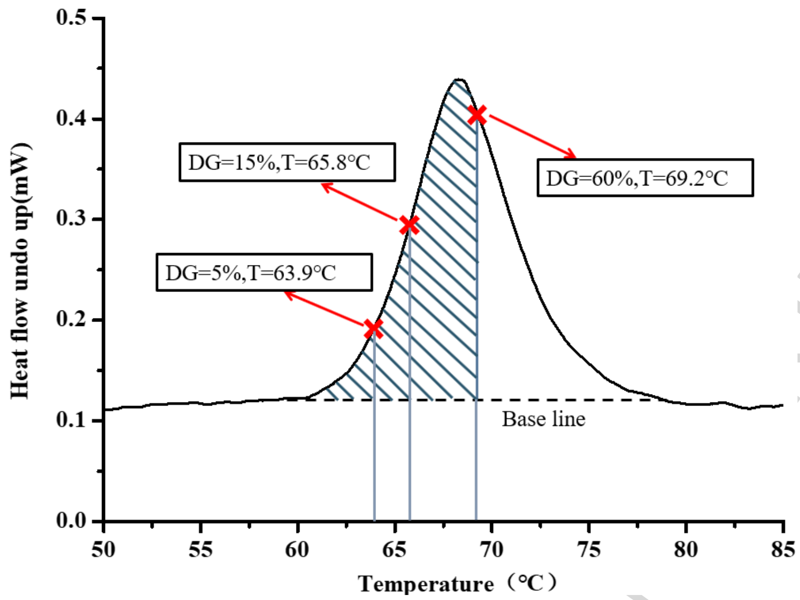

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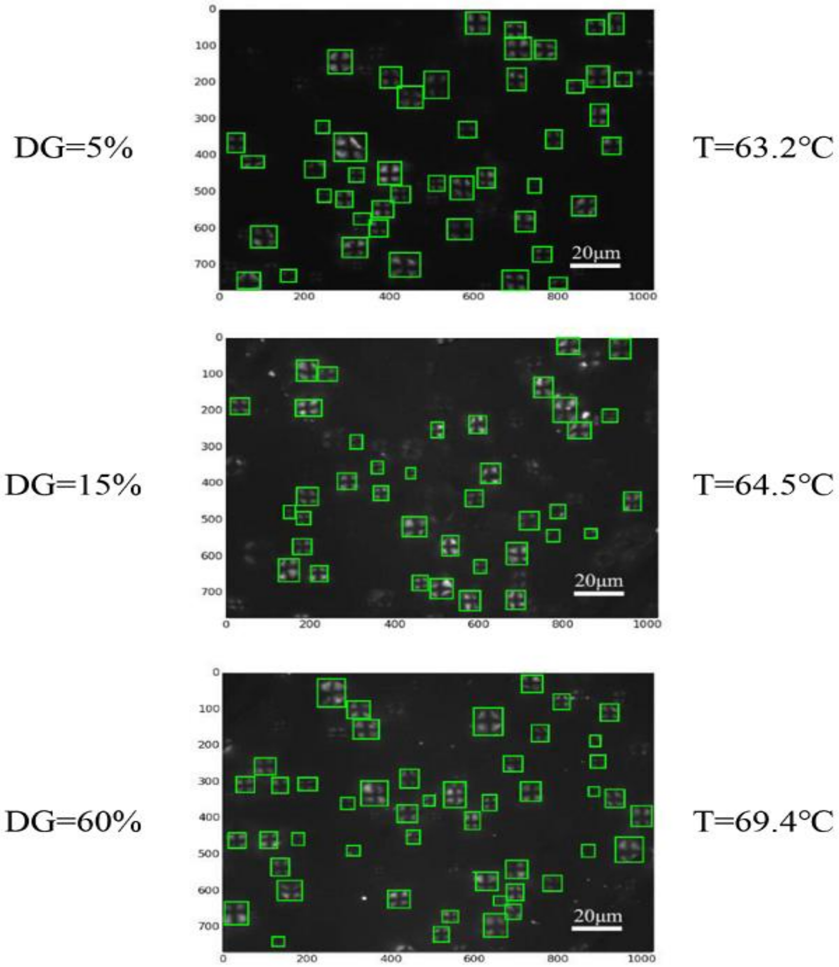
Fig-10



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