Investigating the Effect of Cooking Temperature and Cooking Time on the Thickening Ability, Absorption of Oil, Thermal Properties and Microscopic Property of Roux

by

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Abstract

Roux is made from cooking a mixture of wheat flour and butter, and this is commonly used to create a thick sauce by cooking the roux with excess water. Roux were made from mixtures of canola oil and one of four types of starch or flour (wheat starch, wheat flour, pea starch, and pea flour) which was cooked at one of five temperatures (100, 116, 134, 153, 175 °C) for 0, 4, 8, 12, or 16 min. The oil content of the roux, and the pasting properties of the sauce made from the roux (by rapid viscoamylograph) were determined. The thermal properties (determined in excess water by differential scanning calorimetry (DSC)) and microscopic analyses of roux made from wheat starch and canola oil cooked at one of three temperatures (100, 134, 175 °C) for 0, 8, or 16 min were also determined.

The thickening abilities of all types of roux decreased with an increase in cooking time when the roux was cooked at 154 and 175 °C, but cooking time did not influence the thickening ability at the three lower cooking temperatures. The pasting viscosity of roux made from wheat flour, pea starch, and pea flour was enhanced when cooking temperature increased from 116 to 134 °C, but such an enhancement was not observed with the roux made from wheat starch. Roux made from flour, regardless of the flour type, had significantly higher oil content than roux made from starch.

DSC property changes were slight with changes in roux making conditions. Starch granule integrity and birefringence was maintained regardless of cooking time and temperature.

In conclusion, the cooking process changed the thickening ability of a roux, but no starch gelatinization was observed during the roux making process. Ingredients of a roux, especially protein, significantly affected some physicochemical properties, such as the thickening ability and residual oil content of the roux during the cooking process.

Pea starch and flour are suitable thickening ingredients for making a roux when cooking temperatures were at 134 or 154 °C, but not at higher temperatures.

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1. Introduction

Roux is commonly prepared from cooking a mixture of wheat flour and butter, which is then cooked with milk or water to make a sauce that provides flavor and viscosity (Heyman et al., 2010). Cooking brings about complex changes to roux systems, which markedly affects the properties of the sauce made from a roux (Krasnow et al., 2011). Nonetheless, although there have been several studies investigating the effect of the cooking conditions on the physicochemical properties of a roux (Alvarez-Ramirez et al., 1973), how the cooking conditions affect the properties of the sauce made from a roux is not well understood.

A typical roux is composed of starch, protein, fat, and moisture. Cooking promotes interactions/reactions between ingredients and also brings about changes to these ingredients. The most noticeable interaction between roux ingredients is the non-enzymatic browning Maillard reaction. Such reactions impart a brown color to the roux system (Kato, 2005; Shimada et al., 1973). Another interaction between the roux ingredients is the interaction of fat and starch, especially those that lead to formation of the amylose-lipid complex (Alvarez-Ramirez et al., 2018). As the lipid-amylose complex influences the pasting properties of the starch granules in a roux (Tang & Copeland, 2007), such starch-oil interactions will affect the physical properties of a sauce made from the roux. Cooking also alters the properties of ingredients in a roux. For example, although starch in general retained its granular shape after cooking, some starch granules were found to swell and gelatinize in a cooked roux (Alvarez-Ramirez et al., 2018). Proteins in a roux can be divided into wheat proteins from the wheat flour and animal proteins from the butter. Cooking a roux resulted in the structural change of proteins in a roux (Keiko et al., 1979). In general, cooking altered some properties of ingredients in a typical roux system, and these changes of properties overall result in the cooked roux having different properties. However, how each ingredient plays its role in influencing the overall physical properties of a roux has not been well studied.

The “thickening ability,” defined as the ability of a cooked roux to thicken a sauce made from the roux, was seen to be changed by cooking at different temperatures (Krasnow et al., 2011). Krasnow et al. (2011) found that the thickening ability of a cooked roux decreased as the cooking temperature increased from 120 to 200 °C. The decrease of thickening ability with increasing cooking temperature could be because of multiple changes involving the roux ingredients. Although cooking time is critical to the quality of a roux (Alvarez-Ramirez et al., 2018; Shimada et al., 1973), knowledge about how cooking time affects the thickening ability of a roux is limited. Knowledge about the thickening ability of a roux is important as it is helpful when preparing the sauce based on a cooked roux.

A typical roux contains starch, protein, fat, and moisture. Previous studies have attempted to develop new formulations by replacing one or some of the ingredients of a roux (Heyman et al., 2010; Kato, 2005). Pea is a widely cultivated crop in Canada, enriched with proteins that can provide additional nutritional value for food products (Ratnayake et al., 2001). Pulse starch or flours have not been used to make a roux in previous studies. However, pea and wheat starch are different in particle size distribution, amylose/amylopectin ratio, and crystalline structure (Bajaj et al., 2018; Ratnayake et al., 2001; Ratnayake et al., 2002). Moreover, the composition and structure of pea and wheat proteins are different (Delcour et al., 2012; Lam et al., 2018). Therefore, it is expected that the thickening ability of roux made from pea starch or flour could be different from that of roux made from wheat starch or flour.

In terms of the composition of a roux, the simplest roux system could contain only maize starch and soybean oil (Krasnow et al., 2011). Therefore, it would be possible to investigate how each component in the ingredients influenced the thickening ability of a roux by comparing the thickening ability of the roux with different compositions. This kind of approach had some success as Krasnow et al. (2011) showed, by comparing the

roux systems which contained or did not contain protein, that the color change of a cooked roux was related to its protein.

The thickening ability change of a roux could be due to the structural changes of starch, proteins, and the effect of oil (Tang & Copeland, 2007). Water content in a roux is low, coming mainly from the butter, and this contributes to partial starch gelatinization in a cooked roux (Alvarez-Ramirez et al., 2018). When starch is heated in the presence of moisture, the swelling of amorphous regions in starch and the breakdown of hydrogen bonds between starch molecules results in disorder of the starch's crystalline structure (Chen et al., 2019a). The pasting viscosity of pre-heated starch is lower than that of the uncooked starch (Chen et al., 2018a). It could be speculated that cooking resulted in disorder of the crystalline structure for the starch in a roux, leading to lower thickening ability. However, it has been reported that the thickening ability still changed even if the roux contained minimal moisture since the roux have been made from only soybean oil and wheat starch (Krasnow et al., 2011). Therefore, it seems that the change in the thickening ability of a typical roux is due to the disorder of starch crystalline structure. Consequently, to investigate the reason for the change in a roux's thickening ability, it is better to remove the effect of partial gelatinization, i.e., replacing the butter with oil to remove moisture from the roux system.

The rapid visco analyzer (RVA) is commonly used to determine the pasting properties of starch or flour (Balet et al., 2019). The starch or flour sample is mixed with an excess of water and gelatinizes under a programmed transitions of temperature and stirring speed. Therefore, when the cooked roux is mixed with excess water and heated in the RVA canister, the outcome is similar to preparing the sauce from a roux with a well-controlled temperature and stirring regime. Therefore, the pasting viscosity of the sauce made from a cooked roux by the RVA analysis reflects the thickening ability of the cooked roux.

Due to the essential role that cooking temperature and time play in the physical properties of a roux and how the ingredients affect the properties of a roux, this thesis research aimed to systematically investigate the influence of the ingredients, cooking temperature, and time on the physical properties of a cooked roux. To investigate the effect of the types of ingredients, a cereal and a pulse flour and their corresponding starches were used to make the roux. The specific objectives of this thesis were as follows:

1. To determine the effect of cooking temperature and time on the thickening ability of roux prepared from a cereal and a pulse flour, and their corresponding starches.
2. To determine the effect of cooking temperature and cooking time on the residual oil content of roux prepared from a cereal and a pulse flour, and their corresponding starches.
3. To explain the mechanisms responsible for changes in thickening ability and oil content of the different roux by comparing roux made from starches with those made from flours, and from a cereal and a pulse source.

2. Literature Review

2.1. Introduction

2.1.1. What is a Roux

Roux is a traditional thickening agent which is commonly used in French cuisine. Roux is the base for many foodstuffs, such as gravy, sauces, soups, and stews. The unique flavor provides better quality and taste. The term “roux” is commonly referred to as the system containing wheat flour and butter after heating, while the term “sauce” usually refers to the mixture of the roux with milk or water (Krasnow et al., 2011). Roux can be classified as white or brown roux depending on the heating temperature, as the color of a roux turns from white to brown with increasing cooking temperature. White sauce, or also referred to as bechamel sauce, is made from cooking the mixture of white roux and milk to achieve a preferable mouthfeel and flavor (Heyman et al., 2010).

2.1.2. Use in Industry

Production of roux faces some challenges. One of the challenges is destabilization. Roux is not a continuous phase as the starch does not fully gelatinize because of the limited moisture content, ungelatinized starch granules would separate from a roux system over time (Alvarez-Ramirez et al., 2018). Research has been done on developing new formulations (Arocas et al., 2009; Herranz et al., 2019), but little attempts have been made to adjust the processing method or conditions to solve the destabilization problem because understanding of processing effects on roux is limited.

Another challenge is developing new formulations of roux to meet the increasing demand for novel functional products. Red sweet pepper was added to the roux to improve the functionality and nutritional values (Hernández-Carrión et al., 2015). A new formulation of white sauce by replacing wheat flour with soy protein and maize starch was developed to obtain white sauces that were free of gluten and with high

protein content (Quiles et al., 2012).

2.1.3. Characteristics of Roux

Roux is a complex system containing starch, oil, and protein, with the change of the constituents and interactions among them during cooking resulting in roux's unique properties. For example, the brown color is a result of non-enzymatic browning Maillard reactions between the polysaccharides (leached starch chains) and proteins (Alvarez-Ramirez et al., 2018). The flavor change is due to the formation of the flavor compounds during cooking. It is noticeable that cooking condition has a significant impact on the properties of roux. The term “thickening ability” is defined as the ability of a roux to thicken the sauce made from a roux. When roux was heated under a mild temperature range (<160°C), increasing the cooking temperature increased thickening ability; when roux was heated at a higher temperature range (>160°C), increasing cooking temperature resulted in the decrease of the thickening ability of roux (Krasnow et al., 2011).

2.2. Ingredients of a Roux System

Roux is commonly made from cooking a mixture of the same portion in weight of butter and wheat flour. Other types of lipids such as soybean oil (Krasnow et al., 2011), maize oil, and a mixed oil of soybean and rapeseed (Kato, 2005) have been used to replace butter, and maize starch (Krasnow et al., 2011) is a potential replacer for wheat flour. The key components of a roux include starch, protein, lipids, and water, which is a component of butter. The sources of ingredients have a considerable impact on the flavor and color of the roux (Kato, 2005; Lagrain et al., 2005).

2.2.1. Starch

Starch, as one of the key ingredients of a roux system, is responsible for color and texture development. Wheat flour is the most common ingredient of roux, but other flour or starch sources such as maize starch (Lagrain et al., 2005) can be used to make roux instead of wheat flour. Starch granules have a structure with alternating semi-crystalline and amorphous growth rings. Essentially linear amylose and highly branched amylopectin are two types of polyglucan in starch. Amylopectin forms double helices, and the double helices pack together forming clusters (Shevkani et al., 2017). According to the arrangement of the amylopectin clusters, starch can be classified as A-, B-, and C-type (Wang & Copeland, 2013). Wheat starch has an A-type crystalline structure (Shevkani et al., 2017), while pea starch has a C-type crystalline structure (Barron et al., 2000; Bogracheva et al., 1998). The crystallinity of wheat starch (around 33%) is higher than the crystallinity of most varieties of pea starch (varies from 18.9% to 36.5%) (Barron et al., 2000). The degree of polymerization (DP) of amylopectin impacts the viscosity development of the starch during gelatinization. A higher portion of long-chain amylopectin contributes to higher pasting viscosities (Shevkani et al., 2017). The DP of amylopectin in wheat starch is around 10,000, which is larger than the DP of amylopectin in pea starch (Ratnayake et al., 2002). The ratio of amylose/amylopectin is also a critical property determining the functionality of starch. Wheat starch has a lower amylose/amylopectin ratio compared to that of pea starch (Czuchajowska et al., 1998).

Wheat starch granules can be classified as A- (diameter > 9.9 μm) or B- (diameter < 9.9 μm) granules depending on the granular particle size; these two types of starch granules have different functional properties, such as thermal properties and pasting properties (Zeng et al., 2014). The particle size distribution of hard wheat starch is typically bimodal because hard wheat starch contains both A- and B- type of starch granules (Park et al., 2009). In some cultivars of soft wheat starch, the typical bimodal particle size distribution is not obvious, and only one peak shows on the particle size

distribution profile (Stasio et al., 2007). On the other hand, the particle size distribution profile of wheat flour shows a trimodal size distribution. The first two modes on the particle size distribution profile relate to starch with different granular sizes, and the third mode relates to gluten and particle clusters (Kim, 2004). The first mode representing wheat starch had a smaller particle size which was less than 10 μm, and the second mode representing wheat starch with a larger granular size ranged between 10 and 40 μm. The particle size distribution profile of pea starch only showed one mode, and the diameter of most of the pea starch granules ranged from 20 μm to 40 μm, according to Huang et al. (2007).

2.2.2. Protein

Protein in wheat flour can be classified into gluten and non-gluten-forming proteins. Gluten is further divided into gliadins and glutenins. After being treated with a disulfide-bond reducing agent, glutenins resolve into two groups of subunits, including the high molecular weight glutenin subunits (HMW-GS) and the low molecular weight glutenin subunits (LMW-GS) (Delcour et al., 2012).

The crude protein content of raw pea seeds is around 20 to 25% (Lallés, 1993). The pea proteins consist of albumins (10% to 20%) and globulins (70% to 80%). Globulins can be further classified as legumin and vicilin proteins (Lam et al., 2018).

Wheat proteins are introduced in a roux system as an ingredient from wheat flour, and they play a critical role in affecting the qualities of a roux, such as participating in the Maillard reaction (Alvarez-Ramirez et al., 2018). The moisture content in an oil is much lower than in a commercial butter (maximum 16%) (Wilbey, 2009). Since oil is sometimes used to make a roux to replace the butter (Krasnow et al., 2011; Kato, 2005), cooking roux made from wheat flour and oil would leave the roux as a low moisture system, so that the wheat proteins are heated in a dry manner. Previous studies have

shown that food products made from dry heated wheat flour would have a different quality compared to those made from non-dry heated wheat flour (Nakamura et al., 2008; Ozawa and Seguchi, 2006). It is reasonable to assume that when the roux made from oil was cooked, the wheat proteins would be affected by the cooking process, resulting in the alteration of the properties of the roux.

Zhang et al. (2012) found that dry heat treatment (120 °C, 20 min) altered the secondary structure of wheat gluten by increasing β-sheet from 41.5 to 51.2 % and slightly increasing the α-helices content from 24.5 to 26 %. González et al. (2021) also reported that the content of β-type configurations (β-sheet and β-turn) in wheat proteins increased after dry heating wheat flour at 50, 100, 150, or 200 °C. Dry heat treatment would also result in other property alterations, such as reduced solubility (Mann et al., 2012) and increased surface hydrophobicity (Zhang et al., 2012). Keppler et al. (2018) observed that when soft wheat flour was dry heated between 110 and 200 °C for various times (1-30 min), the protein network formed in the untreated sample disappeared in the dry heated wheat proteins; Keppler et al. (2018) claimed that the lack of protein network was due to the significant denaturation of wheat proteins during dry heating. Gonzále et al. (2021) also observed that wheat starch granules aggregated after dry heat treatment, and the aggregation structure was cemented by the wheat protein matrix. Studies related to dry heating on pea proteins are limited, but a recent study showed that pea protein isolates resuspended in water were affected by drying at 165, 180, and 195 °C (Burger et al., 2020).

2.2.3. Lipids

The lipids in a roux system mainly come from the added butter, which contains a considerable amount of triglycerides. Some lipids exist in the starch, but the content is low. Butter is a dairy product consisting of fat (≥80%), water (≤16%) and nonfat solids

(proteins 0.6–0.7%, lactose 0.7–0.8%, minerals ≈ 0.2%) (Frede, 2002), which is the source of fat for a roux. Most of the lipids in cow milk (98.3%) are triacylglycerols (Huppertz et al., 2009), which implies that the almost all the fatty acids in butter are bound on glycerol molecules. According to Pădureţ (2021), who analyzed the fatty acid composition of butter made from cow milk, butter fat consisted of short and middle-chain saturated (16.78%), long-chain saturated (52.34%), monounsaturated (25.48%), and polyunsaturated fatty acid (5.30%). Specifically, most of the fatty acids in butter are palmitic acids (≈ 32%), stearic acids (≈ 13%), and oleic acids (≈ 21%) and there are still a little portion of fatty acids such as lauric acid, myristic acid or linoleic acid.

Canola oil is recognized as a source of healthy edible oil, as canola oil contains a low amount of saturated fatty acids (Aukema & Campbell, 2011). The triacylglycerol content accounts for 94.4 to 99.1 % of the total lipid content in canola oil (Przybylski, 2011). Canola oil contains about 12% α-linolenic acid (omega-3), 65% oleic acids, and less than 7% saturated fatty acids (Ghazani & Marangoni, 2016). Therefore, canola oil is a potential healthy replacement for butter to make a roux. The type of fat or oil used to make a roux impacted the flavor. Kato (2005) compared the flavors of roux made from three different fat as butter, maize oil, and a mixed oil consisting of soybean and rape. Roux made from butter had a pungent sweet odor compared to roux made from maize oil, while the flavors of butter roux (traditional roux) and maize oil roux were generally preferred compared to roux made from the mixed oil.

2.2.4. Moisture

The moisture within a roux system mainly comes from the butter. The moisture content of a roux system is a critical factor determining the gelatinization temperature. Alvarez-Ramirez et al. (2018) determined that the moisture content in a roux system using butter was insufficient to allow full gelatinization of starch, as most of the starch kept the

integrity of granular structure after cooking, showing that most of starch did not gelatinize in a cooked roux.

2.3. Influence of Heating on Physicochemical Properties of Ingredients in Roux Systems

2.3.1. Starch

Starch is insoluble in water but gelatinizes when heated with sufficient water. Moisture content and heating temperature are two dominant factors influencing the starch gelatinization process. Starch granules in a starch-based system with limited moisture content do not completely gelatinize during heating (Wang & Copeland, 2013). However, limited moisture still allowed some degree of gelatinization of the starch (Jin & Wang, 2020; Pérez-Santos et al., 2016; Qiu et al., 2015). When starch was heated in roux as an ingredient, the moisture level was insufficient to allow full starch gelatinization—only part of the starch granules gelatinized during the cooking process (Alvarez-Ramirez et al., 2018). Therefore, cooking still caused an irreversible change to the starch structure and functional properties.

2.3.1.1. Starch Thermal Degradation

Heating reduced the crystallinity of starch, and increasing heating temperature and heating time resulted in the loss of crystallinity to a more considerable extent (Oh et al., 2018; Sun et al., 2014). Alvarez-Ramirez et al. (2018) also reported that heating disrupted the crystalline structure of starch granules in a roux, which was associated with partial or total gelatinization. Furthermore, higher heating temperature resulted in starch granules gelatinized to a greater extent. Sun et al. (2014) concluded that the decrease in crystallinity of starch with limited moisture content might be due to:

1. the degradation of crystalline structure,

2. melting of the crystalline region, or
3. reorientation of the crystalline structure.

The crystalline structure of starch from different sources is generally divided into three types as A-, B-, or C-type. Commonly, cereal and legume starch have an A-type and C-type crystalline structure, while root and tuber starch have a B-type crystalline structure. Heating starch in a system with limited moisture resulted in alteration of crystalline type. The crystalline type of starch with A-type crystalline structure remained unchanged during heating (Qiu et al., 2015), while starch with B- type crystalline structure changed into A-type (Liu et al., 2019). Noticeably, the alteration of crystalline type was still impacted by the moisture content of the system; Chen et al. (2019b) compared the crystalline type of starch-water-oil mixtures with low (25 %) and high (50 %) moisture content after frying (180 °C, 20 min). They found that the typical X-ray diffraction peaks of B-type crystals of potato starch from the high moisture system disappeared, while the crystal type of samples with low moisture remained the same as the native sample.

Amylose-lipid complexes might form during cooking when starch and lipids are heated together, resulting in a V-type crystalline structure (Yang et al., 2019). When making a roux with butter and wheat flour, the moisture content in the roux system was sufficient to allow the formation of amylose-lipid complexes (Alvarez-Ramirez et al., 2018). However, whether amylose-lipid complexes form during roux making when the butter is replaced by oil, which means low moisture content in the roux system, has not been examined previously. When starch with low moisture content (25%) was fried, the V-type crystal peak was not apparent in the X-ray powder diffraction (XRD) profiles suggesting that the amylose-lipid complex did not form when a hydrated starch system was fried (Chen et al., 2019a; Chen et al., 2019b).

Molecular weight analysis showed that the molecular weight distribution of starch changed after cooking as macromolecules thermally broke down into smaller molecules (Chen et al., 2019a; Chen et al., 2018a; Shi et al., 2018). Chen et al. (2018a) reported that the portion of macromolecules (amylopectin) with a higher degree of polymerization was reduced while the portion of lower molecular weight molecules (amylose) with a lower degree of polymerization increased, and they claimed that such a change of the molecular weight distribution was because some amylopectin molecules decomposed to small molecules during frying (Lei et al., 2020; Shi et al., 2018; Yang et al., 2020; Yang et al., 2019). Increasing the cooking temperature or time accelerated amylose depolymerization and amylopectin debranching (Chen et al., 2019a).

2.3.1.2. Influence of Starch Thermal Degradation on Starch Properties

The crystalline content of starch is the primary factor determining its gelatinization temperature. Starch with a higher degree of crystallinity requires a higher temperature to start gelatinization (Liu et al., 2019). There was almost no difference between the thermal properties of normal wheat starch and wheat starch after dry heating at 120 °C for 30, 60, 90, and 120 min (Ozawa et al., 2009). However, when wheat starch was heated in a roux, the moisture content of a common roux (~15%) was sufficient to cause partial starch gelatinization. Alvarez-Ramirez et al. (2018) determined the change of enthalpy of wheat flour in a roux with changes in cooking temperature and time. They reported that higher cooking temperatures to prepare the roux resulted in a lower enthalpy of gelatinization. They also reported that there was a peak on the curve representing the change of gelatinization enthalpy as a function of time at low cooking temperature (80°C); only a small peak was observed at the higher cooking temperature (100°C). Alvarez-Ramirez et al. (2018) claimed that the change in gelatinization enthalpy was probably due to the combination of starch gelatinization and decomposition of Type I amylose-lipid complexes. In a limited water system, the gelatinization of starch followed a first-order kinetics model (Jin & Wang, 2020). An

increase of cooking temperature and cooking time resulted in a greater degree of starch gelatinization, which meant that more of the crystalline structure in the wheat starch was disrupted. Therefore, the gelatinization enthalpy of wheat starch in a roux decreased.

To best of my knowledge, pea flour has not been previously used to make roux. Thus, the thermal properties of pea flour in a roux are unknown. However, when starch from other botanical sources was heated with limited moisture, the enthalpy of starch gelatinization was reduced after heating (Lei et al., 2020; Liu et al., 2019; Oh et al., 2018; Sun et al., 2014), indicating that the crystalline structure of starch granules in a roux was disrupted during the cooking process. The inner structure of pea starch contains more water molecules compared to wheat starch (Ratnayake et al., 2002). Considering that roux is low-moisture system, higher water molecules in pea starch would promote pea starch gelatinization to a greater extent. Consequently, the enthalpy of pea starch in a roux is expected to decrease to a greater extent as pea starch molecular structure would be more easily affected by heating compared to wheat flour.

The profiles of the Rapid Visco Analyzer (RVA) analysis of starch provide an understanding of viscosity changes during gelatinization. The type of starch or flour used to make a roux, the heating conditions and the existence of oil are three significant effects impacting the pasting properties of a cooked roux.

Krasnow et al. (2011) re-heated the cooked roux with excess water and recorded the change of viscosity as a function of time. They found that cooking conditions had a significant effect on the change of viscosity. RVA is a common piece of analyzing equipment for studying the pasting viscosities of starch. While Krasnow et al. (2011) did not analyze pasting properties with an RVA, they used a rotational rheometer to measure the viscosity change of a slurry (mixture of cooked roux and excess water) at the re-heating phase, to investigate the pasting properties of the cooked roux. To the

best of my knowledge, no other studies have addressed the pasting properties of a cooked roux. However, similar systems to roux have been studied, as Ozawa et al. (2009) reported that heating wheat starch with limited moisture resulted in a decrease in peak viscosity compared to native starch. The peak viscosity decreased from 740 RVU to 694 RVU when wheat starch was dry heated at 120 °C for 30 min. When heating time increased at the same temperature (120 °C), the peak viscosity further decreased slightly. When starch was heated with limited moisture, interactions between starch molecules formed, resulting in resistance to swelling of starch granules. The subsequent swelling ability of starch granules was related to the viscosity development (Ai & Jane, 2015).

Oil had a substantial effect on the pasting properties of gelatinized starch, as the well-known amylose-lipid complex restricted swelling of starch granules and resulted in a reduction of pasting properties (Yang et al., 2019). Devi et al. (2020) added different types of fat or lipid (butter, hydrogenated fat, palm oil, coconut oil, groundnut oil and sunflower oil) to a wheat flour suspension and analyzed the pasting properties and found that all types of fat or oil added resulted in a decrease in the peak viscosity of wheat flour. Similar research was conducted by Desai et al. (2020) with adding salmon oil, cod oil, or coconut oil into the mixture of wheat starch and wheat gluten; they also reported the same result as the peak viscosity was significantly reduced by adding these three types of oil. However, there are no previous studies investigating the effect of starch/flour type and oil on the pasting properties of a cooked roux.

2.3.2. Protein

Keiko et al. (1979) first claimed that cooking of a roux resulted in the denaturation of gluten in the roux. They reported that the degrees of denaturation differed depending on the wheat flour protein fractions. Denaturation was minimal at 130 °C and only observed in the albumin, globulin and gliadin fractions. However, at 180 °C, not just

these fractions, but also the glutenin fraction, underwent significant denaturation. Alvarez-Ramirez et al. (2018) reported that cooking at 80 °C and 100 °C also resulted in a decrease in protein content suggesting that protein in a roux was disrupted by cooking.

Heating also induced the formation of amylose-protein-lipid complexes (Wang et al., 2020), but such amylose-protein-lipid complexes have not been examined previously in a roux. Quiles et al. (2012) reported the distribution of proteins in a formula of white sauce consisting of wheat starch, sunflower oil, soybean proteins and excess water. They claimed that the white sauce consisted of a continuous phase containing water, leached amylose, and soybean protein and a discontinuous phase (swollen starch granules). Heating protein in suspension resulted in protein aggregation (Megha & Grant, 1986; Peng et al., 2016; Rahaman et al., 2016; Sirtori et al., 2012; Stathopoulos et al., 2008). Heating-induced aggregation of protein was because of the covalent and non-covalent interactions in proteins such as disulphide bonds and hydrophobic effects (Singh & MacRitchie, 2004).

2.3.3. Lipids

Lipids in a roux have a critical role in the development of the flavor of a roux (Kato, 2005). Research on the oil distribution and heating effects on the oil in a roux is limited. However, some researchers investigated a starch-water-oil system that was similar to a roux system, and the results from their research could explain the heating effect on oil in a roux system. Quiles et al. (2012) claimed that most of the lipids in a white sauce was in the form of fat bubbles distributed in the continuous phase of a white sauce. Chen et al. (2018b) studied the oil distribution of a mixture consisting of limited moisture, starch, and oil, and they found that the majority of the oil was distributed near the surface of the starch granules (external oil) and part of the oil was absorbed into the starch granules (internal oil). Moreover, Chen et al. (2019a) studied the effect of heating

conditions on the mixture of water, starch, and oil. They found that increasing heating temperature and time contributed to an increase in the fraction of external oil. Therefore, most of the lipids in a roux are expected to surround ungelatinized or swollen starch granules. The free fatty acids can form complexes with amylose or protein (Wang et al., 2020). Amylose-lipid complexes in a roux system were located 1) in the continuous phase, 2) near the surface of the ungelatinized starch granules, and 3) inside the ungelatinized starch granules (Alvarez-Ramirez et al., 2018; Putseys et al., 2010; Wokadala et al., 2012).

Canola oil, which is a stable frying oil, does not hydrolyze and release free fatty acids at the cooking temperatures used to make a roux (Adjonu et al., 2019). Therefore, there would not be an effect on the starch or flour pasting properties through the amylose-lipid complex. However, the addition of oil to a wheat flour suspension was still observed to result in a reduction in the pasting viscosity of wheat flour (Devi et al., 2020; Desai et al., 2021). According to Chen et al. (2018b), vacuum filtration was unable to remove all frying oil from fried maize starch, and the residual oil mainly surrounded the surface of maize starch granules, and a small portion of the frying oil was absorbed and located near the surface of the maize starch granules. It can be assumed that if starch granules in a roux made from canola oil were separated by vacuum filtration, the residual oil would also surround the surface of the starch granules. When these starch granules in a cooked roux are resuspended with water and heated again (in an RVA analysis), the residual oil will interfere with water molecules entering the starch granules and so affect their gelatinization behavior. A later study from Chen et al. (2018a) also confirmed that the pasting viscosity of fried maize starch decreased because of a decrease in swelling power and leached amylose content. Effects such as oil restricting water entry to starch granules might explain why the addition of oil resulted in a decrease in pasting viscosity. Therefore, if roux is made from canola oil, the canola oil may not affect the pasting property of a roux through the amylose-lipid complex, but it is still possible that canola oil affects the pasting viscosity of roux by

affecting the interactions of water molecules with the starch granules in a roux, resulting in a reduction in the pasting viscosity of the roux.

2.4. Analysis Methods

2.4.1. Soxhlet Extraction

Soxhlet extraction is a technique used for a long time that has been a standard technique to determine a food product's crude fat content (Señoráns & Luna, 2012). There are two steps in a Soxhlet extraction, namely extraction of the fat from a sample and isolating the fat from the extraction solvent. During the extraction process, extraction solvent vaporizes from the distillation flask, gradually condenses at the thimble and extracts fat from the sample. When the extraction solvent reaches the overflow level, a siphon aspirates the thimble's solute and unloads the extraction solvent back into the distillation flask. The process repeats until the extraction process is completed (Luque de Castro & García-Ayuso, 1998). After the extraction process is complete, the mixture of extraction solvent and fat is heated at mild temperature allowing the extraction solvent to evaporate. The weight difference of the distillation flask before and after extraction is compared to calculate the content of extractable fat in the sample (Gfrerer et al., 2004).

2.4.2. Determination of Pasting Properties

2.4.2.1. Starch Gelatinization During Rapid Visco Analyzer (RVA) Analysis

The RVA is used to measure the pasting properties of starch-based systems (Figure 2.1). The mixture of starch granules and excess water is typically heated at a controlled heating temperature from 50°C to 95 °C and then with a fall back to 50°C, and the viscosity of the mixture is recorded as a function of time (Balet et al., 2019). The pasting properties of four types of starch/flour is shown in Table 2.1. Comparison of the pasting properties of these four types of starch/flour is discussed in the following section.

Figure 2.1 A generalized pasting curve of wheat starch showing the pasting parameters including pasting temperature, peak viscosity, peak time, breakdown, trough viscosity, setback, and final viscosity. This figure is adopted from Balet et al. (2019).

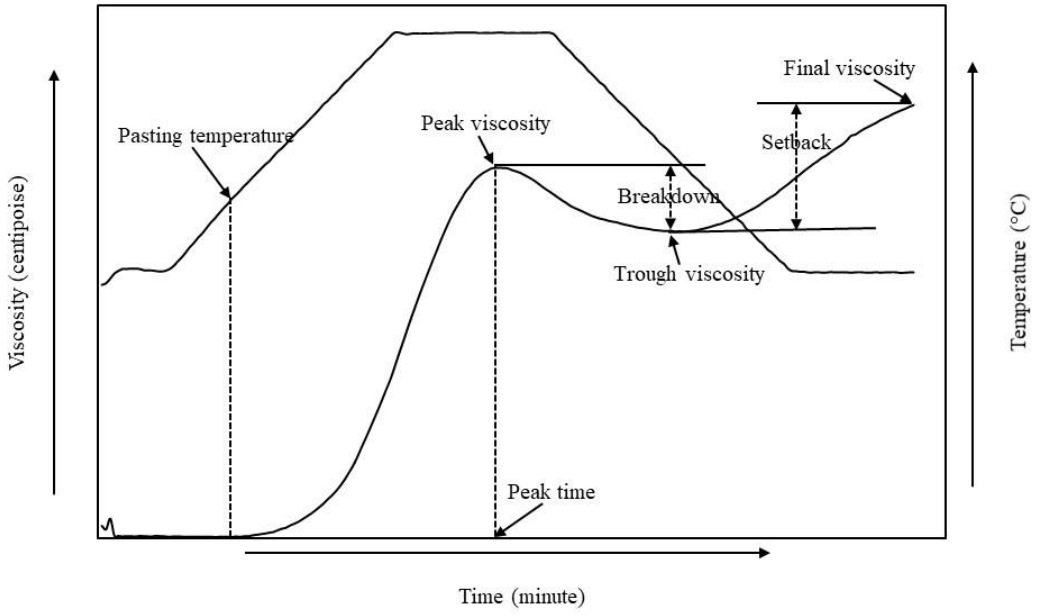


Table 2.1 The pasting properties of different types of starch and flour (wheat starch, wheat flour, pea starch, and pea flour).

	Type	Peak viscosity (cP)	Trough viscosity (cP)	Breakdown (cP)	Final viscosity (cP)	Setback (cP)	Pasting temperature (°C)	Reference
Wheat starch	Hard	4102	1400	2702	3276	1876	70.9	Li et al., 2013
	Soft	4893	1119	3846	3197	2078	65.5	Li et al., 2013
Wheat flour	Hard	1971	1125	N/A	2118	994	N/A	Garimella et al., 2015
Pea starch		736	688	48	933	245	78.2	Wang et al., 2011
		2526	1870	656	3355	1485	74.4	Liu et al., 2015
Pea flour		2215	1957	3487	258	1530	75.83	Qi et al., 2021

2.4.2.2. Pasting Properties of Wheat Flour, Wheat Starch, Pea Flour, and Pea Starch

Simsek et al. (2009) reported the peak viscosity (336-513 cP), trough viscosity (302.4-488.4 cP), breakdown (33.6-42 cP), final viscosity (597.6-862.8 cP) and setback viscosity (295.2-422.4 cP) of dry pea starch (*Pisum sativum L.*) from different cultivars grown in the USA. The pasting properties of pea starch isolated from cultivars grown in Canada was measured, but with a Brabender Viscoamylograph (BVA) (Ratnayake et al., 2001). Pea starch varieties grown in China showed higher pasting viscosities, indicating that the growing environment could affect the pasting properties of pea starch. (Liu et al., 2015). For wheat starch, the pasting properties are dependent are on the type of wheat starch. The pasting temperature, peak viscosity, trough viscosity, breakdown, final viscosity, and setback viscosity of hard wheat starch were 70.9 °C, 4102 cP, 1400 cP, 2702 cP, 3276 cP, and 1876 cP, respectively. And the pasting temperature, peak viscosity, trough viscosity, breakdown, final viscosity, and setback viscosity of soft wheat starch were 65.5 °C, 4983 cP, 1119 cP, 3864 cP, 3197 cP, 2078 cP, respectively (Li et al., 2013).

The pasting temperature of wheat starch is higher than pea starch because the high content of endogenous lipids of wheat starch restricts the swelling of starch granules and leaching of amylose (Yuan et al., 2021). The higher peak viscosity of wheat starch was due to the higher amylopectin content; as amylopectin is responsible for the starch swelling (Srichuwong & Jane, oct2007), peak viscosity increased with increased amylopectin content (Yuan et al., 2021). One noticeable characteristic of pea starch pasting was the low value of setback viscosity. The setback is related with the association and recrystallization of amylose during the cooling phase and an essential indicator for starch quality for storage and other purposes (Balet et al., 2019).

Pea and wheat flour also showed different pasting properties. Starch is the prime factor

impacting the pasting properties of flour, but other compounds such as proteins and lipids in flour also influence the pasting properties. In wheat flour, gluten could combine with water molecules under heat treatment, resulting in decreased available water for starch gelatinization (Morris et al., 1997). The proportion of monomeric or polymeric proteins was also reported to affect the peak viscosity of wheat flour (Singh et al., 2016).

2.4.2.3. Influence of Exogenous Oil on Pasting Properties

When considering the effect of exogenous oil on the pasting properties of starch or flour, the amylose-lipids complex should be noticed. Amylose-lipid complexes can form in the native starch, during processing, and in the RVA analysis. Native cereal starch contains around 1% of lipid, and so only 15% to 55% of amylose molecules formed a complex with lipids (Copeland et al., 2009).

Tang and Copeland (2007) found that the amylose-lipid complex was directly formed in RVA analysis when wheat starch was analyzed for its pasting properties. They observed that adding lipids resulted in reducing trough viscosity but increasing final viscosity. They also revealed that final viscosity would not continuously increase with rising lipid content, as lipid tended to self-associate compared to forming a complex with amylose. Blazek and Copeland (2008) found that the increase of final viscosity was positively correlated with the amylose content of wheat starch. Since fatty acids can form complexes with amylose through the aliphatic tail and associate with the protein through the negatively charged carboxyl group (Wang et al., 2020), ternary interactions among fatty acids, proteins, and starch had an influence on the pasting properties of starch. Wang et al. (2017) found that the addition of fatty acids increased the final viscosity and led to formation of a new peak during the cooling phase in the pasting profile of starch.

Chao et al. (2018) reported that the type of lipid was found to influence the formation of the maize starch-lipid complex during the RVA analysis by examining the peak in the cooling phase of the RVA profiles. The complexing ability varied among different added lipid sources (monomyristyl glycerol, monopalmityl glycerol, monostearyl glycerol, monooleyl glycerol, and monolinoleyl glycerol), and some of the lipids did not even affect the pasting profiles. However, the promoting effect of protein on interactions between starch and lipid was found to be not affected by lipid type. To further understand how starch-lipid complexes formed during pasting in the RVA, Chao et al. (2020) claimed that most of the starch-lipid complex formed during the setback stage.

Oil and starch are two common ingredients used together in the food industry. The peak, trough, breakdown, and final viscosity of wheat flour decreased when adding fat and oil to a flour-water suspension (Desai et al., 2021; Devi et al., 2020). The type of starch was also a factor governing the starch-lipid complex's effect on the pasting properties of starch, as Cai et al. (2021) reported a new peak only showed in the pasting profiles of wheat and non-waxy maize starch but did not show in other sources of starch.

3. Experimental Materials and Methods

3.1. Materials

Four types of starch or flour, including wheat flour, wheat starch, pea flour, and pea starch were used in this study. Wheat flour and wheat starch were purchased from the Archer Daniels Midland Agri-Industries Company (Decatur, Illinois, US) and MilliporeSigma Company (Oakville, Ontario, Canada), respectively. Alliance Grain Traders Food and Ingredients Company (Regina, Saskatchewan, Canada) provided the pea flour and pea starch.

The moisture content of wheat starch, wheat flour, pea starch, and pea flour was determined according to Approved Method 44-15.02 (AACC International, 2010). The protein contents of wheat flour and pea flour were determined according to Approved Method 46-13.01 (AACC International, 2010) using a Kjeldahl 1002 distilling unit (Tecator, Prabin and Co AB, Sweden). The particle size analysis of wheat starch, wheat flour, pea starch, and pea flour were analyzed using a Malvern Mastersizer 2000 (Malvern, U.K.) according to the method described by Davies-Hoes et al. (2017).

3.2. Sample Preparation

Each roux system was prepared from cooking the mixture of one of these four kinds of starch/flour and canola oil at one of five temperature levels (100, 116, 134, 153, and 175 °C) for come up time (15 min) plus one of five cooking time intervals (0, 4, 8, 12, and 16 min). Triplications of roux samples at each formulation were prepared.

Specific preparing steps are described as following. 15 g of starch or flour and 13.5 g of canola oil (starch/flour: canola oil = 10: 9) (Krasnow et al., 2011) were weighed and mixed by a vortex mixer and a steel spatula until the mixture was homogenous. The raw roux sample was mixed in a glass tube. The diameter and length of the glass tube

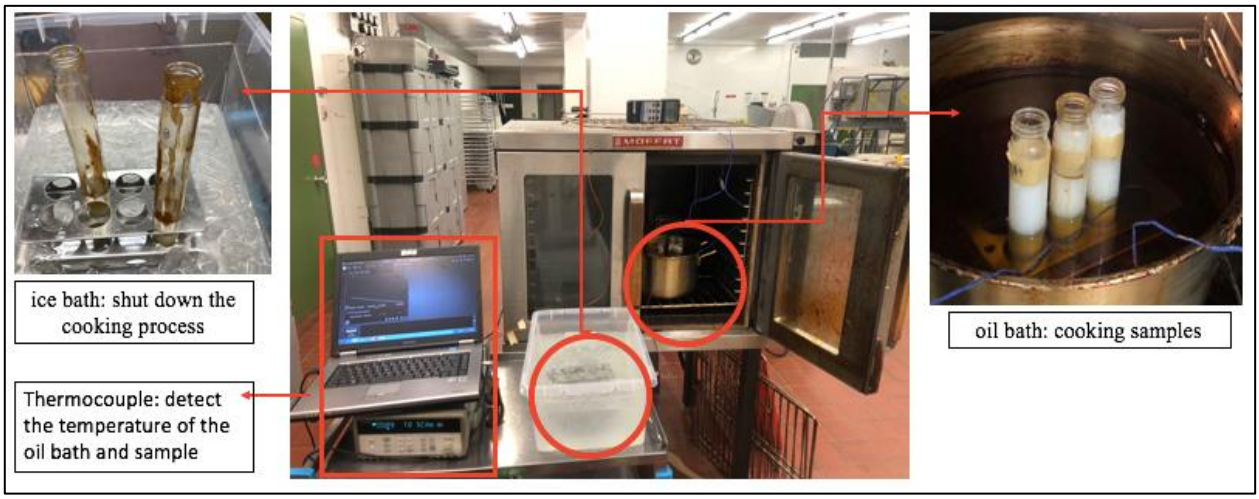
was 2.6 cm and 17.8 cm, respectively. A plastic dropper was used to wash down the residues of mixture of canola oil and starch or flour on the tube wall following the mixing step using 13.5 g of canola oil. The extra amount of canola oil also worked as a sealing material to separate the raw roux from atmospheric air to prevent adverse oxidation effects (Figure 3.1).

Five raw roux samples were transferred into an oil bath, which was previously heated to a specific target temperature in a convection oven. The cooking time consisted of two processes as pre-heating time (15 min), which was necessary as the roux samples needed time to reach target temperatures (see results), plus the target heating time (0, 4, 8, 12, or 16 min). Once heating samples for 15 (15 + 0), 19 (15 + 4), 23 (15 + 8), 27 (15 + 12), and 31 (15+16) minutes had occurred, one of these five samples was removed at random out of the oil bath and immersed into an ice bath quickly to stop the cooking process. Samples were kept in the ice bath for 30 minutes and then kept at ambient temperature. Figure 3.2 shows the experimental set-up. Foaming, that can be seen in Figure 3.2, was evident for flour samples as moisture evaporated during the initial stages of the cooking process.

A vacuum filtration technique, running for 5 minutes to create a negative pressure of 12.3 psi, was used to separate solid materials from excess canola oil after cooking these samples. The solid part of the roux was carefully collected from the filtration paper and stored in a beaker, which was then sealed with parafilm to prevent the dry sample from coming into contact with ambient conditions.

Figure 3.1 Typical sample showing how well the double amount of canola oil washed down residues and covered the roux prior to heating.

Figure 3.2 Roux cooking apparatus set-up.



3.3. Experimental Methods

3.3.1. Rapid Visco Analyzer (RVA) Analysis

Measuring the pasting properties of the cooked roux was done on a Rapid Visco-Analyser (RVA) (RVA-4, Newport Scientific Pty. Ltd., Warriewood, Australia). The pasting properties were measured according to the modified standard AACC International method 76-21.02 (AACC International, 1997). The pasting properties measured are essentially those of a low oil-content roux sauce since excess water is added to create a sauce from the roux (Quiles et al., 2012). Only one RVA analysis was conducted for each replication of a treatment.

When a starch/flour-oil system with low moisture content was subjected to heating at high temperature, the moisture in the system evaporated rapidly and the moisture content of the starch was very low after frying (Chen et al., 2019b; Chen et al., 2019c). The moisture content for all cooked roux was recognized as essentially 0% due to evaporation of water during the previous cooking process. 3.115 g of the vacuum-filtered roux and 25.385 g of deionized water at ambient temperature were mixed in a standard aluminum RVA canister.

The overall RVA measuring process lasted for 13 min. The rotation speed of the paddle inserted in the RVA canister was kept at 160 rpm throughout the measuring process except for a period of 10 s at the start, when the rotation speed was as high as 960 rpm, which allowed the slurry to mix sufficiently. The slurry was heated from 50 °C to 95 °C over 4.7 min and kept at 95 °C for 2.5 min before dropping back to 50 °C over 3.8 min. The slurry was held at 50 °C for 2 min.

The change of the viscosity of the slurry (sauce) was recorded, and the viscosity was expressed in centipoise (cP). The RVA parameters included: 1) peak viscosity (PV, maximum viscosity at 95 °C), 2) trough viscosity (TV, minimum viscosity at 95 °C), 3)

breakdown (peak minus trough viscosity), 4) final viscosity (FV, viscosity at the end of the holding period), 5) setback (final minus trough viscosity), 6) peak time, and 7) pasting temperature.

When conducting the RVA analysis, the viscosity of the mixture (wheat starch, wheat flour, pea starch, or pea flour in cooked roux with excess water) was measured and recorded every 4 s. There were 195 points in total recorded during the 13 minutes of measuring in the RVA analysis. Therefore, these 195 points on the pasting curves represented the viscosity development and retention of the roux sauces. These points associated with viscosity development were used to calculate the Euclidean distance for specific roux treatments relative to control samples. Euclidean distance was calculated based on the analysis of the results of the pasting properties of raw starch/flour in comparison to the pasting properties of a cooked roux. Firstly, the difference between the raw starch/flour and a cooked roux at each time point (from one of these 195 points on the pasting curves) was calculated. Then the difference was squared, followed by summing all the squares of difference. The sum of squares of the difference was determined as the Euclidean distance representing the extent of the shift of the pasting curve of the roux cooked at a specific cooking temperature and time compared to the pasting curve of the raw starch/flour used to make the roux. The following equation shows how Euclidean distance was calculated:

Euclidean distance (ED)= $\sum_{i=195}(\eta_{raw\ starch/flour,i} - \eta_{roux,i})^2$

Two sets of control RVA pasting profiles were analyzed. Firstly, the pasting properties of raw wheat flour, wheat starch, pea flour, and pea starch were measured according to AACC International method 76-21.02 (AACC International, 1997). Raw starch or flour (3.115 g) was mixed with 25.385 g of distilled water (the same ratio as for the cooked roux to distilled water) and the pasting parameters and pasting curves were measured. RVA analysis was done in triplication.

Secondly, the pasting properties of a mixture of canola oil and either raw wheat flour, wheat starch, pea flour, or pea starch were also examined according to AACC International method 76-21.02 (AACC International, 1997). The total weight of the starch/flour-canola oil mixture was 3.115 g, and this was mixed with the 25.385 g of distilled water. The ratio of canola oil to starch or flour was determined based on the results for determination of the crude fat content of the cooked roux. The detailed ratio of starch/flour and canola oil is shown in Table 4.6. Each measurement was done in triplicate.

3.3.2. Crude Fat Content Determination

Crude fat content determination of the roux after cooking and vacuum filtration was conducted by the Soxhlet extraction technique using a Soxhlet extraction apparatus (Gfrerer et al., 2004; Luque de Castro & Priego-Capote, 2010). Thimbles containing 3.5 g of vacuum-filtered roux were inserted in the Soxhlet units. Hexane was used as extraction solvent. The extraction process lasted for 16 h. The crude fat contents of samples were calculated based on comparing the weight of flasks before and after oil extraction.

3.3.3. Differential Scanning Calorimetry (DSC) Analysis

DSC analysis was carried out to determine the thermal properties of the cooked roux samples made from wheat starch, which had been cooked at one of the temperatures 100, 134, and 175 °C for either 0, 8, or 16 min. DSC analysis of samples was carried out using a DSC Q2000 (TA Instruments, New Castle, DE, USA) following the procedure of Zhang et al. (2019), with some modification. An analytical balance (± 0.0001 g) was used to weigh 2.0 mg of the vacuum-filtered roux. The sample was transferred into a DSC pan followed by careful addition of 8 mg of deionized water with a pipette. The DSC pan was then hermetically sealed and allowed to stand for one

hour before measurement, so that the mixture inside reached equilibrium. An indium DSC pan containing the same amount of deionized water was repeatedly used as the reference pan for calibrating the DSC analysis for all samples (Morikawa & Nishinari, 2000).

The slurry was heated from 40 °C to 95 °C, the heating temperature range was applied by referencing to the study of Morikawa & Nishinari (2000), at a heating rate of 10 °C/min. Thermal characteristics of samples measured were the gelatinization onset temperature (T_o), the peak temperature (T_p), and the conclusion temperature (T_c) for the peak (Oh et al., 2018), and the enthalpy change (ΔH, J/g) was obtained and analyzed using the TA Universal Analysis 2000 software.

3.3.4. Light Microscopy Analysis

The microscopical images of the roux made from wheat starch cooked at one of the temperatures 100, 134, and 175 °C for 0, 8, or 16 min (the same treatment as in the DSC analysis) were obtained using a polarizing microscope (Zeiss, Oberkochen, Germany). The steps to prepare the samples subjected to the light microscopy analysis were those of Xiao et al. (2020). First, around 1 mg of roux samples was dispersed in 20 mL of distilled water with stirring for 2 min. Then, a drop of solution containing the roux sample was transferred onto a glass slide by a plastic dropper, which was then carefully covered by a cover glass, and quickly examined microscopically under polarized light. The microscopical images of the roux samples were recorded by a cellphone, which was directly taken from the eyepiece of the microscope.

3.4. Statistical Analysis

Individual dependent variables were analyzed as a three-way analysis of variance (ANOVA) according to the Proc Mixed procedure of JMP (Version 15) (Toomer et al.,

2020). The types of starch/flour, cooking temperature, and cooking time were fixed effects. Least significant difference (LSD) analysis with setting the significance level at $p<0.05$ was conducted to compare the mean values of treatments. An example figure of the JMP code used for the analysis is shown in Appendix A.

4. Results and Discussion

4.1. Characteristics of Raw Starch or Flour

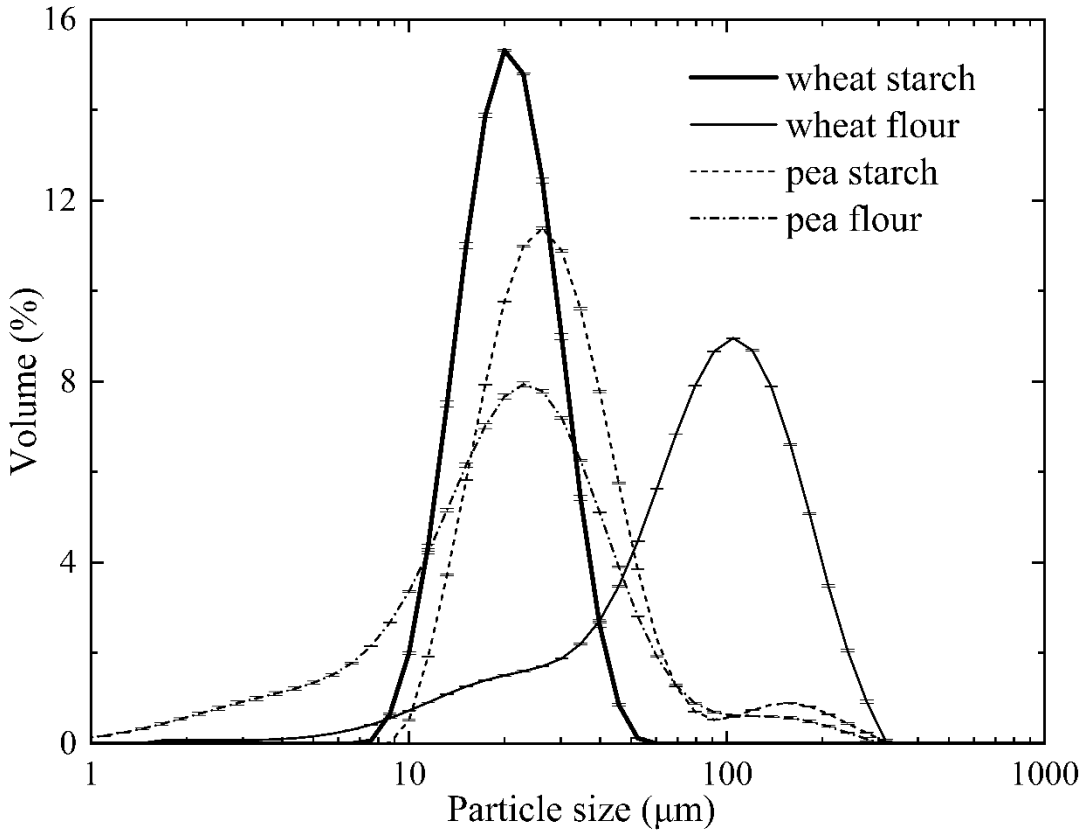
4.1.1. Moisture and Protein Content

The moisture content of wheat starch, wheat flour, pea starch, and pea flour was 12.6 ± 1.1, 12.5 ± 0.0, 7.5 ± 0.3, and 9.8 ± 0.5 %, respectively. The moisture content of wheat starch commonly ranges between 8 to 11 % (Ratnayake et al., 2002) which was slightly lower than the moisture content in the present study. The crude protein content of the wheat flour and pea flour was 11.4 ± 0.4 and 17.5 ± 0.2 %, respectively. Pea flour contained higher protein content than the wheat flour. The protein content in wheat and pea starch were very low as 0.44–0.63 % and 0.52–0.70 %, respectively (Ratnayake et al., 2002; Shevkani et al., 2017), and so it was not determined in the present study.

4.1.2. Particle Size Distribution

The results of an analysis of the particle size distribution for the experimental materials is shown in Figure 4.1. It can be seen that the particle size distribution of starch was more concentrated over a narrower size compared to flour. Previous studies (Kim & Qin, 2014; Valencia et al., 2015) had reported that the particle size distribution of flour was broader than that of starch. Valencia et al. (2015) claimed that fibre lead to the aggregation of starch granules in the flour. The particle size of wheat starch and pea starch mainly ranged from 10 to 50 μm which was similar to the particle size of wheat and pea starch reported in previous studies (Ratnayake et al., 2002; Shevkani et al., 2017).

Figure 4.1 Particle size distribution of wheat starch, wheat flour, pea starch, and pea flour (with representative error bars for three replicates).



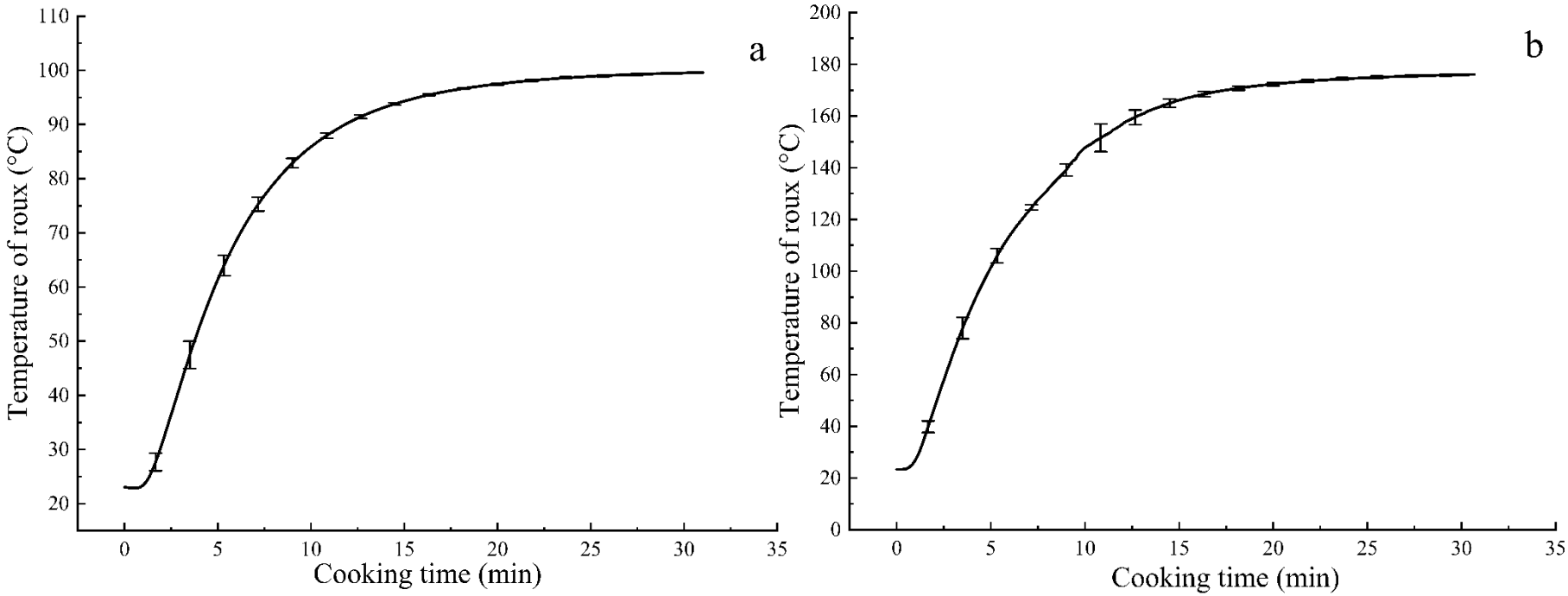
4.2. Heating Rate

Different heating methods to determine the effect of cooking conditions on the properties of roux have been applied in previous studies (Alvarez-Ramirez et al., 2018; Krasnow et al., 2011). To determine the come-up time for a roux to reach target cooking temperature levels, the increase of roux temperature as a function of cooking time was measured. The roux sample (made from wheat starch and canola oil) was placed in an oil bath previously set at 100 °C (Figure 4.2a) or 175 °C (Figure 4.2b). As can be seen in Figure 4.2, there was a rapid increase in the roux temperature in the initial 15 min of heating. The temperature of the roux only slightly increased after this come-up time. Both heating at 100 °C and 175 °C had the same trend as the temperature of the roux rapidly increased during the come-up time and then slightly increased to approach the oil bath temperature after the come-up time.

As 100 °C and 175 °C were the lowest and highest cooking temperatures, respectively, for the study, the increase of roux temperature at 116, 134, or 154 °C was expected to follow the same trend as 100 °C and 175 °C. In other words, the roux temperature of samples was expected to rapidly approach the target cooking temperatures during the come-up time and then to slightly increase.

Based on the results of Figure 4.2, all roux samples were first heated for 15 min, which was recognized as the come-up time, followed by continuing to heat the roux for the selected target time (0, 4, 8, 12, or 16 min). The target cooking time was chosen according to Jin and Wang (2020) with modification based on preliminary experiments. Jin and Wang (2020) conducted a kinetic study of a starch-based system with limited moisture content directly heating samples to target heating temperatures and holding for some time. The heating times chosen in the present study were 0, 4, 8, 12, and 16 min and these correspond to total heating times of 15, 19, 23, 27, and 31 min, respectively.

Figure 4.2 Increase of temperature of roux made from wheat starch and canola oil as a function of cooking time. The roux was cooked in an oil bath pre-set at 100 °C (a) or 175 °C (b). Each temperature curve was constructed by averaging the three replications.



4.3. RVA Analysis

4.3.1. Pasting Curves

The purpose of the analysis of pasting properties was to determine the effect of starch/flour type (flour or starch, wheat or pea), cooking temperature, and cooking time on the pasting properties of the cooked roux. The pasting curves of wheat starch and wheat flour in a roux cooked at different temperatures (100, 116, 134, 154, and 175 °C) for a series of cooking times (0, 4, 8, 12, and 16 minutes) are shown in Figure 4.3-4.4 and for pea starch and pea flour in Figure 4.5-4.6. Detailed RVA pasting curves with roux cooking times for each temperature are shown in Appendix B and detailed pasting properties are shown in Appendix C.

The pasting viscosity of the roux made from wheat starch was not affected by the cooking time when the cooking temperature was lower than 134 °C, but significantly decreased when the cooking temperature was 154 °C or higher. The pasting viscosity of the roux made from pea starch was slightly affected by the cooking time when the cooking temperature was at 100 and 116 °C, but was pronounceably enhanced when the cooking temperature increased from 116 to 134 °C. Moreover, the roux made from pea starch completely degraded when the cooking temperature was as high as 175 °C, but pasting viscosity still showed time dependence at 175 °C. When the roux made from wheat flour was cooked at 100, 116, or 134 °C, the pasting viscosity was slightly enhanced by cooking; such enhancement in pasting viscosity was more noticeable when the cooking temperature was at 134 °C.

For the roux made from pea flour, the pasting viscosity was also enhanced by cooking at 116 and 134 °C; this increase in pasting viscosity showed time-dependence as pasting viscosity generally increased as the cooking time increased from 0 to 12 minutes. When cooking temperature further increased to 154 and 175 °C, the pasting viscosity was enhanced when the cooking time was 0 minutes, then decreased as the cooking time

further increased from 4 to 16 minutes, regardless of whether the roux was made from wheat or pea flour. Noticeably, the pasting viscosity of wheat flour-roux and pea flour-roux had a pronounced time-dependence at these higher temperatures.

What can be concluded from above was that: 1) the type of starch or flour significantly affected the pasting profiles of a roux; 2) when the roux was cooked at lower cooking temperatures, the pasting viscosity was enhanced by the cooking, but when the roux was cooked at a higher cooking temperature, the pasting viscosity was diminished by cooking. Krasnow et al. (2011) reported that cooking temperature at moderate temperatures (120 and 140 °C) increased the thickening power of roux made from wheat flour and soybean oil compared to the thickening ability of uncooked roux, while higher cooking temperatures (>160 °C) resulted in a significant reduction of roux thickening power. The current experiment confirmed their findings and further revealed that the starch or the flour used to make the roux was responsible for the change of thickening power.

Figure 4.3 Pasting curves of wheat starch in the cooked roux.

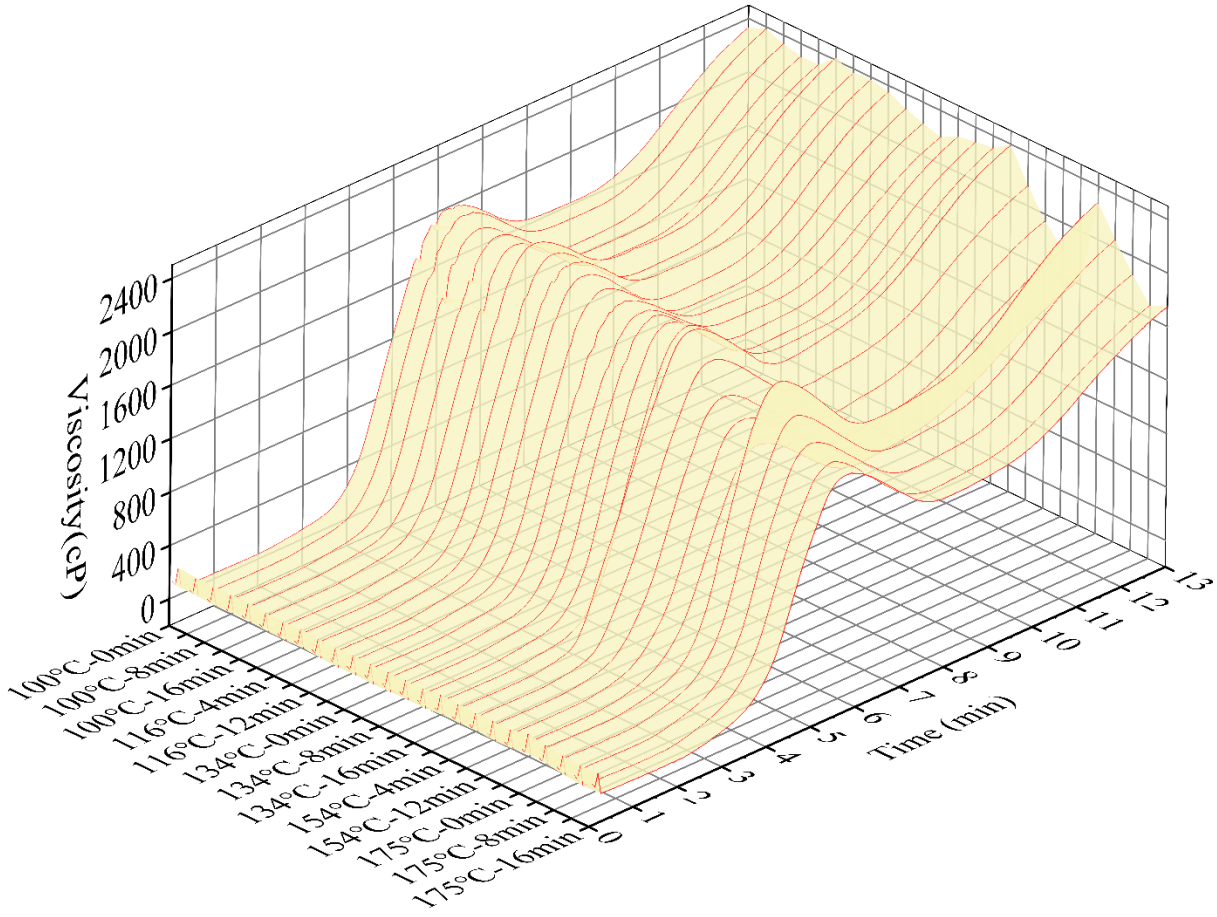


Figure 4.4 Pasting curves of wheat flour in the cooked roux.

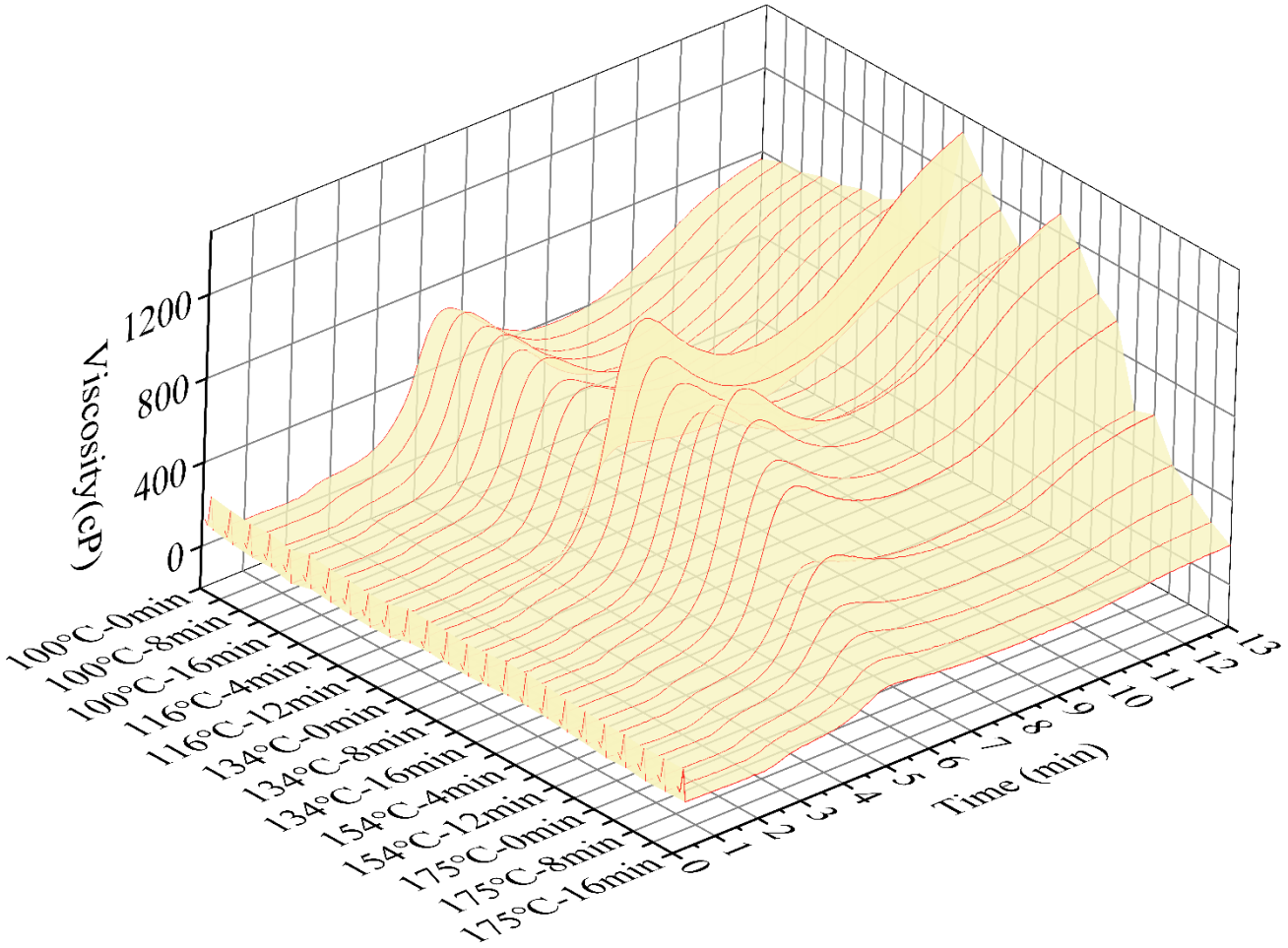


Figure 4.5 Pasting curves of pea starch in the cooked roux.

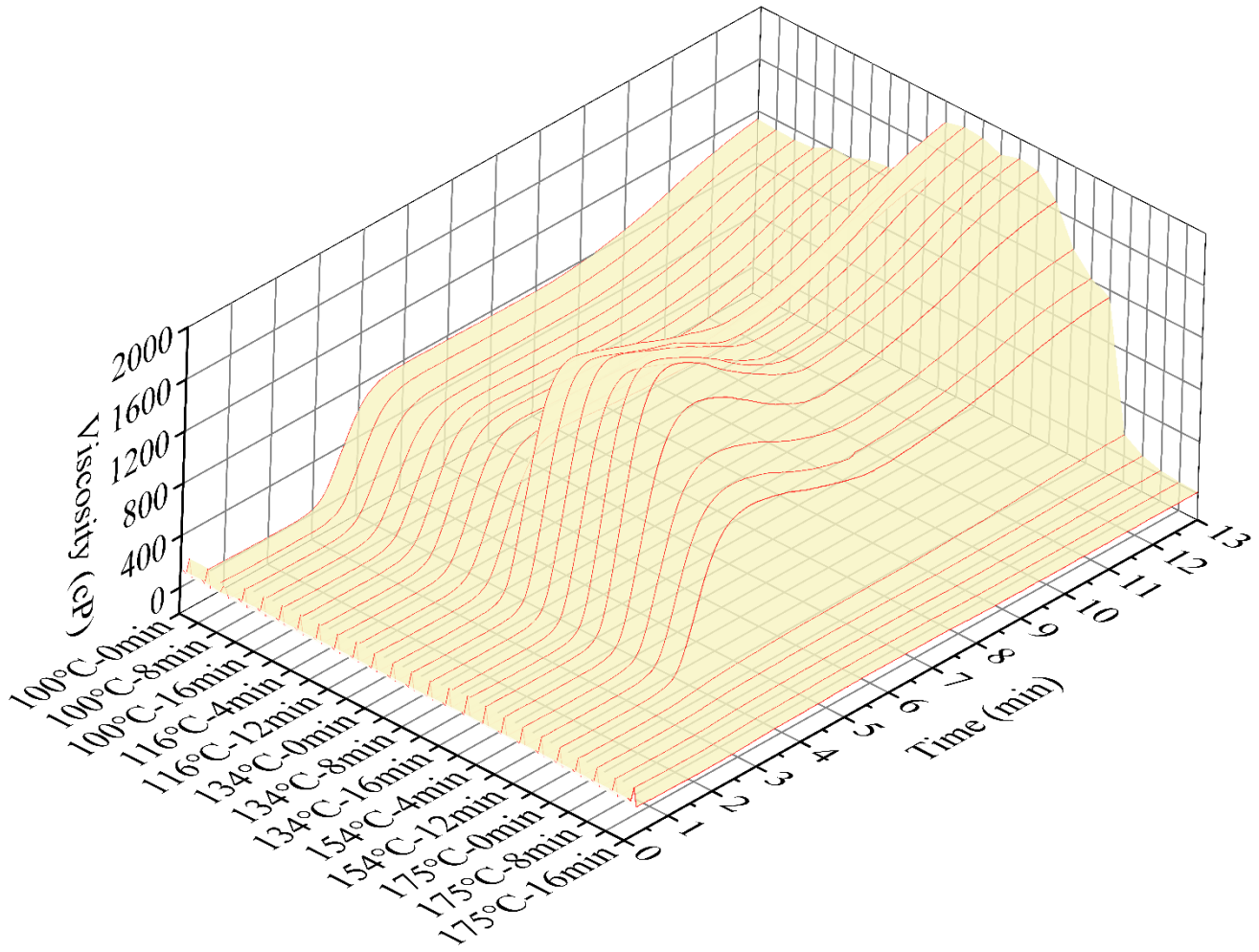
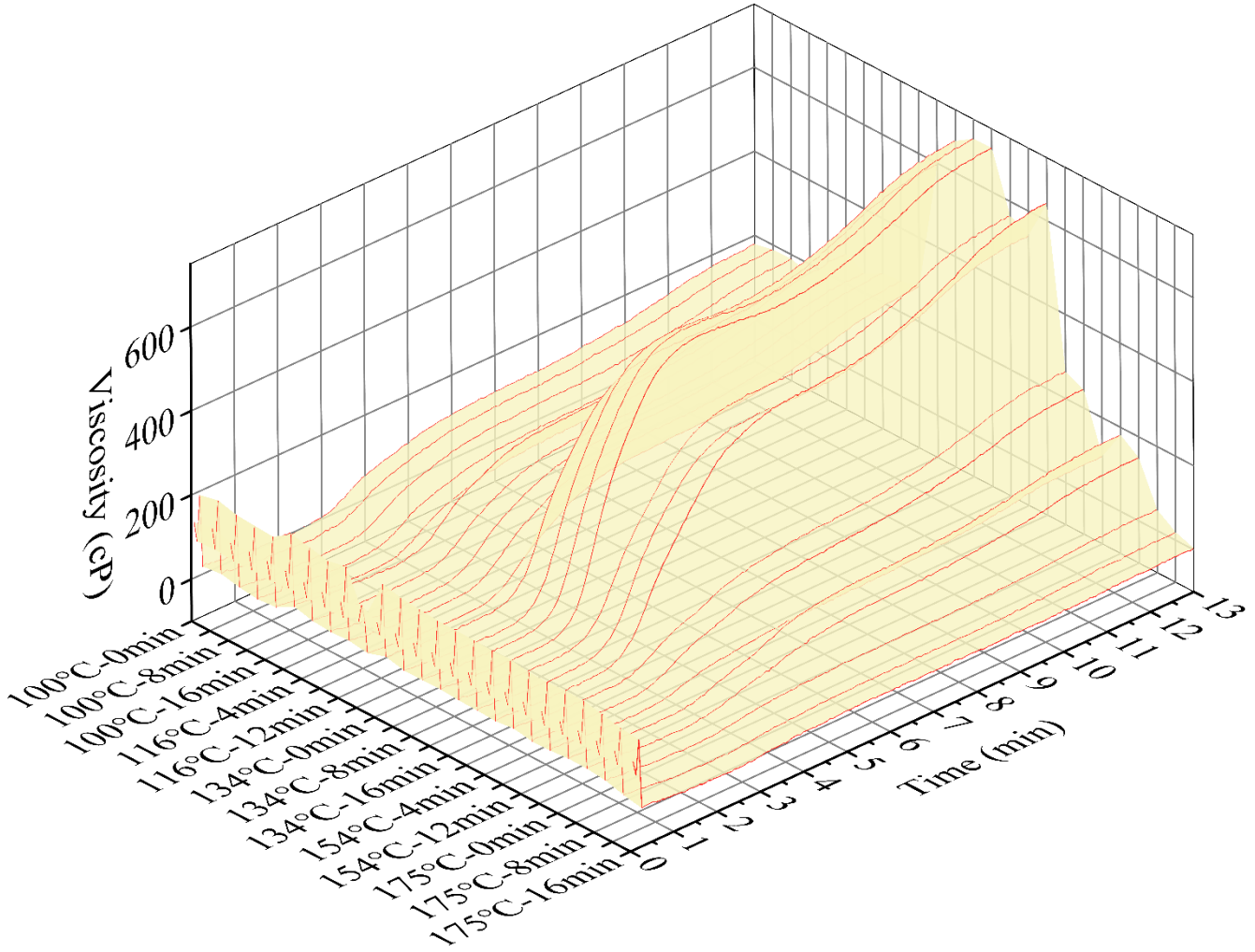


Figure 4.6 Pasting curves of pea flour in the cooked roux.



4.3.2. Peak Viscosity, Trough Viscosity, and Setback

The base material used to make the roux had a significant (p<0.0001) (Table 4.1) impact on the peak viscosity of the cooked roux (Figure 4.7 and Figure 4.8). Wheat starch (Figure 4.7a) and pea flour (Figure 4.8b) in cooked roux showed the highest and lowest peak viscosities, respectively. Wheat starch in the cooked roux had higher pasting viscosities compared to wheat flour because the proportion of starch in wheat starch was higher than in wheat flour (Juhász & Salgó, 2008). On the other hand, wheat starch or flour showed higher pasting viscosities than pea starch or flour, and this could be attributable to the lower amylopectin content in pea starch or flour as amylopectin was responsible for the viscosity development during the heating stage in the RVA analysis (Morris et al., 1997).

Table 4.1 Statistical analysis (P value) of experimental design parameters on the peak viscosity, trough viscosity, and setback of the RVA profiles of starch or flour from the cooked roux. The effects highlighted were statistically significant at $P<0.05$. (A more traditional ANVOA table is given in Appendix D)

Effect	P value		
	Peak viscosity	Trough viscosity	Setback viscosity
Starch/flour type	<0.0001	<0.0001	<0.0001
Cooking temperature	<0.0001	<0.0001	<0.0001
Cooking time	0.0253	0.0049	0.0977
Starch/flour type*cooking temperature	<0.0001	<0.0001	<0.0001
Starch/flour type*cooking time	0.9989	0.9937	0.9987
Cooking temperature*cooking time	0.1834	0.1526	0.6809
Starch/flour type*cooking temperature*cooking time	1.0000*	1.0000*	1.0000*

* Because the statistical software (JMP, Version 14) kept four significant digits after the decimal point, the P values were approximately equal to 1.0000.

Figure 4.7 Peak viscosity of wheat starch (a) and wheat flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.

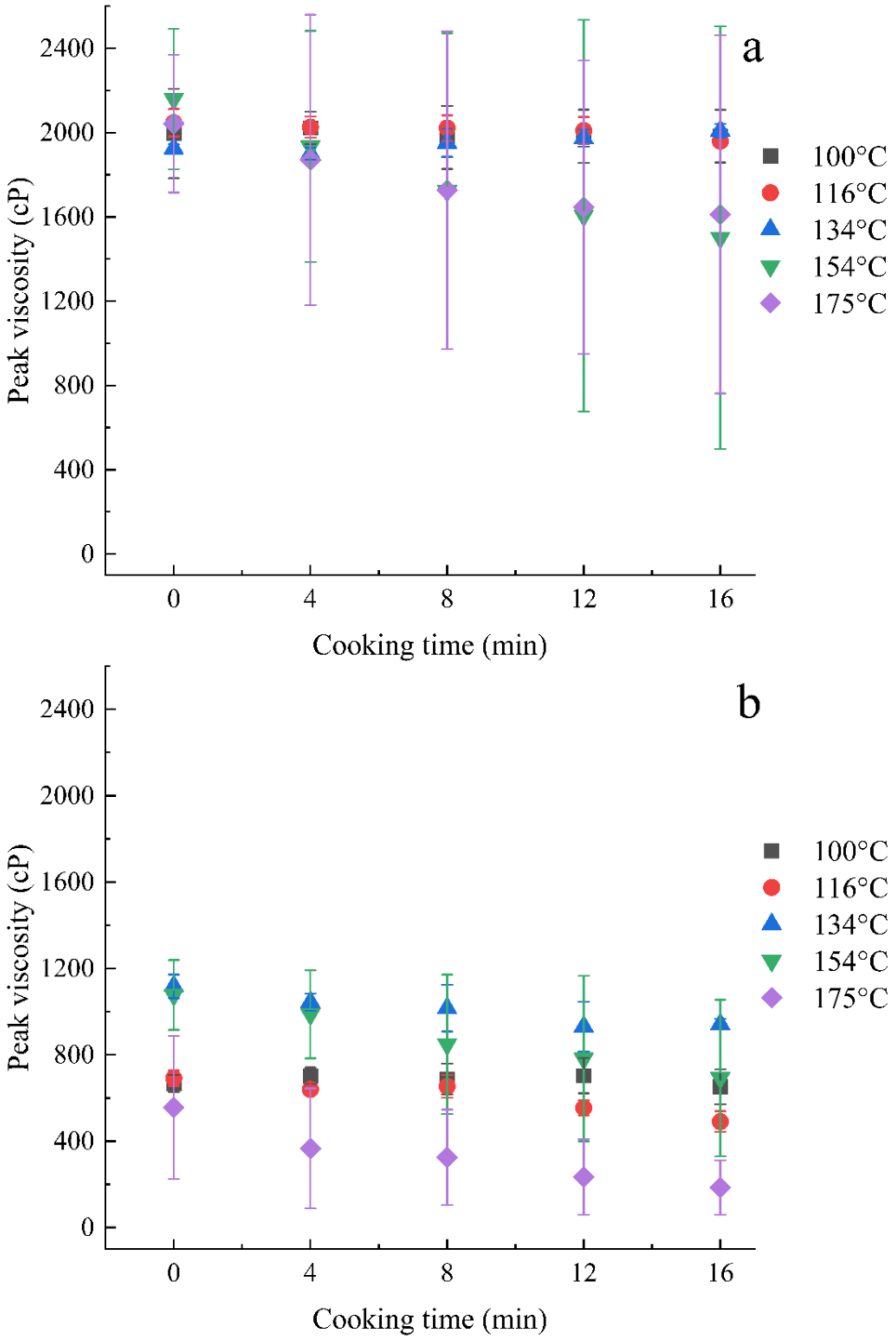
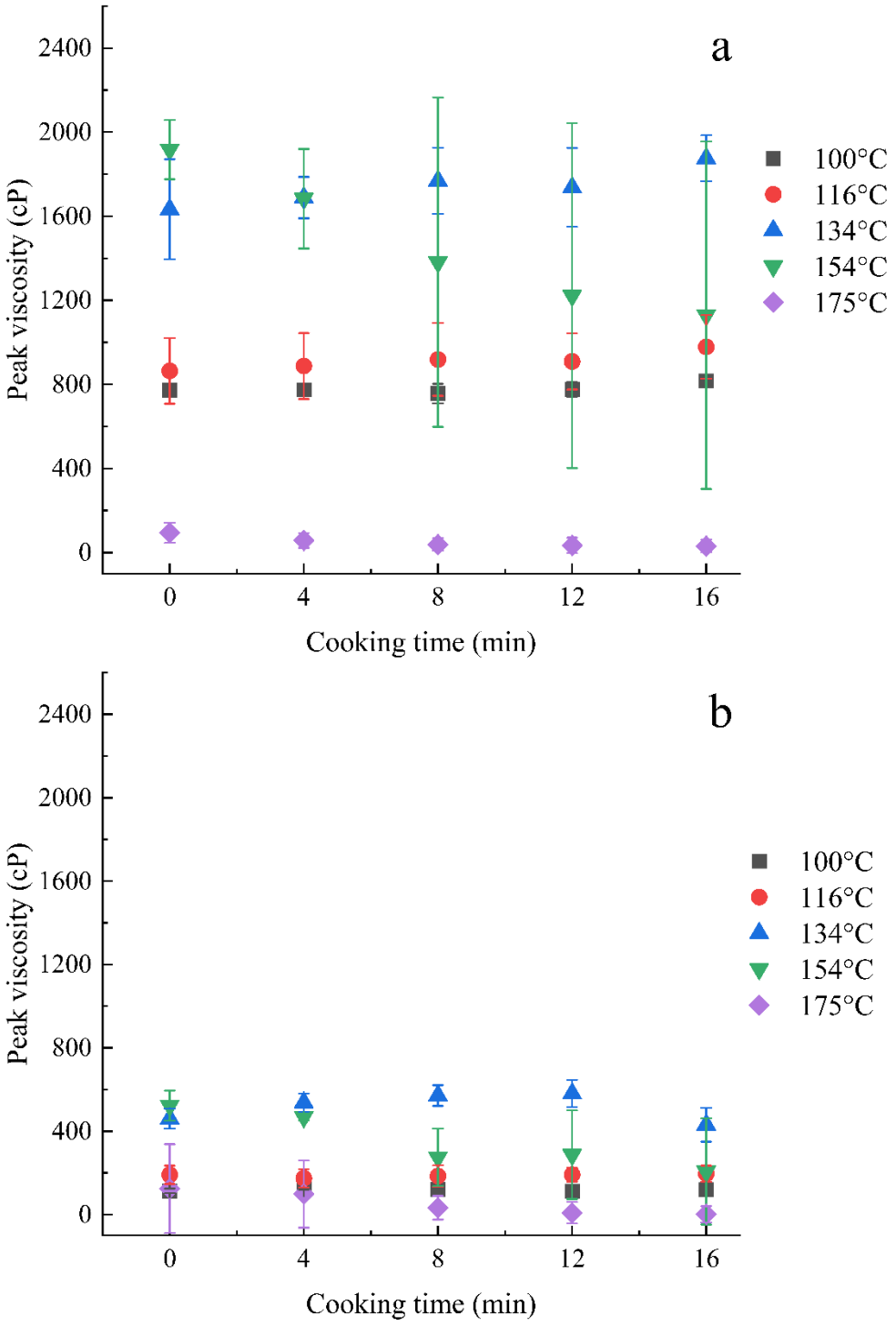


Figure 4.8 Peak viscosity of pea starch (a) and pea flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.



Cooking temperature significantly (p<0.0001) altered the peak viscosity of all four types of roux, as shown in Table 4.1. For wheat starch (Figure 4.7a), when cooking temperature increased from 100 to 116 °C, the average peak viscosity over five cooking times (0, 4, 8, 12, and 16 minutes) increased significantly from 1992 to 2013 cP. Conversely, the average peak viscosity over the same cooking times continuously decreased to 1779 cP with further increase in the cooking temperature to 175 °C. For wheat flour, pea starch, and pea flour, the average peak viscosities over five cooking times peaked when the cooking temperature was at 134 °C. Cooking time significantly (p<0.05) changed the peak viscosity of the cooked roux (Table 4.1). At 100 °C and 116 °C, the peak viscosity of the four kinds of roux systems was only slightly affected by the cooking time. At 134 °C, the peak viscosity of the roux made from wheat flour, pea starch, and pea flour decreased, while the peak viscosity of wheat starch slightly increased, with an increase in cooking time. At 154 °C and 175 °C, increasing cooking time led to a decrease in peak viscosity for all types of starch and flour in a cooked roux. Peak viscosity has been associated with the ratio of amylose to amylopectin content in the starch granules (Juhász & Salgó, 2008; Zhu, 2018).

In short, the peak viscosity of the cooked roux was not statistically affected by cooking at low cooking temperature levels but constantly decreased with an increase of cooking time at the two higher cooking temperatures.

The trough viscosities of the cooked roux were statistically different (p<0.0001) among the four types of roux in the current study, as shown in Figure 4.9 and Figure 4.10. Cooking temperature (p<0.0001) significantly affected the trough viscosity of the cooked roux (Table 4.1). The influence of cooking temperature on trough viscosity was the same as the effects on the peak viscosity, as the averaged trough viscosity over five cooking times of wheat flour, pea starch, and pea flour peaked at 134 °C and for wheat flour peaked at 116 °C. Cooking time significantly affected the trough viscosity (Table 4.1). What should be noticed was that the effect of cooking time was more significant

on trough viscosity compared to peak viscosity (Table 4.1). To be specific, increasing cooking time from 0 to 16 minutes brought about a greater decrease in the trough viscosity than the decrease of trough viscosity due to cooking temperature. This could be due to the different degree of integration of starch granules during roux making (Chen et al., 2018a). In general, low cooking temperatures (100 and 116 °C) had a limited impact on the trough viscosities of the cooked roux. However, trough viscosity decreased with the increase of cooking time at high temperatures (134, 154, and 175 °C). Shevkani et al. (2017) reported that starch with a higher proportion of long amylopectin chains (DP>36) showed higher trough viscosities, and this was because of the ability of amylopectin molecules to form intermolecular linkages with amylose. Lei et al. (2020) reported that dry heating of maize starch at 190 °C resulted in an increase of amylose content but a decrease in amylopectin content. Therefore, it might be speculated that the higher cooking temperatures (134 °C and 175 °C) applied in the present study caused the breakdown of amylopectin molecules resulting in the reduction of amylopectin content but increase in amylose content in the starch granules.

Figure 4.9 Trough viscosity of wheat starch (a) and wheat flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.

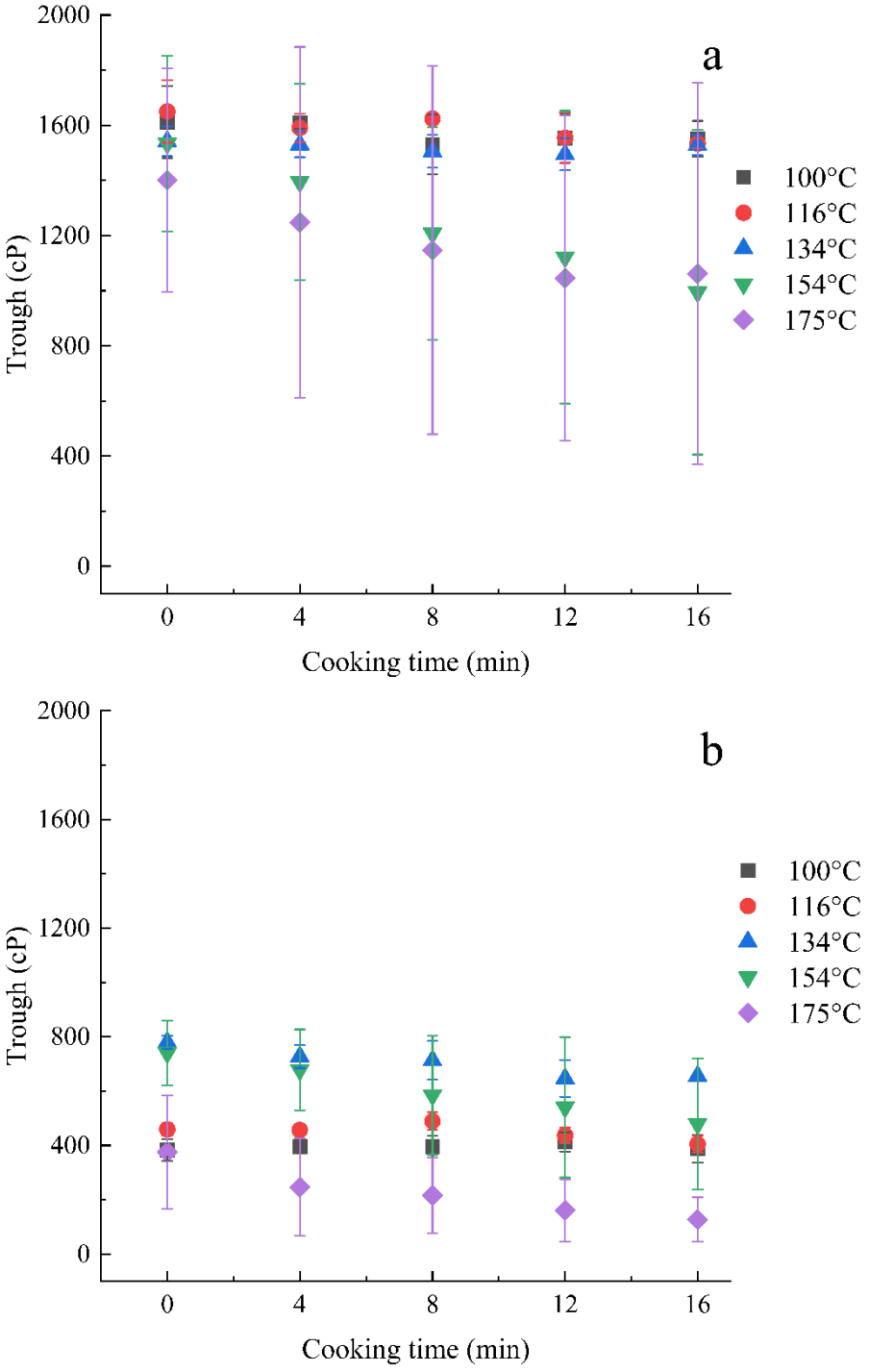
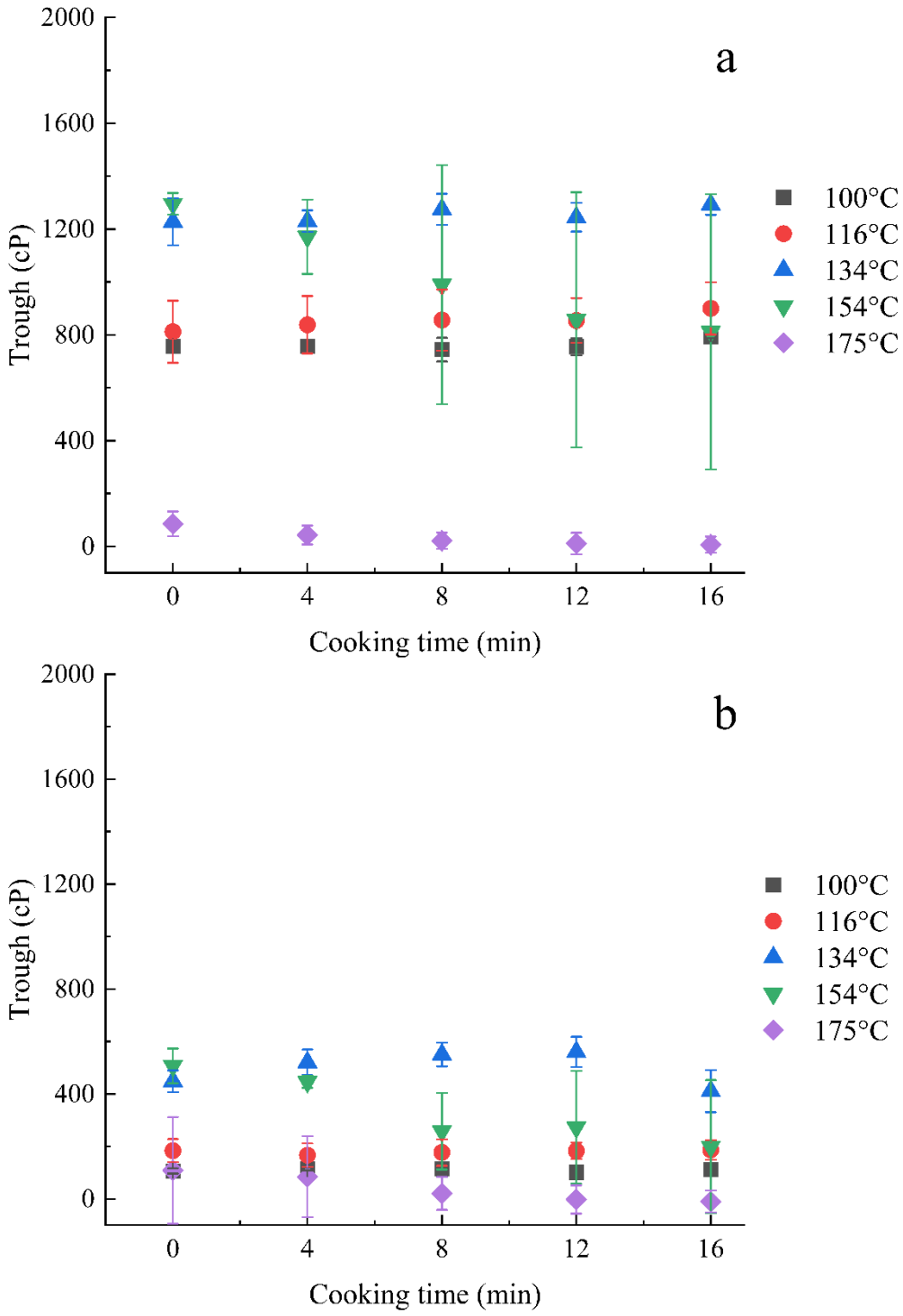


Figure 4.10 Trough viscosity of pea starch (a) and pea flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.



The setback values for the cooked roux are shown in Figure 4.11 and Figure 4.12. Starch/flour type and cooking temperature had a highly significant (p<0.0001) impact on the setback, as for peak and trough viscosities. However, cooking time had no effect on setback values. Cooking temperature had a different influence on setback depending on the starch/flour type. It can be seen in Figure 4.12a that the setback of pea starch in the cooked roux was not affected at the two lower cooking temperatures (100 and 116 °C), but the setback increased when cooking temperature was 134 °C and continuously decreased when the cooking temperature was further increased to 175 °C.

Figure 4.11 The setback of wheat starch (a) and wheat flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.

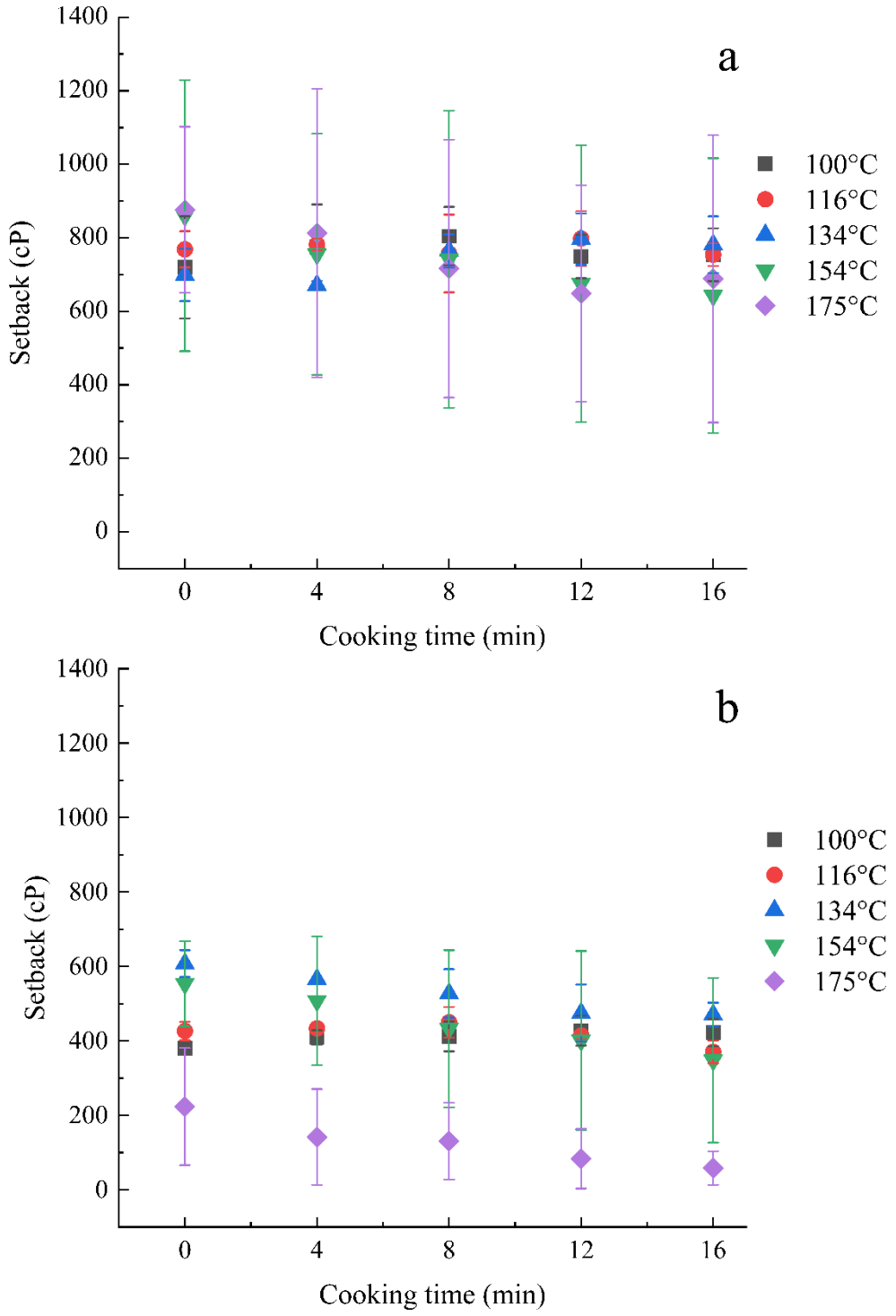
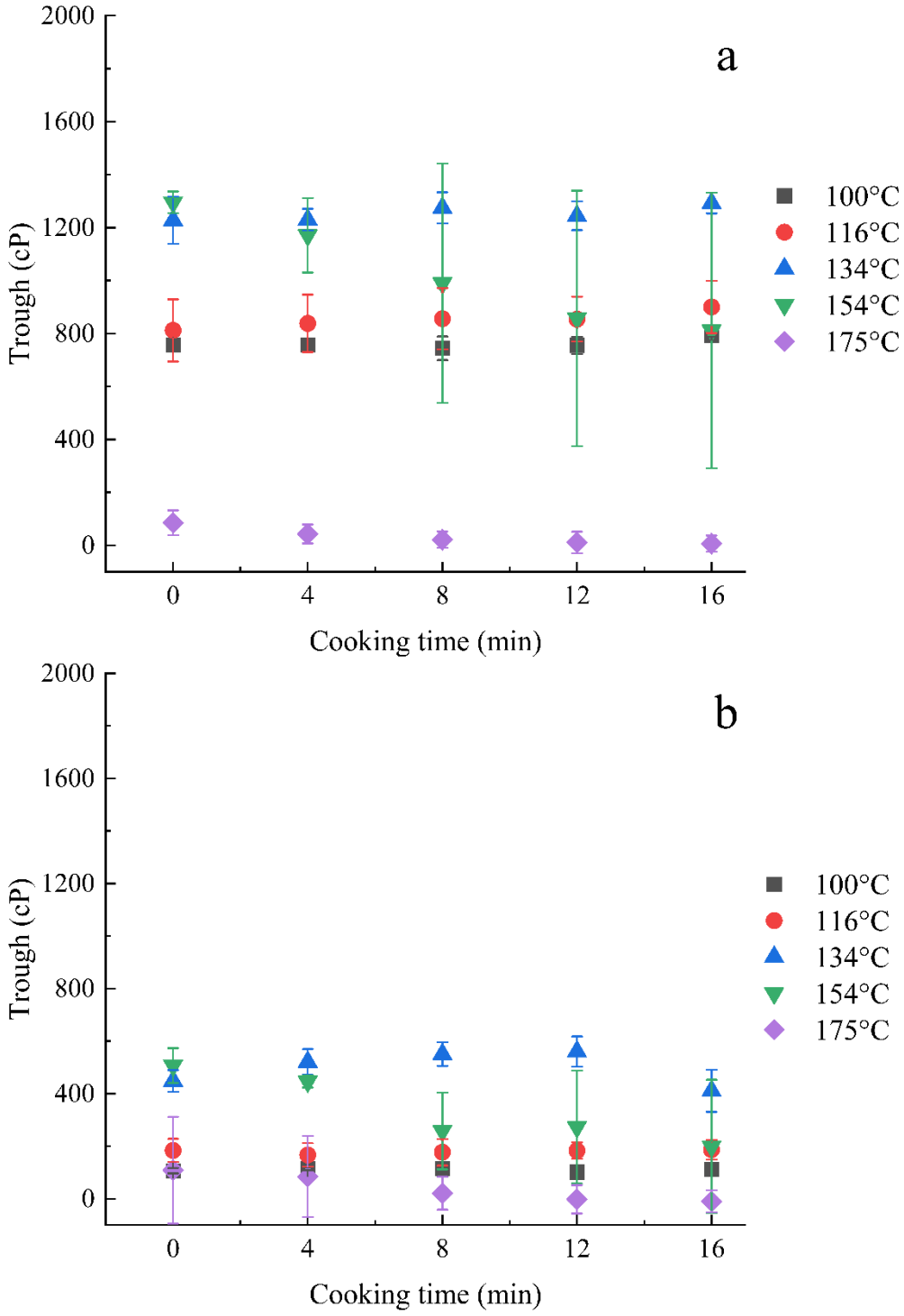


Figure 4.12 The setback of pea starch (a) and pea flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes. Error bars represent standard deviation.



In conclusion, the pasting properties of the roux were altered by the cooking conditions. The cooking temperature was critical to the change of pasting properties. Pasting properties decreased more rapidly with increased cooking time when the roux was cooked at high-temperature levels.

When starch or flour (containing no or limited moisture) was heated and then analyzed by the RVA, the pasting properties were reported to be affected by the heating conditions (Chen et al., 2018a; Lei et al., 2020; Ozawa et al., 2009; Qiu et al., 2015; Shi et al., 2018). Our results agreed with the previous finding as the pasting properties of the roux were altered by the cooking process. Shi et al. (2018) reported that the pasting property of pre-fried potato starch (180 °C, 3 min) changed due to heating even when the moisture content was as low as 5%. However, the pasting properties of wheat starch without moisture were only slightly affected by heating (120 °C, 120 min) according to Ozawa et al. (2009). The larger changes observed in this study were probably because the higher cooking temperatures (154 and 175 °C) applied were higher than the temperature (120 °C) Ozawa et al. (2009) applied. One interesting thing was that our results were consistent with the report of Oh et al. (2018), in which rice starch was dry heated at different temperatures (110, 130, and 150 °C) and times (0, 1, 2, and 4 h). Oh et al. (2018) reported that the pasting curves shifted down with increasing cooking time only when the cooking temperatures were above 130 °C. Oh et al. (2018) claimed that the decrease of pasting viscosity was probably due to the structural rearrangement of starch and the thermal degradation of starch molecules.

The change of pasting property can be further explained by two reasons as: 1) the melting of the crystalline region in starch granules and 2) the thermal degradation of starch molecules. According to Biliaderis et al. (1986), starch granules can be recognized as semi-crystalline spherulites owning a specific melting point. When starch is mixed with water and subjected to heating treatment, water works as a plasticizer reducing the melting point. The relationship between the moisture content and melting

point of starch could be predicted by the Flory-Huggins equation, and the starch with lower moisture content has a higher melting point (Donovan, 1979; Farhat & Blanshard, 1997; Whittam et al., 1990).

In the present study, since the moisture level of the roux was highly limited, it could be speculated that the starch in the roux only melted when cooking temperatures were higher than 134 °C. Conversely, when the roux was cooked at two lower temperatures (100 and 116 °C), which were lower than the melting point of starch, the starch structure was only slightly affected by the cooking process. Once the starch in the roux melted, the starch structure was irreversibly altered, resulting in the change of pasting properties.

At higher cooking temperatures (above 134 °C), the amylopectin molecules thermally degraded into smaller molecules and longer cooking time resulted in more amylopectin molecules that were thermally degraded, while the thermal degradation of amylopectin at the two lower cooking temperatures was not obvious, since the pasting curves were only slightly affected by the cooking process (Chen et al., 2019a; Juhász & Salgó, 2008; Lei et al., 2020).

4.4. Euclidean Distance

Because the responses to the temperature profile of the RVA differed according to the type of starch or flour, the Euclidean distance (ED) was calculated for pasting curves comparing the curve for a raw starch or flour (see Appendix E) against the cooked roux. This helps to elucidate how cooking conditions affected the overall pasting properties of the cooked roux. The ED was used to quantify the shift down of pasting curves due to cooking as a roux compared to the pasting profile of the raw starch or flour, which are shown in Figure 4.13 and Figure 4.14 for wheat and pea, respectively. Starch/flour type significantly (p<0.0001) affected the ED (Table 4.2). The cooked roux made from wheat starch showed the highest ED while the pea flour cooked roux showed the lowest

ED. A higher value for the ED indicates that the measured pasting curve viscosities decreased farther compared to the original pasting curves (native starch or flour). Therefore, pasting curves with larger ED were altered to a greater extent by cooking. When roux was heated at 134, 154, and 175 °C, the ED increased with prolongation of cooking time. This trend was seen for all types of starch/flour in the current study. The change of ED confirmed the findings from the single points of peak, trough, and setback viscosities in the RVA analysis. The change of ED clearly showed that not only were the measured RVA pasting parameters impacted by cooking, but also the whole pasting curves were diminished due to cooking. What should be noticed was that even minimal cooking (at 100 °C for 0 min), where the roux was cooked at 100 °C for come-up time (15 min), resulted in a large ED, i.e., the curve for roux at minimal cooking was different to that of the uncooked starch or flour. This was because of the residual oil in the cooked roux samples and the thermal effect over the 15 min of come up time.

The increase of pasting viscosity during the heating and cooling phase was because starch granules swelled (arising from starch gelatinization) and leached amylose associated with water molecules to form a network structure, respectively (Blazek and Copeland, 2008). Chen et al. (2018a) found that fried maize starch had lower swelling power and leached amylose content, which resulted in the decrease of the pasting viscosity of fried maize starch. Therefore, frying reduced the swelling power and the leached amylose content, so that both outcomes were responsible for the decrease in pasting viscosity. In the present study, cooking for the initial 15 min (come-up time) would result in the decrease of swelling power, contributing to a decrease in pasting viscosity or a change in ED. On the other hand, residual oil in the cooked roux would also affect the pasting properties. Chen et al. (2018b) analyzed the distribution of residual oil in the fried maize starch; the fried maize starch and frying oil were separated by vacuum filtration. They found that residual oil mainly surrounded the starch granules with a little portion of frying oil being absorbed in the maize starch granules. Therefore, it is expected that residual oil in cooked roux would surround the starch granules

(protein in the roux may also entrap part of the residual oil). Kim and Walker (1992) also found that adding sugars and emulsifiers to wheat starch resulted in a decrease of pasting viscosity and claimed that such a decrease in the pasting viscosity was because sugar or emulsifier competed for water molecules with starch granules, resulting in a delay of pasting time. It was also reported in previous studies that addition of oil resulted in a decrease of pasting viscosity of wheat starch or flour (Devi et al., 2020; Desai et al., 2021). Therefore, in the present study, part of the residual oil would surround wheat or pea starch granules which would restrict water entry into the starch granules delaying pasting time and decreasing pasting viscosity.

Figure 4.13 Euclidean distance of wheat starch (a) and wheat flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes.

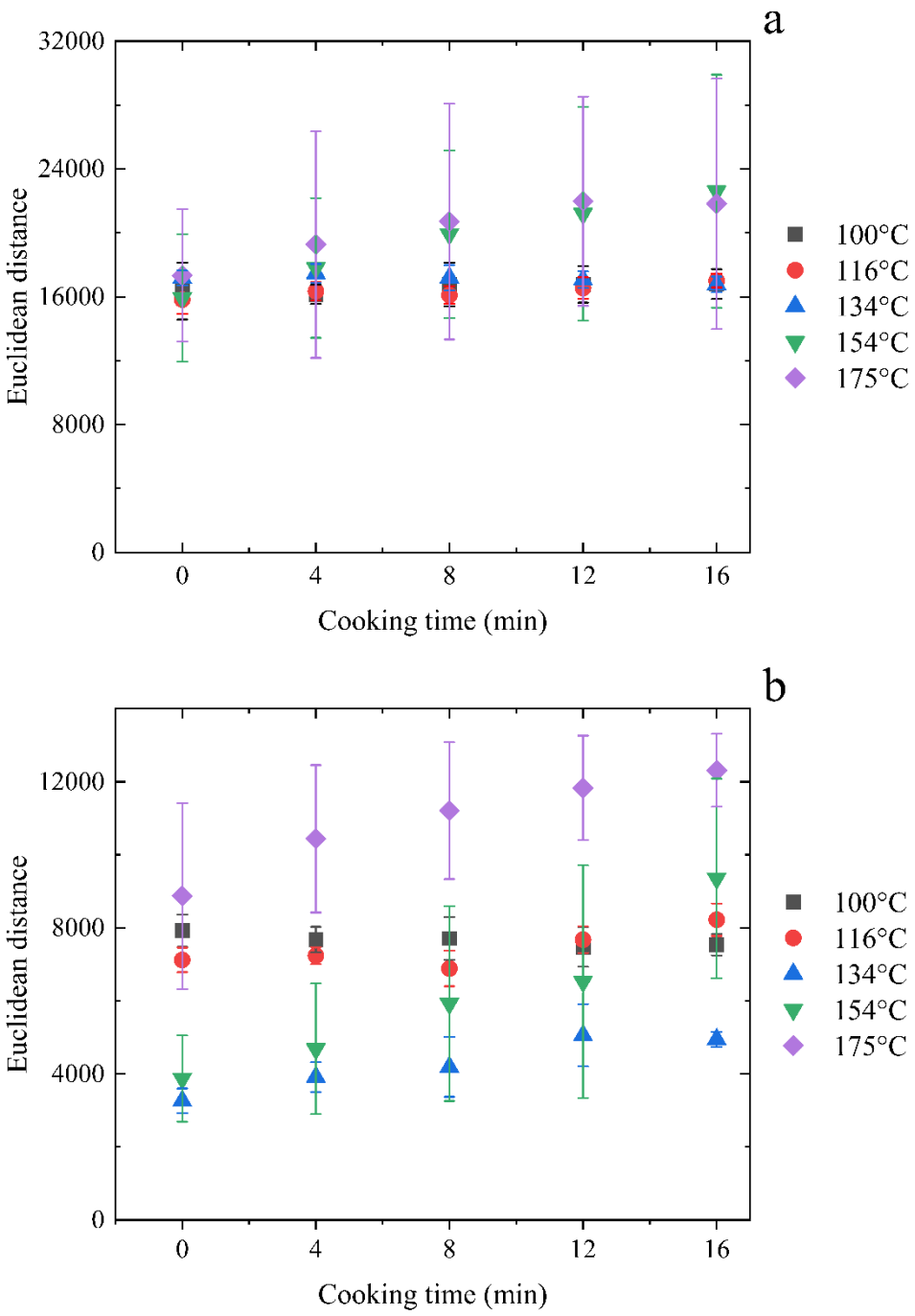


Figure 4.14 Euclidean distance of pea starch (a) and pea flour (b) in the roux cooked at one of the five temperature levels (100, 116, 134, 154, and 175 °C) for 0, 4, 8, 12, and 16 minutes.

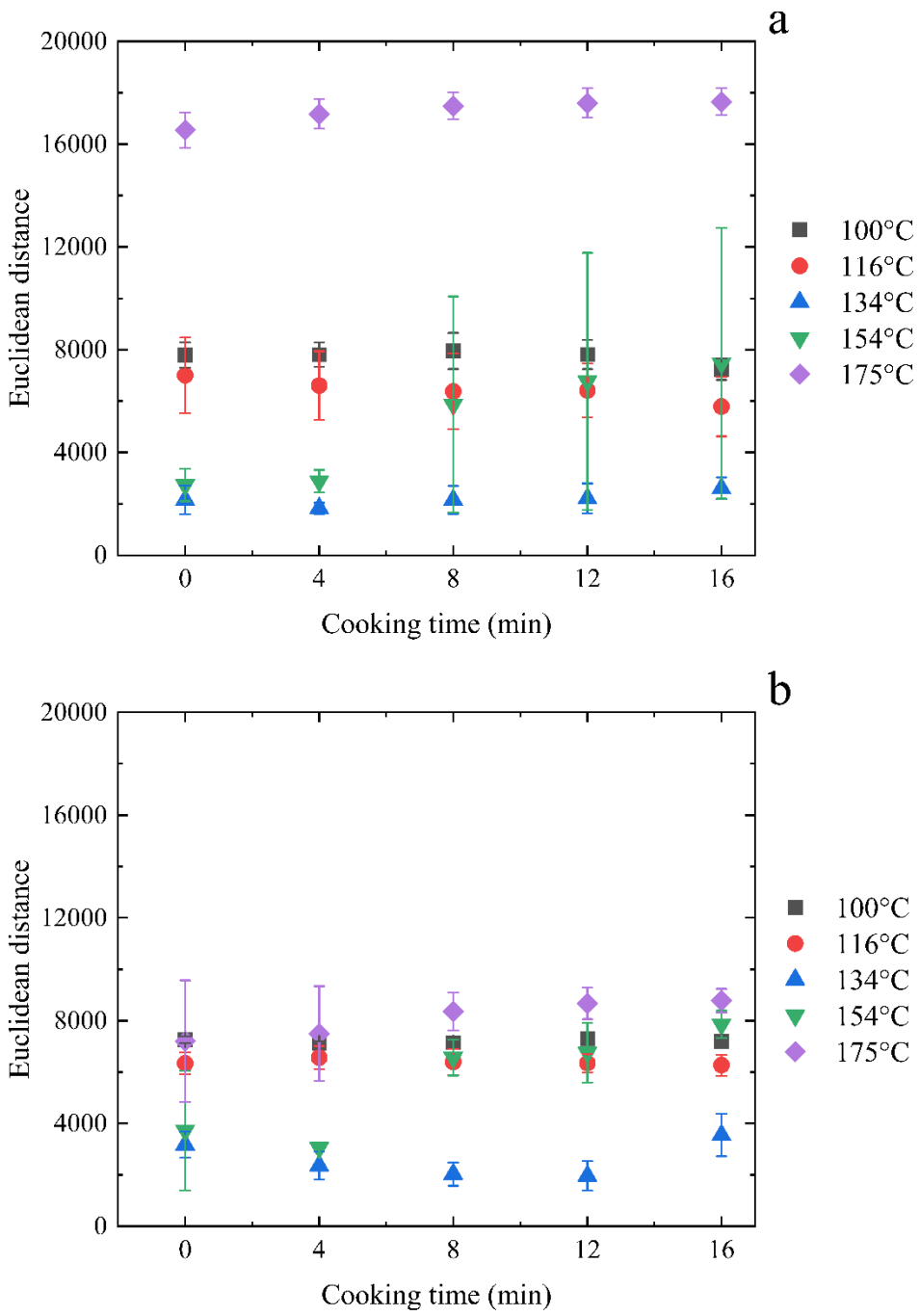


Table 4.2 Statistical analysis (P value) of Euclidean distance (ED) of the pasting curves for the wheat starch, wheat flour, pea starch, and pea flour in the cooked roux. The effects highlighted were statistically significant at P=0.05.

Effect	Euclidean distance
Starch/flour type	<0.0001
Cooking temperature	<0.0001
Cooking time	<0.0001
Starch/flour type*cooking temperature	<0.0001
Starch/flour type*cooking time	0.5999
Cooking temperature*cooking time	<0.0001
Starch/flour type*cooking temperature*cooking time	1.0000*

* Because the statistical software (JMP, Version 14) kept four significant digits after the decimal point, the P values were approximately equal to 1.0000.

4.5. Kinetic Study on the Change of Trough Viscosity

During the RVA analysis, pasting viscosity starts to decrease at peak time because of gelatinization of starch granules, i.e., breakdown of starch granules, then increases again because amylose exuded from starch granule forms a network structure (Copeland et al., 2009). The composition of a cooked roux, including the amylopectin, amylose, residual oil, and proteins, were all dispersed in the paste at the point where the trough viscosity is measured. Therefore, differences in trough viscosity among samples could represent the effect of cooking on starch molecules or on the integrity of the granules. Based on the assumption above, kinetic studies on the change of trough viscosity was conducted, which could potentially show the effect of cooking on the starch molecules or starch structure. The kinetic study was conducted through the following steps:

1. The value of trough viscosity for each sample was transformed into a logarithm

value. For each type of starch or flour, the logarithm of trough viscosity was plotted as a function of cooking time (Appendix F).

2. Linear regression analysis was used to fit the data (logarithm of trough viscosity against time) into a linear kinetic model to obtain the reaction constant (k) as shown in Table 4.3.
3. The k value was plotted as a function of the reciprocal of absolute cooking temperature ($\frac{1}{T}$) (Figure 4.15-4.18).
4. The k value and the $\frac{1}{T}$ were fitted in the Arrhenius expression by a nonlinear fitting. The activation energy (E_a) was determined based on the results of the nonlinear fitting. Fitting curves were determined using the mean values of k without error bars.

The E_a values for the change of the trough viscosity for the four roux were determined based on the nonlinear prediction functions which are shown in Fig. 4.15 to 4.18. Pea starch (Figure 4.17) showed the best fit to the Arrhenius expression (R^2=0.9988). The k value against $\frac{1}{T}$ were fitted in the Arrhenius equation and the E_a value of each type of roux was obtained from the prediction equation as shown in Figure 4.15-4.18. The E_a of the trough viscosity change for a roux made from pea starch was 169231 $kg \cdot m^2 \cdot s^{-2} \cdot mol^{-1}$(J/mol). Wheat flour (Figure 4.16) also showed excellent fit to the Arrhenius expression (R^2=0.9878) and the E_a of the trough viscosity change for a roux made from wheat flour was 62998 J/mol. For wheat starch (Figure 4.15), the data still reasonably fitted to the Arrhenius expression despite a lower R^2 (0.7376), but it is clearly not a good model for the experimental data. The E_a of wheat starch was 43985 J/mol, which was lower than the E_a of wheat flour. For the pea flour (Figure 4.18), it seemed that the data could not be fitted to the Arrhenius expression, but a value for E_a for pea flour was 52986 J/mol. Kinetic studies to different roux systems showed that the change of trough viscosity generally followed a first order reaction. Such finding helps to understand that effect of cooking temperature and time on pasting properties of a roux.

Table 4.3 Results of the linear fitting for the logarithm of trough viscosity as a function of cooking time. Values are means ± SD.

Starch/flour type	Temperature (°C)	$10^{-4}k$ (s^{-1})	Intercept	R^2
wheat starch	100	0.5739 ± 0.1787	7.3957 ± 0.0069	0.7747
	116	0.9413 ± 0.2247	7.4279 ± 0.0176	0.8541
	134	0.0935 ±0.1492	7.3339 ± 0.0098	0.1158
	154	5.7614 ± 0.4041	7.3312 ± 0.0139	0.9855
	175	5.7357 ± 0.6298	7.1985 ± 0.0237	0.9651
wheat flour	100	-0.4191 ± 0.4183	5.963 ± 0.0207	0.2508
	116	0.9931 ± 0.6828	6.1493 ± 0.0257	0.4136
	134	1.7311 ± 0.1239	6.6522 ± 0.0117	0.9849
	154	5.6088 ± 0.4282	6.6067 ± 0.0125	0.9828
	175	11.5000 ± 1.2564	5.7736 ± 0.0713	0.9652
pea starch	100	-0.5945 ± 0.1459	6.6190 ± 0.0130	0.8469
	116	-0.9640 ± 0.2044	6.6941 ± 0.0134	0.8811
	134	-0.5762 ± 0.1982	7.1043 ± 0.0134	0.7381
	154	5.6876 ± 0.8926	7.1673 ± 0.0073	0.9312
	175	54.6000 ± 20.1000	3.6836 ± 0.9526	0.7115
pea flour	100	-0.2969 ± 0.9419	4.7261 ± 0.0403	0.0321
	116	-0.5973 ± 0.6046	5.1495 ± 0.0390	0.2455
	134	-1.4167 ± 1.8609	6.1805 ± 0.0903	0.1619
	154	8.0136 ± 3.6058	6.2784 ± 0.0829	0.6221
	175	5.9129 ± 4.8475	4.1463 ± 0.3013	0.3315

Figure 4.15 The kinetic study of change of trough viscosity of wheat starch in the roux cooked at 100, 116, 134, 154, and 175 °C. The symbols represent the reaction rate constant (k) obtained at each cooking temperature. The solid lines were the fitting curves obtained from a nonlinear regression by applying the Arrhenius equation to the data.

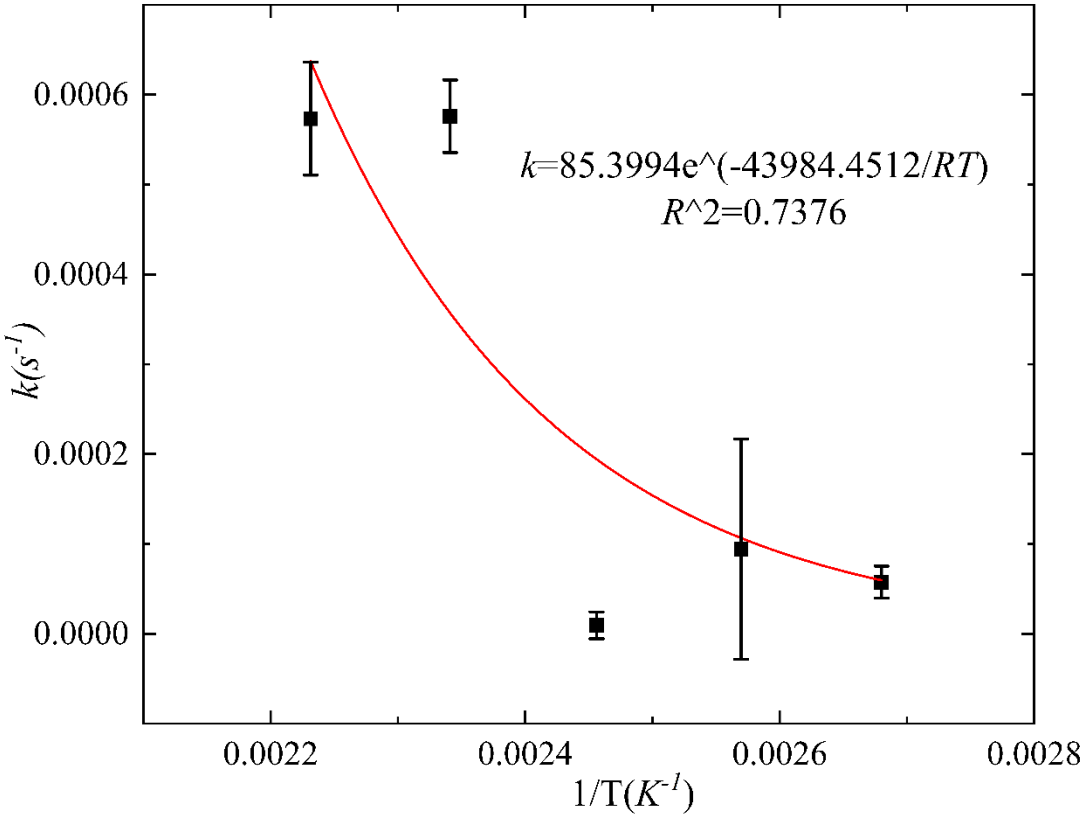


Figure 4.16 The kinetic study of change of trough viscosity of wheat flour in the roux cooked at 100, 116, 134, 154, and 175 °C. The symbols represent the reaction rate constant (k) obtained at each cooking temperature. The solid lines were the fitting curves obtained from a nonlinear regression by applying the Arrhenius equation to the data.

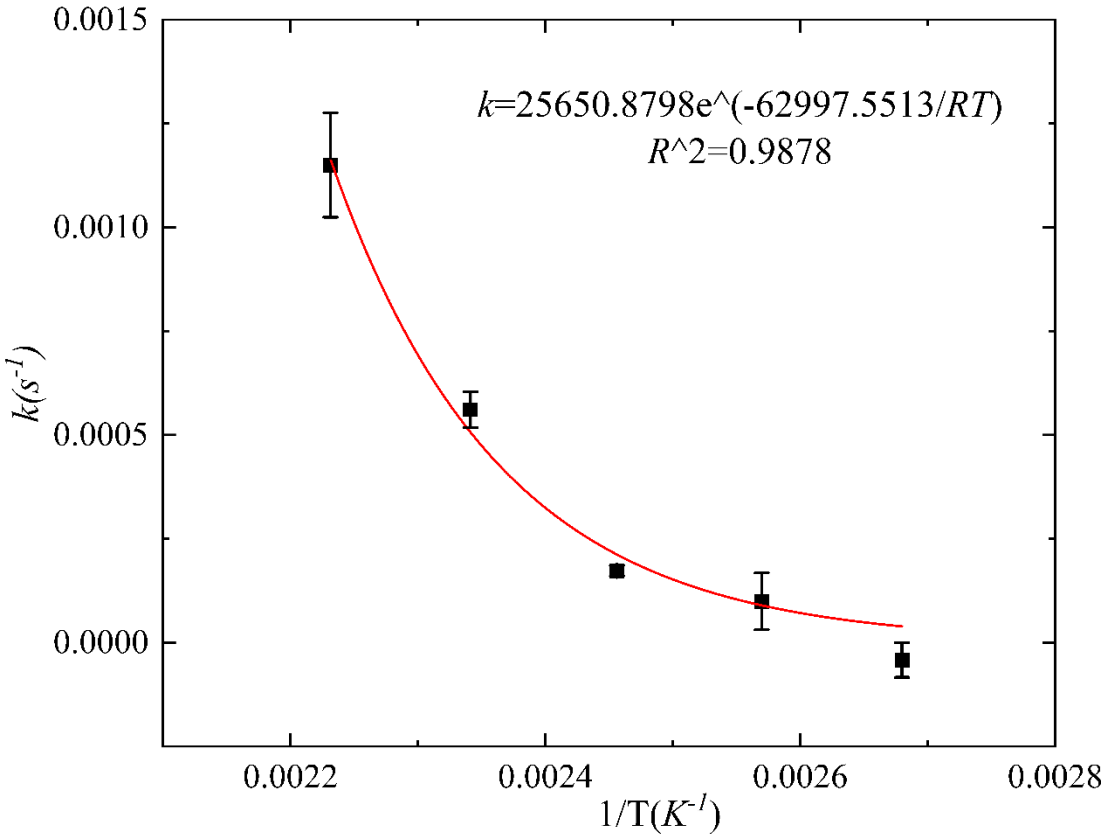


Figure 4.17 The kinetic study of change of trough viscosity of pea starch in the roux cooked at 100, 116, 134, 154, and 175 °C. The symbols represent the reaction rate constant (k) obtained at each cooking temperature. The solid lines were the fitting curves obtained from a nonlinear regression by applying the Arrhenius equation to the data.

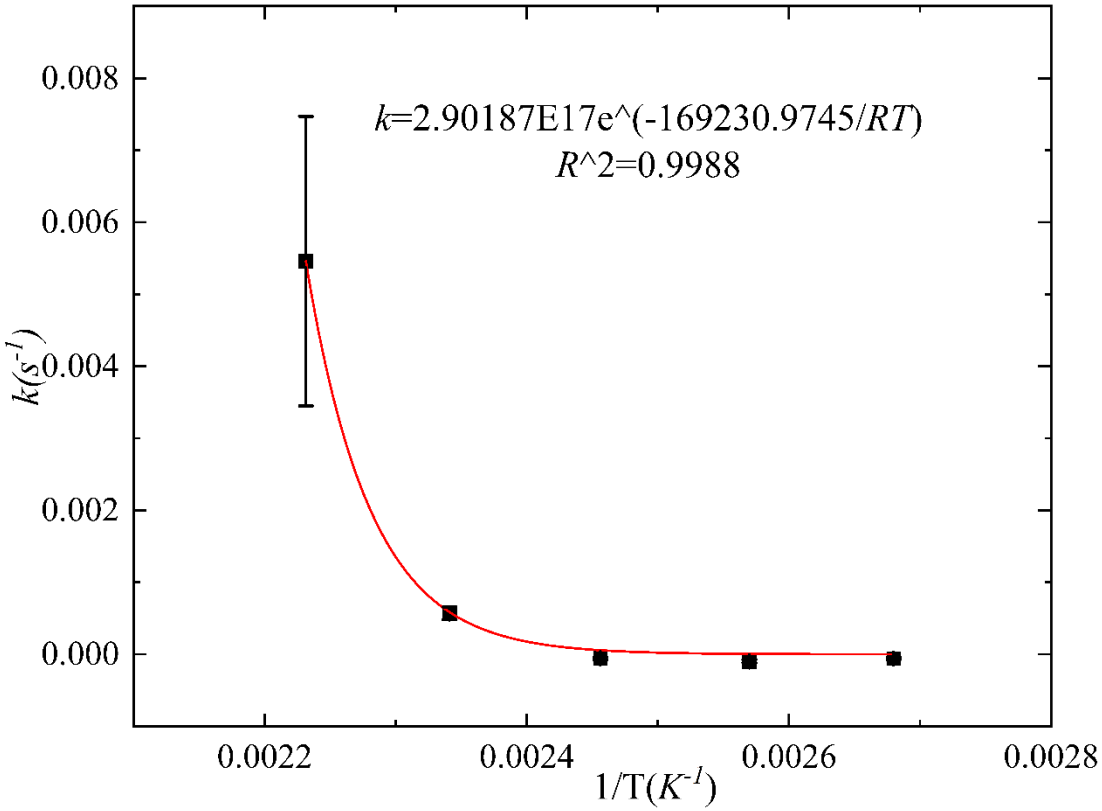
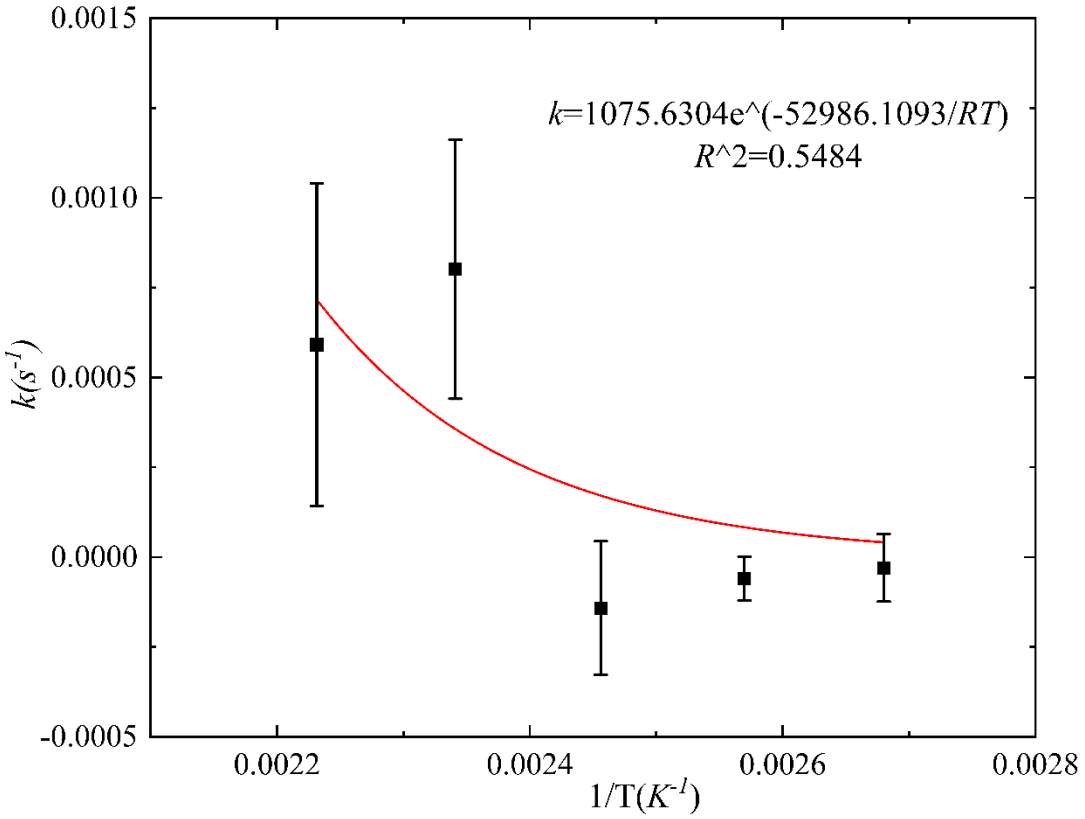


Figure 4.18 The kinetic study of change of trough viscosity of pea flour in the roux cooked at 100, 116, 134, 154, and 175 °C. The symbols represent the reaction rate constant (k) obtained at each cooking temperature. The solid lines were the fitting curves obtained from a nonlinear regression by applying the Arrhenius equation to the data.



4.6. Oil Content in Cooked Roux

In the present study, butter was replaced by canola oil with the aim of avoiding addition of moisture to the roux system. Chen et al. (2018b) found that fried maize starch (180°C for 20 min) with limited moisture (10%) kept its granule integrity, and the frying oil was located near the surface of the starch granules, but a small portion of the oil was absorbed into the starch granules. Proteins in starch would also combine with some oil (Zayas, 1997). After the roux was cooked, considerable amounts of oil were extracted by vacuum filtration, while a proportion of oil was still left in the solid part of the roux

as shown in Figure 4.19-20, with means and standard deviations in these figures reported in Appendix G. Starch or flour type showed a significant (p<0.0001) impact on the oil content of the cooked roux (Table 4.4). When roux made from wheat flour was cooked at 116 °C and 134 °C for 8 min, the oil content in roux was much higher compared to roux cooked at all other conditions (Figure 4.19b).

Wheat flour (Figure 4.19b) had higher oil content compared to wheat starch (Figure 4.19a) in the cooked roux, and pea flour (Figure 4.20b) had higher oil content than pea starch (Figure 4.20a). In general, the flour absorbed substantially more oil than the starch. Zhou et al. (2019) reported that rice flour had a higher oil-binding ability than rice starch and claimed that this was because of the protein difference between rice flour and starch. Protein was able to form interactions with oil which were mainly physical, with oil entrapped in the protein structure (Zayas, 1997). In the present study, wheat (11.4% protein content) and pea flour (17.5% protein content) contained much more protein than the wheat and pea starch, which allowed the roux made with flour to incorporate more oil. The average oil content of roux made of wheat and pea flour for all cooking temperatures and times was 25.78 ± 6.60% and 25.91 ± 4.61%, respectively. Though the average oil content of roux made from pea flour was slightly higher than that of roux made from wheat flour, the paired T-test showed that the averaged oil content of each roux was not significantly different (P>0.0001). The oil binding capacity of pea protein and wheat gluten was close according to Naczk et al. (1986), which might explain why there was no significant difference between the oil content of roux made from wheat flour and pea flour.

Three-way ANOVA showed significantly effects of starch/flour type and cooking temperature. Accordingly, individual two-way ANOVA tests was done on the oil content of roux made from each type of cooked roux (Table 4.5). It can be seen that cooking temperature significantly affected the oil content of roux made from wheat starch, pea starch, and pea flour. Oil content was not changed when the roux was cooked

at 100 and 116 °C, but decreased when the roux was cooked at the three higher temperatures (134, 154, and 175 °C). Heating the starch-oil mixture resulted in the loss of starch's porous surface, reducing the effective contact area of starch granules to oil (Chen et al., 2019a), resulting in a reduction in oil content. What was unexpected was that cooking time did not statistically impact the oil absorption (Table 4.5). This could be because the diminishment of the porous surface happened in a short time, i.e., during the come-up time. The effect of cooking temperature on the oil content in a roux could be because cooking resulted in breakdown of amylopectin, resulting in more amylose (Yang et al., 2019); the content of amylose would affect the oil content absorbed by starch in a roux through forming interactions with oil (Chen et al., 2019c).

Figure 4.19 Oil content of wheat starch (a) and wheat flour (b) in the roux cooked at 100, 116, 134, 154, and 175 °C for 0, 4, 8, 12 or 16 minutes. Error bar in Figure b exceeds out of the figure.

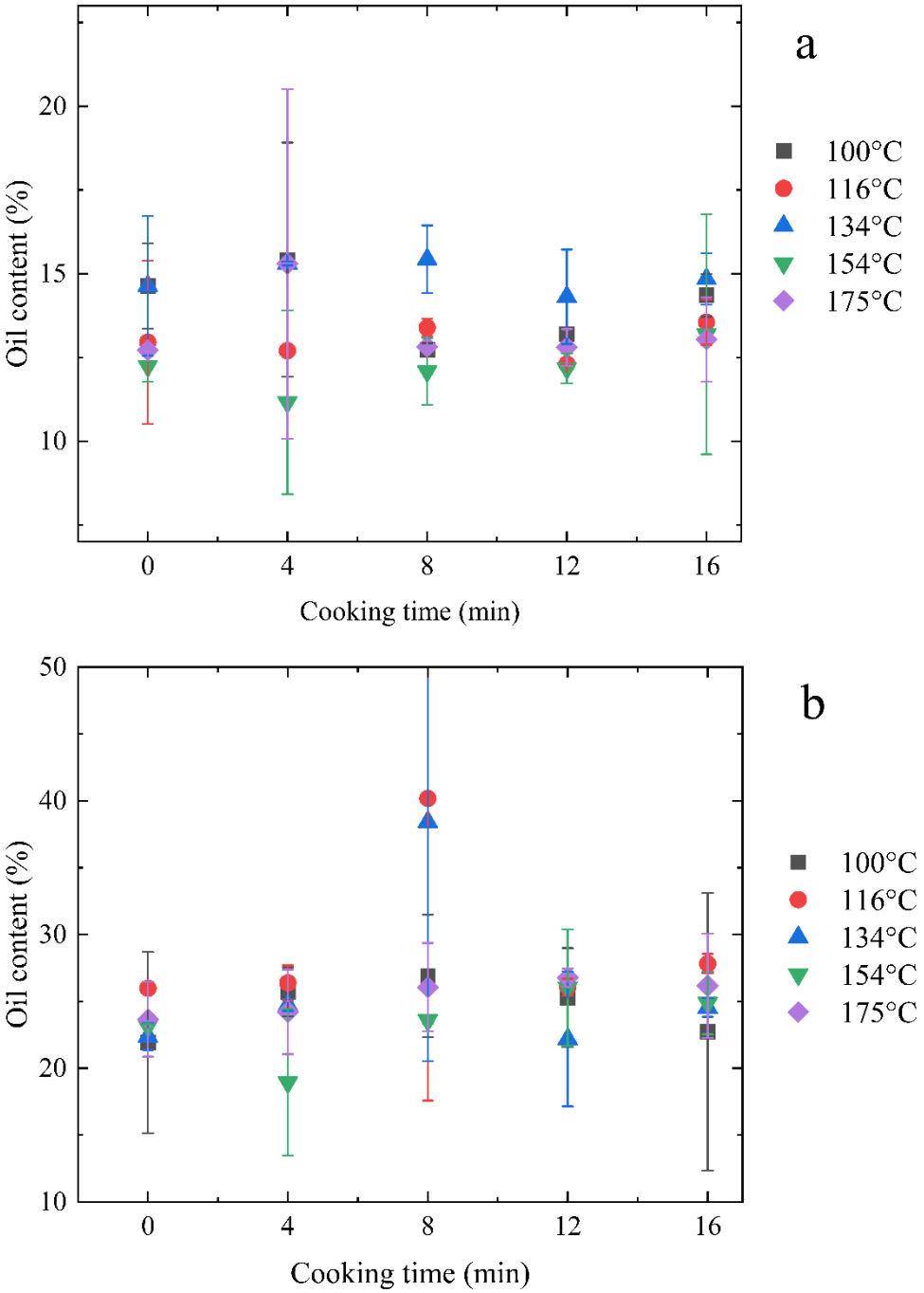


Figure 4.20 The oil content of pea starch (a) and pea flour (b) in the roux cooked at 100, 116, 134, 154, and 175 °C for 0, 4, 8, 12, or 16 minutes.

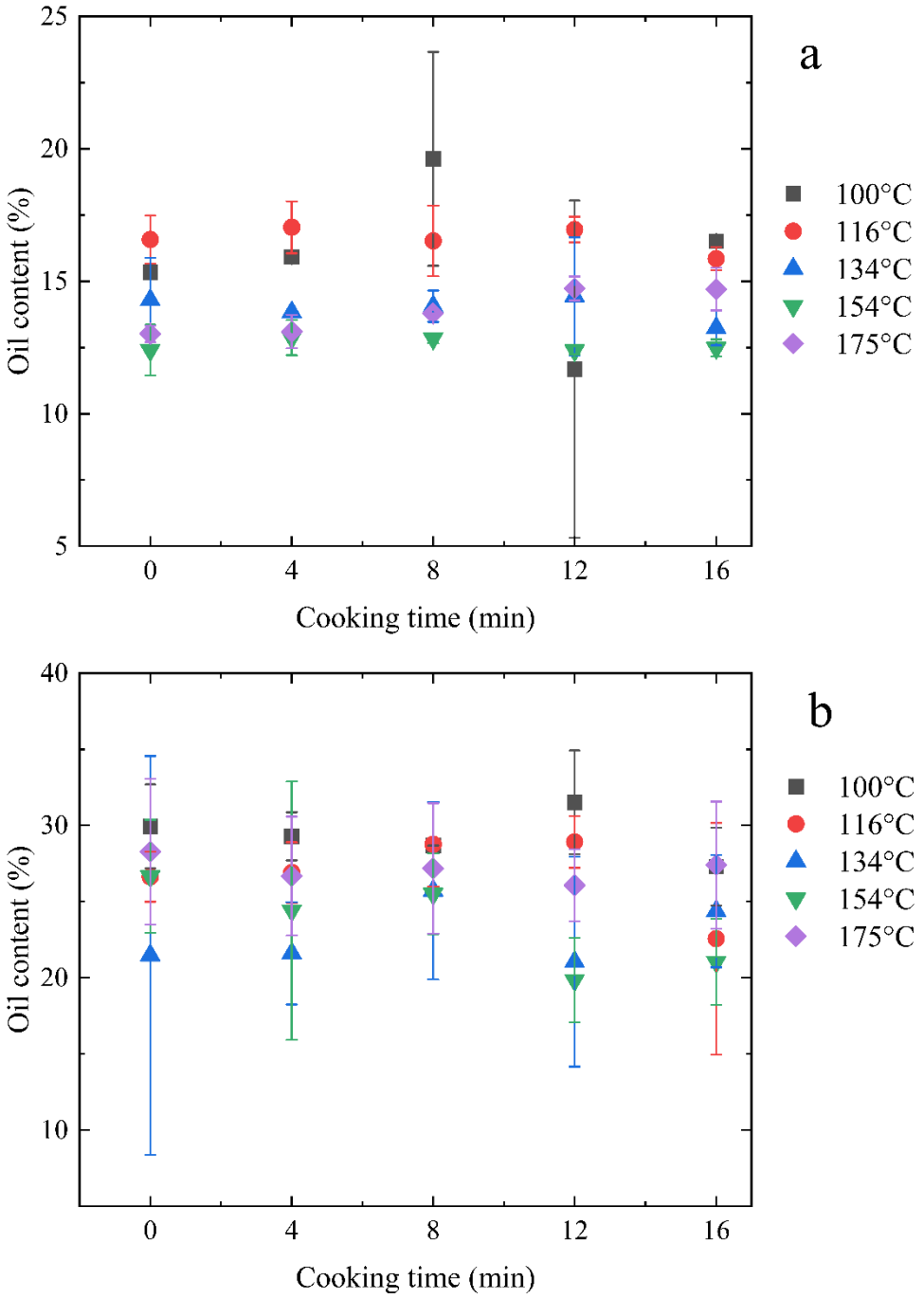


Table 4.4 Statistical analysis of the effect of starch or flour type, cooking temperatures, cooking time and their interactions on the oil content of the roux.

Effect	P value
Starch/flour type	<0.0001
Cooking temperature	0.0048
Cooking time	0.0854
Starch/flour type*cooking temperature	0.1212
Starch/flour type*cooking time	0.2968
Cooking temperature*cooking time	0.9322
Starch/flour type*cooking temperature*cooking time	0.9915

Table 4.5 P values (p<0.05) of the two-way ANOVA of cooking temperature and time on the oil content of roux made from wheat starch, wheat flour, pea starch, or pea flour.

Effect	Wheat starch	Wheat flour	Pea starch	Pea flour
Cooking temperature	0.0229	0.3703	0.0001	0.0294
Cooking time	0.7258	0.1252	0.5080	0.7674
Cooking temperature*time	0.9675	0.8865	0.2223	0.9751

4.7. Effect of Oil on the Pasting Profiles of Raw Starch and Flour

Exogenous oil has an impact on the pasting properties of starch or flour. In the present study, there was residual oil left within the roux after vacuum filtration, and this could influence the pasting profile of a roux. To distinguish the effects of residual oil and cooking effects in oil on the change of the pasting properties of a roux, the effect of oil alone on the pasting properties of raw starch was determined. The RVA profile of raw starch or flour mixed with oil was analyzed. The ratio of starch or flour : oil : water which was used for the RVA analysis is shown in Table 4.6. The total weight of starch/flour and canola oil was maintained as 3.115 g which was the same as the weight

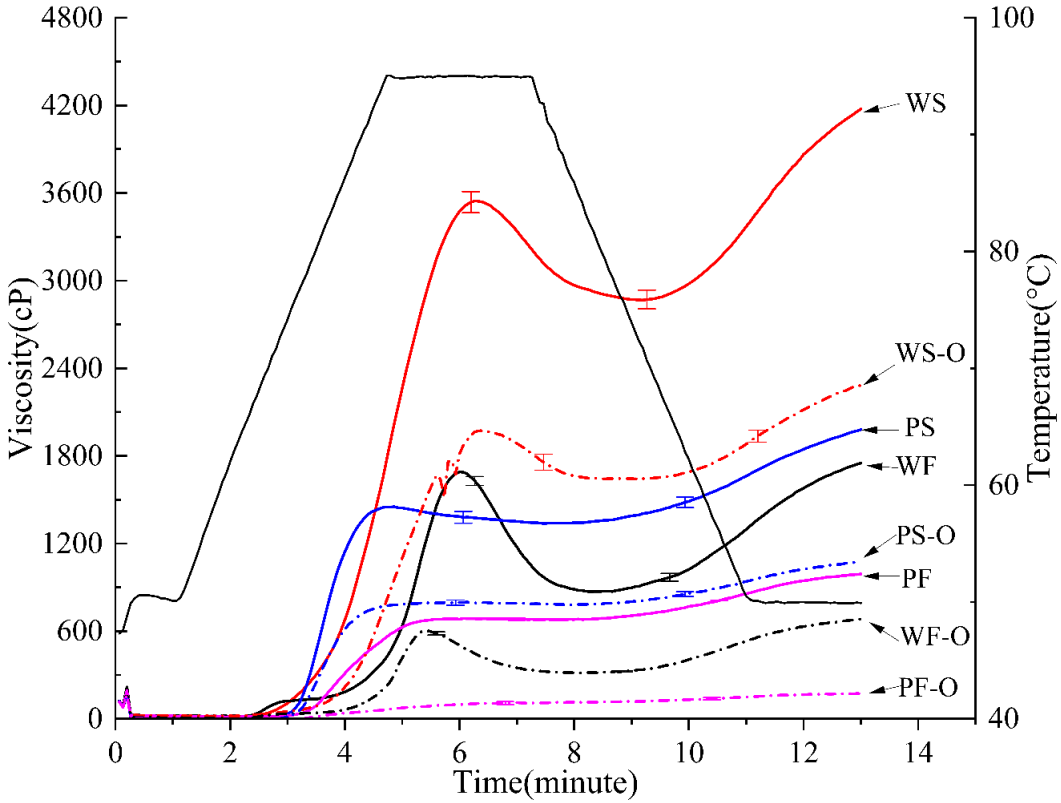
of roux sample used in the RVA analysis. The weight of canola oil was calculated based on the oil content in a cooked roux. To be specific, the average oil content in the roux made from wheat starch, wheat flour, pea starch, and pea flour over all (five) cooking temperatures and (five) times was 13.5, 25.8, 14.6, and 25.9%, respectively. Therefore, the four systems represent the roux with its absorbed amount of oil but in its uncooked state.

Table 4.6 The RVA samples ratio of raw starch or flour, canola oil, and water used to analyze the effect of oil content on the pasting properties of raw starch or flour.

Sample	Weight of canola oil (g)	Weight of starch/flour (g)	Weight of water (g)
Wheat starch-canola oil	0.421	2.694	25.385
Wheat flour-canola oil	0.804	2.311	25.385
Pea starch-canola oil	0.455	2.660	25.385
Pea flour-canola oil	0.807	2.308	25.385

It can be seen from Figure 4.21 that the existence of canola oil diminished the pasting viscosity of raw starch and flour. Desai et al. (2021) reported that a mixture of wheat starch, oil, and wheat gluten had a significantly lower peak viscosity than that of a mixture of raw wheat starch and gluten. Desai et al. (2021) claimed such decrease in peak viscosity was due to the formation of amylose-lipids complex. However, there was no evidence showing that fatty acids could be formed in RVA analysis. In addition, the starch-triglyceride complex was not detected in RVA analysis (Li et al., 2020) suggesting that the decrease of peak viscosity was due to other reasons.

Figure 4.21 Pasting profiles of the raw starch/flour (wheat and pea starch, wheat and pea flour) and the mixture of one of the four starch or flour and canola oil. Each pasting profile is constructed from three replications.

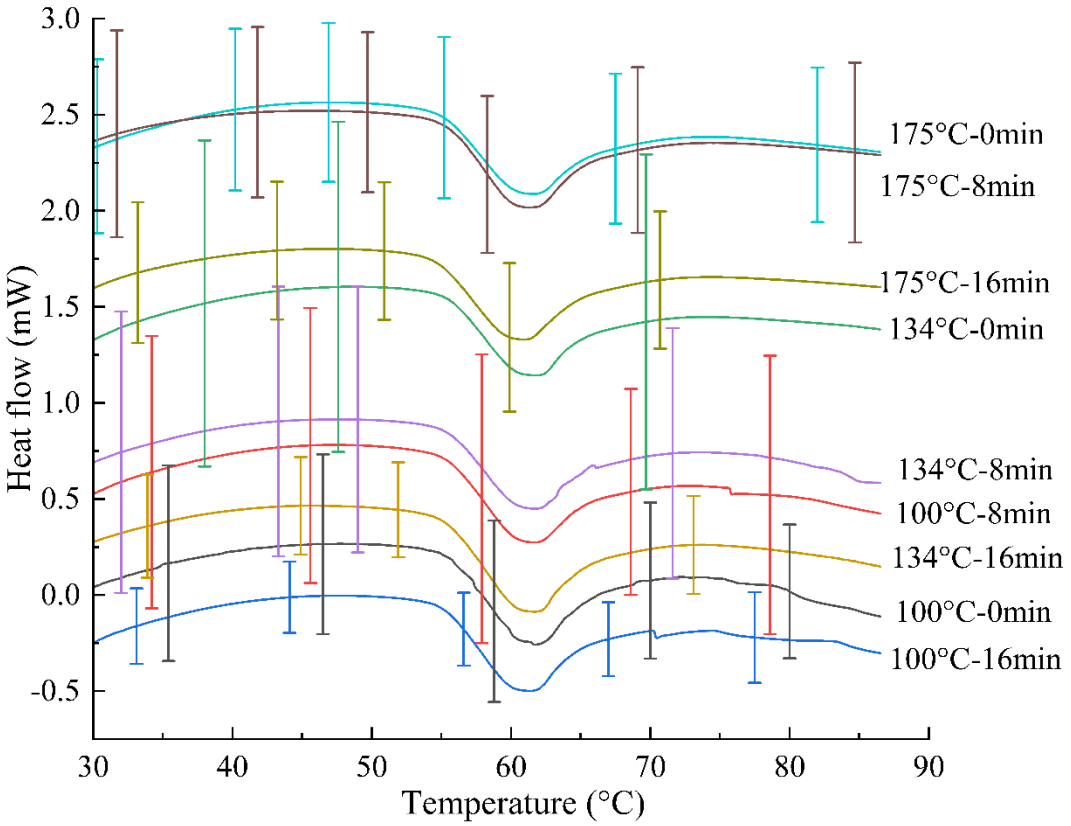


*Characters from top to bottom represent: WS: wheat starch; WS-O: wheat starch-oil; PS: pea starch; WF: wheat flour; PS-O: pea starch-oil; PF: pea flour; WF-O: wheat flour-oil; PF-O: pea flour-oil.

4.8. DSC Properties

To determine the effect of cooking on the crystalline structure in the cooked roux, the thermal properties of wheat starch in a cooked roux were measured. Conducting DSC analysis on the roux system made from wheat starch was able to elucidate the mechanism of how cooking conditions affected pasting properties and oil content, which was one of the research objectives in the present study. Therefore, only the DSC properties of roux made from wheat starch was examined. The preparation procedure of roux studied in the thermal analysis was the same as the RVA analysis but fewer samples were analyzed by the differential scanning calorimetry (DSC) compared to the pasting properties analysis. The thermal properties of the roux made with wheat starch, which were cooked at one of three temperatures (100, 134, and 175 ℃) for 0, 8, or 16 minutes, were measured. The thermograms of the roux that had been cooked at the different temperatures and times are shown in Figure 4.22, and the thermal properties of the roux are summarized in Table 4.7.

Figure 4.22 Endothermic heat flow traces for wheat starch in cooked roux prepared at one of three cooking temperatures (100, 134, and 175 °C) for 0, 8, or 16 minutes.



When the roux was cooked at the higher temperature, the thermogram curves of the roux shifted up. However, increasing cooking time from 0 to 16 minutes at each cooking temperature resulted in a downward shift of the thermogram curves except for the roux cooked at 100 °C for 8 min (Figure 4.22).

Cooking temperature showed a significant (p<0.05) impact on the start temperature (T_s), onset temperature (T_o), and peak temperature (T_p) of the cooked roux (Table 4.8). Conclusion temperature (T_c) was not statistically affected by cooking (Table 4.8). The average of T_s over three cooking times increased from 49.50 °C to 51.95 °C, while the T_o decreased from 56.05 °C to 55.29 °C, when the cooking temperature increased from 100 to 175 °C (Table 4.7). The averaged T_p over three cooking times increased slightly from 61.16 °C to 61.48 °C, when cooking temperature increased from 100 °C to 134 °C,

then decreased to 60.85 °C when cooking temperature further increased to 175 °C (Table 4.7). Cooking time only significantly affected the T_s (Table 4.8) as increasing cooking time resulted in the decrease of T_s (Table 4.7).

The interesting finding was that the enthalpy of wheat starch in a cooked roux was not affected by the cooking procedure (Table 4.8). However, according to Shevkani et al. (2017), the enthalpy of common wheat starch (heating rate: 10 $°C \cdot min^{-1}$; ratio of starch/water=1:4) was 5.31 $J.g^{-1}$. The enthalpy of wheat starch in the cooked roux, as reported in the current study, ranged between 1.41 and 1.76 $J \cdot g^{-1}$, which is substantially lower than normal wheat starch. It seemed that cooking resulted in a decrease of crystallinity of the starch in the roux. When starch granules with limited or without moisture were heated to high temperatures, starch granules did not gelatinize. However, the crystalline structure inside the heated starch melted due to heating (Parker & Ring, 2001). The residual oil in the samples would also result in a decrease of enthalpy. Wang et al. (2016) reported that addition of fatty acids (lauric acid, myristic acid or palmitic acid) into wheat starch resulted in the decrease of gelatinization enthalpy from 11.7 to 8.6 or 7.7 $J \cdot g^{-1}$ depending on the fatty acid added, and they claimed that they inhibited the melting of starch crystallites. Previous studies had reported a second peak on the DSC thermograms of starch occurred at around 104 °C, which was recognized as the melting of amylose-lipid complex (Alvarez-Ramirez et al., 2018; Yang et al., 2019). The scanning temperature applied in the present study ranged 20 to 90 °C, so melting of amylose-lipid complexes was not observed in the DSC thermograms.

Table 4.7 Thermal properties of wheat starch in the cooked roux.

Sample	T_s / ℃	T_o / ℃	T_p / ℃	T_c / ℃	ΔH (J/g)
100℃ 0min	48.84 ± 2.83	56.22 ± 1.11	61.55 ± 0.19	72.86 ± 0.50	1.76 ± 0.12
100℃ 8min	50.09 ± 0.98	56.47 ± 0.71	61.10 ± 0.20	70.88 ± 3.09	1.41 ± 0.11
100℃ 16min	49.56 ± 2.51	55.46 ± 0.21	60.82 ± 0.10	71.07 ± 1.63	1.50 ± 0.24
134℃ 0min	50.11 ± 0.07	55.82 ± 0.11	61.37 ± 0.11	72.73 ± 0.11	1.49 ± 0.11
134℃ 8min	50.72 ± 0.15	55.79 ± 0.78	61.75 ± 1.06	72.60 ± 0.98	1.50 ± 0.25
134℃ 16min	47.40 ± 1.61	55.65 ± 0.23	61.33 ± 0.11	73.42 ± 0.65	1.70 ± 0.13
175℃ 0min	52.37 ± 0.23	55.35 ± 0.39	61.05 ± 0.23	71.34 ± 0.78	1.56 ± 0.08
175℃ 8min	52.80 ± 0.82	55.66 ± 0.77	60.85 ± 0.55	71.57 ± 0.94	1.63 ± 0.03
175℃ 16min	50.68 ± 0.25	54.86 ± 0.16	60.64 ± 0.16	72.00 ± 0.68	1.66 ± 0.14

*T_s, T_o, T_p and T_c refer to the start, onset, peak and conclusion temperature, respectively and ΔH refers to the gelatinization enthalpy.

Table 4.8 Statistical analysis (P value) of the thermal properties of wheat starch in the cooked roux. Roux was cooked at one of three temperatures (100, 134, and 175 °C) for 0, 8, and 16 minutes.

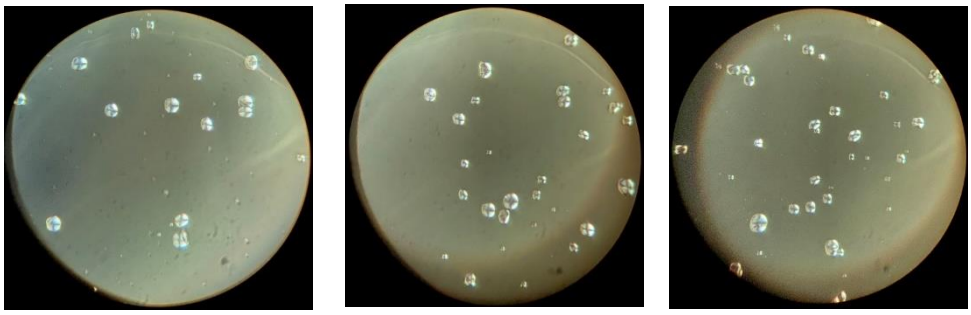
	T_s	T_o	T_p	T_c	ΔH
Cooking temperature (T)	0.0042	0.0458	0.0345	0.2327	0.6237
Cooking time (t)	0.0345	0.1050	0.1107	0.3213	0.0693
Cooking T*t	0.2180	0.6780	0.5124	0.5290	0.1133

*T_s, T_o, T_p and T_c refer to the start, onset, peak and conclusion temperature, respectively, and ΔH refers to the gelatinization enthalpy.

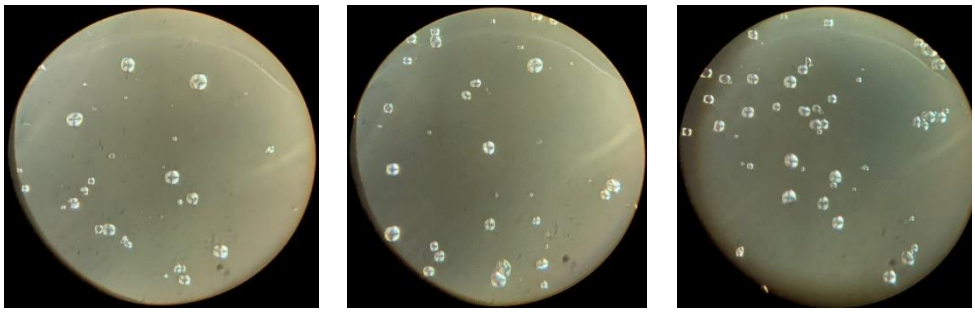
4.9. Microscopy Analysis

The microscopy images of roux cooked at 100, 134, and 175 °C for 0, 8, and 16 minutes are shown in Figure 4.23. The materials (wheat flour and canola oil) and cooking conditions in the microcopy analysis were the same as in the thermal analysis. The most noticeable finding was that all starch granules kept their integrity, and birefringence was observed in the starch granules under polarized light (Figure 4.23). Alvarez-Ramirez et al. (2018) indicated that most of the starch granules in the roux lost birefringence due to gelatinization after cooking. However, considering that butter contained much more water compared canola oil, moisture in the roux system from Alvarez-Ramirez et al. (2018)'s study was higher than the moisture content in the roux system studied in the current study. Dry heating starch (no moisture) or heating starch-oil mixture at high temperatures did not result in the breakdown of granular integrity nor starch gelatinization (Chen et al., 2018a; Liu et al., 2019). The moisture content of the roux studied was too low to allow the gelatinization of starch. Therefore, the birefringence and visual integrity of the starch were not affected by the cooking.

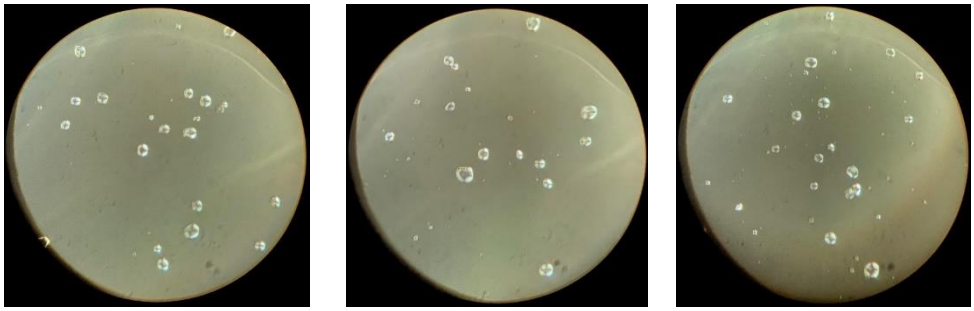
Figure 4.23 Polarized light microscopy images of wheat starch in the roux cooked at one of three cooking temperatures (100, 134, and 175 °C) for 0, 8, or 16 minutes, with larger magnification for the two extremes of roux cooking.



100°C-0min 100°C-8min 100°C-16min

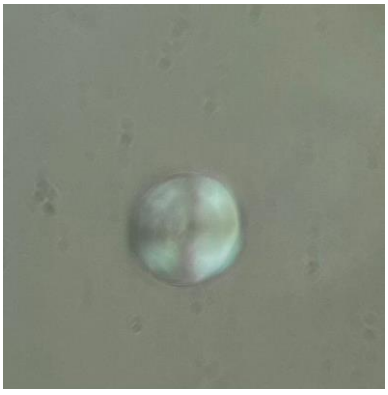


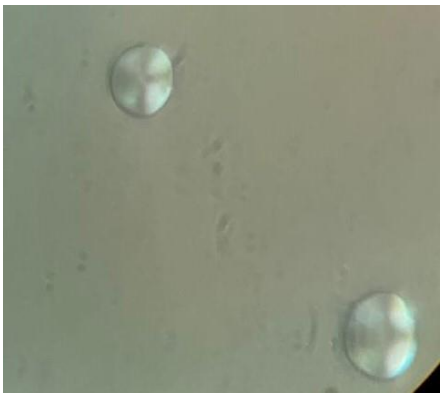
134°C-0min 134°C-8min 134°C-16min



175°C-0min 175°C-8min 175°C-16min

175°C-16min 100°C -0min





5. General Discussion

A key objective of the thesis research was to determine the effect of starch or flour type and cooking conditions on roux pasting properties. Perhaps the most meaningful finding in the current study is that cooking conditions significantly influenced the pasting viscosity of a roux (Figure 4.3-6). To be specific, the pasting viscosity of all cooked roux samples (wheat starch, wheat flour, pea starch, and pea flour) decreased with increasing cooking time (0-16 min) at the two higher cooking temperatures (154 and 175 °C). Conversely, cooking time only slightly affected the pasting viscosity at the lower cooking temperatures (100-134 °C) (Figure 4.13-14). Besides the cooking conditions, the type of starch or flour used to make a roux resulted in a significant difference in the properties of a roux which could be due to the molecular structural difference between different types of starch/flour.

From the kinetic study to the change of trough viscosity, it can be seen that for the reaction constant (k) values at higher cooking temperatures (154 and 175 °C) were much higher than k values at lower cooking temperatures (100 and 116 °C) (Figure 4.15-4.18); such difference was more apparent for pea starch (Figure 4.17). Thus, it seemed that cooking-induced change to the trough viscosity only happened at higher cooking temperatures, and lower cooking temperatures did not result in a significant change in the trough viscosity. Therefore, outside of the kinetic model, the change of the trough viscosity is more like a phase transition model. Donovan (1979) claimed that the crystalline structure of starch with low moisture content only melts when cooking temperature increased to a specific temperature. In terms of the low moisture content of the roux in the present study, the melting of the crystalline structure in starch within the roux would melt only when high cooking temperatures were applied. In other words, the crystalline structure of starch within the roux was not affected by the cooking until the cooking temperature reached to or over 134 °C; once the crystalline structure melted, the roux properties changed with increasing thermal time causing greater change in properties.

The pasting viscosity of wheat starch (Figure 4.3) in the cooked roux was higher than pea starch (Figure 4.5) at every cooking temperature and time. The noticeable similarity for the pasting profiles of wheat starch and pea starch was that cooking resulted in no obvious change in the pasting profiles when cooking temperature was lower than 134 °C, but profoundly altered the pasting profiles when cooking temperature was at or over 154 °C. When cooking temperature was at 154 or 175 °C, the pasting profiles shifted down (pasting viscosity decreased) with an increase in cooking time for both the wheat and pea starch. The influence of cooking conditions on the starch pasting properties seemed not to be affected by the type of starch.

It can be seen from Figure 4.5 that the pasting curves of pea starch in a roux were slightly affected by cooking at 100 and 116 °C, but shifted up in a pronounced manner when cooking temperature increased to 134 °C. However, such an increase in pasting viscosity at 134 °C was not seen in the wheat starch in a roux (Figure 4.3). Such heating-induced increase of pasting viscosity was also reported in previous studies (Lei et al., 2020; Liu et al., 2019; Oh et al., 2018; Qiu et al., 2015). Qiu et al. (2015) found that the crystallinity of rice starch increased after dry heating and claimed that such an increase in pasting viscosity was due to starch molecular structural rearrangement. The crystalline structure of wheat and pea starch is different as wheat starch has A- type while pea starch has C- type crystalline structure (Ratnayake et al., 2001; Shevkani et al., 2017). Starch with a C-type crystalline structure contains more water molecules, which was more easily affected by heating with low moisture. Previous studies reported that heating would result in the transformation of C-type crystalline structure into A-type crystalline structure, while the crystalline nature of starch with A-type crystalline structure was not affected by heating (Ambigaipalan et al., 2014; Bogracheva et al., 1998; Chen et al., 2019a; Shi et al., 2018). Therefore, it was likely that pea starch in a roux was more easily affected by the cooking process, leading to structural rearrangements in the granule, and therefore, increasing the pasting properties after

cooking at 134 °C.

The pasting curves of wheat flour also shifted up when the cooking temperature reached 134 °C. Unlike the oil-starch and starch molecules-starch molecules interactions of pea starch, the shift up of the pasting curves of wheat flour was probably because of the interactions between non-starch components, especially the proteins. The SS linkages formed during roux cooking resulted in protein-protein bonding in wheat flour, which resulted in the significant increase of pasting viscosity at 134 °C (Ozawa et al., 2009; Qiu et al., 2015). As wheat starch is lacking in protein compared to wheat flour, this phenomenon was not observed in wheat starch. Another significant difference between the wheat starch and wheat flour was the oil content, as the roux made from wheat flour contained higher oil content than those made from the wheat starch. Similarly, roux made from pea flour also showed higher oil content than those made from pea starch. Overall, flour roux had higher oil content than starch roux. Oil-starch interactions might be one of the reasons explaining why wheat flour had lower pasting viscosity than wheat starch. The current study further proved that the non-starch components had a critical impact on other properties as protein molecules might promote the thickening ability of a roux that has been cooked at mild temperatures.

Roux made from wheat flour and pea flour had the same change of pasting viscosity as the pasting curves shifted at 134 °C. However, the difference between the wheat flour and pea flour was that the wheat flour still showed clear pasting curves (Figure 4.4) while the typical characteristics of pasting curves was obviously diminished for the roux made from pea flour (Figure 4.6) cooked at a temperature of 175 °C. It can be concluded that roux made from wheat flour had better thickening ability than roux made from pea flour when cooked at high temperatures. It is suggested that the type of flour used to make a brown roux, which is commonly cooked over 170 °C (Kato, 2005), should be properly chosen in terms of thickening ability.

In general, cooking resulted in the starch crystalline region melting during roux making at temperatures greater than 134 °C, contributing to the decrease of starch or flour pasting viscosity in the cooked roux. This finding has important implications for understanding the effect of cooking conditions and differences in the ingredients of a roux. The current study was successful as it was able to identify the impact of cooking conditions on the properties of a cooked roux made from a given starch or flour. As the present study investigated the roux system made from controlled moisture content, it is suggested to use different moisture content to study the effect of moisture content on the roux system. Moreover, investigation of the role of residual oil content on the roux's physicochemical properties is also suggested.

6. Conclusion

The present study was designed to determine the effect of cooking temperature and time on the physicochemical properties of different types of starch or flour in cooked roux. I determined the effect of cooking conditions on the pasting properties of a roux. The present study shows that cooking for longer times resulted in a decrease of pasting properties, but only at higher temperatures. The present study also showed that the different type of starch and protein influenced the pasting properties of a roux during the cooking, as the viscosities of the pasting curves of the roux made with wheat flour, pea starch, and pea flour increased when cooking temperature increased to 134 °C, a phenomenon that was not evident in wheat starch. The results of determining residual oil content showed that flour had higher residual oil than that of starch. This is likely due to the higher oil binding capacity of the protein in the flour. The analysis of oil content indicates that protein, as one of the ingredients for a roux made with flour was important. The study of thermal and microscopic properties of the roux made from wheat starch showed that even though the granular structure of starch in a roux was not altered, the thermal properties of the roux were still changed. Results showed that pea starch and flour were good thickening agent at the cooking temperature range of 134 to 154 °C.

The present study is meaningful as it showed that the type of ingredients used had a critical influence on the properties of a roux. The finding from the current study can help us to understand the role that each component played in the roux properties during the cooking process. The present study systematically investigated the effect of cooking conditions on the thickening ability of a roux; such knowledge would be useful for preparing a roux in the domestic or commercial kitchen. As water is one of the ingredients of a roux, further research is suggested to study the effect of moisture content on the change of properties during roux making.

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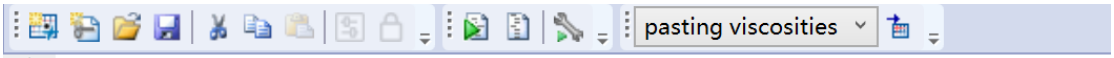
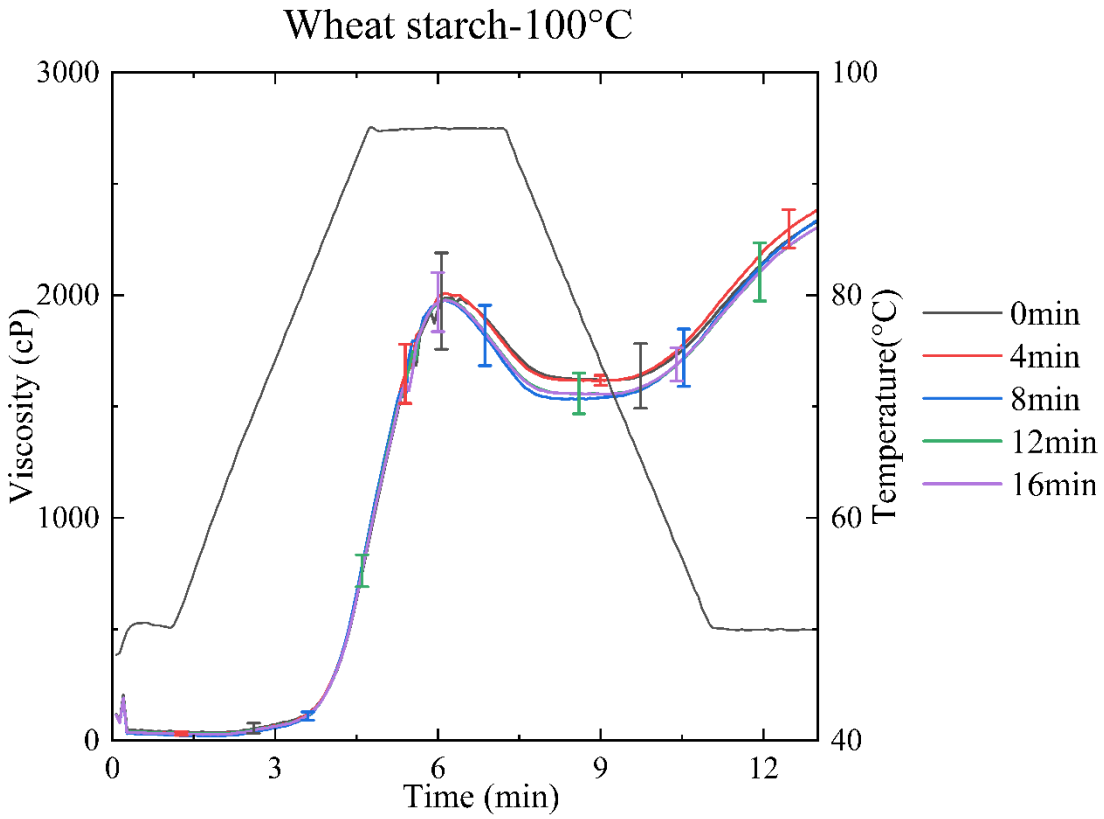
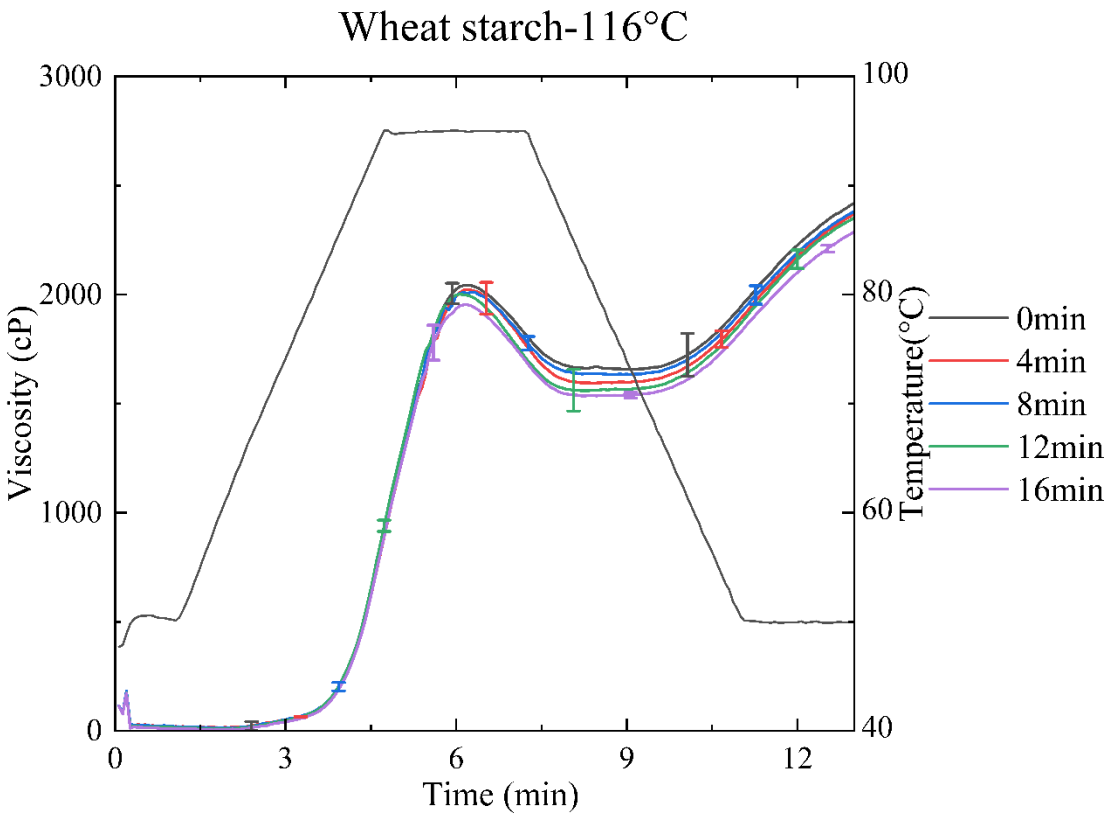
Appendix A: Example of JMP code.

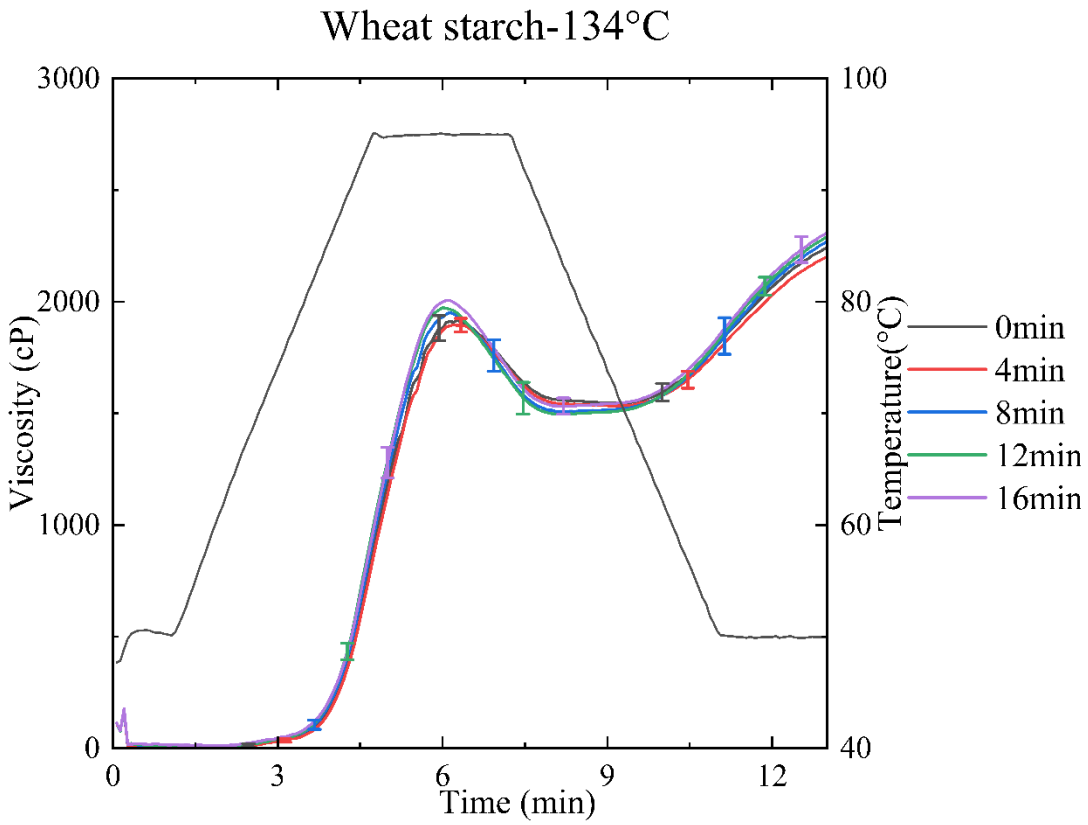


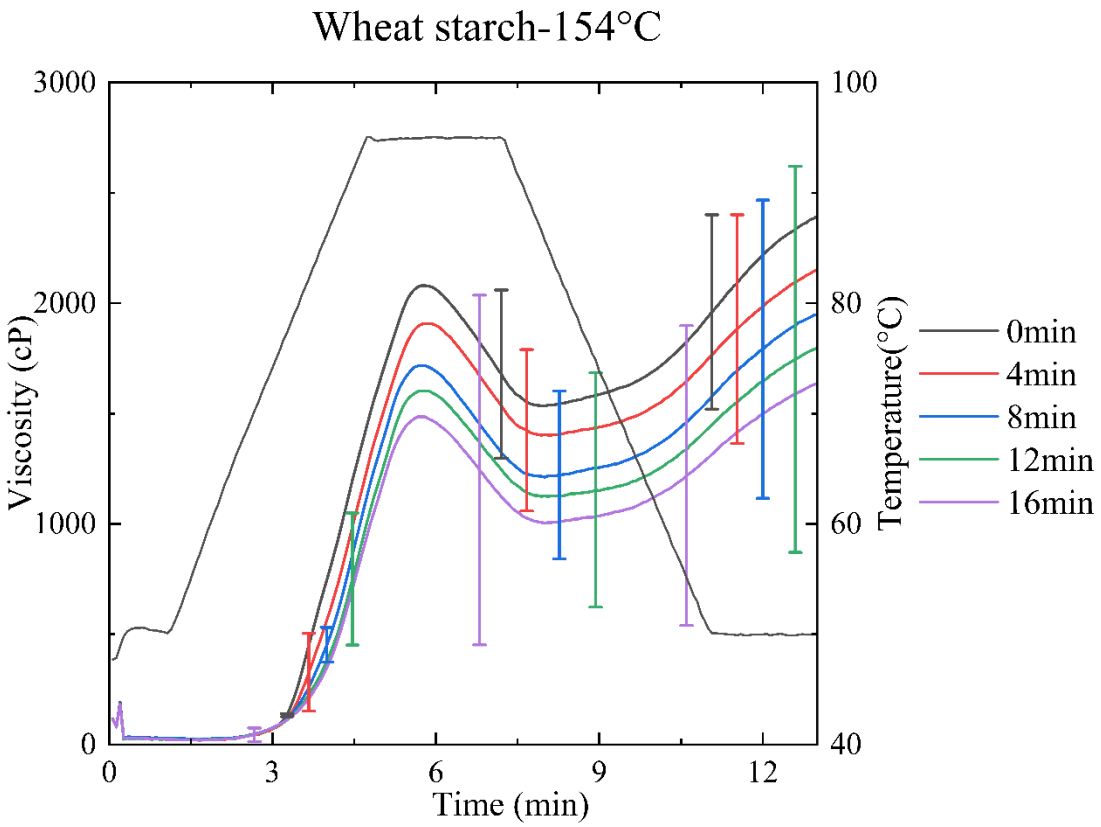

```
Fit Model(
    Y( :Peak viscosity, :Trough, :Setback ),
    Effects(
        :Powder type, :Cooking temperature, :Powder type * :Cooking temperature,
        :Cooking time, :Powder type * :Cooking time,
        :Cooking temperature * :Cooking time,
        :Powder type * :Cooking temperature * :Cooking time
    ),
    Personality( "Standard Least Squares" ),
    Emphasis( "Effect Leverage" ),
    Run(
        :Peak viscosity << {Summary of Fit( 1 ), Analysis of Variance( 1 ),
        Parameter Estimates( 1 ), Lack of Fit( 0 ), Scaled Estimates( 0 ),
        Plot Actual by Predicted( 1 ), Plot Regression( 0 ),
        Plot Residual by Predicted( 1 ), Plot Studentized Residuals( 0 ),
        Plot Effect Leverage( 1 ), Plot Residual by Normal Quantiles( 0 ),
        Box Cox Y Transformation( 0 )},
        :Trough << {Summary of Fit( 1 ), Analysis of Variance( 1 ),
        Parameter Estimates( 1 ), Lack of Fit( 0 ), Scaled Estimates( 0 ),
        Plot Actual by Predicted( 1 ), Plot Regression( 0 ),
        Plot Residual by Predicted( 1 ), Plot Studentized Residuals( 0 ),
        Plot Effect Leverage( 1 ), Plot Residual by Normal Quantiles( 0 ),
        Box Cox Y Transformation( 0 )},
        :Setback << {Summary of Fit( 1 ), Analysis of Variance( 1 ),
        Parameter Estimates( 1 ), Lack of Fit( 0 ), Scaled Estimates( 0 ),
        Plot Actual by Predicted( 1 ), Plot Regression( 0 ),
        Plot Residual by Predicted( 1 ), Plot Studentized Residuals( 0 ),
        Plot Effect Leverage( 1 ), Plot Residual by Normal Quantiles( 0 ),
        Box Cox Y Transformation( 0 )}
    )
);
```

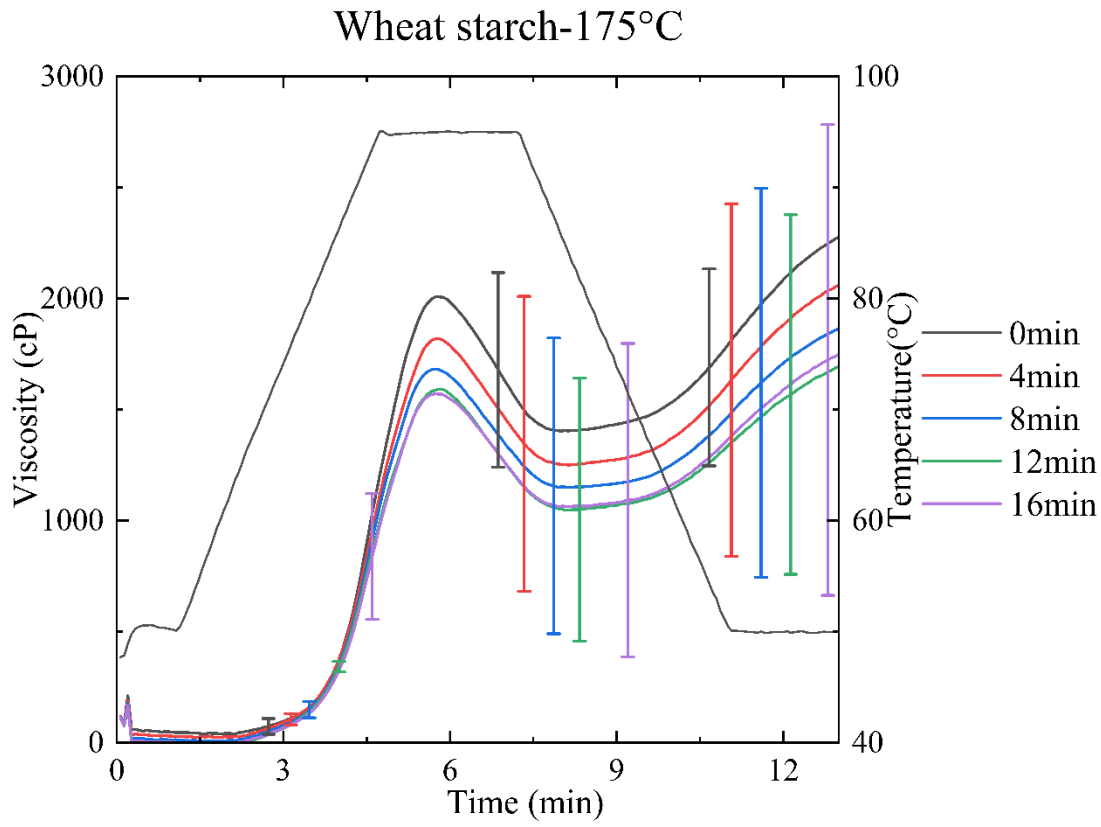
Appendix B: Detailed RVA profiles of wheat starch, wheat flour, pea starch, and pea flour in the cooked roux. The roux was cooked at either 100, 116, 134, 154, and 175 °C for 0, 4, 8, 12, or 16 minutes**.**

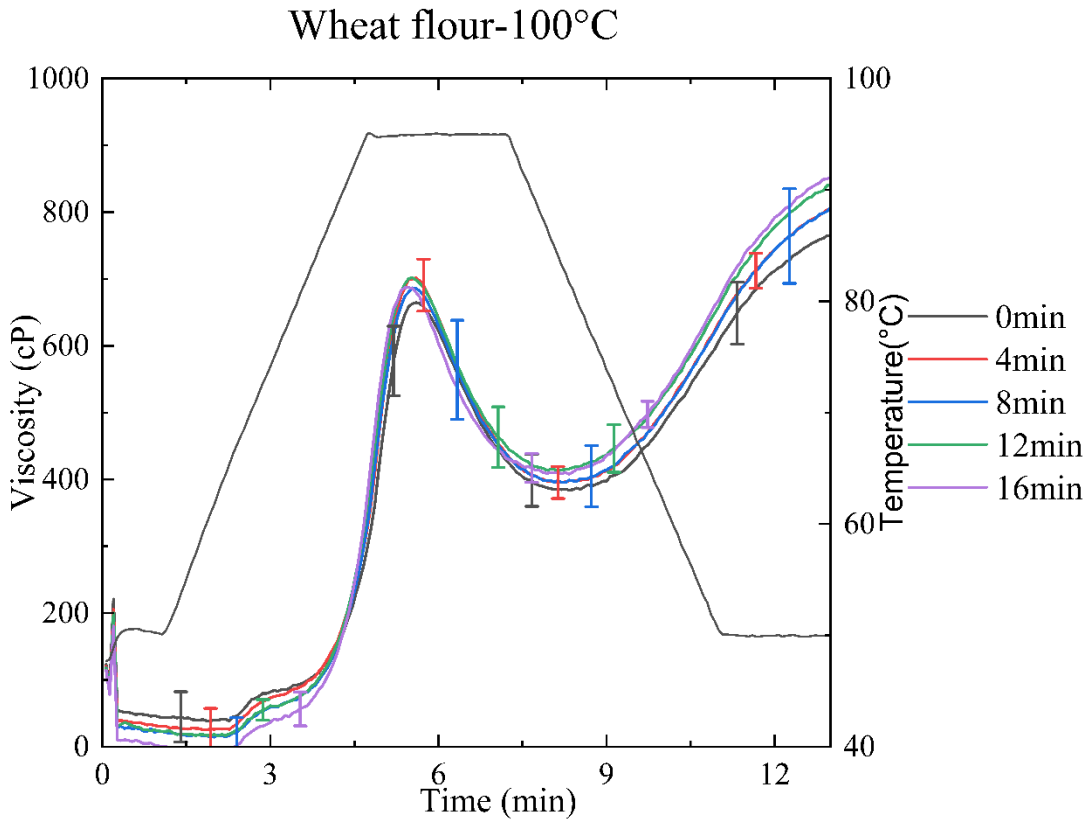


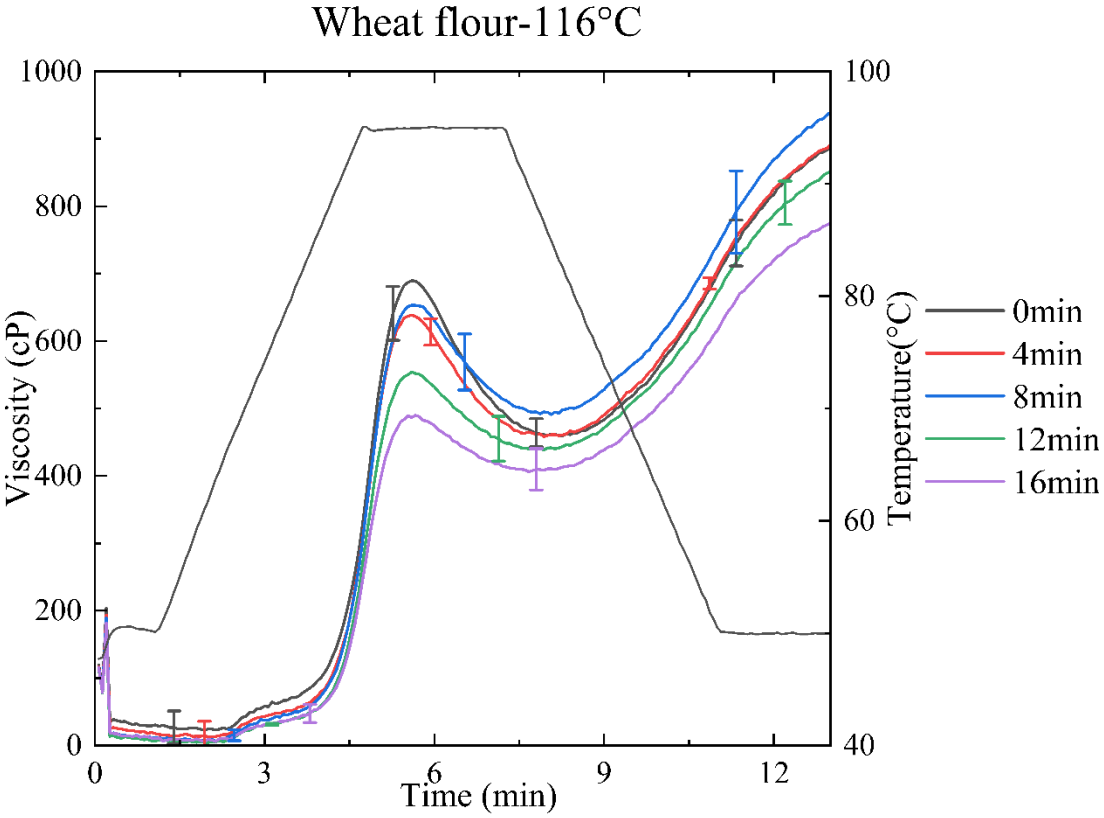

Wheat starch-116°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

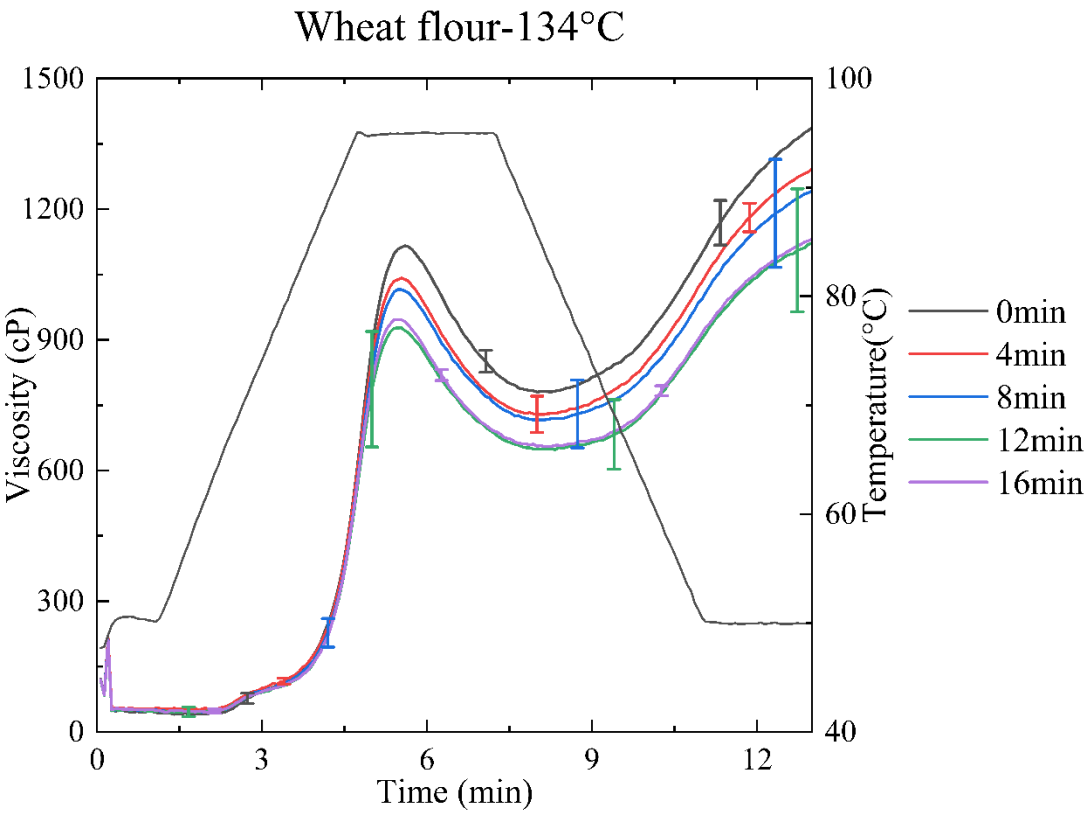

Wheat starch-134°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

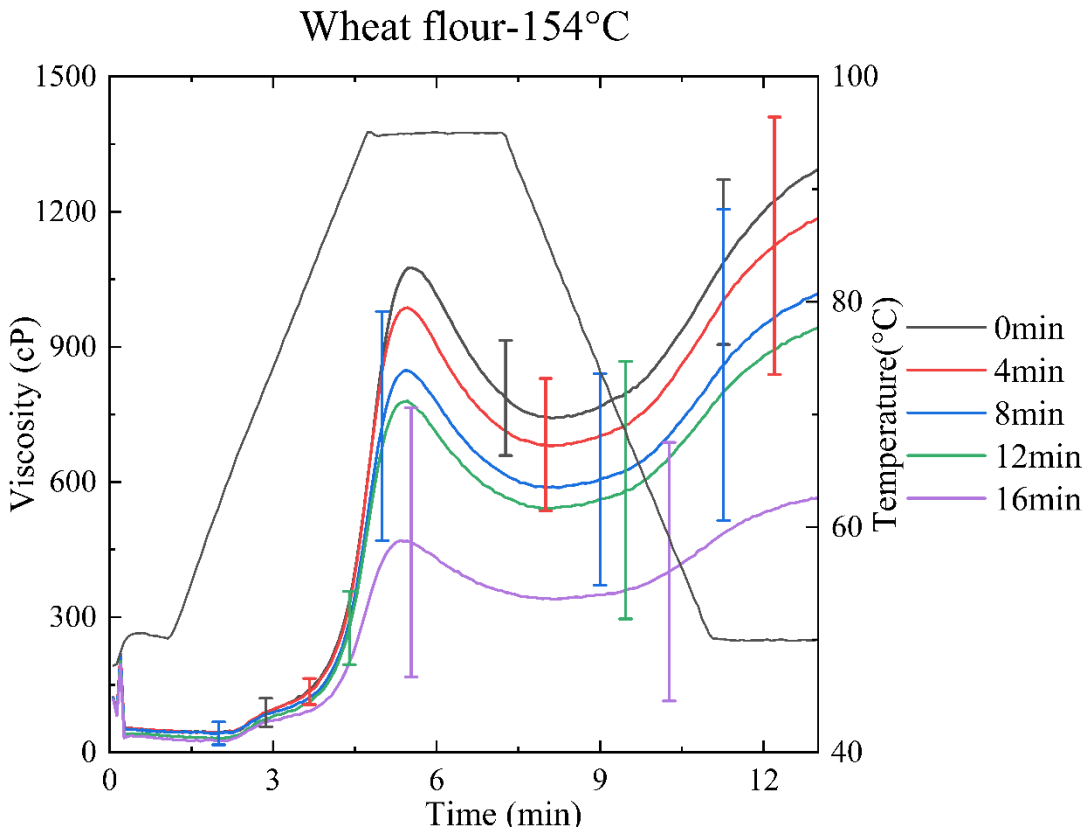

Wheat starch-154°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

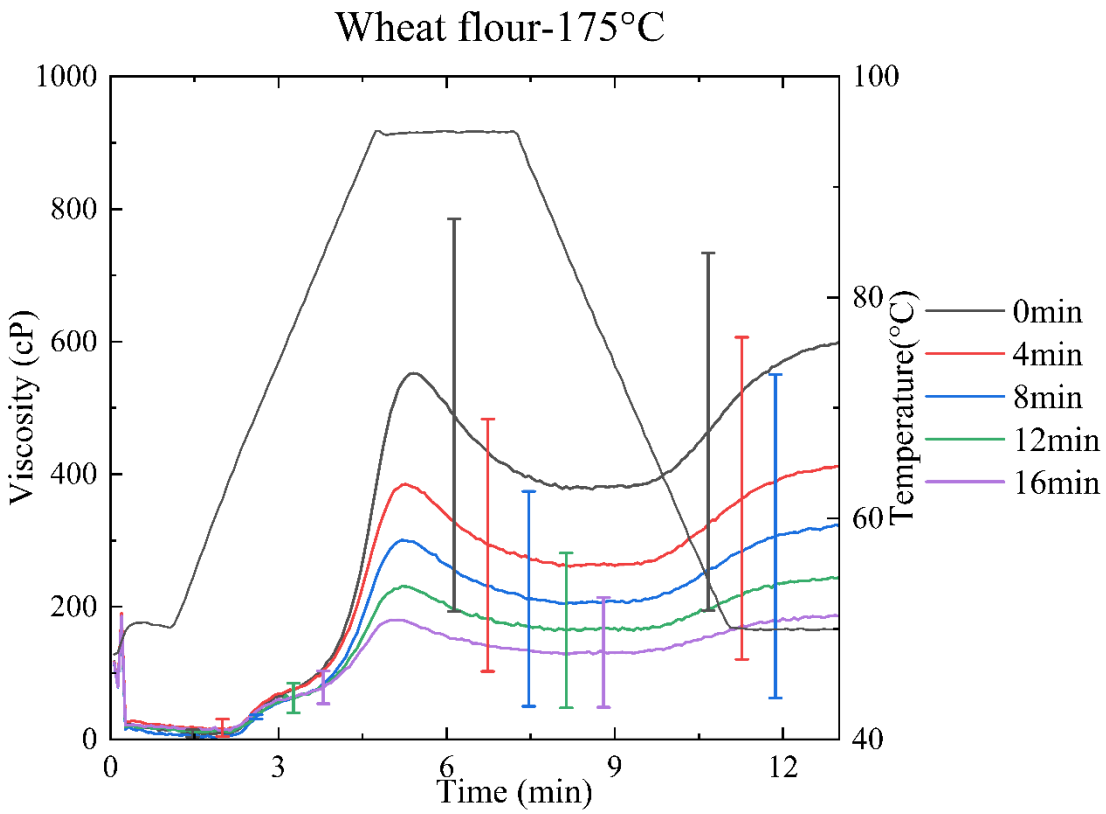

Wheat starch-175°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

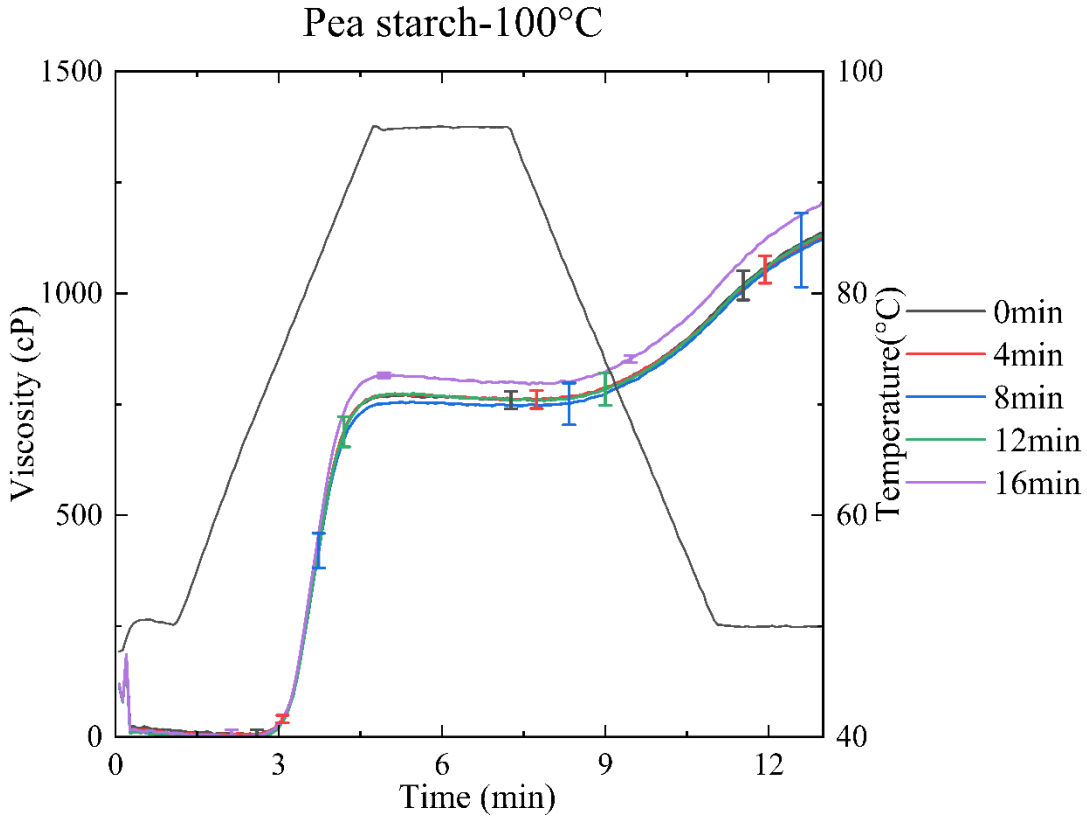

Wheat flour-100°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

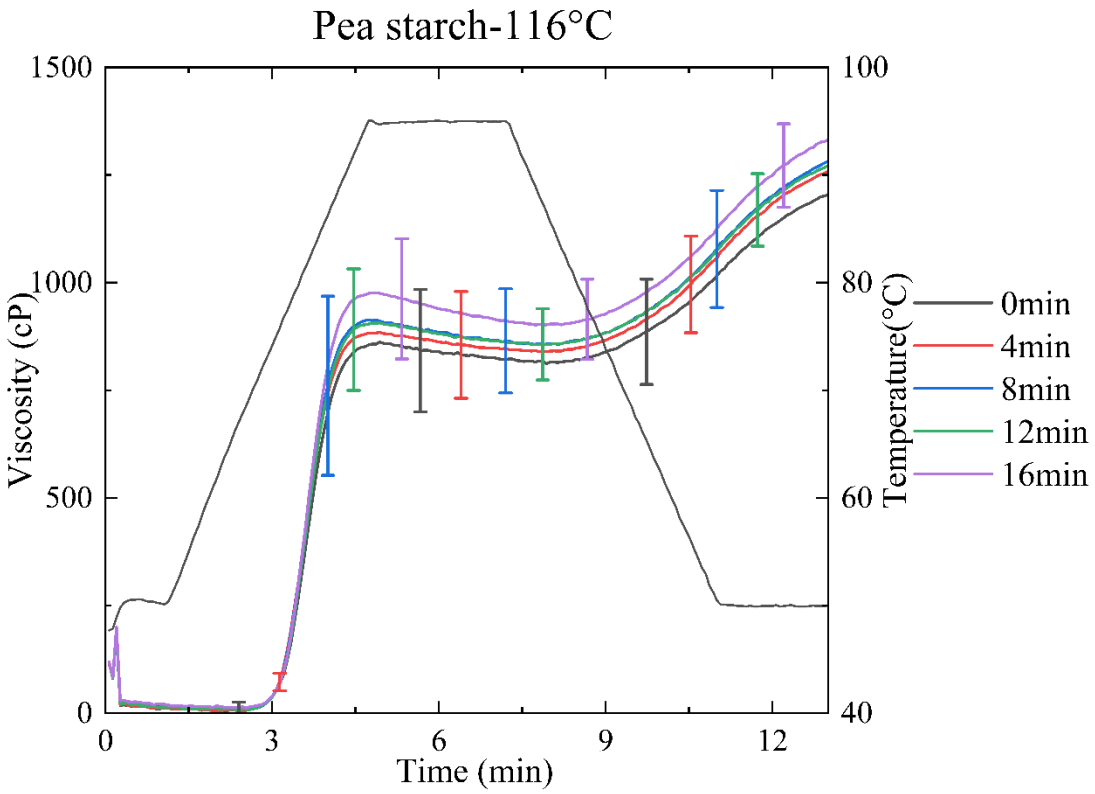

Wheat flour-116°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

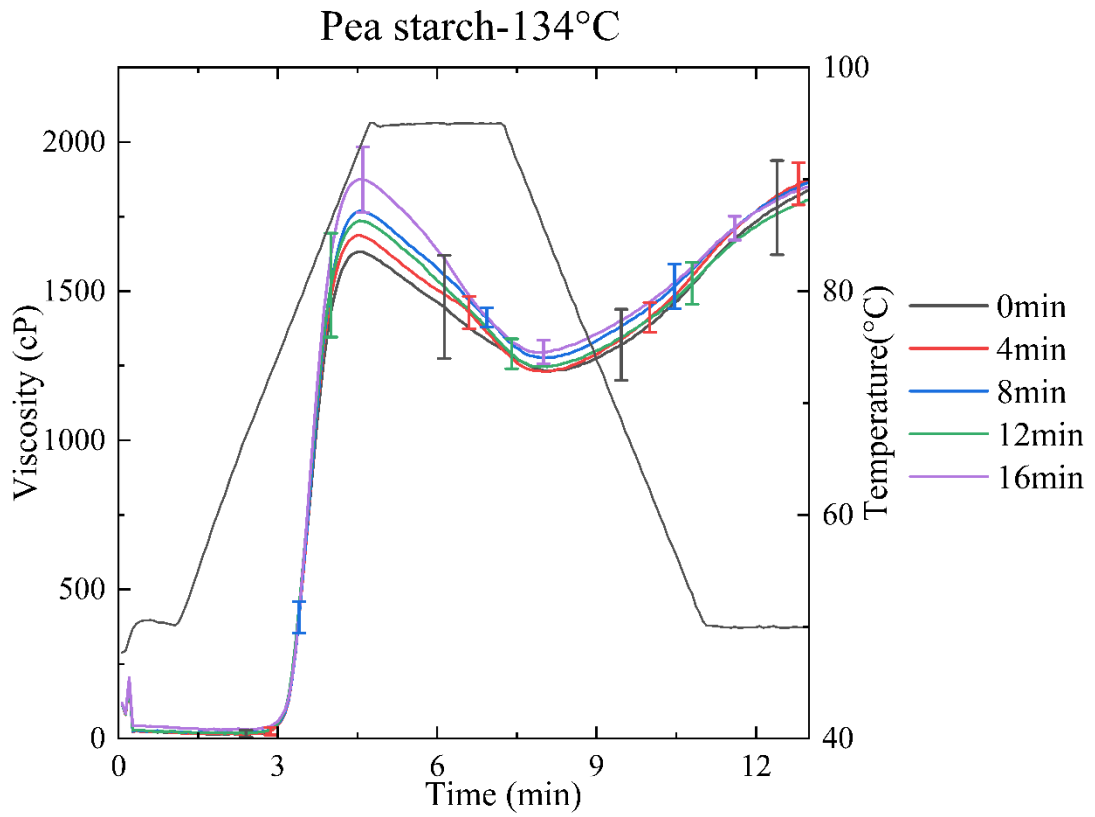

Wheat flour-134°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

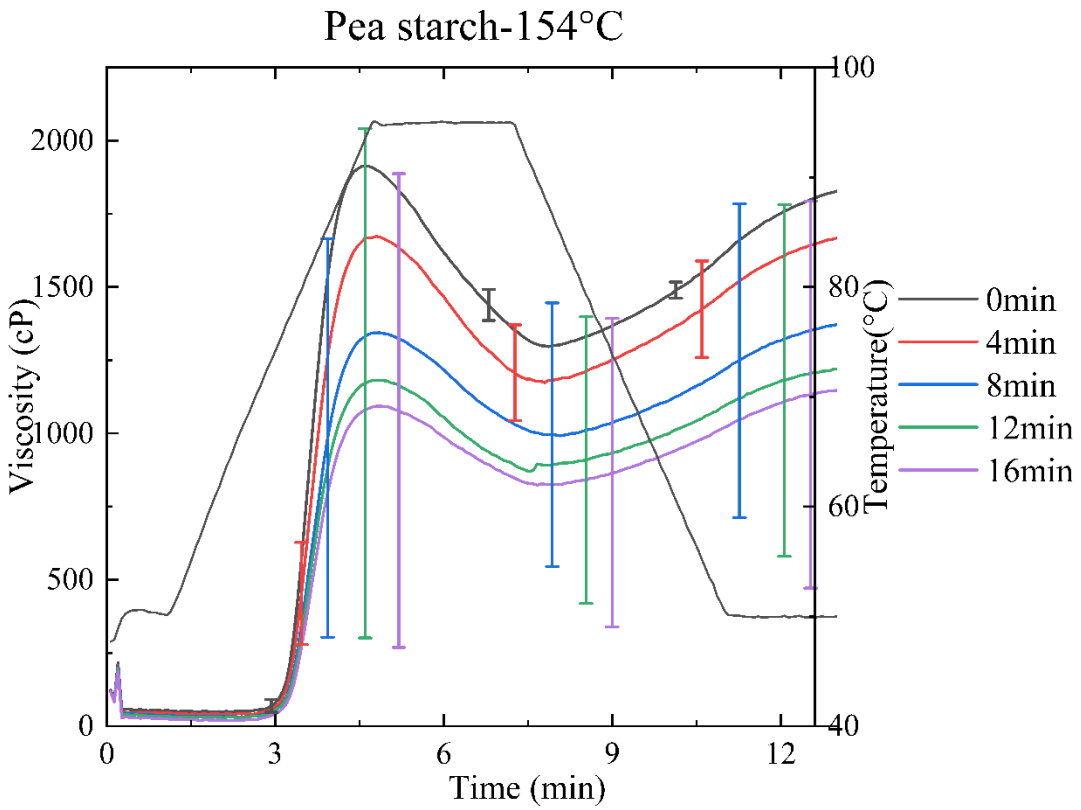

Wheat flour-154°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

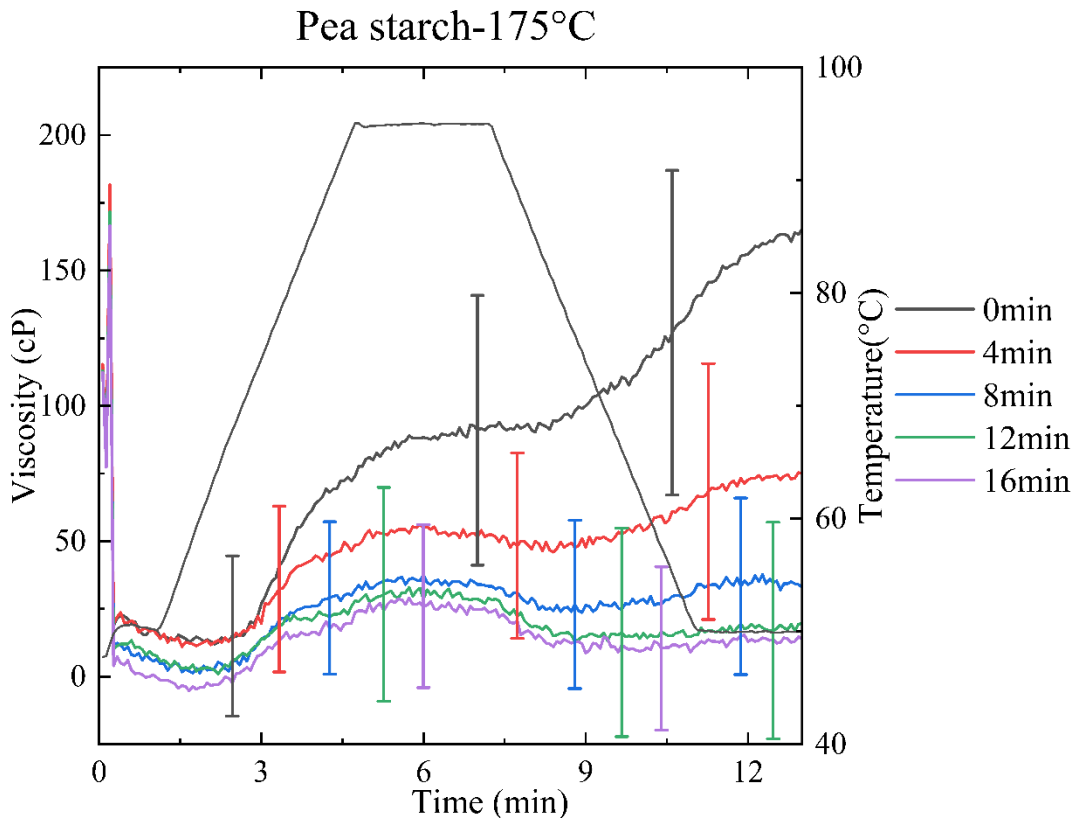

Wheat flour-175°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

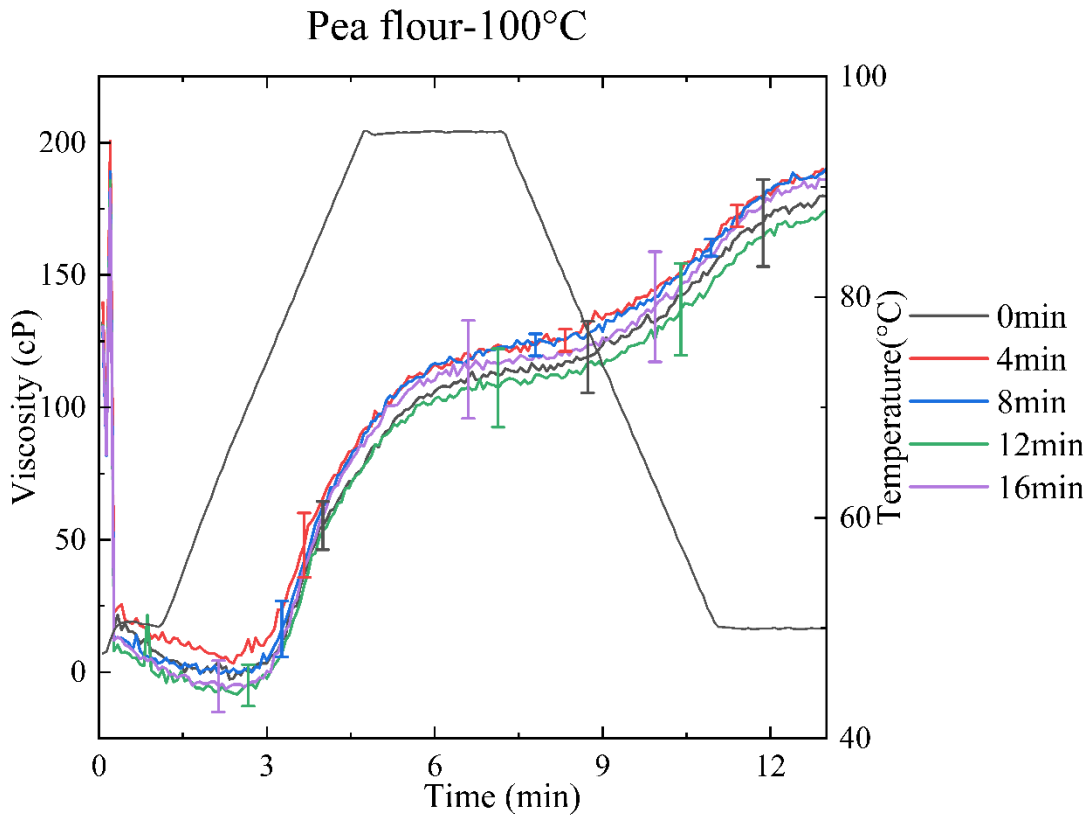

Pea starch-100°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

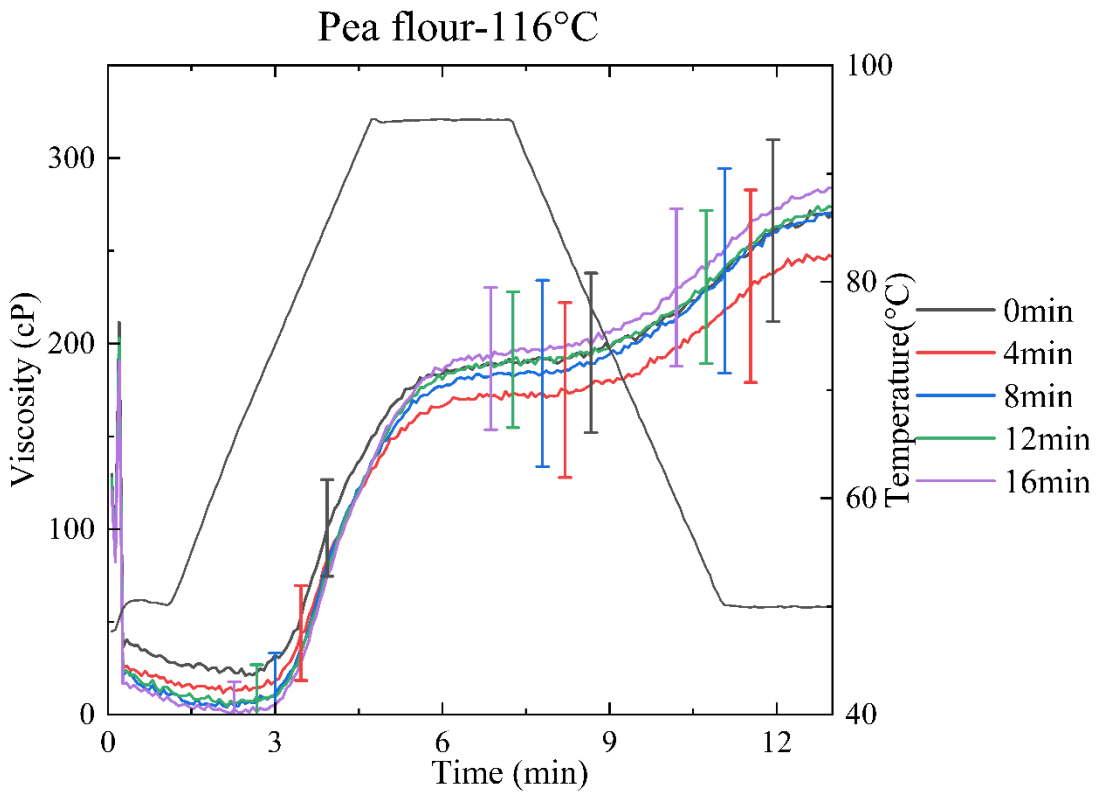

Pea starch-116°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

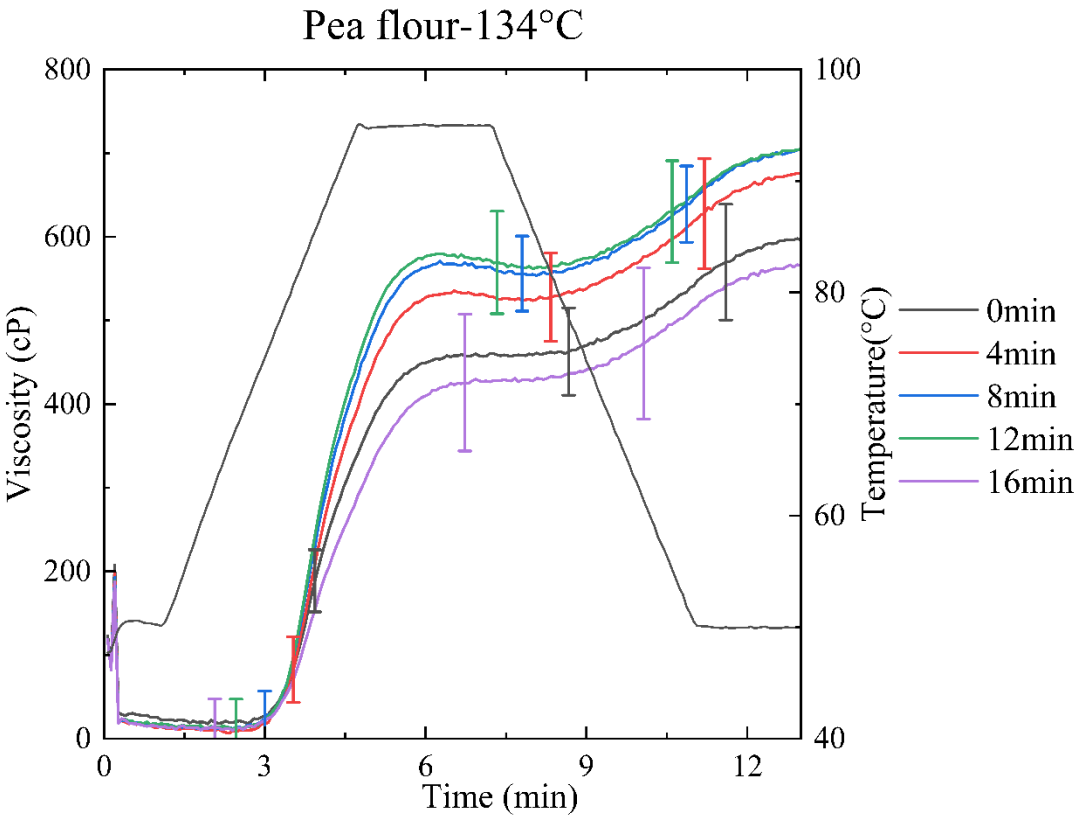

Pea starch-134°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

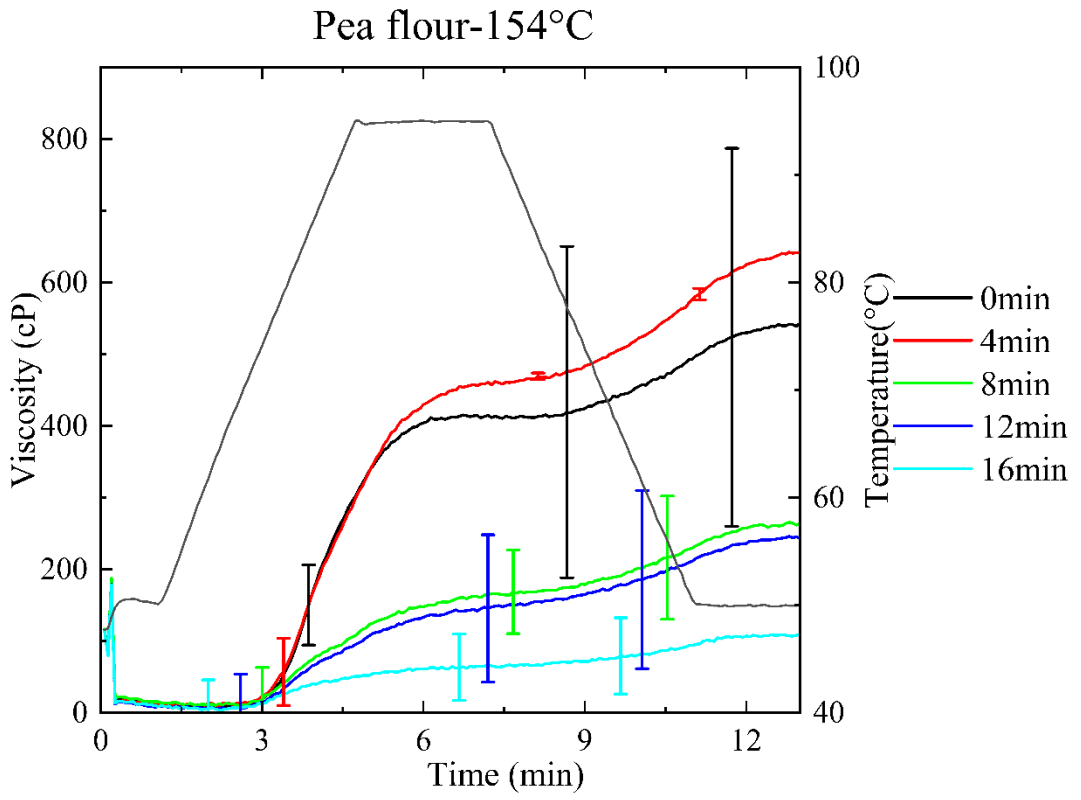

Pea starch-154°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min

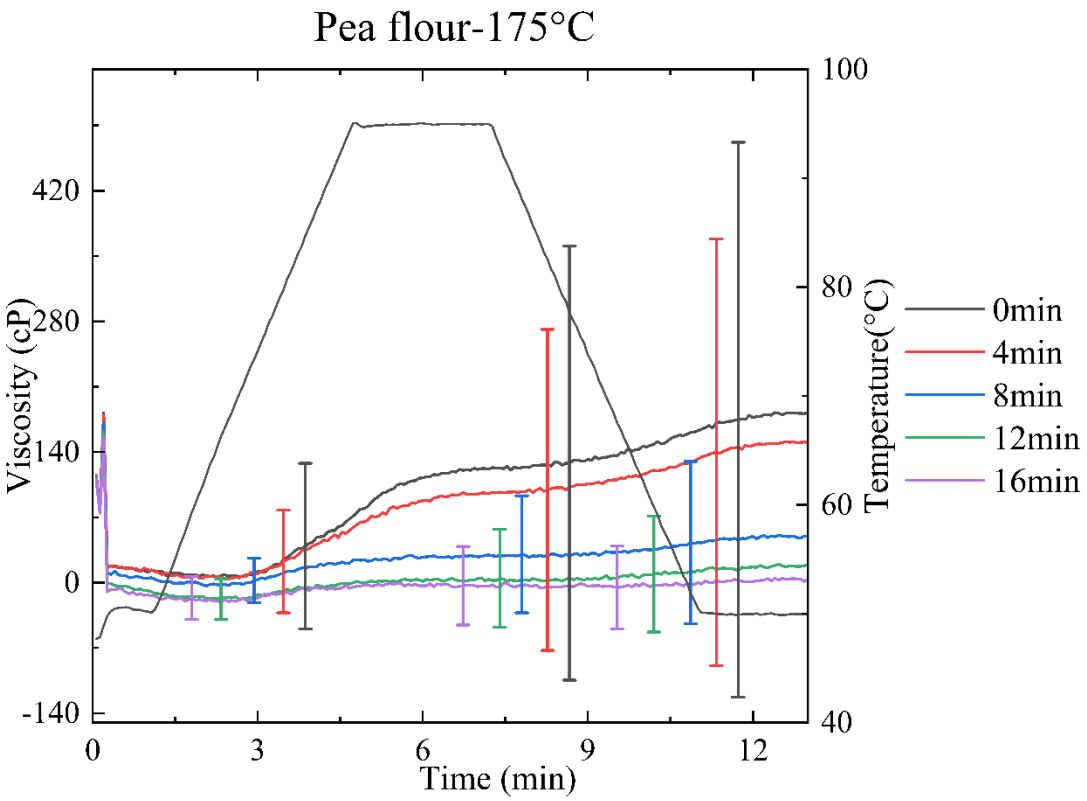

Pea starch-175°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min


Pea flour-100°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min


Pea flour-116°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min


Pea flour-134°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min


Pea flour-154°C
Viscosity (cP)
Temperature(°C)
Time (min)
0min
4min
8min
12min
16min


Pea flour-175°C
Viscosity (cP)
Temperature(°C)
Time (min)
420
280
140
0
-140
100
80
60
40
0
3
6
9
12
0min
4min
8min
12min
16min

Appendix C: Pasting properties of roux.

Table C. 1 Pasting properties of roux made from wheat starch cooked at 100 and 116 ℃ for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
100	0	1996 ± 212	1612 ± 130	384 ± 102	2332 ± 253	720 ± 139	83.05 ± 2.13
	4	2022 ± 77	1609 ± 21	413 ± 95	2384 ± 96	775 ± 116	82.27 ± 1.46
	8	1976 ± 49	1531 ± 109	445 ± 41	2336 ± 186	804 ± 80	82.48 ± 0.45
	12	1982 ± 26	1554 ± 90	428 ± 43	2303 ± 143	749 ± 59	82.73 ± 0.92
	16	1983 ± 125	1551 ± 65	432 ± 83	2305 ± 120	754 ± 72	82.82 ± 0.40
116	0	2047 ± 65	1650 ± 114	396 ± 58	2419 ± 71	769 ± 49	82.80 ± 0.48
	4	2026 ± 50	1591 ± 52	435 ± 10	2372 ± 40	781 ± 16	81.95 ± 0.39
	8	2022 ± 60	1624 ± 21	397 ± 78	2382 ± 90	757 ± 106	82.22 ± 0.03
	12	2010 ± 64	1555 ± 91	455 ± 27	2352 ± 37	798 ± 74	81.73 ± 1.24
	16	1959 ± 14	1534 ± 16	425 ± 26	2288 ± 17	754 ± 31	83.35 ± 0.48

Table C.2 Pasting properties of roux made from wheat starch cooked at 134 and 154 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
134	0	1924 ± 19	1543 ± 54	382 ± 69	2242 ± 31	699 ± 71	82.53 ± 0.40
	4	1900 ± 29	1531 ± 46	369 ± 29	2202 ± 36	671 ± 11	83.32 ± 0.51
	8	1952 ± 67	1506 ± 59	446 ± 14	2270 ± 104	764 ± 45	82.78 ± 0.42
	12	1974 ± 41	1495 ± 58	478 ± 38	2292 ± 54	797 ± 70	81.43 ± 0.73
	16	2009 ± 31	1529 ± 37	480 ± 18	2310 ± 60	781 ± 77	81.42 ± 0.80
154	0	2159 ± 333	1534 ± 318	625 ± 47	2394 ± 687	860 ± 369	77.70 ± 2.76
	4	1934 ± 549	1395 ± 356	539 ± 194	2150 ± 682	755 ± 328	77.97 ± 1.79
	8	1722 ± 749	1208 ± 385	515 ± 365	1949 ± 785	742 ± 404	78.47 ± 1.29
	12	1606 ± 929	1122 ± 531	484 ± 400	1797 ± 907	675 ± 377	78.50 ± 1.23
	16	1501 ± 1003	995 ± 589	506 ± 422	1637 ± 959	643 ± 374	78.55 ± 0.57

Table C.3 Pasting properties of roux made from wheat starch cooked at 175 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
175	0	2043 ± 327	1402 ± 406	641 ± 95	2278 ± 615	876 ± 226	80.88 ± 1.03
	4	1870 ± 689	1248 ± 636	622 ± 55	2061 ± 1025	813 ± 393	79.23 ± 1.20
	8	1727 ± 754	1147 ± 668	579 ± 86	1864 ± 1017	716 ± 351	78.78 ± 3.23
	12	1646 ± 696	1046 ± 589	600 ± 110	1694 ± 884	648 ± 295	79.25 ± 1.72
	16	1612 ± 850	1062 ± 692	550 ± 165	1750 ± 1079	688 ± 391	80.07 ± 1.21

Table C.4 Pasting properties of roux made from wheat flour cooked at 100 and 116 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
100	0	666 ± 40	383 ± 40	283 ± 3	764 ± 45	381 ± 5	90.33 ± 0.83
	4	702 ± 40	395 ± 24	307 ± 18	804 ± 41	409 ± 20	89.02 ± 0.46
	8	687 ± 72	395 ± 41	292 ± 32	807 ± 80	412 ± 40	88.95 ± 0.48
	12	703 ± 82	413 ± 35	290 ± 51	840 ± 72	427 ± 40	88.50 ± 1.24
	16	652 ± 80	388 ± 51	264 ± 31	810 ± 84	422 ± 36	89.23 ± 0.88
116	0	691 ± 36	459 ± 23	231 ± 16	886 ± 40	427 ± 25	89.80 ± 0.39
	4	640 ± 16	457 ± 10	183 ± 6	891 ± 19	434 ± 16	89.47 ± 0.83
	8	654 ± 53	490 ± 34	164 ± 21	940 ± 74	450 ± 42	89.53 ± 0.06
	12	554 ± 34	437 ± 29	117 ± 6	852 ± 37	415 ± 10	90.08 ± 0.42
	16	491 ± 48	405 ± 31	86 ± 17	776 ± 60	371 ± 30	90.82 ± 0.49

Table C.5 Pasting properties of roux made from wheat flour cooked at 134 and 154 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
134	0	1117 ± 55	780 ± 24	338 ± 31	1387 ± 60	608 ± 36	86.57 ± 0.51
	4	1044 ± 40	727 ± 42	317 ± 3	1293 ± 41	566 ± 5	86.28 ± 0.78
	8	1016 ± 108	714 ± 71	302 ± 37	1241 ± 136	528 ± 65	86.55 ± 1.28
	12	931 ± 115	647 ± 68	284 ± 47	1122 ± 144	475 ± 77	87.08 ± 1.63
	16	942 ± 23	656 ± 3	286 ± 27	1126 ± 29	470 ± 32	84.95 ± 0.43
154	0	1077 ± 162	740 ± 120	337 ± 49	1294 ± 226	554 ± 115	86.82 ± 0.54
	4	987 ± 204	678 ± 149	309 ± 55	1186 ± 314	508 ± 173	86.03 ± 1.85
	8	849 ± 322	585 ± 219	264 ± 106	1017 ± 428	433 ± 211	87.08 ± 2.18
	12	782 ± 383	540 ± 258	242 ± 128	942 ± 498	401 ± 240	88.07 ± 2.58
	16	692 ± 363	479 ± 242	213 ± 126	827 ± 462	348 ± 221	88.98 ± 2.56

Table C.6 Pasting properties of roux made from wheat flour cooked at 175 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
175	0	556 ± 331	375 ± 208	181 ± 123	599 ± 366	224 ± 157	88.67 ± 2.83
	4	366 ± 278	246 ± 180	120 ± 98	388 ± 308	142 ± 129	N/A
	8	326 ± 220	216 ± 140	109 ± 81	347 ± 243	131 ± 103	N/A
	12	234 ± 175	161 ± 115	74 ± 60	244 ± 194	84 ± 80	N/A
	16	185 ± 125	127 ± 82	58 ±4	186 ± 26	59 ± 5	N/A

Table C.7 Pasting properties of roux made from pea starch cooked at 100 and 116 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
100	0	774 ± 27	757± 21	16 ± 11	1137 ± 43	380 ± 22	75.87 ± 0.08
	4	775 ± 21	758 ± 18	17 ± 6	1128 ± 35	370 ± 17	75.82 ± 0.08
	8	756 ± 46	744 ± 44	12 ± 5	1123 ± 86	378 ± 42	75.90 ± 0.80
	12	777 ± 35	756 ± 33	21 ± 3	1133 ± 55	377 ± 25	75.88 ± 0.03
	16	817 ± 6	794 ± 6	23 ± 5	1207 ± 15	413 ± 14	74.75 ± 1.33
116	0	864 ± 156	812 ± 117	52 ± 42	1204 ± 172	392 ± 56	75.30 ± 0.48
	4	887 ± 157	838 ± 108	49 ± 54	1258 ± 139	420 ± 31	75.07 ± 0.83
	8	919 ± 173	856 ± 115	63 ± 60	1282 ± 155	426 ±40	75.53 ± 1.16
	12	909 ± 33	854 ± 84	55 ± 49	1272 ± 86	418 ± 3	75.28 ± 0.49
	16	978 ± 51	900 ± 98	78 ± 53	1332 ± 98	432 ± 7	75.30 ± 0.91

Table C.8 Pasting properties of roux made from pea starch cooked at 134 and 154 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
134	0	1633 ± 238	1228 ± 89	405 ± 159	1840 ± 166	612 ± 77	74.48 ± 0.49
	4	1689 ± 99	1230 ± 41	459 ± 59	1876 ± 70	646 ± 34	75.37 ± 0.55
	8	1769 ± 157	1274 ± 59	495 ± 99	1864 ± 72	589 ± 13	75.02 ± 0.03
	12	1737 ± 187	1244 ± 54	493 ± 136	1807 ± 76	563 ± 47	74.57 ± 1.24
	16	1876 ± 109	1292 ± 39	584 ± 72	1850 ± 58	557 ±74	74.47 ± 0.43
154	0	1917 ± 141	1295 ± 41	622 ± 143	1828 ± 103	533 ± 92	74.85 ±0.91
	4	1683 ± 237	1171 ± 141	512 ± 102	1667 ± 228	496 ± 89	75.05 ± 0.78
	8	1381 ± 784	990 ± 452	391 ± 333	1372 ± 586	382 ± 135	76.97 ± 2.58
	12	1222 ± 821	857 ± 482	365 ± 346	1221 ± 620	364 ± 138	75.35 ± 0.57
	16	1129 ± 826	812 ± 521	317 ± 313	1146 ± 672	335 ± 153	75.80 ± 1.20

Table C.9 Pasting properties of roux made from pea starch cooked at 175 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
175	0	58 ± 36	43 ± 36	14 ±5	75 ±50	32 ± 20	N/A
	4	39 ± 30	22 ± 31	17 ± 1	33 ± 33	11 ± 8	N/A
	8	34 ± 37	11 ± 41	23 ± 4	20 ± 35	8 ± 6	N/A
	12	31 ± 30	7 ± 31	24 ± 1	14 ± 32	N/A	N/A
	16	114 ± 11	107 ± 10	7 ± 1	180 ± 14	73 ± 4	N/A

Table C.10 Pasting properties of roux made from pea flour cooked at 100 and 116 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
100	0	123 ± 4	114 ± 5	9 ±1	189 ± 2	74 ± 5	N/A
	4	122 ± 3	116 ± 4	7 ± 1	190 ± 8	75 ± 11	N/A
	8	112 ± 14	101 ± 15	10 ± 1	174 ± 22	73 ± 8	N/A
	12	119 ± 19	111 ± 19	8 ± 1	186 ± 26	75 ± 8	N/A
	16	191 ± 44	184 ± 44	7 ± 1	271 ± 49	87 ± 9	N/A
116	0	174 ± 44	167 ± 45	8 ± 1	247 ± 51	80 ± 8	N/A
	4	185 ± 52	177 ± 51	7 ± 2	268 ± 55	91 ±5	N/A
	8	192 ± 33	183 ± 31	9 ± 2	274 ± 44	91 ± 13	N/A
	12	196 ± 39	187 ± 38	9 ± 1	284 ± 48	97 ± 11	N/A
	16	461 ± 48	449 ± 41	12 ± 7	596 ± 72	147 ± 40	81.13 ± 0.88

Table C.11 Pasting properties of roux made from pea flour cooked at 134 and 154 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cP)	Setback (cP)	Pasting Temperature (°C)
134	0	538 ± 44	521 ± 49	17 ± 6	676 ± 68	155 ± 22	80.92 ± 0.51
	4	571 ± 50	550 ± 45	21 ± 9	704 ± 49	153 ± 6	80.32 ± 1.17
	8	581 ± 65	560 ± 58	21 ± 8	705 ± 65	144 ± 10	79.57 ± 0.89
	12	431 ± 81	411 ± 81	20 ± 7	566 ± 98	154 ± 44	83.03 ± 2.13
	16	523 ± 72	507 ± 66	15 ± 6	657 ± 83	149 ± 23	80.88 ± 1.69
154	0	68 ± 16	447 ± 23	22 ± 8	627 ± 21	181 ± 43	80.25 ± 1.63
	4	274 ± 138	258 ± 146	16 ± 9	402 ± 124	143 ± 26	N/A
	8	287 ± 213	273 ± 215	14 ± 4	427 ± 222	154 ± 27	N/A
	12	207 ± 255	199 ± 254	8 ± 1	294 ± 302	95 ± 50	N/A
	16	125 ± 213	109 ± 203	16 ± 11	182 ±307	74 ± 104	N/A

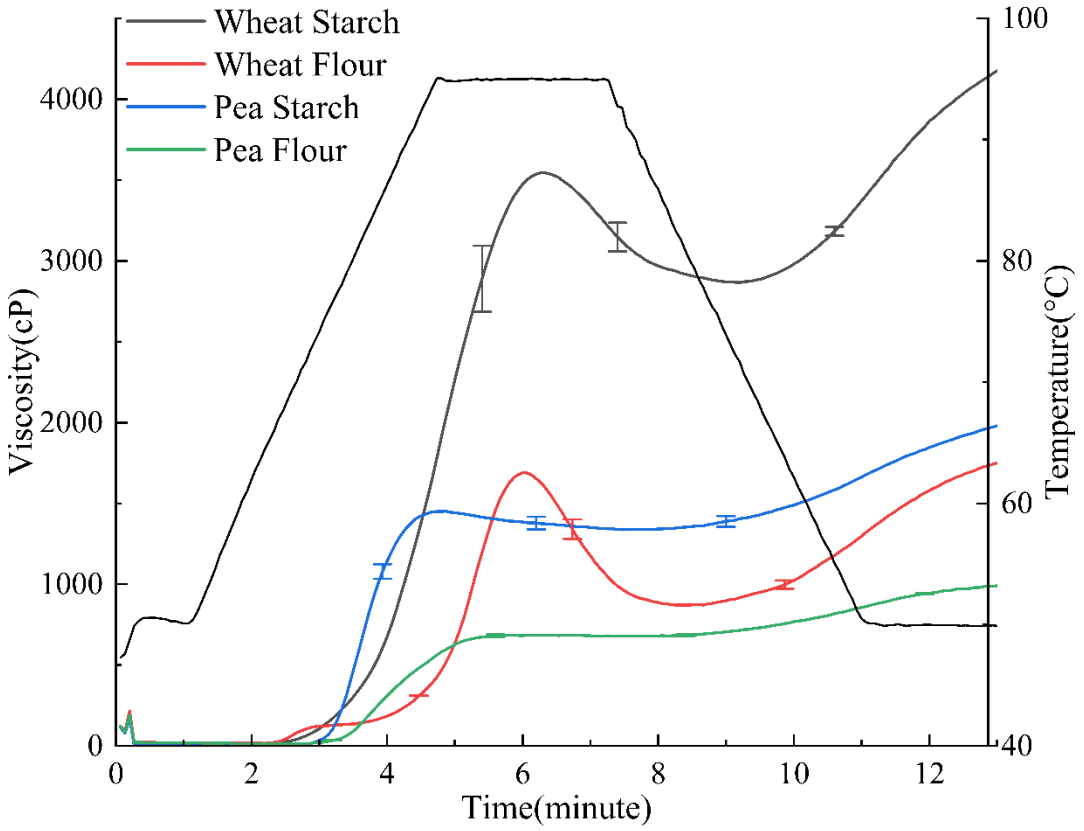
Table C.12 Pasting properties of roux made from pea flour cooked at 175 °C for 0, 4, 8, 12, and 16 min. Values are mean ± SD.

Cooking Temperature (°C)	Cooking time (min)	Peak viscosity (cP)	Trough (cP)	Breakdown (cP)	Final Viscosity (cp)	Setback (cP)	Pasting Temperature (°C)
175	0	99 ± 161	84 ± 154	14 ± 7	150 ± 251	66 ± 97	N/A
	4	33 ± 57	21 ± 63	11 ± 6	50 ± 99	28 ± 38	N/A
	8	9 ± 51	2 ± 54	11 ± 3	19 ± 78	21 ± 25	N/A
	12	1 ± 39	10 ± 42	13 ± 3	7± 40	15± 21	N/A
	16	1 ± 36	5± 27	11 ± 3	4 ± 59	14 ± 17	N/A

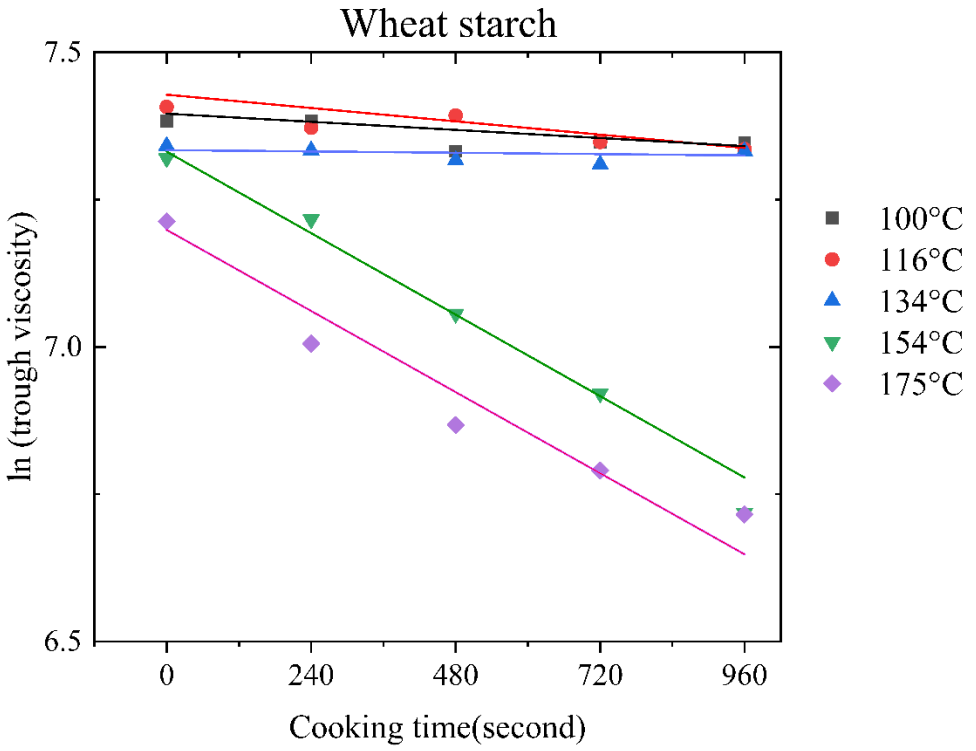
Appendix D: A more traditional ANOVA table for the effect test of experimental design parameters on the peak viscosity, trough viscosity, and setback of the RVA profiles of starch or flour from the cooked roux.

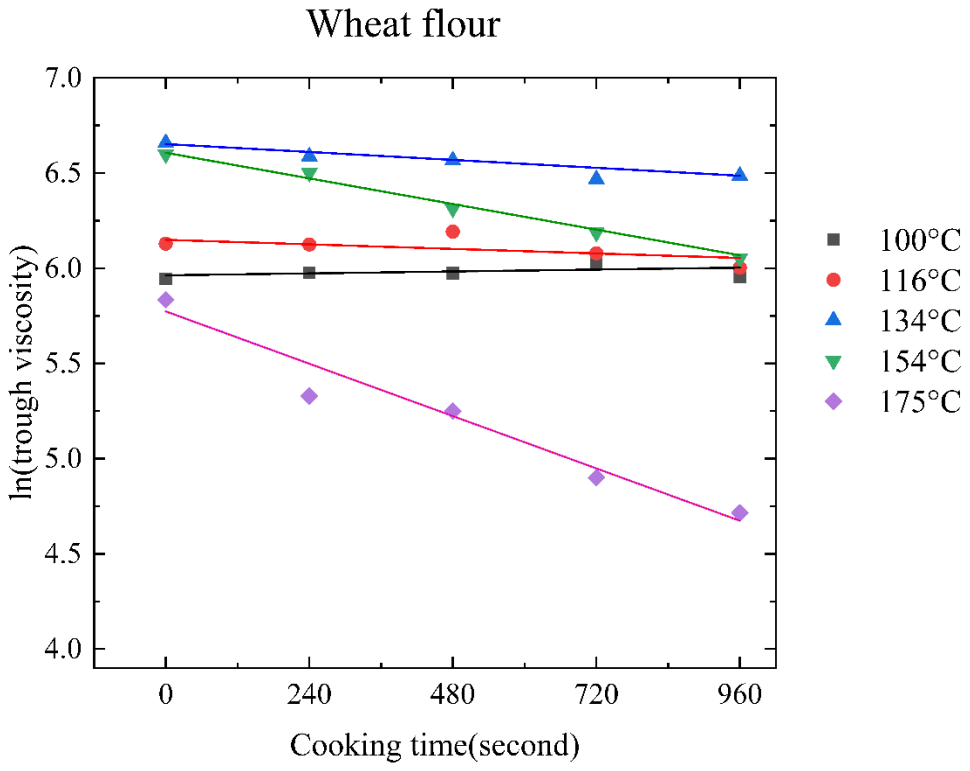
	Peak viscosity				Trough viscosity				Setback			
Source	**DF**	**SS**	**F -value**	**Pr>F**	**DF**	**SS**	**F -value**	**Pr>F**	**DF**	**SS**	**F value**	**Pr>F**
Starch/flour type	3	110194592	423.5433	<.0001*	3	59609064	459.4982	<.0001*	3	16118294	306.1773	<.0001*
Cooking temperature	4	18869843	54.3960	<.0001*	4	12408327	71.7375	<.0001*	4	2383402	33.9557	<.0001*
Cooking time	4	986374	2.8434	0.0253*	4	666776	3.8549	0.0049*	4	139541	1.9880	0.0977
Starch/flour type*Cooking temperature	12	13931344	13.3866	<.0001*	12	6642065	12.8001	<.0001*	12	1739862	8.2624	<.0001*
Starch/flour type*Cooking time	12	190823	0.1834	0.9989	12	137281	0.2646	0.9937	12	39922	0.1896	0.9987
Cooking temperature *Cooking time	16	1841397	1.3270	0.1834	16	957158	1.3834	0.1526	16	225572	0.8034	0.6809
Starch/flour type*Cooking temperature *Cooking time	48	671152	0.1612	1.0000	48	238430	0.1149	1.0000	48	169117	0.2008	1.0000
Error	200	17344877			200	8648430			200	3509578		

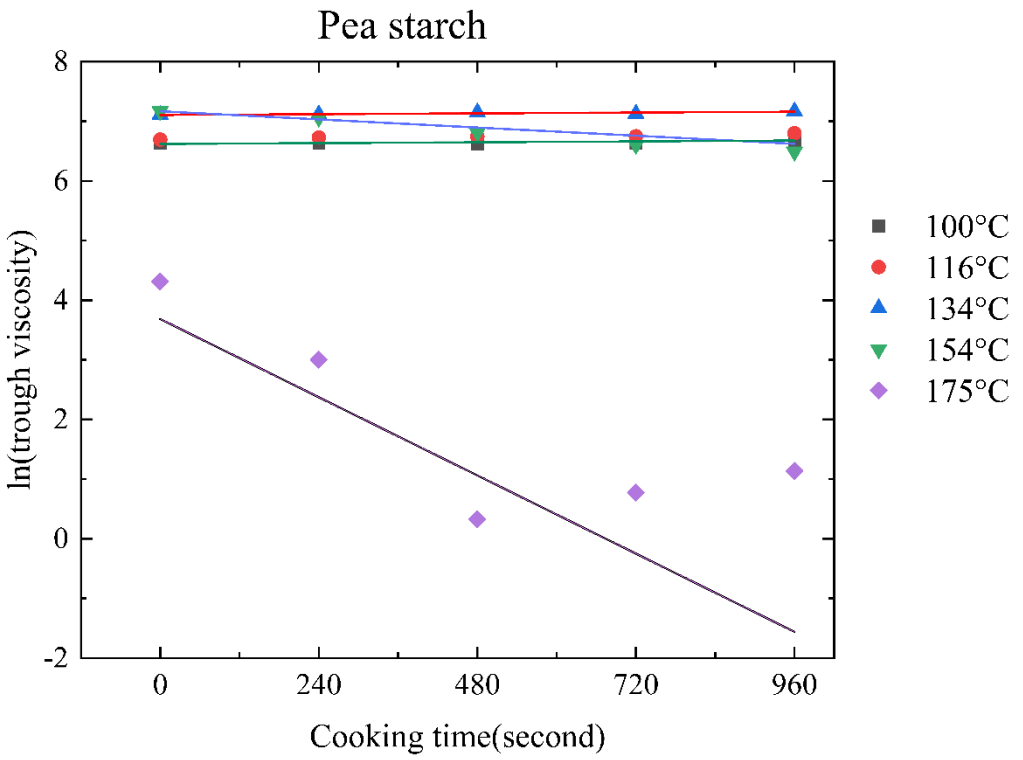
Appendix E: Pasting profiles of raw materials (wheat starch, wheat flour, pea starch, and pea flour).

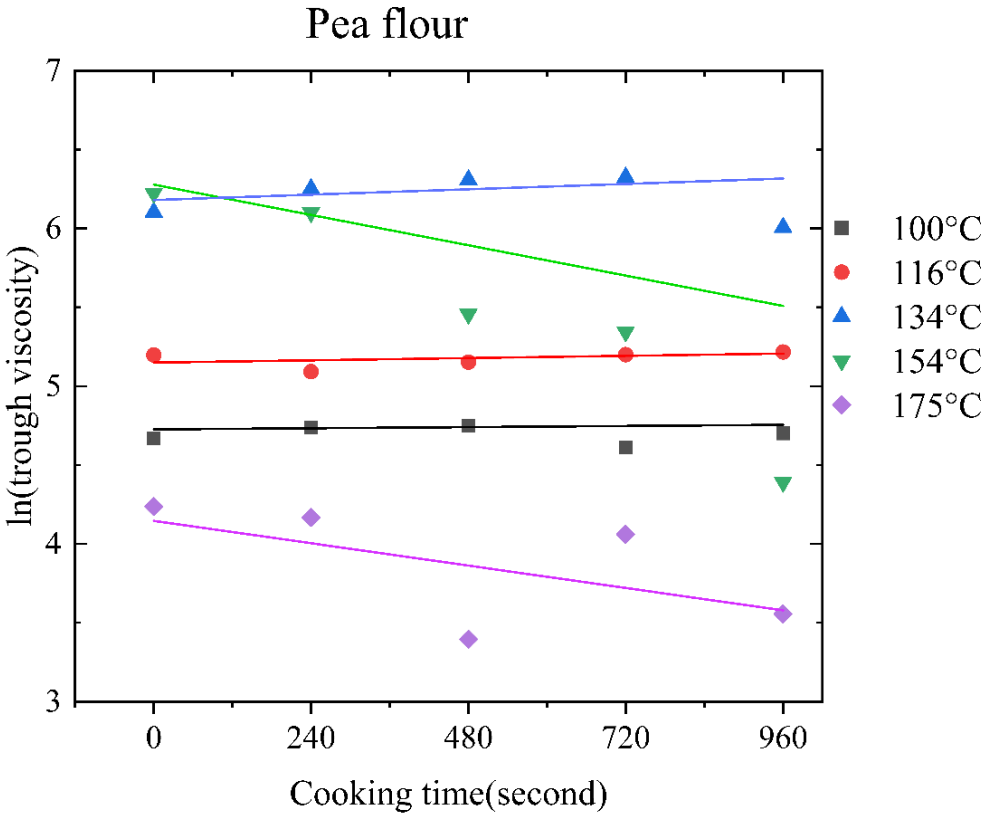


Appendix F: Figures of the plots for the logarithm of trough viscosity as a function of cooking time and the linear fitting.






Pea starch
ln(trough viscosity)
Cooking time(second)
100°C
116°C
134°C
154°C
175°C


Pea flour
ln(trough viscosity)
Cooking time(second)
100°C
116°C
134°C
154°C
175°C

Appendix G: Oil content of the roux made from different starch and flour and cooked different conditions. Values are mean ± SD.

Cooking Temperature (˚C)	Cooking Time (min)	Wheat Starch	Wheat flour	Pea starch	Pea flour
	0	14.63 ± 1.27	21.91 ± 6.78	15.34 ± 0.23	29.95 ± 2.75
	4	15.42 ± 3.50	25.72 ± 1.83	15.92 ± 0.08	29.29 ± 1.58
	8	12.72 ± 0.20	26.89 ± 4.58	19.62 ± 4.03	28.68 ± 0.00
	12	13.19 ± 0.10	25.28 ± 3.70	11.69 ± 6.36	31.52 ± 3.41
100	16	14.36 ± 0.64	22.72 ± 10.37	16.51 ± 0.19	27.30 ± 2.57
	0	12.96 ± 2.44	25.97 ± 0.39	16.58 ± 0.91	26.63 ± 1.64
	4	12.70 ± 0.11	26.38 ± 1.31	17.04 ± 0.98	26.91 ± 1.99
	8	13.39 ± 0.27	40.17 ± 22.60	16.53 ± 1.33	28.77 ± 2.78
	12	12.30 ± 0.00	26.06 ± 0.65	16.95 ± 0.48	28.92 ± 1.70
116	16	13.55 ± 0.67	27.82 ± 0.74	15.86 ± 0.44	22.56 ± 7.60
	0	14.64 ± 2.09	22.37 ± 1.00	14.30 ± 1.59	21.48 ± 13.08
	4	15.31 ± 0.06	24.72 ± 0.45	13.83 ± 0.00	21.60 ± 3.36
	8	15.43 ± 1.01	38.43 ± 17.91	14.06 ± 0.59	25.72 ± 5.82
	12	14.31 ± 1.41	22.19 ± 5.05	14.44 ± 2.22	21.05 ± 6.88
134	16	14.85 ± 0.77	24.53 ± 0.69	13.24 ± 0.65	24.38 ± 3.68
	0	12.25 ± 0.46	23.15 ± 0.13	12.40 ± 0.96	26.66 ± 3.71
	4	11.16 ± 2.74	18.93 ± 5.49	12.87 ± 0.66	24.40 ± 8.48
	8	12.08 ± 1.00	23.57 ± 0.23	12.84 ± 0.18	25.51 ± 2.64
	12	12.17 ± 0.45	26.02 ± 4.36	12.39 ± 0.14	19.84 ± 2.78
154	16	13.19 ± 3.58	24.87 ± 2.34	12.48 ± 0.32	21.04 ± 2.83
	0	12.72 ± 0.03	23.67 ± 2.81	13.02 ± 0.31	28.27 ± 4.80
	4	15.29 ± 5.22	24.21 ± 3.16	13.10 ± 0.63	26.68 ± 3.91
	8	12.82 ± 0.04	26.05 ± 3.31	13.79 ± 0.00	27.18 ± 4.28
	12	12.80 ± 0.55	26.77 ± 0.67	14.73 ± 0.45	26.07 ± 2.37
175	16	13.03 ± 1.26	26.16 ± 3.89	14.71 ± 0.81	27.40 ± 4.17