SUCROSE CRYSTALLIZATION BY USING MICROWAVE-VACUUM EVAPORATION

A THESIS SUBMITTED TO
THE GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES
OF
MIDDLE EAST TECHNICAL UNIVERSITY

BY

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IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR
THE DEGREE OF MASTER OF SCIENCE
IN
FOOD ENGINEERING

SEPTEMBER 2023

Approval of the thesis:

SUCROSE CRYSTALLIZATION BY USING MICROWAVE-VACUUM EVAPORATION

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ABSTRACT

SUCROSE CRYSTALLIZATION BY USING MICROWAVE-VACUUM EVAPORATION

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September 2023, 76 pages

Sucrose crystallization is carried out by conventional evaporation methods, using evaporators, resulting in very long processing times in the food industry. In addition, process efficiency is very important in this process. Therefore, in this study, the sucrose crystallization process was achieved using a tailor-made microwave vacuum evaporation system in order to obtain the final product with shorter processing times and with higher efficiency. The aim of the thesis is to design a semi-batch crystallization process on a laboratory scale starting from beet syrup concentration to crystal mash formation. The effects of the microwave power levels (20%, 30% and 40%) used in the process and the feeding time (5 and 15 minutes) used in the crystallization stage on the efficiency and finished product properties (crystal morphology, crystallinity, particle size, solution color and turbidity and L*a*b* color values) were investigated. Microwave treatment has been shown to affect the sucrose crystal morphology and quality parameters significantly. Process yield was affected by crystallization power and feeding time significantly ($p<0.05$). Longer feeding time (15 minutes) and the use of medium microwave power (30%) resulted in the highest process efficiency (~ 57%). Microwave proccessed sucrose samples were smaller (~460 μm) than commercial (~695 μm) sucrose. Product solution color and b* values of the microwave processed sucrose samples increased with increasing microwave power. Turbidity of the microwave processed sucrose samples increased

with increasing feeding time when 30% and 40% microwave power used. Yet, the trend was opposite in 20% microwave power. As a result of this study, it has been revealed that microwave processing can be a promising alternative to the industrial crystallization process.

Keywords: Sucrose Crystallization, Crystal Properties, Microwave Processing, Microwave Vacuum Evaporation

ÖZ

MİKRODALGA VAKUM EVAPORASYONU TEKNİĞİ İLE SAKAROZ KRİSTALİZASYONU

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Eylül 2023, 76 sayfa

Endüstride sükroz kristalizasyonu, geleneksel buharlaştırma yöntemleri ile, evaporatörler kullanılarak gerçekleştirilmekte ve bu da son ürün eldesinde çok uzun işlem süreleri ile sonuçlanmaktadır. Bunun yanı sıra endüstride işlem verimi çok önemlidir. Bu nedenle bu çalışmada, son ürünü daha kısa sürede elde edebilmek ve potansiyel olarak daha yüksek verim ile çalışabilmek amacıyla sükroz kristalizasyonu işlemi, özel olarak tasarlanmış bir mikrodalga vakumlu buharlaştırma sistemi kullanılarak gerçekleştirilmiştir. Proses, pancar şurubu konsantrasyonundan başlamakta olup, kristal lapa oluşumuna kadar bir yarı – kesikli kristalizasyon prosesini laboratuvar ölçeğinde simüle edecek şekilde tasarlanmıştır. İşlemde kullanılan mikrodalga güç seviyesinin (%20, %30 ve %40) ve kristalizasyon aşamasında kullanılan besleme süresinin (5 ve 15 dakika) işlem verimi ve ürün özellikleri (kristal morfolojisi, kristalinite, parçacık boyutu, çözelti rengi ve bulanıklık ve L*a*b* renk değerleri) üzerindeki etkisi araştırılmıştır. Mikrodalga işleminin sükroz kristal morfolojisini ve kalite parametrelerini önemli ölçüde etkilediği gösterilmiştir. İşlem verimi, hem kristalizasyon gücünden hem de besleme süresinden önemli ölçüde etkilenmiştir ($p<0.05$). Daha uzun besleme süresi (15

dakika) ve orta mikrodalga gücü (%30) kullanımı en yüksek işlem verimiyle (~%57) sonuçlandırılmıştır. Mikrodalgada işlenmiş sükroz numunelerinin (~460 µm), ticari sükrozdan (~695 µm) daha küçük parçacık boyutuna sahip olduğu tespit edilmiştir. Mikrodalga ile işlenmiş sükroz örneklerinin ürün solüsyonu renk ve b* değerleri, artan mikrodalga gücü ile artmıştır. Mikrodalga ile işlenmiş sükroz numunelerinin bulanıklığı, %30 ve %40 mikrodalga gücü kullanıldığında artan besleme süresi ile artmıştır. Ancak, %20 mikrodalga gücünde eğilim tam tersi olarak gözlemlenmiştir. Bu çalışma sonucunda, mikrodalganın endüstriyel kristalizasyon işlemine umut vaat eden bir alternatif olabileceği ortaya konulmuştur.

Anahtar Kelimeler: Sükroz Kristalizasyonu, Kristal Özellikleri, Mikrodalga, Mikrodalga Vakumla Buharlaştırma

for my grandparents Ömer İbiş (1932-2023) and Nedim Baltacı (1941-2018)

ACKNOWLEDGMENTS

This study was funded by the Scientific and Technological Research Council of Turkey under 1005 Program with the grant number of 120R024.

First, I would like to share my gratitude to my supervisors Prof. Dr. Servet Gülüm Şumnu and Assoc. Prof. Dr. Mecit Halil Öztop. I want to thank to Yağmur Balabanlı and Bodoor Aljurf Nabil for their cooperation. Moreover, I would like to thank to OztopLab members especially Ece Göktayoğlu, Berkay Berk and Şirvan Uğuz for their friendship and support.

A special appreciation should be stated to my teaching assistants Cansu Kabakçı, Eda Yıldız, Esmanur İlhan, Elif Gökçen Ateş, Gökcem Tonyalı Karslı, Serap, and Şahin Namlı for their friendly attitude, help, and guidance starting from my undergraduate study. They always tried their best to contribute when I had problems and helped me stay solution oriented throughout my journey at METU FDE.

The most important support came from my dear friend and partner Murat Kiriş, who never lost his faith in me. His support was vital for me to continue this challenging work. I am so grateful for having him by my side. Another special gratitude should go to Ecem Kaya, who is my best friend and my colleague, for always being with me during the sleepless days of hardwork. I want to say that finally, we made it and we will always make it, together! Moreover, I would like to thank to my closest friends Aykut Alagöz, Şule Koçak, Berk Okutan, İdil Kit, and Melike Dönmez for their friendship and endless support.

Last, but not least, I would like to thank to my beautiful family for their support. My mother Naile İbiş deserves a thousand gratitudes. She has always been there by my side, being a perfect role model for me to believe in myself and stand by myself. Nejmi İbiş, who is probably the best dad ever, has always been a friendly father with whom I easily shared my struggles. Thank you! And the best of the blessed, my sister Özgün İbiş is my lucky charm in this world. I would not turn out to be the person I

am right now If it were not for her. Thank you, you are one of the best things that happened to me! My grandparents Mercan İbiş and Rukıye Baltacı also deserve gratitude for being there, especially during my childhood. Thank you all!

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ABBREVIATIONS

AC: Activated carbon

ANOVA: Analysis of variance

GHz: Giga Hertz

HPLC: High performance liquid chromatography

MHz: Mega Hertz

NMR: Nuclear magnetic resonance

RID: Refractive index detector

RPM: Revolution per minute

SE: Solid echo

SEM: Scanning electron microscopy

XRD: X-ray diffraction

LIST OF SYMBOLS

SYMBOLS

A: Overall crystal surface area

a*: red/green chromaticity (color)

b = cuvette height (cm)

b*: yellow/blue chromaticity (color)

C: Sucrose concentration in the syrup

C_0: Sucrose concentration on the crystal surface

°C: The degree Celcius

d: density (kg/m^3)

$\overline{D_S}$: Sauter mean diameter

$\overline{D_{pi}}$= the arithmetic mean of the smallest and largest diameters of the increment

ΔG: Gibbs free energy

i = subscript showing the increments

K_D: Diffusion coefficient

kPa: Kilopascal

Kss: Supersaturation Coefficient

kW: Kilowatt

L*: lightness (black to white color)

μL: microliter

μm: micrometer

nm: nanometer

Rc: Growth rate

S: °Brix value of the solution

$(S/W)_{actual}$: The sucrose water ratio in the sample solution (g/g)

$(S/W)_{theoretical:}$ The sucrose water ratio in a particular impure solution (g/g)

T_1: Relaxation time

TiO_2: Titanium dioxide

W: Watt

W/g: Watt per gram (unit of microwave density)

w/v: weight per volume ratio

w/w: weight per weight ratio

X_i^w = mass fraction of a given increment

LIST OF EQUATIONS

CHAPTER 1

INTRODUCTION

1.1 Sucrose

Sucrose (sugar, $C_{12}H_{22}O_{11}$) is a sweet tasting carbohydrate in disaccharide form, composed of one glucose and one fructose units, which can be found naturally in various plant based sources (Asadi, 2007). Haworth projection of a sucrose molecule is given in Figure 1.1. Marketable sucrose is white in color and has crystalline structure. It is widely used in food formulations for its various functionalities such as improving taste and flavor, adding color and texture in bakery products extending the shelf-life of foods by lowering the water activity (Asadi, 2007).

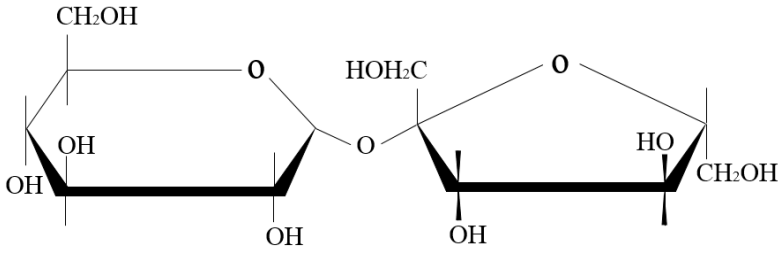


Figure 1.1 Hawort Projection of a Sucrose Molecule

Sucrose is an extremely stable molecule in room temperature in its crystalline form. Its melting point is as high as 185 °C (Asadi, 2007). By having many hydroxyl groups attached to its molecular body, sucrose is able to form multiple hydrogen bonds which enhances the stability of molecules which also makes it immensely soluble in water. Since sucrose is a nonpolar and a nonionic compound, it does not form ionic compounds when dissolved in water.

Crystal sucrose can be provided in a wide range of particle sizes depending on the area it is going to be used. Depending on the mean particle diameter, there are five main crystal groups defined as extra fine (mean particle diameter ≈ 200 μm), fine (mean particle diameter ≈ 300 μm), medium size (mean particle diameter ≈ 500 μm), coarse (mean particle diameter ≈ 1000 μm) and extra coarse (mean particle diameter > 1000 μm) (Asadi, 2007). For instance, the extra fine crystals are used in the bakery products for obtaining well mixing in the dough. Mostly the medium size crystals are considered as standard marketable sucrose.

1.2 Sugar Production

Sugar can either be produced from sugar beet or sugarcane. The sugar obtained from both sources are compatible in all kinds of properties and can be used interchangeably (Asadi, 2007). These raw materials are similar in sugar content but their non-sugar solid content is significantly different (Asadi, 2007). These compositional differences evoke a need to use different methods for processing, which results in the distinction of beet sugar and cane sugar industries.

Both beet and cane can be considered as ancient products discovered early in the history yet sugarcane cultivation is known to be much older (Rajaeifar et al., 2019). It was first cultivated in India, developed and used as a sugar source in Persia (Iran) then finally introduced in Mediterranean region around the 7^{th} century (Rajaeifar et al., 2019). Its cultivation around Europe did not become common due to its tropical nature (Rajaeifar et al., 2019). Beet is on the other hand, much more available for cultivation in Europe. However, the studies on sugar beet on research scale is considerably recent when compared to cane. The first sugar factory was built in 18^{th} century in Germany (Asadi, 2007). Since then, the beet sugar industry is considered to be a high potential area both qualitatively and quantitatively owing to its high quality output and satisfactory sugar yield.

Sugar production from sugar beet is a detailed operation including lots of unit operations that are serving for the proper extraction of sugar from the beet roots (Asadi, 2007). It is mainly aimed to separate sucrose from non-sucrose particles and to produce considerably pure sugar crystals throughout this procedure. The main unit operations used in this process is shown in Figure 1.2.

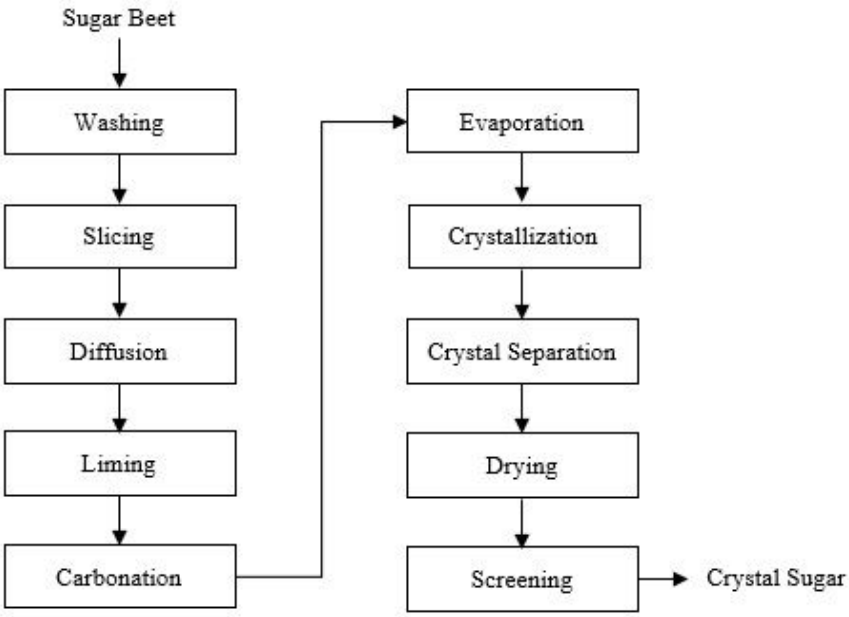


Figure 1.2 Flow Chart of Sugar Production

After the raw material acceptance procedure is completed, sugar beets are immediately washed and sliced to obtain higher extraction efficiencies. As the contact area of diffusion increased, i.e., the surface area of beet slices increases, the diffusion rate increases. Diffusion process can be explained as the movement of sucrose molecules from beet to the water introduced to the diffusers (Asadi, 2007). The driving force in this process is the concentration difference between beet slices and water. Other than the increased contact area, this process is also affected by the physical and chemical conditions of the system such as temperature and pH (Asadi,

2007). Generally, the thermal diffusion is performed at 70–75°C for about 1 hour (Mhemdi et al., 2014).

The exposure to high temperatures causes cell wall disruption, which leads to non sugar components to co-diffuse with sugar. These components are undesirable since they cause color formation and affect crystallization efficiency. Liming and carbonation are used to get rid of these components in sugar beet industry. Lime reacts with the impurities and forms insoluble compounds that can be separated from the juice by filters (Mhemdi et al., 2014). Carbonation serves for the removal of the excess lime in the juice. The product after this multistage purification procedure is the thin juice, which is generally 85-92% pure and has 14-15% dry substance (Asadi, 2007).

The thin juice is concentrated before crystallization to obtain saturation and increase the driving force for crystal formation. In the evaporation operation, juice is generally heated under vacuum (which is called flashing evaporation) and the water is removed at temperatures below the boiling point of water at atmospheric conditions, which is energy efficient and provides higher quality output. The thin juice is concentrated up to 60-70% dry substance before crystallization and the resulting semi-finished product is called thick juice (Asadi, 2007).

The thick juice is passed to the crystallizer, which is mostly an evaporator as well. Similarly, this operation can be performed either under vacuum or at ambient pressure. In addition, cooling crystallization is also an option. Crystallization is the operational step where the sugar with highest possible quality and the yield is obtained by the movement of dissolved sucrose into solid phase. The product of crystallization is called massecuite, which is the mixture of solid crystals with juice.

After crystallization, the crystals in massecuite are covered with juice therefore a separation process is required. Centrifugation is used in sugar industry for that separation. Separation performance is affected by the centrifugal force, crystal-massecuite mixture temperature and massecuite concentration. Mostly,

centrifugation is performed at around 1200 RPM (Asadi, 2007).. There is also a washing step included during centrifugation to provide higher separation performance. Washing provides higher purity, lower ash and color in the final product. In sugar industry, this is mostly a batch operation (Asadi, 2007).

The wet sugar obtained after separation is dried in sugar dryers. The moisture content of crystal sugar should be below 0.03%. Therefore, this process can take 24-72 hours. One of the four types of dryers are used in sugar industry which are rotary-drum, rotary-louver, rotary-tray, or fluidized bed dryer. The main difference in these methods is the movement types of sugar and drying air (Asadi, 2007).

Drying process can be accompanied by crystallization due to the supersaturation obtained on sucrose surface. This supersaturation creates a driving force for the remaining dissolved sugar on the wet surface to move onto the crystal surface (Asadi, 2007).

It is important to keep the drying rate at an optimum level to prevent case hardening which can be defined as the hard crystal surface formation in case of fast drying. When case hardening is observed, it becomes much harder to dry the interior of the crystals. Moreover, it affects the quality by causing the crystals to look dull. On the other hand, slow drying might cause improper and heterogeneous drying (Asadi, 2007). In this case, sugar might agglomerate during its shelf life.

Finally, sugar is screened for size separation. The main objective of this operation is to collect the sugar with the desired particle size. However, the removal of the lumps and very fine particles are also an important aims of screening. Sugar is passed through an inclined vibrating screen for higher separation performance. Screen mesh size might differ depending on the area of use of the final product. Smaller particle size can be required for industrial use while larger size is required for the marketable sugar. This operation might be performed before or after storage depending on the facility.

1.3 Sucrose Crystallization

Crystallization is a separation process which can be defined as the transfer of dissolved particles in a solution into the solid phase (Asadi, 2007). The thick juice is required to reach supersaturation for crystallization. Either cooling or evaporation can provide the supersaturation required for this process (Jones, 2003).

Since crystallization is related with solubility in a great extent, it is important to understand the phase equilibria in a solution to comprehend the mechanism of this process. Phase diagram of a sucrose – water system is given in Figure 1.3.

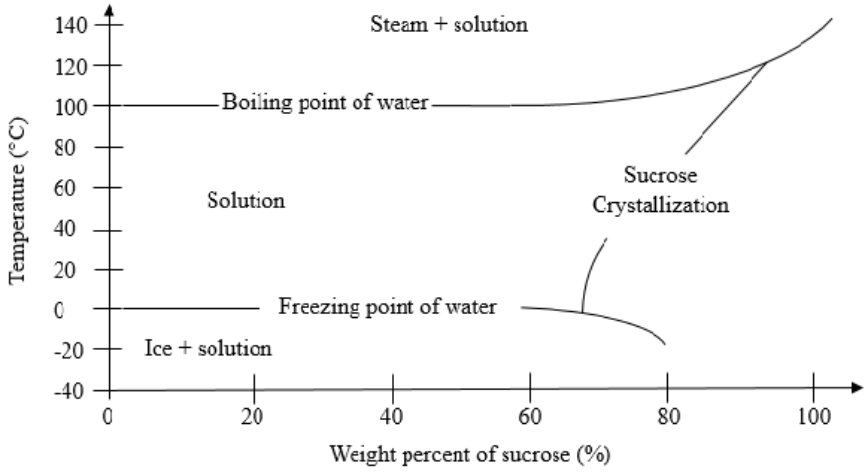


Figure 1.3 Phase Diagram of Sucrose- Water System (Coupland, 2014)

The maximum amount of any substance in a solvent at a given condition is defined as solubility (Jones, 2003). For aqueous solutions, sucrose is a highly soluble compound. Solutions are defined based on the solubility as unsaturated, saturated and supersaturated. When an aqueous sucrose solution is considered, unsaturated solution is the one in which the amount of dissolved sugar is smaller than the maximum amount water can solve. Saturation means the amount of dissolved sugar reached to its maximum limit in water and for supersaturation to occur, there must be excess sugar in the water that it cannot dissolve and crystallize.

Crystallization can be thermodynamically explained by Gibbs free energy (ΔG) which is denoting that there is a chemical potential difference (the driving force of crystallization) between the crystal formed and the solution it lies within triggering the crystal formation (Coupland, 2014). Thus, as crystals are formed and grown, the free energy of the solution system drops, which is called Ostwald ripening (Kabalnov & Kabalnov, 2007). There are three crystallization zones based on this phenomenon; stable, metastable and unstable zones (Asadi, 2007). A solubility diagram showing crystallization zones is given in Figure 1.4. For proper sucrose crystallization, it is important to keep the system at metastable zone during nucleation to prevent false grain formation. In other words, the system goes from a metastable zone to a stable zone as the crystals are formed when the Gibbs free energy is smaller than zero (ΔG < 0).

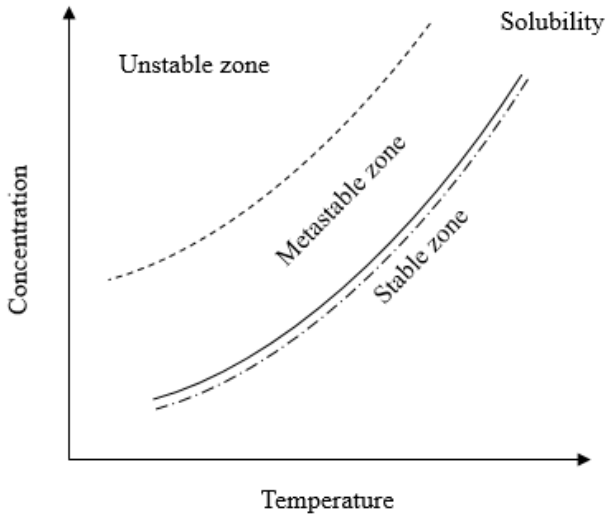


Figure 1.4 Solubility Diagram

Supersaturation coefficient (Kss) is defined to identify the sample state (Asadi, 2007): unsaturated (Kss < 1), saturated (Kss = 1) or supersaturated (Kss > 1). Kss values greater than 1.5 means that the solution enters the unstable zone (Asadi,

2007). It can be used for samples with various purity levels and at various temperatures. Its formula is given in Equation 1.1 (Asadi, 2007);

$$Kss = (S/W)_{actual} / (S/W)_{theoretical}$$ **Equation 1.1**

This coefficient is mainly affected by purity, concentration and the temperature of the sample; namely, it can be the same for many different combinations of these parameters. Therefore, it is important to note that not only Kss but also the concentration and temperature should be considered to understand whether the right conditions to start crystallization are met or not. For a typical concentrated syrup used in the industry, temperatures around 74°C combined with Kss in 1.1–1.2 range are the suitable conditions for the initiation of sucrose crystallization (Asadi, 2007).

Nucleation and crystal growth are the two main steps in crystallization procedure. In sucrose solutions, it is known that the metastable zone is wide leading to a seeding requirement for nucleation to take place (Jones, 2003). Seeding process can be defined as the addition of small particles in solution to stimulate the crystallization process (Asadi, 2007). This process can be performed by using various sources of seed materials such as powdered sugar, sugar slurry, massecuite and seed magma. Powdered sugar is not commonly used since it is challenging to control the seeding process and provide homogeneous seed distribution within the slurry while using powdered sugar. Sugar slurry provides better control over particle size in crystallization process. This slurry mostly consists of powdered sugar and isopropyl alcohol with 0.4–0.5%(w/v) concentration. Massecuite and seed magma (a mixture of crystal sugar with syrup) can be used as seeding medium when the crystallization is a continuous process.

Seed addition at a low supersaturation level (Kss < 1.1) leads to less crystal formation and growth since the metastable zone has not been reached yet in that case. Similarly, seeding at high supersaturation levels (Kss > 1.3) causes various problems such as higher viscosity, conglomeration and undesirable crystal formation. Moreover, seeding the solution with an optimized amount and particle size is crucial for obtaining a uniform size distribution among the final crystals. Particles added in the

syrup creates a basis for the dissolved sucrose to attach and to form crystal structures. As particle size gets smaller and the amount of those small particles increases in the syrup, the crystallization rate directly increases due to the increased surface area (Asadi, 2007). Seed size of around 5-10 microns provides the optimum product formation after crystallization. Having a uniform particle size distribution in the seed used is also important to reach the equilibrium state during crystallization (Asadi, 2007). When the particle size distribution is non-uniform, the smaller particles tend to dissolve and only larger particles can grow.

There are two main nucleation mechanisms observed in crystallization: homogeneous and heterogeneous nucleation. In the absence of fine crystals (seed), the dissolved molecules in a pure solution may randomly come together and form a conformation resembling a small crystal (Coupland, 2014). If these small crystals can reach a threshold particle radius and remain in the solution without being dissolved, ΔG starts to decrease which will result in homogeneous nucleation (Coupland, 2014) Heterogeneous nucleation on the other hand requires a surface for crystallization to start. The presence of a surface drastically lowers the free energy of the system, generating a more suitable environment for crystallization. The crystallization pan surface provides that surface as well as the added crystal particles.

Crystal growth is a complex mechanism that occurs as the dissolved sucrose molecules move from the solution to the crystal surface. The free energy difference is the main driving force in crystal growth as well as in the nucleation (Coupland, 2014). There are two mechanisms providing crystal growth: diffusion and surface reaction (Asadi, 2007). First, the dissolved sucrose molecules in the system diffuses to the crystal surface. After that process, sucrose molecules align themselves in the crystal structure with the mechanism called surface reaction (Asadi, 2007). In sucrose crystallization, these processes are enhanced by the heat introduced to the system to provide the required energy for the movements of sucrose molecules.

The diffusion process is precisely related to the concentration difference between the diffusion layer and the crystal surface, whereas the surface reaction occurs due to the

concentration gradient between the absorption layer and crystal surface (Asadi, 2007). The sucrose concentration at the absorption layer is directly equal to the corresponding saturated concentration for the syrup. During growth, the sucrose molecules diffuse to the crystal surface and attach themselves on that surface, continue their movement within the crystal structure at the same rate they diffuse. The mechanisms can be expressed in the crystallization rate equation given in Equation 1.2 (Asadi, 2007):

$$Rc = K_D A(C-C_0) \qquad \textbf{Equation 1.2}$$

Diffusion coefficient is directly proportional to the solution temperature and viscosity. The increase in temperature enhances the diffusion, however the increase in viscosity is an opposing factor. In other words, temperature rise favors the crystallization rate whereas increase in viscosity hinders it. Other than these factors, seed size, purity, supersaturation and agitation also affect the crystallization rate. For purity, as supersaturation and agitation rate increase, the crystallization rate also increases. However, as it was mention in nucleation part, the smaller seed size is required for higher crystallization rates.

1.3.1 Crystallization by Cooling

In cooling crystallization, supersaturation is achieved due to the decreased solubility at lower temperatures. The method is most commonly used in the pharmaceutical industry (Choong & Smith, 2004) and can be performed as either a batch or continuous process. In sugar industry, it is most commonly used in the recovery of crystals from the juice after crystal separation. The main reason why cooling crystallization is being used in crystal recovery is the higher crystal concentration it provides. However, this method is less energy efficient than evaporation crystallization due to its long retention time (Asadi, 2007).

Continuous cooling crystallizers can be either horizontal or vertical. In the horizontal systems, there are numerous stages connected through the channels. Massecuite

enters the system from the first stage, gradually cools down while going to the last one (Asadi, 2007). Viscous input cannot be handled in every horizontal crystallizer due to the limitations of its mechanism (Asadi, 2007). Vertical systems on the other hand, typically have two stages and the cooling cycle is different from horizontal crystallizers (Asadi, 2007). They are faster and their operation temperature is lower than the horizontal crystallizers (Asadi, 2007). Moreover, they take up less space so it can be said that vertical crystallizers are more advantegous.

There are numerous factors affecting this process such as cooling rate, seeding, particle size, temperature (Gros et al., 2001; Myronchuk et al., 2013). It is critical to modify the cooling rate throughout the crystallization process to maintain the desired supersaturation level in the system. At the beginning of the crystallization, slow cooling is required while at the later stages it should be faster (Bohlin & Rasmuson, 1992). The reduced concentration of solution by crystallization can be compensated by this cooling rate control (Bohlin & Rasmuson, 1992). In sucrose crystallization, since cooling is not the main process, seeding is mostly skipped in industrial applications and cooling should be gradual and fast (Asadi, 2007). For other applications of cooling crystallization, seeding provides enhanced crystallization and less time dependence (Choong & Smith, 2004).

Crystal size obtained in cooling crystallization processes is highly dependent on supersaturation level and the process temperature (Asadi, 2007; Choong & Smith, 2004). Keeping the supersaturation low results in larger particle size and higher crystallization yield (Asadi, 2007; Choong & Smith, 2004). For sucrose cooling crystallization, the most efficient process temperatures are stated as 45-50 °C (Asadi, 2007). At lower temperatures, high viscosity hinders the crystallization process (Asadi, 2007). Moreover, when a minor heating is performed as a final step of cooling crystallization, better crystallization might be observed (Asadi, 2007).

1.3.2 Crystallization by Evaporation

Evaporative crystallization can be performed either under vacuum or at ambient pressure (Asadi, 2007). However, ambient pressure results in higher process temperatures leading to quality defects in both semi-finished and final products. Therefore, evaporative crystallization is performed mainly under vacuum which is called flashing evaporation (Asadi, 2007). The operation can be batch or continuous.

Batch crystallizers are most commonly vacuum pans with stirrers. Steam or hot water can be used as heating medium. A tubular radiator is used for the heating medium to go through (Asadi, 2007). The tubes of the radiators are positioned vertically and the height may change depending on the capacity of the pan (Asadi, 2007).

Continuous crystallizers have similar design with batch crystallizers but as the name implies, there is a continuous inlet and outlet from the system. Continuous crystallization is more efficient in terms of energy and process yield and easier to control (Asadi, 2007). Yet, there are some disadvantages as well. The major disadvantages can be listed as having more challenges for cleaning, causing more non-uniform crystal distribution and requiring high purity (higher than 97%) to operate properly (Asadi, 2007).

It is crucial to mix the boiling medium properly to provide homogeneous temperature and concentration distribution (Asadi, 2007). Impeller-type, top driven stirrers are commonly used in the industry (Asadi, 2007). Moreover, there are studies in which the most accurate impeller speed for the best crystal size was determined (Zhang et al., 2020). It is required to design the impeller in a way that it can stir all of the volume in the pan in a proper period of time (Asadi, 2007). Both fast and slow stirring have negative outcomes; dissolving the newly formed small crystals and generating heterogeneous temperature and concentration distribution within the material, respectively.

Other components in the crystallizer design also significantly affects the product quality. For instance, the pan diameter is known to be one of the major parameters

that affects the circulation velocity as well as the agitation level (Zhang et al., 2020). Sugar level, radiator tube diameter and length are also important design parameters that should be considered to obtain the most homogeneous process possible (Zhang et al., 2020).

1.4 Microwave Processing

Microwaves lie in the electromagnetic spectrum at frequencies from 300 MHz to 300 GHz. The specific frequencies that are allowed to be used in microwave processing for domestic and industrial use are 2450 MHz and 915 MHz, respectively (Raghavan, 2005). Microwave heating occurs due to the molecular friction of dipoles and/or the ion acceleration in the subject material trying to reorient themselves with the electrical field of waves (Regier et al., 2004). When materials are exposed to microwave, the energy is transmitted, reflected and absorbed by the material (Regier et al., 2004). The absorbed energy is then dissipated into heat within the material (Regier et al., 2004) so the heating occurs volumetrically.

The ability of the food material to dissipate microwave energy into heat is defined by the material specific attributes called dielectric properties (Guo et al., 2017), namely dielectric constant and dielectric loss factor (Regier et al., 2004). The dielectric constant is a numerical expression of the ability of a material to store the energy when exposed to an electromagnetic field whereas the dielectric loss factor shows the ability of a material to convert that stored electrical energy into heat. These attributes of a food material are affected by both the process parameters (such as frequency and temperature) and composition of the material (Sumnu, 2001).

Microwave ovens consist of three main parts that are called magnetrons and waveguides. Microwaves are produced in the magnetron and delivered to the microwave cavity by waveguide (Regier et al., 2004).

Microwave processing technology provides many advantages such as short process time, less start up and being user-friendly. Moreover, microwave heating affects the

flavor and nutrition value less than conventional heating does (Chandrasekaran et al., 2013). Owing to these benefits, microwave has been significantly studied in drying, cooking, pasteurization and baking processes (Guo et al., 2017). However, the non-uniform heating observed in microwave processing might result in under or over processed products (Ekezie et al., 2017). The reasons why non-uniform heating may occur are the shape of the food material, oven design and dielectric properties of food (Ekezie et al., 2017). Designing process specific packaging materials and moving the subject material by turning table or stirring might help overcoming non-uniform heating (Chandrasekaran et al., 2013).

1.4.1 Microwave Vacuum Evaporation & Crystallization

Vacuum evaporation and crystallization are used mainly to obtain a concentrated material. In the absence of vacuum, the process temperature should be so high that superheating (the phenomenon in which the material is heated to temperatures higher than its boiling point) might occur. As mentioned before, high process temperature affects the product quality in sugar production as well as in many other food materials' productions. Although vacuum evaporation improves the process by providing lower process temperatures, the process time needs to be improved since long process duration can lead to the loss of valuable nutritive compounds. Using microwave vacuum systems have the advantage of both lower process temperature and shorter process time.

Microwave vacuum evaporation has been used in the literature for the concentration of many fruit juices owing to its potential to eliminate the disadvantages of traditional concentration methods. For instance, apple (Bozkir & Baysal, 2017) and pineapple juice concentration (Assawarachan & Noomhorm, 2011), syrup production from sugar solutions (glucose, maltose monohydrate and trehalose anhydrate solutions) prepared by (Tao et al., 2021) was studied and the effect of process time and microwave application was investigated. It was found that microwave vacuum evaporation resulted in higher final product quality with shorter

process time when compared to rising film and rotary evaporation methods in apple juice concentration (Bozkir & Baysal, 2017). Similarly, in pineapple juice concentration, it was found that microwave vacuum evaporation was more efficient as compared to the conventional evaporation (Assawarachan & Noomhorm, 2011). At 50kPa vacuum pressure, sugar solutions reached boiling point in a minute with 40 W/g microwave power density showing that the sugar solutions can be concentrated much faster than it is in conventional methods (Tao et al., 2021).

In the literature, there was only one study about sucrose crystallization by microwave processing (Lin et al., 1998). In this study, sucrose solutions with a determined concentration were crystallized with a microwave vacuum setup and the results showed that microwave interactions could increase the crystal growth rate from approximately 20 to 40 μm/min. However, the experimental setup used in this study is not comparable with the conventional sucrose crystallization. The system was designed in a way that only a single sucrose crystal formation was observed and evaluated. In other words, the crystallization was not fully performed and terminated at early stages.

In chemical engineering, microwave crystallization of various inorganic materials was studied. For instance, TiO_2 crystallization with and without vacuum was performed with microwave processing (Danty et al., 2020; Tian et al., 2015). In microwave assisted crystallization under vacuum, it was found that microwave processing was more efficient and provided enhanced crystal characteristics (Tian et al., 2015). It was also found that as microwave power increased from 100 W to 700 W, the efficiencies increased from 42% to 98%. Similarly, at atmospheric pressure, the study showed that TiO_2 crystallization could be a potential alternative due to the fast processing which led to less exposure to high temperature (Danty et al., 2020). Moreover, zeolite crystallization by microwaves was performed in various studies and it was found that microwave processing was faster and eliminated the impurity formation (Bonaccorsi & Proverbio, 2003).

1.4.2 Previous Microwave Vacuum Evaporation Designs

Microwave vacuum systems were designed for evaporation, crystallization, and drying purposes in the literature. The designs mainly consist of microwave oven, a vacuum system, and a component to provide agitation. There are slight technical differences observed such as vacuum capacity and microwave power. In addition, some of the designs have automation connections for better process control.

In the only study performed on sucrose crystallization with microwave, the crystallizer design was performed as shown in Figure 1.5 (Lin et al., 1998). A frequency of 2450 MHz and a microwave power of 750 W were combined with a vacuum condenser. It is stated that the aim of this design was to provide constant temperature and super saturation (Lin et al., 1998).

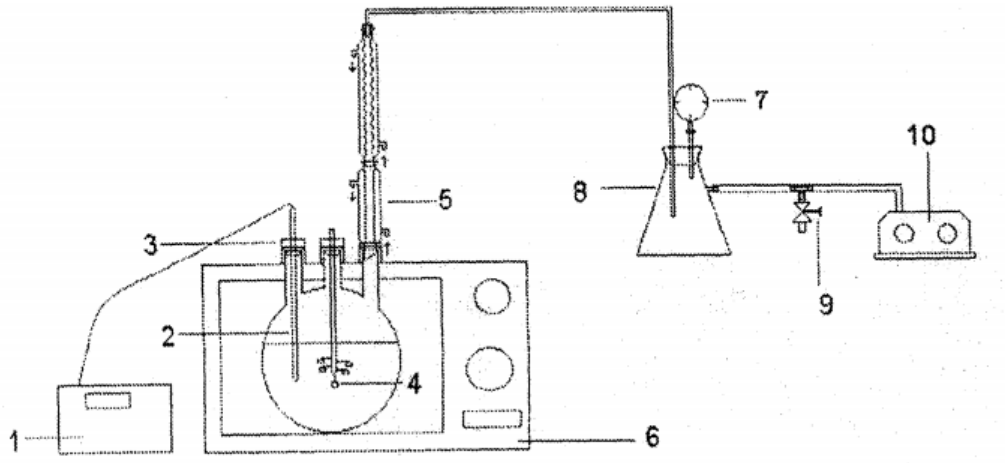


Figure 1.5 Schematic of Microwave Vacuum Crystallizer (Lin et al., 1998): (1) Digital temperature display, (2) K – type thermocouple probe, (3) Metal shield, (4) Single sucrose crystal, (5) Condenser, (6) Microwave oven, (7) Vacuum gauge, (8) Buffer flask, (9) Valve, (10) Vacuum Pump

Another similar system was designed for the evaporation of various sugar solutions (Tao et al., 2021). In that microwave vacuum evaporator, also a domestic microwave was used with maximum 2.3 kW capacity. The vacuum was connected to a rotating quartz sample tube to reduce the possible heterogeneous heating. An infrared temperature probe was connected to the system to track temperature rise throughout the process. 50 kPa vacuum at 40 W/g microwave power density was determined as fixed independent variables in the study. The original schematic of this design is given in Figure 1.6.

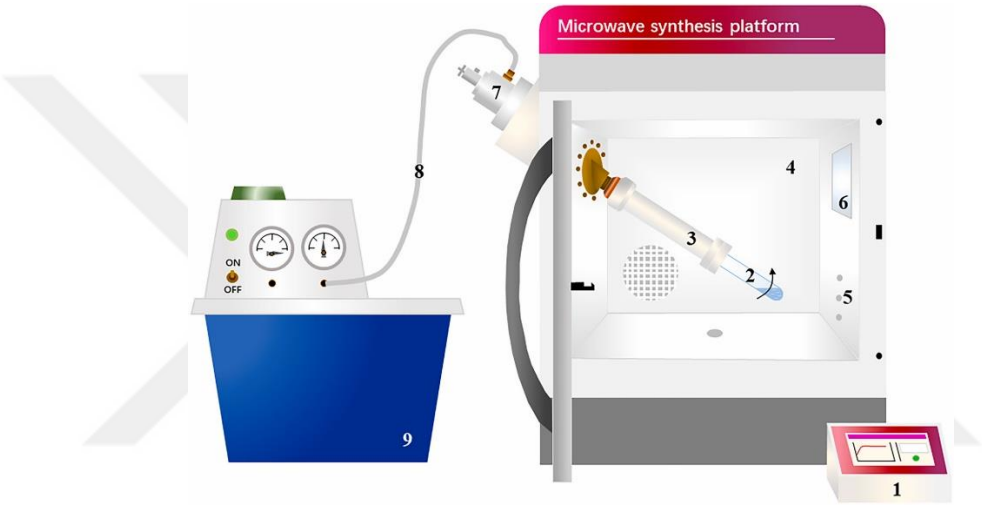


Figure 1.6 Schematic of The Microwave Vacuum Evaporator for Sugar Solutions (Tao et al., 2021): (1) Microwave system terminal, (2) Rotating quartz tube, (3) Teflon tube, (4) Microwave cavity, (5) Infrared sensor, (6) Monitoring window, (7) Rotating motor and output connections

In the two microwave assisted juice concentrator designs (Assawarachan & Noomhorm, 2011; Bozkir & Baysal, 2017), standard microwave ovens were modified with vacuum condenser attachments and automation connections so that the process parameters can be controlled from computers. Assawarachan's design is different from Bozkir's by means of the vacuum system they used which is a rotary vacuum system. The process parameters used are also different in the two studies.

For apple juice concentration 668 W at 500 mbar used whereas for pineapple juice concentration various microwave power densities (0.451, 0.925 and 1.536 W/g) and vacuum pressures (200, 300, 400 and 500 mbar) were used. The schematic of the two systems is given in Figure 1.7 and 1.8.

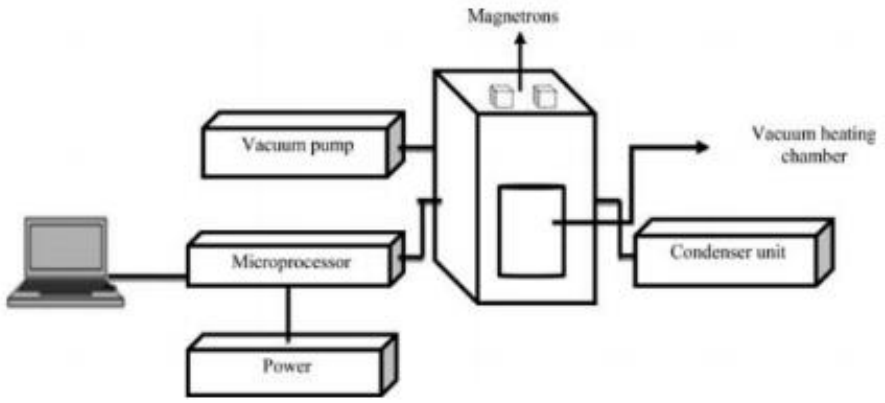


Figure 1.7 Schematic of The Microwave Vacuum Evaporator for Apple Juice Concentration (Bozkir & Baysal, 2017)

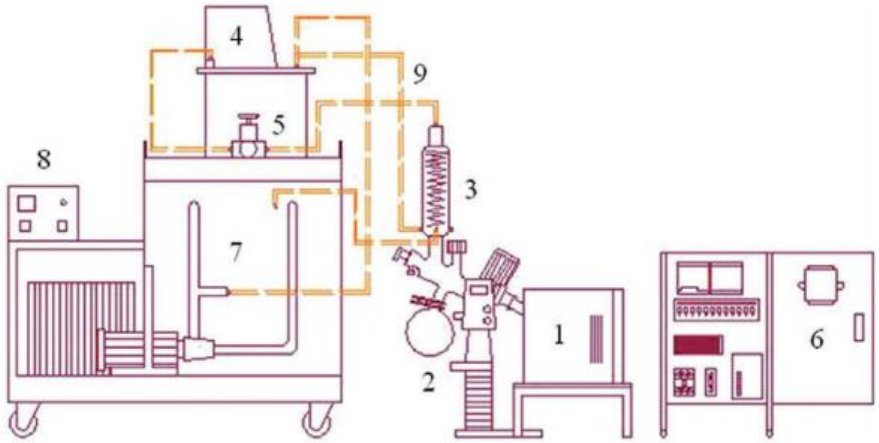


Figure 1.8 Schematic of The Microwave Vacuum Evaporator for Pineapple Juice Concentration (Assawarachan & Noomhorm, 2011): (1) Microwave oven, (2) Rotary vacuum, (3) Condenser, (4) Vacuum pump, (5) Vacuum regulator, (6)

Programmable logic controller, (7) Cooling tank, (8) Temperature controller, (9) Controlling line

Two microwave vacuum drying systems are given in Figure 1.9 and 1.10 which were designed for similar purposes. The design in Figure 1.9 was used for banana drying and a domestic microwave with 650 W capacity was selected for the process (Mousa & Farid, 2002). In this design, no stirrer or turning table were used and the process power was determined as 168 W. Moreover, air conditioning was used to improve the drying performance of the system. The design in Figure 1.10 was used for artichoke drying (Muştu & Eren, 2019)and unlike the previous system, it did not contain an air conditioning unit. A domestic microwave with maximum 1000 W capacity was used. Different microwave powers (450 and 800 W) and vacuum pressures (31.2 kPa and 12.4 kPa) were determined as independent variables of the study.

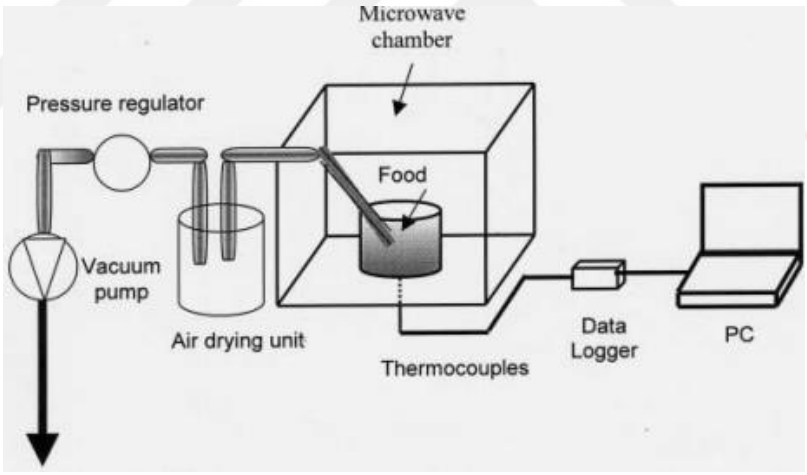


Figure 1.9 Schematic of Microwave Vacuum System for Banana Drying (Mousa & Farid, 2002)

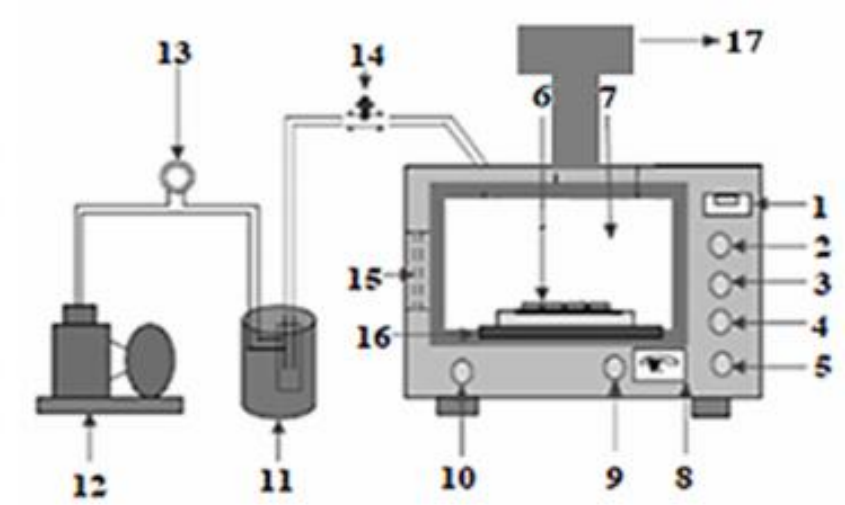


Figure 1.10 Schematic of Microwave Vacuum System for Artichoke (Muştu & Eren, 2019): (1) Display panel, (2) End-off button, (3) Short setting, (4) Manual power adjustment, (5) Electric power switch, (6) Product, (7) Vacuum cabinet, (8) Analog vacuum meter, (9) Timer, (10) Sensor indicator, (11) Condenser, (12) Vacuum pump, (13) Digital pressure meter, (14) Pressure control valve, (15) Heating resistance, (16) Turntable, (17) Magnetron

1.5 Objectives of The Study

This study mainly aims to develop a novel processing method for sucrose crystallization by using microwave vacuum evaporation method. Since sucrose crystallization is a time consuming procedure, it can be significantly beneficial for the industry to develop a faster methodology for this operation. Thereby, it is hypothesized that microwave vacuum evaporation can provide a faster operation while increasing the process yield since these are the main benefits microwave processing. In the literature, there is only one study concentrating on the microwave crystallization of sucrose (Lin et al., 1998). However, in that study the focus is being able to observe the crystallization instead of performing a completed crystallization process compatible with the industrial crystallization. Consequently, it can be said

that there is a gap in the literature to investigate the novel sucrose crystallization methods.

As a result, this study is focused on procuding crystal sucrose efficiently with shorter process time by using microwave vacuum processing. Developing a novel, industrially applicable sucrose crystallization method as well as detecting the crystal quality, crystallinity, crystal morphology and process yield can be considered as the sub-goals of the study.

CHAPTER 2

MATERIALS AND METHODS

2.1 Materials

The 70 °Brix beet syrup, the seeding solution (isopropyl alcohol – powdered sugar mixture), control samples and antifoaming agent (polyether polyol) were supplied from Kayseri Şeker Inc., Kayseri, Turkey. The microwave vacuum evaporator was manufactured by IFTECH Food R&D Consulting, Ankara, Turkey. The apparent magnetron power was determined as 335 Watts by IMPI 2L test. (Buffler, 1993). The device includes a vacuum pump connected to the sample chamber, in addition to a standard home type microwave oven. The pump power is 0.25 kW and the pressure levels were in the range of 80 – 360 mmHg. The sample chamber was made of polytetrafluoroethylene and its lid was produced from polyethylene since these materials do not reflect or absorb microwaves. There was a reverse T shaped polytetrafluoroethylene – glass rod connected to the lid to let sample to be stirred inside the chamber as the turntable operated (~10RPM). The schematic of the microwave vacuum system is given in Fig. 2. A specially designed, frusto – conical container (Ildam Glass Laboratory Materials Trade and Industry Inc., Ankara, Turkey) was used for the solutions to prevent the so-called ‘focusing effect’ observed in liquid samples in the microwave oven.

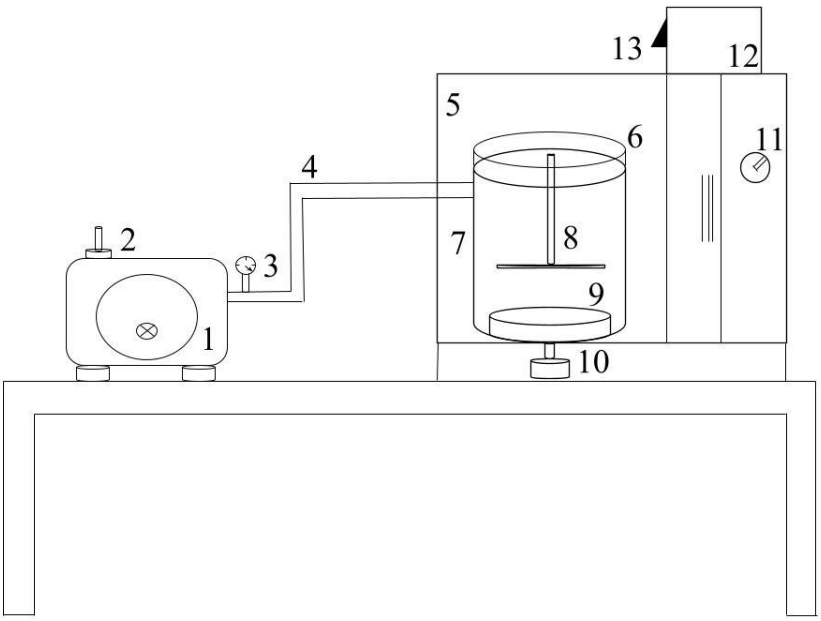


Figure 2.1 Schematic of The Microwave Vacuum Oven System (1) Vacuum Pump, (2) Pump On-Off Switch, (3) Vacuum Regulator, (4) Polytetrafluoroethylene Tube, (5) Microwave Oven, (6) Chamber Lid, (7) Sample Chamber, (8) Stirring Rod, (9) Turning Table, (10) Turning Motor, (11) Microwave Oven On-Off Switch, (12) Control Panel, (13) Motor On-Off Switch

2.2 Methods

2.2.1 Sample Preparation

Vacuum evaporation was performed by using the microwave vacuum system given in Figure 2.1 for sucrose crystallization. Before initiating the crystallization, the beet syrup was boiled to obtain the required super saturation level. When the boiling was completed, the crystallization started by seeding. During crystallization, there was a feeding procedure which aimed to keep the sample at a constant super saturation level to obtain controlled crystal nucleation and growth. After that, there was a

continuous evaporation procedure called concentration. The flow chart of sucrose crystallization procedure is given in Figure 2.2. When massecuite was obtained, sucrose crystals were separated from the mother liquor by centrifugation and immediately dried.

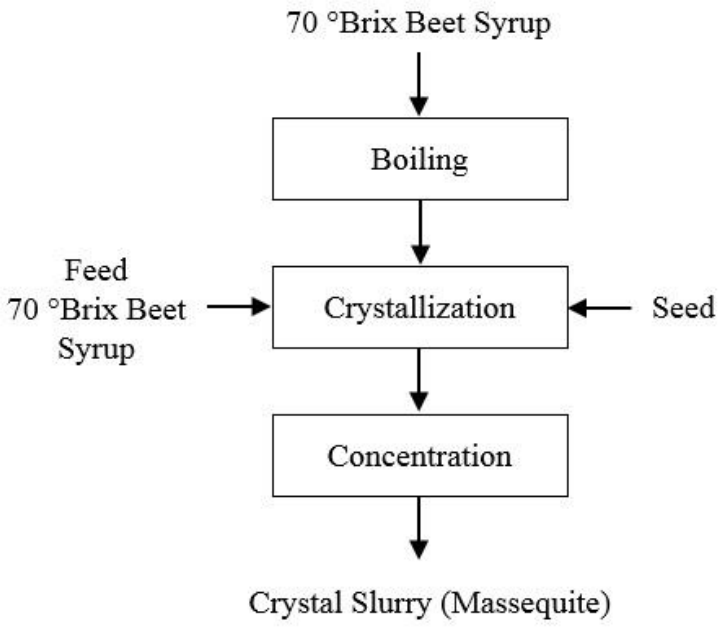


Figure 2.2 Flow Chart of Sucrose Crystallization Procedure

2.2.1.1 Boiling Process

Boiling was performed at 50% microwave power for 15 minutes with 190 g sample. The process conditions were determined by preliminary experiments, considering the prevention of spill out of solution during boiling and in further processing. Boiling was continued until the syrup reached to 1.1 – 1.2 super saturation levels at 73 ± 2°C (Asadi, 2007; Verma et al., 2021). The pressure was adjusted to 210 ± 10 mmHg to obtain constant temperature throughout the process.

2.2.1.2 Crystallization Process

Crystallization process was performed at three different microwave power levels: 20, 30 and 40%. Process conditions were determined as explained in the boiling process. When the beet syrup reached to the required super saturation level at the end of boiling, 5% (w/w) seeding solution was introduced to the system to initiate the crystallization. The amount of seeding solution used was determined by trial and error.

During this process, feed (70 °Brix beet syrup) was introduced to the system to keep the sample at the same super saturation level for a while to obtain controlled nucleation. The evaporation rate at each microwave power level was modelled and the models shown in Figures 2.3, 2.4 and 2.5 were obtained and used to determine the amount of feed required in each microwave power level. Feeding was performed for 5 and 15 minutes, which was adapted from the literature (Asadi, 2007).

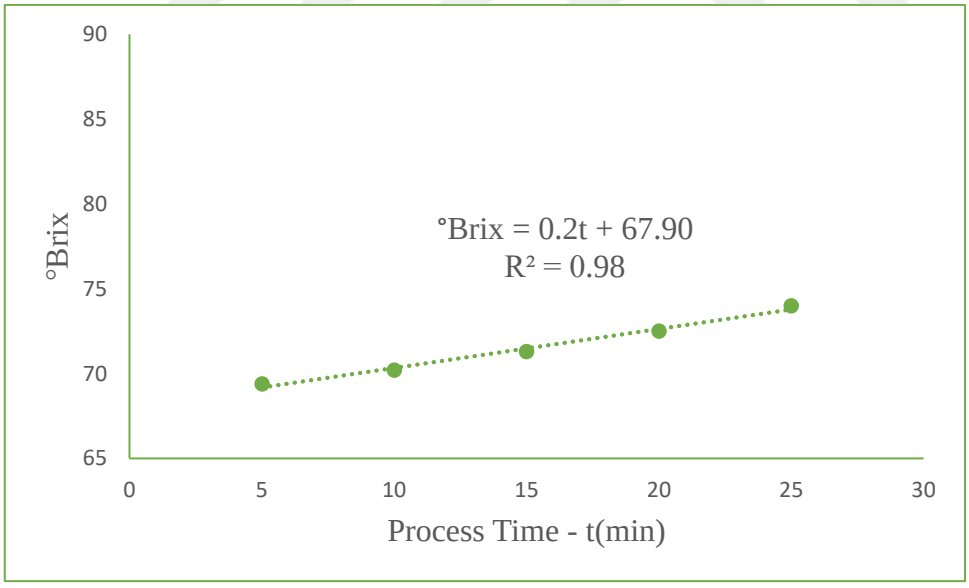


Figure 2.3 Evaporation Rate of Beet Syrup with 20% Microwave Power

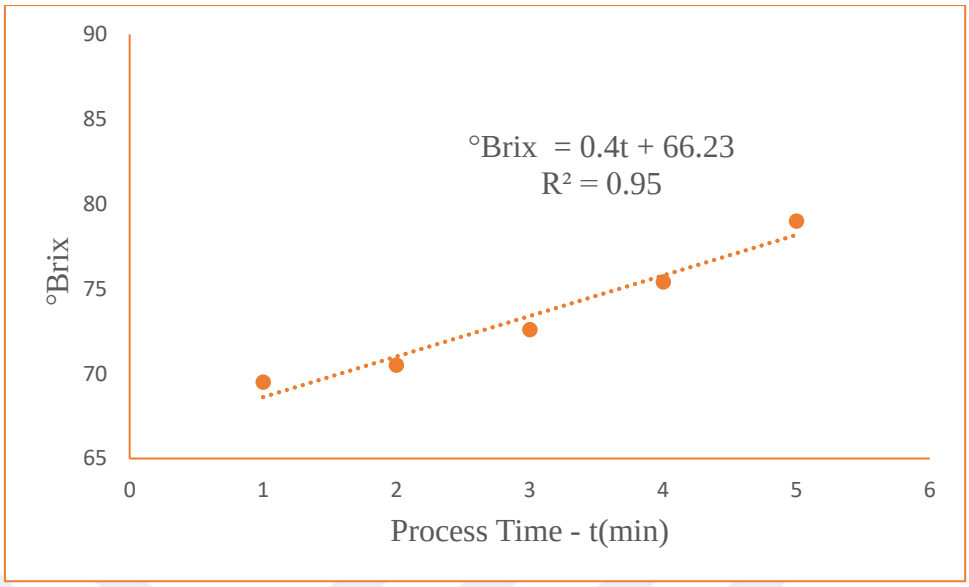


Figure 2.4 Evaporation Rate of Beet Syrup with 30% Microwave Power

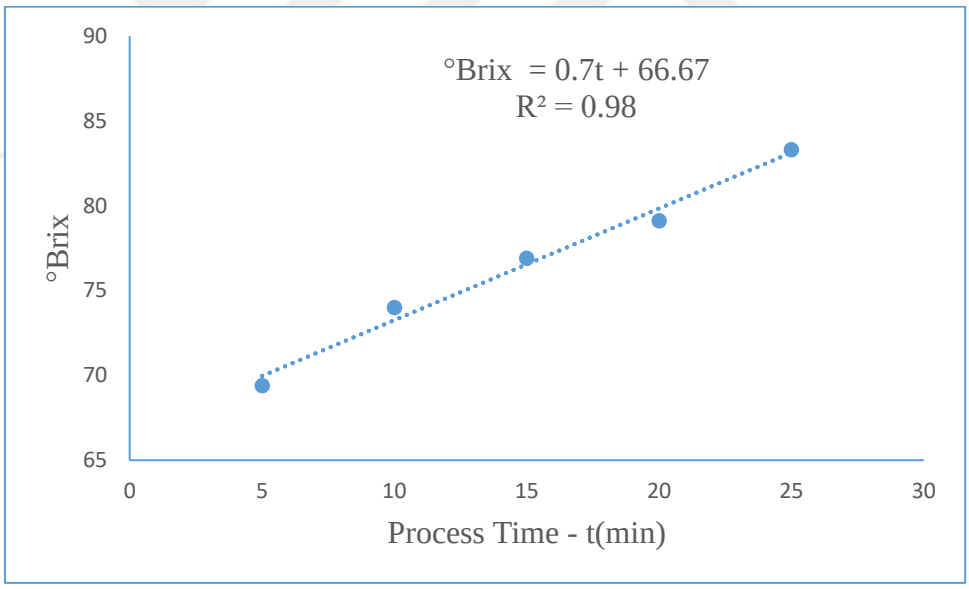


Figure 2.5 Evaporation Rate of Beet Syrup with 40% Microwave Power

2.2.1.3 Concentration Process

This process is the continuum of crystallization and the only difference is stopping the feed. In this part, feed was stopped for further crystal growth and massecuite concentration up to ≈90 °Brix. The same microwave power levels in crystallization process were used.

2.2.1.4 Crystal Separation and Drying

When massecuite was obtained, it was immediately centrifuged (Nuve-Bench Top Centrifuge, NF 1200R, Turkey) at 1500 RPM for 5 minutes. The centrifugation was repeated to mimic industrial sugar washing procedure where massecuite was washed simultaneously with centrifugation. The washing was mimicked by attaching wetted sponges on the tube lids that allowed water release during the centrifugation. After separation, sugar particles were dried at 35 °C in an incubator (Digital incubator WIG-32, Daihan Scientific Co. Ltd., Korea) overnight.

2.2.2 X-Ray Diffraction (XRD) Analysis

XRD diffractometer (Rigaku Ultima IV X-Ray Diffractometer, Japan) was used to determine the crystallinity of both microwave processed and commercial (control) sucrose samples. Data analysis was performed by using Origin 9 (OriginPro 9.0, Origin Lab Corporation, Northampton, USA). Crystallinity values were quantified by assuming that among the total area under the XRD curve, the area under sharp peaks represented the crystalline area, and the percent crystallinity of the samples was calculated by proportioning the area under the sharp peaks directly to the total area under the XRD graph (Grunin et al., 2019).

2.2.3 Time Domain Nuclear Magnetic Resonance (NMR) Analysis

TD-NMR measurements were performed to compare the crystallinity results obtained with XRD. It has been shown in several studies that T_1 spin-lattice (Le Botlan et al., 1999) and solid echo measurements can be used to predict crystallinity of sugar samples (Grunin et al., 2019). T_1 measurements were performed on a 20.34 MHz system (Spin Track, Resonance Systems GmbH, Kirchheim/Teck, Germany) using a saturation recovery pulse sequence with 400-ms repetition time and 16 scans. The data were analyzed by using Relax8 (Resonance Systems GmBH, Kirchheim/Teck, Germany) software (Grunin et al., 2019; Pocan et al., 2019). Solid echo (SE) analyses were also carried out by using the same system with repetition time set to 10 s for 64 scans. Crystallinity values were determined by using a special module installed on Relax8 software (Grunin et al., 2019).

2.2.4 Scanning Electron Microscopy (SEM)

Scanning electron microscopy (FEI Nova NanoSEM 430, USA) was used to determine the crystal morphology. A magnification level of 300x was used for all the samples including the commercial sucrose (control) sample to detect the particles properly.

2.2.5 Solution Color and Turbidity Analysis

Solutions with 50% (w/w) sucrose content were prepared with distilled water. The °Brix value of the solutions were measured with HI 96801 Refractometer (Hanna Instruments, USA) for the turbidity analysis. An ultrasonic bath (Elmasonic P30H ultrasonic cleaning unit, Elma Schmidbauer GmbH, Germany) was used for 3 minutes at 37 Hz for the disruption of the air bubbles in the solutions. After ultra-sonication, absorbance value at 420 nm was determined by using UV-visible

spectrophotometer (Optizen UV/Vis spectrophotometer, Mecasys Co. Ltd. Seoul, South Korea). The turbidity value was calculated with the formula below:

$$Turbidity\ (or\ Solution\ Color) = 10^8 \times \frac{Absorbance}{S \times d \times b} \qquad \textbf{Equation 2.1}$$

For the solution color measurement, solutions with the same method were prepared and before °Brix measurements, the solutions were filtered from a membrane with 0.45 μm porosity. After filtration, the same procedure explained for turbidity was followed and the same formula was used. A similar approach was also used in Schlumbach's study (Schlumbach et al., 2017).

2.2.6 L*a*b* Color Measurements

The color values of the microwave processed samples as well as the commercial sucrose samples (control) were measured using the CIE L*a*b* color scale. The values were directly measured and recorded using the Konica Minolta Chromameter (Konica Minolta Ltd., Tokyo, Japan) device.

2.2.7 Sieve Analysis

Sieve analysis was performed to determine the particle size distribution of the samples. Sieves with 1000, 710, 500, 400, 250 and 180 μm mesh sizes were stacked in the given order on the automatic shaker (Fritsch Analysette 3 Spartan, Fritsch Milling & Sizing Inc., USA) and the analysis was performed by 5 minutes of shaking with 20 g of sample. The remaining samples on each sieve was weighted and the Sauter mean diameter ($\overline{D_S}$) was calculated with the given formula (Sahin & Sumnu, 2006):

$$\overline{D_S} = \frac{1}{\sum_{i=1}^{n} \frac{X_i^W}{\overline{D_{pi}}}} \qquad \textbf{Equation 2.2}$$

2.2.8 Process Yield Determination

Process yield was estimated by calculating the percent sugar obtained from the massecuite. To detect the sucrose content of the massecuite, HPLC analysis was performed by using the HPLC-RID (Shimadzu Scientific Instruments, Japan) device that is equipped with an auto-sampler (SIL-20A HT), degasser (DGU-20A_5), pump (LC-20AD), oven (CTO-20A) and refractive index detector (RID-20A). The method explained in Namli's study was used (Namli, 2019). The massecuite sample was diluted in 10% (w/v) by using double distilled water (Milli – Q Water System, Millipore S.A., France). For total dissolution, the solution was stirred at 500 RPM for 5 minutes (MaxTir 500, Daihan Scientific, Seoul, Korea). The solution was filtered by using a 0.45 μm polypropylene filter before being poured into the HPLC vial. Acetonitrile – water mixture (80:20 v/v) was the mobile phase used in the analysis. Injection volume was 20 μL with 1mL/min flow rate at 40°C oven temperature. Percent yield was calculated by using:

$$\%\, Yield = \frac{Amount\ of\ sugar\ collected\ (g)}{Amount\ of\ sugar\ in\ the\ massequite\ (g)} \times 100 \qquad \textbf{Equation 2.3}$$

2.2.9 Statistical Analysis

Statistical analysis (analysis of variance, ANOVA) was conducted for all experimental data by using MINITAB (ver.16.2.0.0, Minitab Inc., State College, USA). Tukey's test was used as 95% confidence interval to determine the statistical difference between results.

2.3 Experimental Design

The procedure was designed with boiling process power level set as a constant variable and two independent variables: crystallization process power levels and feeding duration. The effect of independent variables on the crystallization product was investigated. The experimental design was summarized in Table 2.2.

Table 2.1 Experimental Design

Factors	**Levels**	**Responses**
Boiling Process	50%	X-Ray Diffraction Analysis Scanning Electron Microscopy
Crystallization Process	20%, 30%, 40% Power Levels	NMR Relaxometry Analysis Solution Color & Turbidity
Feeding Time	5 minutes, 15 minutes	Mesh Analysis Process Yield

CHAPTER 3

RESULT AND DISCUSSION

3.1 Preliminary Studies

The process parameters given in the experimental design were determined by the preliminary studies performed. It was aimed to obtain sucrose crystallization within a shorter time. Therefore, the process time was shortened as much as possible by using the highest possible microwave power levels during crystallization. Boiling operation duration is directly linked to the microwave power used since it is required to concentrate the syrup up to supersaturation level 1.1-1.2 at that stage. When higher microwave power levels than 50% was used, sample spill-out problem arised. The main reason could be explained by the focusing effect which can be explained as the focusing of microwaves at particular locations of the sample, especially the center and the corner (Schubert & Regier, 2005). Similarly, during crystallization and concentration operations, using higher power levels were not feasible. As mentioned in the materials section, a frusto-conical sample cup was used to decrease the focusing effect observed within the system since it is mainly affected by the product geometry (Schubert & Regier, 2005) yet this effect was not totally avoidable since the material is liquid and could be conserved in cylinder-like cups.

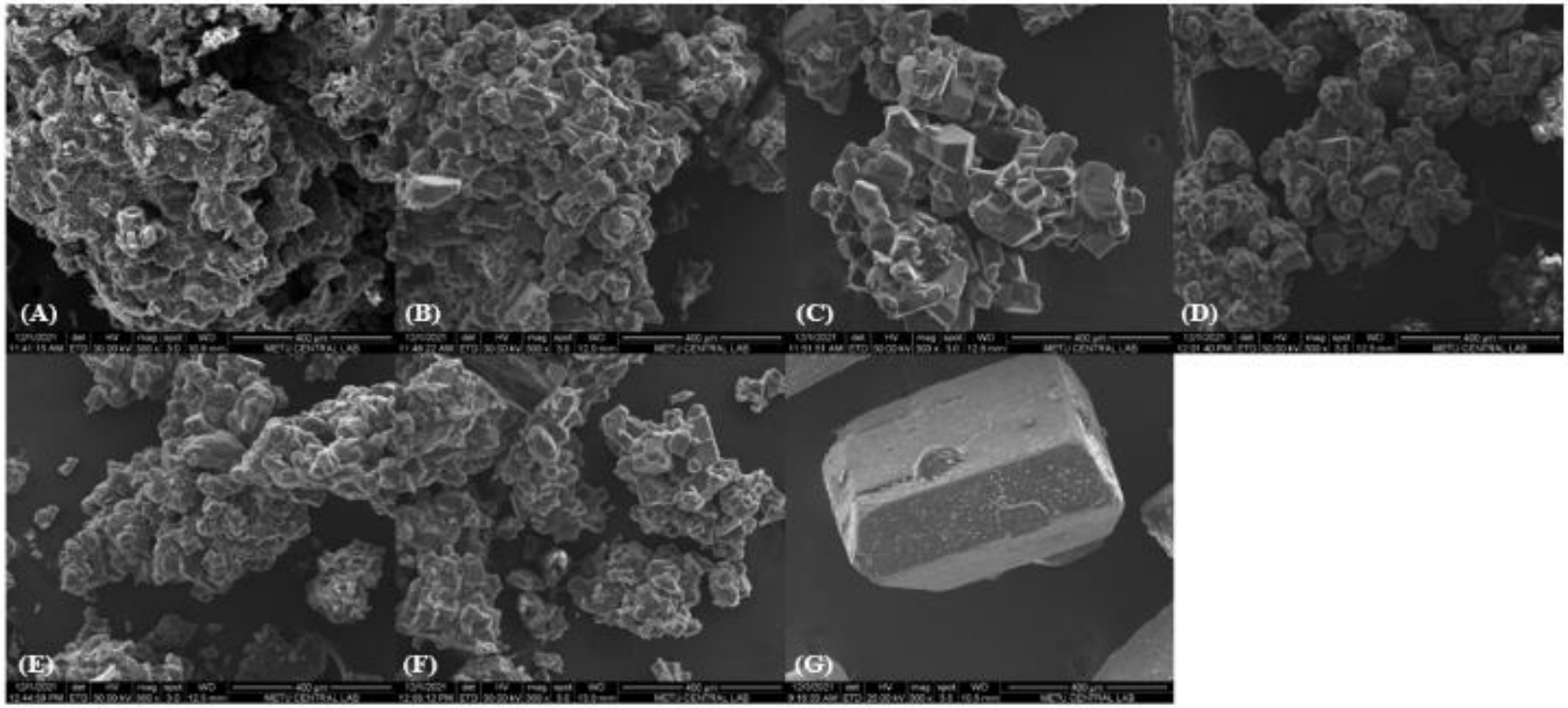
3.2 Effect of Microwave Processing on Crystal Structure and Crystallinity

3.2.1 Crystal Morphology

Crystal morphology is a cumulative term defining both the crystal structure and orientation (Prasad et al., 2018). Number of crystals in a crystal system can be oriented in numerous ways, which is thermodynamically affected by the crystal

growth (M. R. Prasad et al., 2018). Therefore, it is a crucial parameter to understand the crystallization process performed in this study. Crystal morphology of the samples were analyzed by SEM. Images of microwave processed sugar samples as well as the commercial sugar samples at magnification 300x were investigated as shown in Figure 3.1. The SEM images showed that microwave processing had no effect on sucrose crystal shape (which is defined as monoclinic shape). This was also observed in a study where crystallization was performed under ultrasonication (Singh et al., 2019; Zhong et al., 2022). Yet, commercial sugar has a more definite morphology and possessed a single large crystal structure whereas the microwave processed sugar samples developed a clustered structure consisting of many small crystals. Microwave processed sugars showed reduced crystal size, regardless of the process parameters used, as compared to the commercial sugar which could be explained by the difference of the mechanism of microwave and commercial heating and shorter crystallization times used in microwave processing. In a study where AC/TiO_2 crystallization in microwave was investigated, it was found that as microwave power increased, particles tended to agglomerate more due to the stabilization tendency of the small individual particles (Tian et al., 2015) which might also be the reason why microwave processed sugars had smaller crystal size. In microwave crystallization of the sucrose, due to the fast crystallization time, agglomeration tendency might be increased and hindered the crystal growth resulting in the agglomerated small crystal morphology as observed in the similar research. Studies on crystallization under ultrasonication (Singh et al., 2019; Zhong et al., 2022) reported that ultrasound resulted in smaller crystals and this effect was explained by the enhanced micro-scale mixing during ultra-sonication. Similarly, the molecular vibration formed by the microwave energy might lead to an improved micro-mixing effect. Additionally, it is crucial to consider the distribution and alignment of glucose and fructose molecules when examining the process of sucrose crystallization. The coalescence of glucose and fructose molecules is essential for the establishment of requisite hydrogen bonds present in the crystalline form of sucrose thereby could contribute to the desired sucrose crystallization process (Singh

et al., 2019). Notably, the orientation of these molecules in solution is a pivotal factor in determining the success of crystallization. In this regard, the electromagnetic field formed during microwave processing potentially could induce alterations in molecular orientations of glucose and fructose, subsequently bringing an influence on the crystallization process and the ability of small crystals to undergo rapid growth. Moreover, as the microwave exposure increased, crystals forming the cluster became more definite up to a certain exposure level as can be seen from the images. When the images of samples formed at 30% microwave power (Figure 3.1, (C) and (D)) were compared with the ones formed at 20% (Figure 3.1, (A) and (B)), it could be observed that the sugars exposed to higher microwave power had more definite images. However, when the samples from 40 and 30% microwave powers were compared, no such difference was observed. That showed that the crystallization time did not follow a linear trend with the microwave exposure level. It was definitely affected by the microwave power level up to a certain point, which can only be defined by further investigation of the crystallization mechanism of sucrose during microwave exposure.


(A)
(B)
(C)
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(F)
(G)

Figure 3.1 SEM Images of The Different Sucrose Samples, (A) Microwave processed sugars at 20% microwave power and with 5-minute feeding time, (B) Microwave processed sugars at 20% microwave power and with 15 minute feeding time, (C) Microwave processed sugars at 30% microwave power and with 5 minute feeding time, (D) Microwave processed sugars at 30% microwave power and with 15 minute feeding time, (E) Microwave processed sugars at 40% microwave power and with 5 minute feeding time, (F) Microwave processed sugars at 40% microwave power and with 15 minute feeding time, (G) Control (Commercial) Sucrose

The influence of feeding time on the process of sucrose crystallization is visible through the microscopic analysis. Prolonged feeding duration yields more distinct and definite crystal formation, as evidenced by the comparative evaluation of dyadic pairs of images, namely A and B, C and D, and E and F. Feeding, a critical operation in sucrose crystallization, plays a vital role in maintaining a constant level of supersaturation during the crystallization process. Subsequently, feeding is carried out as concentration continues, wherein the supersaturation ratio progressively intensifies, leading to a consequent increase in the viscosity of the solution. It is known that increased viscosity hinders crystallization by slowing down the particle diffusion from solution to crystal surface (Eggleston et al., 2012). Hence, based on the available evidence, it can be understood that the process of controlled nucleation was not fully accomplished by the end of the 5^{th} minute of feeding. This incomplete nucleation could be attributed to the heightened viscosity that occurs during the subsequent concentration stage. The elevated viscosity posessed a potential prevention to the controlled nucleation process, thereby increasing the likelihood of randomization in the crystallization process.

3.2.2 Crystallinity

The molecules forming a compound should be aligned in a specific order in the microscopic scale in order for a material to be defined as a crystal (Coupland, 2014). Sucrose is found in crystalline form in its solid phase. Consequently, one of the most

important parameters that should be evaluated to understand the crystallization performance is crystallinity, which is the percent alignment of the molecules in overall molecular distribution. Crystallinity analysis for commercial and microwave processed sucrose samples were performed both with XRD analysis and NMR analysis. XRD crystallinity results of microwave processed sugars were not significantly different as can be seen in Table 3.1.

Table 3.1 Effect of Microwave Power and Feeding Time on X-Ray Crystallinity

Crystallization Power Levels (%)	**Feeding Time (minutes)**	**X-Ray Crystallinity**
20	5	79.0 ± 0.001^{E}
20	15	85.0 ± 0.000^{A}
30	5	81.0 ± 0.001^{D}
30	15	83.0 ± 0.001^{B}
40	5	82.0 ± 0.001^{C}
40	15	83.0 ± 0.001^{B}
Commercial Sucrose		85.0 ± 0.000^{A}

Coloumns having different letters are significantly different ($p \leq 0.05$).

XRD crystallinity results of microwave processed crystals, were found to be statistically different where all the microwave processed samples revealed around 82% crystallinity which was considerably high. Therefore, it can be concluded that the crystallinity of the samples was far above being amorphous showing that crystallization process was performed properly. It was found that the microwave power did not affect the crystallinity significantly and application of higher feding time increased the crystallinity. That can be explained by the elongated crystal growth process time with longer feeding time. Additionally, for the common peaks appeared in all samples, the peak intensities were higher in commercial sugar as shown in Figure 3.2. In other words, peak intensity increased with increasing single

crystal size. It was stated that the peak intensity increased with the surface area of the diffraction face (Inoue & Hirasawa, 2013) which directly supported the intensity difference in microwave and commercial sugars. Commercial sucrose had much larger crystallite surface than microwave processed sucrose as can be seen from the SEM images in Figure 3.1. Similarly, it was found that when the TiO_2 crystals were examined with XRD, the peak intensity increased with crystal size (Thamaphat et al., 2008).

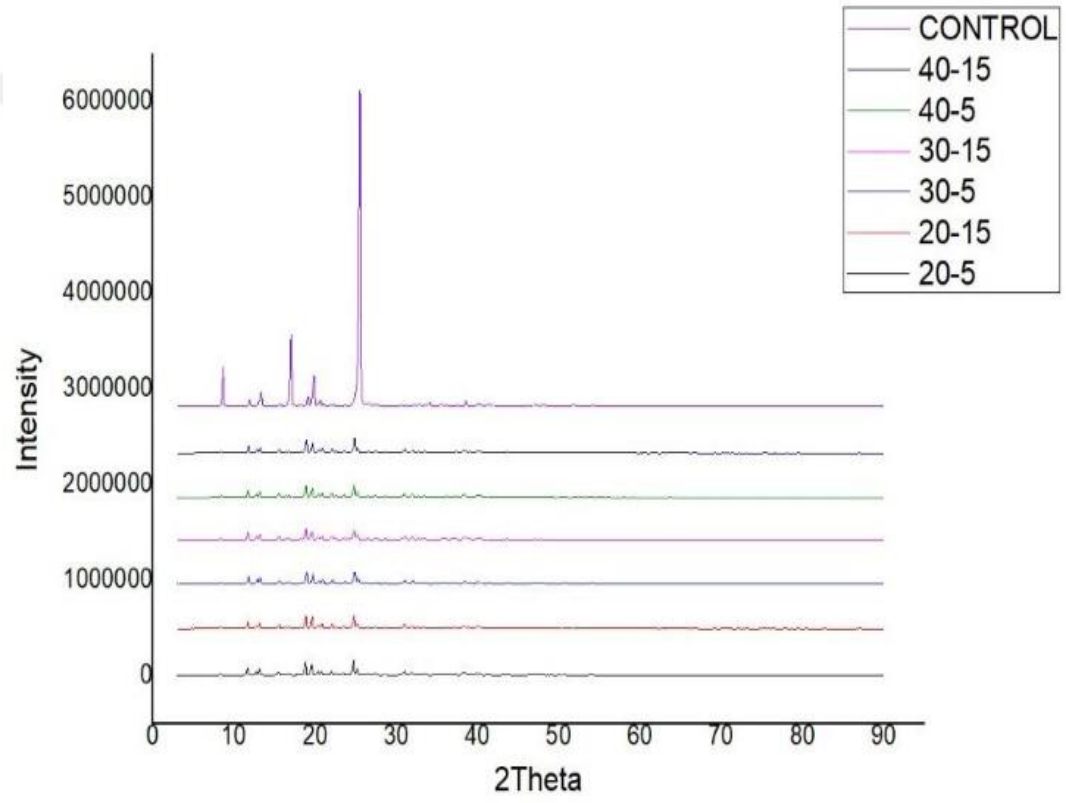

Figure 3.2 X-Ray Diffraction Results of Microwave Processed and Commercial Sucrose Samples (Control)

The legends are numbered as first number representing the microwave power used and the second number representing the feeding time. Control result is indicating the

commercial sucrose. The crystallinity estimated with NMR partially confirmed the XRD crystallinity results. Only the sugar processed at 20% microwave level showed significant difference in NMR-SE crystallinity. According to the **XRD** results, the difference on the crytsallinity values were less than **~2%** for the power levels other than 20%. This showed that the sensitivity of the *SE-Crystallinity* method may not be sufficient to detect these differences.

When microwave power level and feeding time's combined effects were investigated by 2-way ANOVA analysis, it was found that the highest power – longer feeding time combination results in longer T_1 whereas the lowest power – shortest feeding time results in shorter T_1 (Table 3.2). That result can be related to the crystal morphology. As mentioned, using high power for crystallization and long time resulted in more definition of the crystals. It is known that higher crystallinity could possess longer relaxation time (Uguz et al., 2022). Therefore, the higher T_1 values could be explained by the increased crystal definition as observed in SEM results.

Table 3.2 Effects of Feding Time and Crystallization Power Levels on NMR Results

Crystallization Power Levels (%)	**Feeding Time (minutes)**	**T_1**	**SE-Crystallinity**
20	5	1904.2 ± 85.3^{C}	73.7 ± 0.5^{B}
20	15	1911.2 ± 5.1^{BC}	73.0 ± 0.4^{B}
30	5	1977.3 ± 59.1^{ABC}	76.9 ± 0.8^{A}
30	15	1978.0 ± 14.0^{ABC}	76.5 ± 0.4^{A}
40	5	1858.7 ± 12.9^{C}	75.8 ± 0.8^{A}
40	15	2031 ± 20.6^{AB}	75.7 ± 1.2^{A}
Commercial Sucrose		2091.33 ± 38.4^{A}	77.0 ± 0.1^{A}

Coloumns having different letters are significantly different ($p \leq 0.05$).

3.3 Effect of Microwave Processing on Crystal Quality and Process Yield

3.3.1 Particle size

Particle size is a critical feature in sugar industry, and the desired particle size can vary depending on the area in which the product is going to be used. It is known that table sugar with particle size in the range of 200–5000 μm are commercially available (Richardson et al., 2018). Particle sizes of microwave crystallized sucrose (Sauter mean diameter) were detemined in the range of 400–500 μm which are shown in Table 3.3. It was found that, when 20% crystallization power with 15-minute feeding time was performed, particles were significantly smaller. Upon observing the SEM images, it became evident that there was no significant difference in the sizes of crystals produced by using 30% and 40% crystallization powers. However, the microwave-processed sucrose crystals, as shown in the SEM images, were noticeably smaller in comparison to commercial sucrose, which exhibited an average size (Sauter mean diameter) of 695 μm according to the sieve analysis. Another study revealed that the using ultrasound in crystallization process also led to a reduction in crystal sizes (Prasad & Dalvi, 2020). This was attributed to the enhanced micro-mixing and nucleation rates, directly resulting in smaller crystals (Kaur Bhangu et al., 2016; Prasad & Dalvi, 2020). Therefore, the difference between commercial and microwave-processed sucrose can be explained by the induction of a similar micro-mixing effect during microwave processing. Furthermore, an increase in microwave power was found to cause larger TiO_2 particles in microwave-assisted crystallization. This phenomenon was explained by the AC surface crystallization mechanism and the influence of increased surface area (Tian et al., 2015). Consequently, in order to comprehensively understand the microwave crystallization mechanism and kinetics of sucrose crystallization, further investigation of the crystallization kinetics of sucrose is essential. Although no significant differences were observed within the microwave-processed sucrose samples, except for the one produced with 20% microwave power and 15-minute

feeding time, it can be concluded that both microwave power level and feeding time used in the process were crucial for accurately distinguishing the differences within the microwave-processed samples.

Table 3.3 Effects of Feeding Time and Crystallization Power Levels on Sauter Mean Diameter Analysis

Crystallization Power Levels (%)	Feeding Time (minutes)	Sauter Mean Diameter (µm)
20	5	480.4 ± 0.1^{B}
20	15	427.3 ± 7.2^{C}
30	5	482.3 ± 0.7^{B}
30	15	482.3 ± 1.6^{B}
40	5	491.0 ± 4.3^{B}
40	15	482.3 ± 16.5^{B}
Commercial Sucrose		695.7 ± 2.0^{A}

Coloumns having different letters are significantly different ($p \leq 0.05$).

3.3.2 Solution Color and Turbidity

Solution color and turbidity are both highly dependent on the crystallization and crystal separation processes. Turbidity, also known as cloudiness, is mainly caused by the small undissolved particles whereas some other impurities like caramels and decomposition products of invert sugar degradation are the common source of color (Asadi, 2007). Both of these parameters are analyzed in the industry as a part of sugar quality control (Asadi, 2007). It was found that the solution color increased with decreased crystallization power as given in Table 3.4, which might be linked to prolonged process time required at the lower power levels. Turbidity results showed a similar trend. However, there was no significant difference between the turbidity levels of sucrose obtained from 30% and 40% crystallization powers with 5-minute feeding time which might show that during microwave processing, separable

coloring units are being formed rather than the inseparable compounds that are causing the turbidity. In general, increased feeding time resulted an increase in solution color and turbidity (Table 3.4). When compared with the previous studies, solution color values obtained from microwave processing were higher (Schlumbach et al., 2017). Also, the solution color and turbidity of commercially produced sugar were 17.4 and 4.1 IU respectively, which was much lower than the literature. These results might be linked to the difference in the particle size and morphology in the microwave and commercially produced sucrose crystals. The molasses separation from conglomerated microwave crystals could not be as efficient as it was with the large single crystals that the commercial sucrose consisted of. It is also known that the reduction in solution color and turbidity in white sugar is mostly achieved by the bleaching agents used in juice decolorization (Bensouissi et al., 2009). The number of impurities remaining on the sugar crystals depended on the process conditions (Bensouissi et al., 2009). Therefore, it can be concluded that the higher color and turbidity results observed in microwave processing might be affected also from the microwave application and vacuum level which was set slightly at the upper limit given in the literature (Asadi, 2007). Moreover, the surface adsorption mechanism might be affected by the microwave processing, as well is it might be affected by any impurities in the syrup (Eggleston et al., 2012).

Table 3.4 Effect of Feeding Time and Crystallization Power Levels on Solution Color (IU) and Turbidity (IU) Analysis

Crystallization Power Levels (%)	**Feeding Time (minutes)**	**Solution Color (IU)**	**Turbidity (IU)**
20	5	331.4 ± 6.8^{A}	51.7 ± 5.0^{AB}
20	15	287.5 ± 4.3^{B}	38.0 ± 6.6^{B}
30	5	159.5 ± 5.0^{E}	12.2 ± 1.4^{C}
30	15	245.0 ± 0.3^{C}	40.9 ± 4.2^{B}
40	5	152.8 ± 0.1^{E}	12.0 ± 1.2^{C}

Table 3.4 (Continued)

40	15	212.7 ± 2.6^{D}	57.1 ± 3.2^{A}
Commercial Sucrose		17.4 ± 1.4^{F}	4.1 ± 0.0^{C}

Coloumns having different letters are significantly different ($p \leq 0.05$).

3.3.3 L*a*b* Color

In addition to analyzing the solution color of the final products, a more comprehensive evaluation of the sugar color can be performed directly through L*a*b* color analysis. In solution color analysis, the spectrophotometric method gives only a perspective of yellow color as it is performed at 420 nm, whereas L*a*b* provides quantitative assessment on not only the lightness but also the material colors within red-green and yellow-blue chromatograms, respectively. Therefore, to support solution color and to obtain a deeper insight about crystal quality, color results are also given in Table 3.5. b* values and results from solution color analysis were expected to be similar since they both indicate yellowness of the samples. Yet, there are minor differences. For instance, solution color indicates higher degree of yellowness since b* indicates the intensity of yellowness as it deviates from 0 in the positive direction and blueness as it deviates from 0 in the negative direction. This can be a result from the presence of soluble color compounds within the crystal, which are cumulatively hidden by the lightness of the solid surface. Although the L* values of each sample resulted to be statistically different, numerically the difference between microwave processed sucrose and commercial sucrose is quite low so the L* results of microwave processed sucrose might fall in the acceptable range in the sugar quality analysis in the industry. The a* results may be considered similar with the L* results. The difference between microwave processed sucrose and the commercial sucrose might be coming from the heavier impurity load in microwave processed samples. As mentioned, one of the main reasons for color formation is the invert sugar degradation (Asadi, 2007) and it is known that the high temperature also accelarates inversion (Panpae et al., 2008). All

in all, percent degradation of sucrose in the microwave processing might be higher than the commercial sucrose production due to the potential pointwise overheating that might occur in microwave and lead to the notable increase in color a* and b*.

Table 3.5 Effect of Feeding Time and Crystallization Power Levels on L*a*b* Color Measurements

Crystallization Power Levels (%)	**Feeding Time (minutes)**	**L* (Lightness)**	**a* (Redness/ Greenness)**	**b* (Yellowness/ Blueness)**
20	5	85.2 ± 0.0^{C}	1.2 ± 0.0^{D}	15.6 ± 0.0^{A}
20	15	85.9 ± 0.0^{B}	0.8 ± 0.0^{F}	13.2 ± 0.0^{B}
30	5	84.7 ± 0.0^{G}	1.6 ± 0.0^{A}	13.2 ± 0.0^{B}
30	15	85.1 ± 0.0^{D}	1.5 ± 0.0^{B}	12.8 ± 0.0^{C}
40	5	85.0 ± 0.0^{E}	1.3 ± 0.0^{C}	11.6 ± 0.0^{D}
40	15	85.0 ± 0.0^{F}	1.0 ± 0.0^{E}	10.8 ± 0.0^{E}
Commercial Sucrose		86.4 ± 0.0^{A}	-0.4 ± 0.0^{G}	3.7 ± 0.0^{F}

Coloumns having different letters are significantly different ($p \leq 0.05$).

When the effect of process parameters on the values L*a*b* are evaluated, it can be observed that the colors L* and a* increased with increasing feeding time. L* values also increased with increasing microwave power level used in crystallization. Similarly, increased microwave power level resulted an increase in the b* values of the samples. These results suggest that longer processing time might leads to more color development, which might be caused by the extended exposure to elevated temperatures. However, commercial sucrose revealed higher L* and lower a* values which cannot be explained by the exposure to the temperature since the process time in commercial sucrose production is much higher. Less color development was expected in microwave when compared to commercial sucrose as previously observed in juice concentration in a microwave vacuum evaporation system (Bozkir & Baysal, 2017). However, as elaborated in the solution color and turbidity section, the difference between commercially processed sucrose and microwave processed

sucrose might be attributed to the impact of microwave processing on the surface adsorption mechanism of sucrose crystals (Eggleston et al., 2012) as well as the reduced efficiency of molasses separation due to decreased crystal size obtained from microwave processing.

3.3.4 Process Yield

It is known that crystallization yield in beet sugar industry was generally 55–60% (Asadi, 2007). Given the critical importance of reaching the greatest feasible process yield in the industry, there is a significant opportunity for the development of a high-performance process for commercial sucrose production. Hence, the potential of microwave processing to induce the process yield in sucrose crystallization was investigated in this study and yield was calculated for each process conditions given in the experimental design.

In microwave crystallization processes of sucrose, the process yield was varying between 45–55% and in various process conditions the yield from microwave processing was the same as the commercial sucrose yield as interpreted in Table 3.6. The main reason why the process yield values of microwave vacuum evaporation is in a lower percent range might be the absence of a juice recovery after centrifuge and performing the process within a semi-batch design. Continuous flow in the system is known to improve the extraction rate (Shiau, 2003) which is directly linked to the process yield. In addition, the syrup separated from the sugar after crystallization is generally reprocessed in the industry, which increases the crystallization yield as well (Asadi, 2007). It can be understood that the range of process yields attained in microwave crystallization of sucrose primarily caused by the absence of reprocessing and not having a continuous process, rather than resulting from the use of microwave energy throughout the crystallization. Therefore, it can still be said that with further process improvement, microwave might reveal its yield increasing effect as shown in various studies (Asghar et al., 2020; Assawarachan & Noomhorm, 2011; Bozkir & Baysal, 2017) even though the

enhancement in yield was not currently observed. When the effect of crystallization powers used were compared in Table 3.6, the yield in 40% crystallization power was found to be significantly lower. This might be due to the accelerated crystallization caused by microwave radiation. Microwave exposure might result in enhanced micro-scale mixing, which also potentially affected the particle size as explained, thereby accelerated crystallization, which might lead to the decrease in yield. Shorter feeding time also resulted in significantly lower yield. The highest yield was obtained in the combination of 30% crystallization power and 15-minute feeding time. Proper feed flow enhances crystal growth by providing higher diffusion rates (Shiau, 2003). Therefore, having higher process yields while keeping the feed longer is expected. Moreover, the process times are around 120 minutes for commercial crystallization (Asadi, 2007) and microwave processing can shorten this process time up to 45 minutes. In the experimental design, maximum process time was about 70 minutes which showed that microwave processing provided an advantage in this regard.

Table 3.6 Effect of Feeding Time and Crystallization Power Levels on Process Yield

Crystallization Power Levels (%)	**Feeding Time (minutes)**	**Percent Yield (%)**
20	5	47.8 ± 1.5^{BCD}
20	15	54.5 ± 2.5^{AB}
30	5	53.3 ± 1.9^{ABC}
30	15	57.4 ± 1.9^{A}
40	5	44.1 ± 2.9^{D}
40	15	45.7 ± 2.8^{CD}
Commercial Sucrose		57.5 ± 2.5^{A}

Coloumns having different letters are significantly different ($p \leq 0.05$).

CHAPTER 4

CONCLUSION AND RECOMMENDATIONS

The primary objective of this study was to establish a novel sucrose crystallization process capable of achieving a level of efficacy comparable to that of existing commercial methods, thereby revealing the potential of microwave crystallization. Various microwave power levels for crystallization and feeding durations were employed during the microwave vacuum crystallization of sucrose as part of this investigation. Notably, the incorporation of microwave exposure played a crucial role in accelerating the crystallization rate, resulting in the formation of smaller crystalline structures rather than larger individual particles.

When the final products were examined, the results revealed a significant degree of crystallinity, indicating that the formation of amorphous by-products during crystallization was negligible which was supported by NMR data as well. As microwave power level increased, the solution color and b* (which both indicate yellowness) decreased. In general, increased feeding time increased sample quality, resulting in decreased color and turbidity. Similarly, the process yield was higher in 15 minute feeding processes of the same microwave level. When evaluating the impact of different process parameters on the dependent variables as a whole, it was concluded that utilizing a crystallization power of 30% combined with a 15-minute feeding time yielded the most compatible crystallization process. Although the numerical values for product quality parameters and product yield fell below those achieved by the commercial crystallization, it is necessary to perform further investigations to understand the effect of microwave exposure on the crystallization kinetics of sucrose. Such studies would enable the identification of necessary modifications to enhance the microwave crystallization procedure.

Furthermore, the incorporation of microwave vacuum evaporation has the potential to significantly reduce the overall process time, potentially by half or even more. In summary, the utilization of microwave vacuum evaporation can be a potential alternative to commercial sucrose crystallization methods.

In further studies, different process designs may be evaluated for microwave vacuum evaporation of sucrose. Especially designing a continuous system might increase the process yield. The focus might be on the crystallization kinetics of sucrose in microwave processing to identify the reason behind the resulting clustered crystal morphology and small crystal size better as compared to the commercially produced sucrose.

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APPENDICES

A. Statistical Analysis

Table 1. Analysis of Variance for .X-Ray Crystallinity – Comparison of Microwave Processing Conditions

```
Factor                  Type   Levels  Values
Crystallization Power   fixed       3  20; 30; 40
Feeding Time            fixed       2  5; 15


Analysis of Variance for Crystallinity, using Adjusted SS for Tests

Source                               DF     Seq SS     Adj SS     Adj MS
Crystallization Power                 2  0,0000667  0,0000667  0,0000333
Feeding Time                          1  0,0027000  0,0027000  0,0027000
Crystallization Power*Feeding Time    2  0,0014000  0,0014000  0,0007000
Error                                 6  0,0000100  0,0000100  0,0000017
Total                                11  0,0041767

Source                                  F      P
Crystallization Power                20,00  0,002
Feeding Time                       1620,00  0,000
Crystallization Power*Feeding Time  420,00  0,000
Error
Total


S = 0,00129099   R-Sq = 99,76%   R-Sq(adj) = 99,56%


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N  Mean  Grouping
40               4   0,8  A
30               4   0,8    B
20               4   0,8    B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N  Mean  Grouping
15       6   0,8  A
 5       6   0,8    B

Means that do not share a letter are significantly different.
```

```
Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N  Mean  Grouping
20               15       2   0,8  A
40               15       2   0,8    B
30               15       2   0,8    B
40                5       2   0,8      C
30                5       2   0,8        D
20                5       2   0,8          E

Means that do not share a letter are significantly different.
```

Table 2. Analysis of Variance for X-Ray Crystallinity – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF         SS         MS       F      P
Sample ID   6  0,0055429  0,0009238  646,67  0,000
Error       7  0,0000100  0,0000014
Total      13  0,0055529

S = 0,001195   R-Sq = 99,82%   R-Sq(adj) = 99,67%


                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N     Mean    StDev  -------+---------+---------+---------+--
1      2  0,79000  0,00141  (*)
2      2  0,85000  0,00000                                      (*)
3      2  0,81000  0,00141            (*)
4      2  0,83000  0,00141                      (*)
5      2  0,82000  0,00141                  (-*)
6      2  0,83000  0,00141                      (*)
7      2  0,85000  0,00000                                      (*)
                          -------+---------+---------+---------+--
                               0,800     0,816     0,832     0,848

Pooled StDev = 0,00120


Grouping Information Using Tukey Method

Sample
ID      N      Mean  Grouping
7       2  0,850000  A
2       2  0,850000  A
6       2  0,830000    B
4       2  0,830000    B
5       2  0,820000      C
```

```
3       2  0,810000        D
1       2  0,790000          E

Means that do not share a letter are significantly different.
```

Table 3. Analysis of Variance for T_1 – Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for T1, using Adjusted SS for Tests

Source                                DF  Seq SS  Adj SS  Adj MS     F
P
Crystallization Power                  2   14603   14603    7302  3,78
0,053
Feeding Time                           1   16399   16399   16399  8,50
0,013
Crystallization Power*Feeding Time     2   28578   28578   14289  7,41
0,008
Error                                 12   23152   23152    1929
Total                                 17   82733


S = 43,9246   R-Sq = 72,02%   R-Sq(adj) = 60,36%


Unusual Observations for T1

Obs       T1      Fit  SE Fit  Residual  St Resid
  1  1995,70  1904,23   25,36     91,47      2,55 R
  2  1827,00  1904,23   25,36    -77,23     -2,15 R

R denotes an observation with a large standardized residual.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N    Mean  Grouping
30               6  1977,7  A
40               6  1945,2  A B
20               6  1908,0    B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N    Mean  Grouping
15       9  1973,8  A
 5       9  1913,4    B

Means that do not share a letter are significantly different.
```

```
Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N    Mean  Grouping
40               15       3  2031,7  A
30               15       3  1978,0  A B
30                5       3  1977,3  A B
20               15       3  1911,7  A B
20                5       3  1904,2    B
40                5       3  1858,7    B

Means that do not share a letter are significantly different.
```

Table 4. Analysis of Variance for T_1 – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose

Source     DF      SS     MS      F      P
Sample ID   6  115707  19284  10,34  0,000
Error      14   26105   1865
Total      20  141812

S = 43,18   R-Sq = 81,59%   R-Sq(adj) = 73,70%


                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N    Mean  StDev  ---------+---------+---------+---------+
1      3  1904,2   85,2       (----*-----)
2      3  1911,7    5,1        (----*-----)
3      3  1977,3   59,1              (-----*----)
4      3  1978,0   14,0              (-----*----)
5      3  1858,7   12,9   (----*----)
6      3  2031,7   20,6                    (----*-----)
7      3  2091,3   38,4                          (----*----)
                          ---------+---------+---------+---------+
                                1900      2000      2100      2200

Pooled StDev = 43,2


Grouping Information Using Tukey Method

Sample
ID      N     Mean  Grouping
7       3  2091,33  A
6       3  2031,67  A B
4       3  1978,00  A B C
3       3  1977,33  A B C
```

```
2       3  1911,67    B C
1       3  1904,23      C
5       3  1858,67      C

Means that do not share a letter are significantly different.


Tukey 95% Simultaneous Confidence Intervals
All Pairwise Comparisons among Levels of Sample ID
```

Table 5. Analysis of Variance for SE Crystallinity – Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for SE_Average TD Crystallinity, using Adjusted SS
for
     Tests

Source                              DF   Seq SS   Adj SS   Adj MS      F
P
Crystallization Power                2  35,4900  35,4900  17,7450  33,13
0,000
Feeding Time                         1   0,7200   0,7200   0,7200   1,34
0,269
Crystallization Power*Feeding Time   2   0,3033   0,3033   0,1517   0,28
0,758
Error                               12   6,4267   6,4267   0,5356
Total                               17  42,9400


S = 0,731817   R-Sq = 85,03%   R-Sq(adj) = 78,80%


Unusual Observations for SE_Average TD Crystallinity

     SE_Average TD
Obs  Crystallinity      Fit  SE Fit  Residual  St Resid
 16        77,0000  75,6667  0,4225    1,3333      2,23 R

R denotes an observation with a large standardized residual.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N  Mean  Grouping
30               6  76,7  A
40               6  75,7  A
20               6  73,4    B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence
```

```
Feeding
Time     N  Mean  Grouping
 5       9  75,5  A
15       9  75,1  A

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N  Mean  Grouping
30                5       3  76,9  A
30               15       3  76,5  A
40                5       3  75,8  A
40               15       3  75,7  A B
20                5       3  73,7    B C
20               15       3  73,0      C

Means that do not share a letter are significantly different.
```

Table 6. Analysis of Variance for SE – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose

Source     DF      SS     MS      F      P
Sample ID   6  43,945  7,324  15,94  0,000
Error      14   6,433  0,460
Total      20  50,378

S = 0,6779   R-Sq = 87,23%   R-Sq(adj) = 81,76%


                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N    Mean  StDev  ---------+---------+---------+---------+
1      3  73,733  0,493       (-----*----)
2      3  73,000  0,400  (-----*----)
3      3  76,900  0,794                            (-----*----)
4      3  76,533  0,379                          (----*-----)
5      3  75,767  0,833                     (----*-----)
6      3  75,667  1,159                    (----*-----)
7      3  76,967  0,058                              (----*-----)
                          ---------+---------+---------+---------+
                                73,5      75,0      76,5      78,0

Pooled StDev = 0,678


Grouping Information Using Tukey Method
```

```
Sample
ID      N     Mean  Grouping
7       3  76,9667  A
3       3  76,9000  A
4       3  76,5333  A
5       3  75,7667  A
6       3  75,6667  A
1       3  73,7333    B
2       3  73,0000    B

Means that do not share a letter are significantly different.
```

Table 7. Analysis of Variance for Particle Size (Sauter Mean Diameter)– Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for Sauter Mean Diameter, using Adjusted SS for Tests

Source                              DF  Seq SS  Adj SS  Adj MS      F
P
Crystallization Power                2  2538,6  2538,6  1269,3  22,11
0,002
Feeding Time                         1  1273,0  1273,0  1273,0  22,17
0,003
Crystallization Power*Feeding Time   2  1616,8  1616,8   808,4  14,08
0,005
Error                                6   344,5   344,5    57,4
Total                               11  5772,8


S = 7,57712   R-Sq = 94,03%   R-Sq(adj) = 89,06%


Unusual Observations for Sauter Mean Diameter

     Sauter Mean
Obs     Diameter      Fit  SE Fit  Residual  St Resid
 11      470,615  482,261   5,358   -11,646     -2,17 R
 12      493,907  482,261   5,358    11,646      2,17 R

R denotes an observation with a large standardized residual.


Grouping Information Using Tukey Method and 95,0% Confidence


Crystallization
Power            N   Mean  Grouping
40               4  486,6  A
30               4  482,3  A
20               4  453,9    B

Means that do not share a letter are significantly different.
```

```
Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N   Mean  Grouping
 5       6  484,6  A
15       6  464,0     B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N   Mean  Grouping
40                5       2  491,0  A
30               15       2  482,3  A
30                5       2  482,3  A
40               15       2  482,3  A
20                5       2  480,4  A
20               15       2  427,3     B

Means that do not share a letter are significantly different.
```

Table 8. Analysis of Variance for Particle Size (Sauter Mean Diameter) – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF       SS       MS       F      P
Sample ID   6  89478,2  14913,0  299,71  0,000
Error       7    348,3     49,8
Total      13  89826,5

S = 7,054   R-Sq = 99,61%   R-Sq(adj) = 99,28%


                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N    Mean  StDev  --------+---------+---------+---------+-
1      2  480,37   0,06         (*-)
2      2  427,34   7,18   (*-)
3      2  482,30   0,67         (*-)
4      2  482,31   1,62         (*-)
5      2  491,04   4,32          (*-)
6      2  482,26  16,47         (*-)
7      2  695,69   1,96                                    (-*)
                          --------+---------+---------+---------+-
                                480       560       640       720

Pooled StDev = 7,05


Grouping Information Using Tukey Method
```

```
Sample
ID      N    Mean  Grouping
7       2  695,69  A
5       2  491,04    B
4       2  482,31    B
3       2  482,30    B
6       2  482,26    B
1       2  480,37    B
2       2  427,34      C

Means that do not share a letter are significantly different.
```

Table 9. Analysis of Variance for Solution Color – Comparison of Microwave Processing Conditions

```
Factor                Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for Solution Color, using Adjusted SS for Tests

Source                              DF  Seq SS  Adj SS  Adj MS        F
P
Crystallization Power                2   37254   37254   18627  1165,25
0,000
Feeding Time                         1    3438    3438    3438   215,06
0,000
Crystallization Power*Feeding Time   2    9398    9398    4699   293,94
0,000
Error                                6      96      96      16
Total                               11   50185


S = 3,99816   R-Sq = 99,81%   R-Sq(adj) = 99,65%


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N   Mean  Grouping
20               4  309,5  A
30               4  202,2    B
40               4  182,8      C

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N   Mean  Grouping
15       6  248,4  A
 5       6  214,6    B

Means that do not share a letter are significantly different.
```

```
Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N   Mean  Grouping
20                5       2  331,4  A
20               15       2  287,5    B
30               15       2  245,0      C
40               15       2  212,7        D
30                5       2  159,5          E
40                5       2  152,8          E

Means that do not share a letter are significantly different.
```

Table 10. Analysis of Variance for Solution Color – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF        SS       MS        F      P
Sample ID   6  128683,0  21447,2  1534,04  0,000
Error       7      97,9     14,0
Total      13  128780,9

S = 3,739   R-Sq = 99,92%   R-Sq(adj) = 99,86%


                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N    Mean  StDev  ---------+---------+---------+---------+
1      2  331,43   6,76                                    *)
2      2  287,52   4,33                              (*
3      2  159,47   4,94                 (*)
4      2  244,99   0,31                          *)
5      2  152,77   0,13                 *)
6      2  212,73   2,64                       *)
7      2   17,37   1,40  (*
                          ---------+---------+---------+---------+
                                 100       200       300       400

Pooled StDev = 3,74


Grouping Information Using Tukey Method

Sample
ID      N    Mean  Grouping
1       2  331,43  A
2       2  287,52    B
4       2  244,99      C
6       2  212,73        D
3       2  159,47          E
```

```
5       2  152,77          E
7       2   17,37            F

Means that do not share a letter are significantly different.
```

Table 11. Analysis of Variance for Turbidity – Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for Turbidity, using Adjusted SS for Tests

Source                               DF   Seq SS   Adj SS   Adj MS      F
P
Crystallization Power                 2   674,28   674,28   337,14  20,21
0,002
Feeding Time                          1  1199,60  1199,60  1199,60  71,90
0,000
Crystallization Power*Feeding Time    2  1848,53  1848,53   924,26  55,40
0,000
Error                                 6   100,11   100,11    16,68
Total                                11  3822,52


S = 4,08472   R-Sq = 97,38%   R-Sq(adj) = 95,20%


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N  Mean  Grouping
20               4  44,8  A
40               4  34,6    B
30               4  26,5    B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N  Mean  Grouping
15       6  45,3  A
 5       6  25,3    B

Means that do not share a letter are significantly different.
```

Table 12. Analysis of Variance for Turbidity – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
```

```
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF      SS     MS      F      P
Sample ID   6  5393,7  898,9  62,86  0,000
Error       7   100,1   14,3
Total      13  5493,8

S = 3,782   R-Sq = 98,18%   R-Sq(adj) = 96,62%


                         Individual 95% CIs For Mean Based on
                         Pooled StDev
Level  N    Mean  StDev  -+---------+---------+---------+--------
1      2  51,740  5,077                          (--*--)
2      2  37,945  6,569                  (--*--)
3      2  12,200  1,414      (--*--)
4      2  40,855  4,207                   (--*---)
5      2  11,990  1,202      (--*--)
6      2  57,120  3,168                            (---*--)
7      2   4,085  0,007  (--*--)
                         -+---------+---------+---------+--------
                          0        20        40        60

Pooled StDev = 3,782


Grouping Information Using Tukey Method

Sample
ID      N    Mean  Grouping
6       2  57,120  A
1       2  51,740  A B
4       2  40,855    B
2       2  37,945    B
3       2  12,200      C
5       2  11,990      C
7       2   4,085      C

Means that do not share a letter are significantly different.
```

Table 13. Analysis of Variance for L* – Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for L*, using Adjusted SS for Tests

Source                              DF   Seq SS   Adj SS   Adj MS
F
Crystallization Power                2  1,48981  1,48981  0,74491
33520,75
Feeding Time                         1  0,42936  0,42936  0,42936
19321,00
```

```
Crystallization Power*Feeding Time   2  0,48501  0,48501  0,24251
10912,75
Error                               12  0,00027  0,00027  0,00002
Total                               17  2,40444

Source                                  P
Crystallization Power                0,000
Feeding Time                         0,000
Crystallization Power*Feeding Time   0,000
Error
Total


S = 0,00471405   R-Sq = 99,99%   R-Sq(adj) = 99,98%


Unusual Observations for L*

Obs       L*      Fit  SE Fit  Residual  St Resid
 17  84,9200  84,9100  0,0027    0,0100      2,60 R
 18  84,9000  84,9100  0,0027   -0,0100     -2,60 R

R denotes an observation with a large standardized residual.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N  Mean  Grouping
20               6  85,5  A
40               6  85,0    B
30               6  84,9      C

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N  Mean  Grouping
15       9  85,3  A
 5       9  85,0    B

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N  Mean  Grouping
20               15       3  85,9  A
20                5       3  85,2    B
30               15       3  85,0      C
40                5       3  85,0        D
40               15       3  84,9          E
30                5       3  84,7            F

Means that do not share a letter are significantly different.
```

Table 14. Analysis of Variance for L* – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF        SS        MS         F      P
Sample ID   6  6,712829  1,118805  58737,25  0,000
Error      14  0,000267  0,000019
Total      20  6,713095

S = 0,004364   R-Sq = 100,00%   R-Sq(adj) =  99,99%

                          Individual 95% CIs For Mean Based on
                          Pooled StDev
Level  N     Mean   StDev  ------+---------+---------+---------+---
1      3  85,1800  0,0000          (*
2      3  85,8800  0,0000                       (*
3      3  84,7200  0,0000  *)
4      3  85,0500  0,0000        *
5      3  85,0133  0,0058       *
6      3  84,9100  0,0100     *
7      3  86,4200  0,0000                                 *)
                          ------+---------+---------+---------+---
                             85,00     85,50     86,00     86,50

Pooled StDev = 0,0044


Grouping Information Using Tukey Method

Sample
ID      N      Mean  Grouping
7       3  86,42000  A
2       3  85,88000    B
1       3  85,18000      C
4       3  85,05000        D
5       3  85,01333          E
6       3  84,91000            F
3       3  84,72000              G

Means that do not share a letter are significantly different.
```

Table 15. Analysis of Variance for a* – Comparison of Microwave Processing Conditions

```
Factor                Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15
```

Analysis of Variance for a*, using Adjusted SS for Tests

Source	DF	Seq SS	Adj SS	Adj MS	F
Crystallization Power	2	0,98101	0,98101	0,49051	22072,75
Feeding Time	1	0,29902	0,29902	0,29902	13456,00
Crystallization Power*Feeding Time	2	0,06854	0,06854	0,03427	1542,25
Error	12	0,00027	0,00027	0,00002	
Total	17	1,34884			

Source	P
Crystallization Power	0,000
Feeding Time	0,000
Crystallization Power*Feeding Time	0,000
Error	
Total	

S = 0,00471405 R-Sq = 99,98% R-Sq(adj) = 99,97%

Unusual Observations for a*

Obs	a*	Fit	SE Fit	Residual	St Resid
1	1,14000	1,15000	0,00272	-0,01000	-2,60 R
3	1,16000	1,15000	0,00272	0,01000	2,60 R

R denotes an observation with a large standardized residual.

Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization Power	N	Mean	Grouping
30	6	1,5	A
40	6	1,1	B
20	6	1,0	C

Means that do not share a letter are significantly different.

Grouping Information Using Tukey Method and 95,0% Confidence

Feeding Time	N	Mean	Grouping
5	9	1,3	A
15	9	1,1	B

Means that do not share a letter are significantly different.

Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization Power	Feeding Time	N	Mean	Grouping
30	5	3	1,6	A

```
30              15       3   1,5    B
40               5       3   1,3      C
20               5       3   1,1        D
40              15       3   1,0          E
20              15       3   0,8            F

Means that do not share a letter are significantly different.
```

Table 16. Analysis of Variance for a* – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source      DF        SS        MS         F      P
Sample ID    6  7,894914  1,315819  69080,50  0,000
Error       14  0,000267  0,000019
Total       20  7,895181

S = 0,004364   R-Sq = 100,00%   R-Sq(adj) = 100,00%


                              Individual 95% CIs For Mean Based on
                              Pooled StDev
Level  N      Mean    StDev  --------+---------+---------+---------+-
1      3   1,15000  0,01000                                  *
2      3   0,80000  0,00000                        *
3      3   1,57333  0,00577                                      *)
4      3   1,49000  0,00000                                    *
5      3   1,31000  0,00000                                 *
6      3   0,97000  0,00000                           *)
7      3  -0,38000  0,00000  *)
                              --------+---------+---------+---------+-
                                    0,00      0,50      1,00      1,50

Pooled StDev = 0,00436


Grouping Information Using Tukey Method

Sample
ID      N      Mean  Grouping
3       3   1,57333  A
4       3   1,49000    B
5       3   1,31000      C
1       3   1,15000        D
6       3   0,97000          E
2       3   0,80000            F
7       3  -0,38000              G

Means that do not share a letter are significantly different.
```

Table 17. Analysis of Variance for b* – Comparison of Microwave Processing Conditions

```
Factor                 Type   Levels  Values
Crystallization Power  fixed       3  20; 30; 40
Feeding Time           fixed       2  5; 15


Analysis of Variance for b*, using Adjusted SS for Tests

Source                              DF   Seq SS   Adj SS   Adj MS
F
Crystallization Power                2  30,9234  30,9234  15,4617
2783107,00
Feeding Time                         1   6,7589   6,7589   6,7589
1216609,00
Crystallization Power*Feeding Time   2   3,1167   3,1167   1,5583
280501,00
Error                               12   0,0001   0,0001   0,0000
Total                               17  40,7991

Source                                  P
Crystallization Power                0,000
Feeding Time                         0,000
Crystallization Power*Feeding Time   0,000
Error
Total


S = 0,00235702   R-Sq = 100,00%   R-Sq(adj) = 100,00%


Unusual Observations for b*

Obs       b*      Fit  SE Fit  Residual  St Resid
 17  10,8000  10,7933  0,0014    0,0067      3,46 R

R denotes an observation with a large standardized residual.


Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization
Power            N  Mean  Grouping
20               6  14,4  A
30               6  13,0     B
40               6  11,2        C

Means that do not share a letter are significantly different.


Grouping Information Using Tukey Method and 95,0% Confidence

Feeding
Time     N  Mean  Grouping
 5       9  13,5  A
15       9  12,3     B

Means that do not share a letter are significantly different.
```

```
Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization  Feeding
Power            Time     N  Mean  Grouping
20                5       3  15,6  A
20               15       3  13,2    B
30                5       3  13,2    B
30               15       3  12,8      C
40                5       3  11,6        D
40               15       3  10,8          E

Means that do not share a letter are significantly different.
```

Table 18. Analysis of Variance for b* – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
                          Pooled StDev
Level  N     Mean   StDev  --------+---------+---------+---------+-
1      3  15,6100  0,0000                                        *
2      3  13,2300  0,0000                                  *
3      3  13,2300  0,0000                                  *
4      3  12,7800  0,0000                                *
5      3  11,6400  0,0000                           *
6      3  10,7933  0,0058                      *
7      3   3,7400  0,0000  *
                          --------+---------+---------+---------+-
                                6,0       9,0      12,0      15,0

Pooled StDev = 0,0022


Grouping Information Using Tukey Method

Sample
ID      N     Mean  Grouping
1       3  15,6100  A
3       3  13,2300    B
2       3  13,2300    B
4       3  12,7800      C
5       3  11,6400        D
6       3  10,7933          E
7       3   3,7400            F

Means that do not share a letter are significantly different.
```

Table 19. Analysis of Variance for Process Yield – Comparison of Microwave Processing Conditions

Factor	Type	Levels	Values
Crystallization Power	fixed	3	20; 30; 40
Feeding Time	fixed	2	5; 15

Analysis of Variance for Yield, using Adjusted SS for Tests

Source	DF	Seq SS	Adj SS	Adj MS	F
Crystallization Power	2	0,0331741	0,0331741	0,0165871	20,61
Feeding Time	1	0,0077651	0,0077651	0,0077651	9,65
Crystallization Power*Feeding Time	2	0,0020298	0,0020298	0,0010149	1,26
Error	12	0,0096558	0,0096558	0,0008046	
Total	17	0,0526249			

Source	P
Crystallization Power	0,000
Feeding Time	0,009
Crystallization Power*Feeding Time	0,318
Error	
Total	

S = 0,0283663 R-Sq = 81,65% R-Sq(adj) = 74,01%

Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization Power	N	Mean	Grouping
30	6	0,6	A
20	6	0,5	A
40	6	0,4	B

Means that do not share a letter are significantly different.

Grouping Information Using Tukey Method and 95,0% Confidence

Feeding Time	N	Mean	Grouping
15	9	0,5	A
5	9	0,5	B

Means that do not share a letter are significantly different.

Grouping Information Using Tukey Method and 95,0% Confidence

Crystallization Power	Feeding Time	N	Mean	Grouping
30	15	3	0,6	A
20	15	3	0,5	A B
30	5	3	0,5	A B C

```
20                5       3   0,5    B C D
40               15       3   0,5      C D
40                5       3   0,4        D

Means that do not share a letter are significantly different.
```

Table 20. Analysis of Variance for Process Yield – Comparison of Microwave Processed Sucrose with Commercial Sucrose

```
Level(Sample ID)  Process Conditions
1                 20% Crystallization Power*5 minute Feeding Time
2                 20% Crystallization Power*15 minute Feeding Time
3                 30% Crystallization Power*5 minute Feeding Time
4                 30% Crystallization Power*15 minute Feeding Time
5                 40% Crystallization Power*5 minute Feeding Time
6                 40% Crystallization Power*15 minute Feeding Time
7                 Commercial Sucrose
Source     DF        SS        MS      F      P
Sample ID   6  0,043045  0,007174  10,40  0,000
Error      14  0,009656  0,000690
Total      20  0,052701

S = 0,02626   R-Sq = 81,68%   R-Sq(adj) = 73,83%


                            Individual 95% CIs For Mean Based on
                            Pooled StDev
Level  N     Mean    StDev  --------+---------+---------+---------+-
1      3  0,47762  0,01821         (------*-----)
2      3  0,54522  0,03088                    (-----*------)
3      3  0,53264  0,02343                  (------*-----)
4      3  0,57409  0,02373                           (------*-----)
5      3  0,44110  0,03554  (-----*------)
6      3  0,45667  0,03416     (-----*------)
7      3  0,51000  0,00000               (------*------)
                            --------+---------+---------+---------+-
                                  0,450     0,500     0,550     0,600

Pooled StDev = 0,02626


Grouping Information Using Tukey Method

Sample
ID      N     Mean  Grouping
4       3  0,57409  A
2       3  0,54522  A B
3       3  0,53264  A B
7       3  0,51000  A B C
1       3  0,47762    B C
6       3  0,45667      C
5       3  0,44110      C

Means that do not share a letter are significantly different.
```