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Cooling and Evaporative Crystallization of α-D-Galactose from a Highly Viscous Industrial Side Stream

An industrial side stream containing mainly glucose and galactose was used to crystallize α-D-galactose. The dynamic viscosity of samples containing galactose and glucose solutions was measured and compared with pure glucose and galactose solutions. Various crystallization conditions were investigated in terms of temperature range and batch times in cooling crystallization and their influence on the product crystal properties. The obtained results were compared with the results of evaporative crystallization. Several characterization methods were used for studying crystal size and shape, crystal structure, and purity. The overall crystallization time of α-D-galactose was shortened by cooling crystallization based on stepwise temperature decrease and the desired crystal properties were achieved.

Supporting Information available online

Keywords: Cooling crystallization, Crystal size distribution, Evaporative crystallization, Galactose, Viscosity

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1 Introduction

Galactose is a naturally occurring monosaccharide that is found in a variety of natural sources, including milk and other dairy products. Galactose has the same chemical formula as glucose: $C_6H_{12}O_6$. One major dietary source of galactose is lactose, a disaccharide made of one glucose molecule and one galactose molecule [1]. The hydrolysis process of lactose to galactose and glucose can be achieved by using either acid or enzymes. The review by Sienkiewicz and Riedel [2] provides details of these processes, the chemical composition of the different syrups and characterization of the enzymes used, including commercial preparations, and their countries of origin. One of the methods of recovering galactose from a solution is crystallization. The crystallization of galactose is important in the production of food and pharmaceutical products. For example, in the dairy industry, galactose crystals are used to create a sweet, creamy texture in products like ice cream and cheese. In the pharmaceutical industry, galactose crystals are used as a starting material for the production of certain drugs.

Currently, there are only a few scientific publications on the crystallization of galactose and the effects of the parameters. Patent WO2005001145A1 [3] discloses a process based on chromatographic fractionation and crystallization for purifying galactose from a solution. The solution was concentrated by evaporation to 64–72 % of solutes at a temperature of 65 °C. The mass was cooled down from a temperature of 60–68 °C to 20–28 °C. The crystallization cake was recovered after 40 h from seeding by centrifugation, whereby a cake purity of 92.1–98.3 % based on dry substance was obtained. However, the literature is devoid of data on the variations in crystallization methods and synthesis parameters. The process of crystallization can be influenced by a number of factors, including temperature, viscosity, supersaturation level, cooling rate, the equipment employed, and the presence of other molecules in the solution [4]. For example, if a small amount of another sugar, such as glucose, is present in the solution, it can affect the formation of the galactose crystals by competing for the same sites on the crystal surface. In addition, impurities can decrease the nucleation and crystal growth rates and contaminate crystals, thus affecting the purity [5]. Hence, an in-depth study of these crystallization parameters is necessary to deliver high crystallinity and purity in the crystallization of galactose.

The present paper focuses on the investigation of various crystallization methods for the recovery of galactose from an industrial side stream, with the aim of shortening the overall crystallization and incubation times. The main focus is on the method of cooling crystallization by stepwise temperature decrease (CCSTD), where a hot solution concentrated by evaporation is routed to an undercooled cooling crystallizer.

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Further, various filtration and centrifugal methods were investigated to separate the crystals from the highly viscous mother liquor.

2 Experimental Section

2.1 Analysis of the Mother Liquor

2.1.1 Solubility Measurements

The studied material (an industrial side stream) contained a high concentration of glucose and a smaller quantity of galactose. Although the glucose concentration in the industrial side stream was high, the only equilibrium concentration of α-D-galactose could be reached during evaporation. Therefore, the crystalline product was mainly less soluble galactose. A Dionex 3000 ICS high-performance anion exchange (HPAE) chromatography system for quantitative carbohydrate analysis was used to determine the galactose solubility in the initial industrial side stream and the purity of the crystals obtained. Four samples were prepared at 25, 40, 60, and 80 °C and kept for 24 h with an excess of α-D-galactose crystals to study the solubility. Crystal-free samples for HPAE chromatography were obtained using a preheated syringe equipped with a microfilter.

2.1.2 Viscosity Measurements

The presence of glucose in higher amounts leads to high solution viscosity. A Brookfield DV-E viscometer was used to measure the viscosity of α-D-galactose and glucose solutions at given shear rates and temperatures. The principle of operation of the DV-E is to rotate a spindle immersed in the test fluid through a calibrated spring. The viscous drag of the fluid against the spindle is measured by the spring deflection. Spring deflection is measured with a rotary transducer which provides a torque signal. The measurement range of a DV-E (in mPa s) is determined by the rotational speed of the spindle (10–100 rpm), the size and shape of the spindle, the container in which the spindle is rotating, and the full-scale torque of the calibrated spring.

2.2 Crystallization of α-D-Galactose

In the present work, three techniques for α-**D**-galactose crystallization and their operating parameters were evaluated regarding their influence on the product crystal properties: conventional cooling crystallization (CC), CCSTD, and evaporative crystallization (EC). First, the initial solution was concentrated by evaporation. The concentrated industrial side stream solution contained 28.8 g of galactose per 100 g of solution. Fig. 1 illustrates the typical temperature profiles used in CCSTD and Supporting Information Fig. S2 shows all the cooling profiles used in the experiments.

A systematic study was carried out regarding the effect of crystallization parameters such as cooling rate, temperature, and evaporation rate in an EasyMax 402 stirred reactor work-

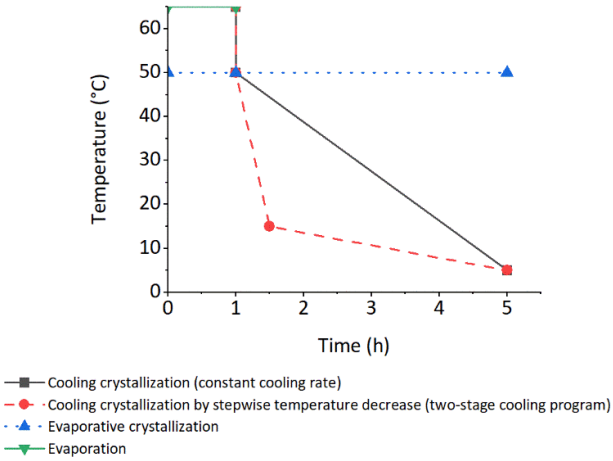

Figure 1. Crystallization methods studied with the industrial α-D-galactose solution.

station from Mettler Toledo. An upward pumping pitched-blade four-blade stirrer with a diameter of 38 mm and a mixing speed of 450 rpm (tip speed of 0.90 m s^{-1}) was used with the baffled 400-mL reactor. A schematic diagram of the experimental setup used in CC and EC is shown in Fig. 2.

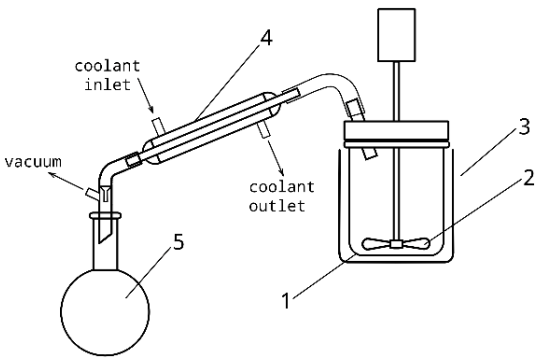

Figure 2. Experimental setup used for CC and EC studies. (1) Reactor, (2) stirrer, (3) jacket, (4) cooling system, (5) water collector.

2.2.1 CCSTD

Concentrated industrial side stream solutions were prepared by evaporation at 65 °C to study the CCSTD. Experiments were performed using different cooling rates and times. In addition, constant (from 1.7 K min^{-1} to 15 K h^{-1}) and two-stage cooling rates were investigated. The temperature range of 50 to 5 °C was studied at a chosen cooling rate and seeding policy. Once the temperature reached 48.0 °C, seed crystals were added. The mass of seed crystals of 50 µm in average crystal size was 1 wt % [6] of the theoretical crystal mass (m_{th}), which was calculated from the theoretical solubility difference between the initial galactose concentration and the solubility at 5 °C.

2.2.2 Evaporative Crystallization

The second method investigated was semi-batch EC. It was performed in a vacuum of 230 mbar at 65 °C, or a constant temperature of 50 °C and atmospheric pressure for 3–12 h. The evaporation rates were varied to change the supersaturation degree. Seeding was carried out 15 min after the beginning of the experiment to ensure that the solution was supersaturated and the pressure in the reactor had stabilized.

2.2.3 Cooling Crystallization

Initial industrial side stream solutions were concentrated by evaporation at 50 or 65 °C for the studies on batch CC. Temperature ranges of 65 to 5 °C and 50 to 5 °C were used. Cooling rates of 1.7, 5, and 10 K h^{-1} were applied. Seed crystals were added to the solution to provoke nucleation according to the same seeding policy as introduced in Sect. 2.2.1. The suspension obtained after crystallization was kept at 5 °C for 48 h to study the effect of the incubation time.

A comparative experiment on the crystallization of galactose from pure water was carried out in a similar way. In this case, the temperature was reduced from 60 to 5 °C with a cooling rate of 15 K h^{-1}.

2.3 Characterization of α-D-Galactose Crystals

2.3.1 Crystal Structure Determination

A PANanalytical X'PERT PRO MPD Alpha1 X-ray powder diffractometer was used for determining the structural properties of the crystallized materials. The diffractometer was operated in the Bragg-Brentano diffraction mode, and monochromatized Cu-Kα1 radiation ($\lambda = 1.540598$ Å) was generated with a voltage of 45 kV and a current of 40 mA. The measured 2θ angle range was 5.0–90.0°, with a step size of 0.026° and a measurement time of 51 s per step. The samples were ground gently before the measurement to minimize the sample size (preferred crystal orientation). The samples were measured on stainless-steel sample holders. It must be pointed out that the phase detection threshold of this kind of X-ray powder diffraction (XRD) measurement is limited to approximately 5 %. The measured diffractograms were analyzed using HighScore and Match! software.

2.3.2 Scanning Electron Microscopy

Scanning electron microscopy (SEM) was used to analyze the morphology, uniformity, and size of the crystals. The main advantage of SEM is the possibility to observe the solid-state topography with sufficient resolution. Measurements were carried out by using a Tabletop TM4000 SEM series from Hitachi High Technologies.

2.3.3 Crystal Size Distribution Measurements

A Malvern Mastersizer 2000 was used to analyze the crystal size distribution (CSD) of the galactose material. The device can be applied to particles in the size range of 0.5–2000 µm.

2.3.4 Crystal Purity Studies

Crystal purity studies were conducted with a Dionex 3000 ICS HPAE chromatography system. A sample of 1 g L^{-1} crystals was diluted to the required concentration of 50 mg L^{-1}, and the resulting difference in galactose content was used to determine the concentration of pure galactose in the sample.

2.3.5 Focused Beam Reflectance Measurement

During the crystallization experiments, an inline probe (Particle Track G400, Mettler Toledo) was immersed in the reactor and used to measure the count rates of various chord length size fractions to obtain kinetic data corresponding to nucleation and the crystal growth rate.

The Mettler-Toledo Particle Track G400 is based on focused beam reflectance measurement (FBRM). A laser beam passes through a set of optics and focuses on a tight beam spot in the sapphire window. The optics rotates at a fixed speed of 2 m s^{-1} to scan the flow of particles through the window. FBRM provides precise and highly sensitive chord length data collection to capture real-time changes. Before the experiments, the probe was cleaned and stabilized in advance with distilled water for zero particle counts. All the FBRM measurements were performed for chord length size fractions between 1 and 1000 µm with a time interval of 10 s.

2.4 Separation of Crystals

The separation of crystals from the mother liquor was investigated by three methods: vacuum filtration with a Buchner funnel, a basket centrifugal filter, and a benchtop centrifuge (Fig. 3). After galactose was separated by the chosen method, the crystals were washed to achieve the highest purity.

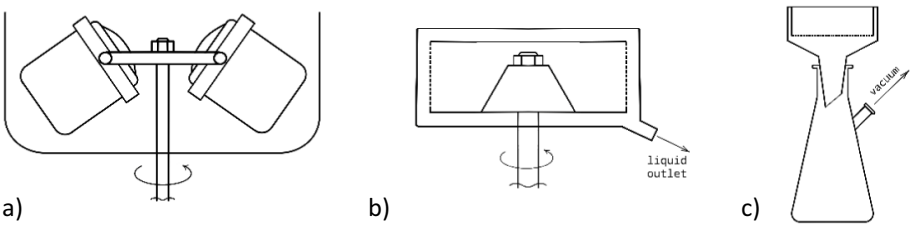

Figure 3. Separation systems: (a) benchtop centrifuge, (b) basket centrifuge, (c) Büchner funnel.

After crystallization was complete, the crystallized material was washed first with an ethanol/water solution (85:15, wt %), then with pure ethanol, and dried in an oven at 55 °C.

3 Results and Discussion

3.1 Solubility and Viscosity Measurements

The obtained solubility data of galactose is shown in Fig. 4. Compared with the pure aqueous galactose solution, the solubility of galactose in the initial industrial side stream is lower.

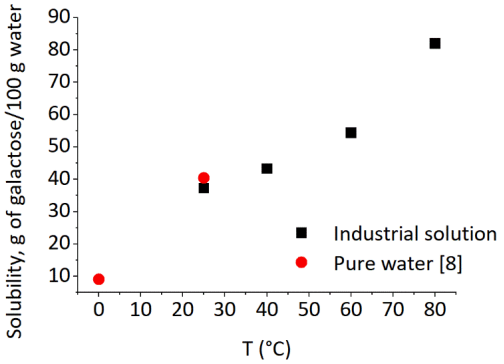

Figure 4. Solubility of galactose in the industrial side stream and in pure water [7].

One of the most significant impacts of viscosity on crystallization is its effect on the rate of nucleation and crystal growth. The results of our previous crystallization research with highly viscous xylitol solutions [8] revealed the effect of viscosity on the crystallization kinetics. In fluids with high viscosity, the mobility of atoms or molecules is hindered, making it more difficult for nuclei to form. As a result, the rate of nucleation is slower in high-viscosity fluids compared with low-viscosity fluids. Another impact of viscosity on crystallization is its effect on the rate of crystal growth. In addition to the above, viscosity can also affect the shape and size of crystals. Thus, viscosity plays a critical role in the crystallization process. High-viscosity fluids have a slower rate of nucleation and crystal growth, leading to the formation of poorly formed and asymmetrical crystals. To achieve optimal crystallization results, it is essential to take the viscosity effects into account in the crystallization process performance.

As can be seen from Fig. 5, Supporting Information Fig. S1, and Supporting Information Tabs. S1 and S2, the viscosity of the concentrated industrial side stream is significantly higher than the viscosity of the pure glucose and galactose solutions, due to the influence of admixtures, such as organic acids and macromolecules with a molecular weight of about 24 kDa. The concentrated industrial side stream containing 28.8 g of galactose per 100 g of solution was estimated to be saturated approximately at 65 °C, based on the obtained solubility data with the initial industrial side stream. The measured dynamic viscosities of pure, saturated, and undersaturated glucose solutions are shown in Supporting Information Tab. S1 and Fig. S1. This indicates that the rheology of the industrial side stream solution differs greatly due to the multi-component composition of the

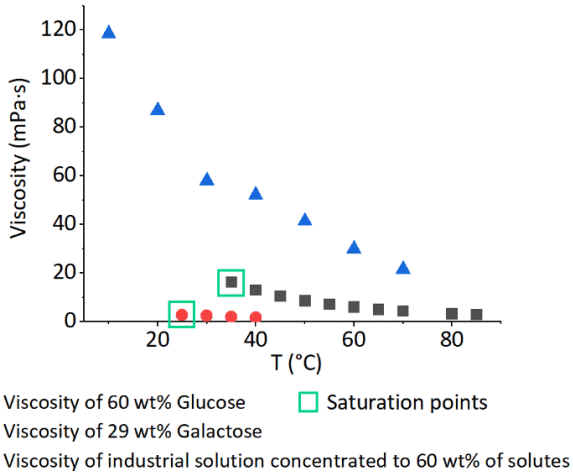

Figure 5. Measured viscosities of the pure glucose and galactose solution and the concentrated industrial side stream.

solution. In addition, the concentrated industrial side stream shows thixotropic and pseudoplastic behavior. It should be noted here that high viscosities can reduce the mass and heat transfer efficiency, as well as the crystallization rate. Thus, when selecting an appropriate temperature range for crystallization, this type of viscosity data is useful.

3.2 Crystallization Results

Galactose crystals were produced by a systematic variation of operation parameters such as the batch time, temperature, cooling rate (*CR*), and evaporation rate, resulting in crystalline products with specific crystal properties. Saturated solutions were prepared based on the measured solubility. The detailed information on the experimental conditions and results can be found in Supporting Information Tab. S3.

3.2.1 CCSTD

The highest m_{obt}/m_{th} was obtained using long crystallization (5.5–26 h) or incubation times of 24–48 h. The incubation period significantly increased the m_{obt}/m_{th} by 27 %. This observation indicates a low crystallization rate. In addition, concentrating the solution to over 65 % of solutes slows down crystallization by concentrating impurities and increasing the viscosity of the solution. The highest m_{obt}/m_{th} was achieved using CCSTD and a two-stage cooling rate, where the temperature dropped rapidly from 50 to 15 °C (in 20 min) and then from 15 to 5 °C at $CR = 2\ K\ h^{-1}$.

3.2.2 Semi-Batch EC

Although EC in 12 h at atmospheric pressure and 50 °C led to the formation of crystals, this experiment required a lot of energy and time compared to CCSTD. Therefore, no further studies of EC at atmospheric pressure were carried out. In

addition, reducing the crystallization time to 5 h or the evaporation rate by vacuum led to the formation of a gel, from which crystals could not be extracted. Moreover, a higher dynamic viscosity also affected the filterability by extending the filtration time significantly when crystals obtained by EC at 50 °C were filtered from the highly viscous mother liquor.

3.2.3 Batch CC

CC experiments were also carried out. The initial temperature was the same as the evaporation temperature used in solution concentration at the beginning. The cooling profiles used are shown in Supporting Information Fig. S2. The cooling crystallization did not result in appropriate crystal properties compared with CCSTD.

3.3 Results of Crystal Characterization

3.3.1 X-Ray Diffraction

According to the XRD analysis, the crystallized material was α-D-galactose. No peaks were observed corresponding to the glucose structure. In addition, some samples were amorphous, most likely because the cooling rate was too high to form the crystals. Supporting Information Fig. S3 shows typical X-ray diffractograms for the crystalline material, amorphous material, and pure galactose.

3.3.3 CSD Measurement Results

The CSD is an important aspect of crystallization that can affect the properties and performance of the final product. The CSD can be affected by various factors such as the nucleation rate, growth rate, dissolution rate, and the presence of impurities. Understanding and controlling these factors can lead to the production of crystals with the desired size and shape, critical for many industries such as the pharmaceuticals, food, and chemical industries. The results of the CSD measurements are shown in Fig. 6.

As can be seen from the obtained results, the experimental conditions significantly influenced the crystal sizes obtained. In this section, the results obtained on the median crystal size (d_{50}) are compared. Among the investigated methods, the CCSTD with a 20-h incubation period (no. 15) produced the smallest crystals of 61 µm in median crystal size, while the CC at 5 K min^{-1} (no. 7) and 10 K min^{-1} (no. 8) resulted in larger crystals (213 and 195 µm, respectively). The EC for 12 h (no. 6) and the CC at 1.7 K h^{-1} (no. 11) resulted in crystals with intermediate sizes of 117 and 367 µm, respectively. Notably, the CCSTD without incubation (no. 10) yielded the largest crystals (445 µm).

The inclusion of an incubation period in the CCSTD process (no. 15) led to a significant reduction in crystal size compared to the CCSTD without incubation (no. 10). This observation suggests that incubation promotes nucleation and growth, leading to the formation of smaller crystals.

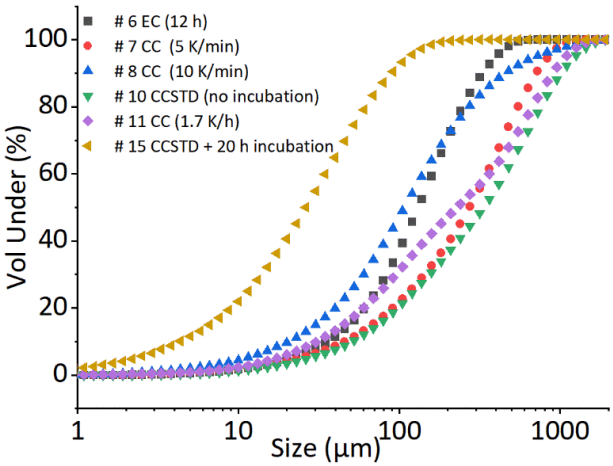

Figure 6. Cumulative particle size distribution curves obtained by laser diffraction analysis.

The cooling rate during the CC experiments (nos. 7 and 8) had a noticeable impact on the crystal size. Faster cooling (10 K min^{-1}) resulted in slightly smaller crystals compared to the lower cooling rate (5 K min^{-1}). This finding indicates that a higher cooling rate leads to smaller particle sizes.

Comparing the crystal sizes obtained, it can be inferred that CCSTD with incubation (no. 15) is the most effective method for producing smaller crystals. However, if the aim is to achieve larger crystal sizes, CC at 5 K min^{-1} (no. 7) or CC at 10 K min^{-1} (no. 8) could be more suitable. These conclusions highlight the importance of selecting the appropriate crystallization method depending on the desired crystal size range.

In summary, the presented data demonstrates the influence of different experimental conditions on the crystal sizes obtained. The findings emphasize the significance of incubation, cooling rate, and the choice of crystallization method in controlling the crystal size. These insights contribute to the understanding of crystallization processes and can aid in the optimization of industrial crystallization operations.

3.3.3 SEM Results

In the present work, SEM was used to study the morphology of crystals. Fig. 7 shows the difference between D-galactose crystallized from the industrial side stream and from pure water.

Based on the SEM image in Fig. 7a, D-galactose crystals obtained from the industrial solution form agglomerates that are much larger than the actual single-crystal size. Thus, the actual size of the D-galactose crystals crystallized from the industrial side stream is about 60 µm. The presence of impurities or other substances in the solution can contribute to the formation of agglomerates. These impurities can act as nucleation sites, leading to the formation of additional crystals. These additional crystals can then stick together with the existing crystals, resulting in agglomerate formation. The high viscosity of the solution can also contribute to the formation of agglomerates. High viscosity slows down the movement of crystals, which can cause them to stick together.

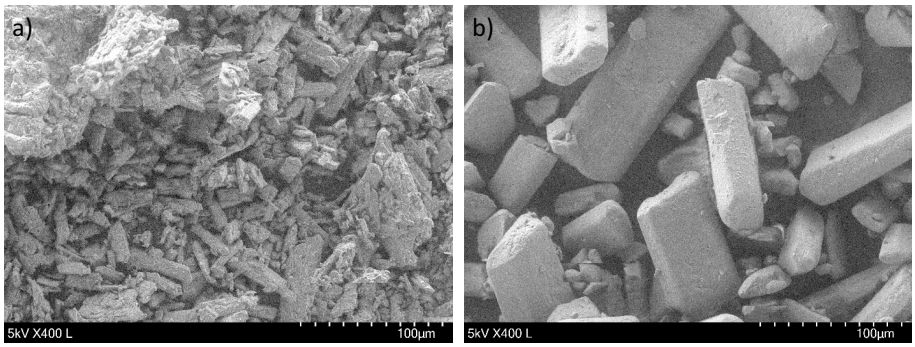

Figure 7. Typical SEM micrographs of (a) D-galactose crystallized from the industrial side stream using fast cooling from 50 to 15 °C and from 15 to 5 °C with a cooling rate of 2 K h^{-1}, and a total batch time of 5.3 h (experiment no. 10) (400-fold magnification, scale bar 100 µm), and (b) pure water (400-fold magnification, scale bar 100 µm).

3.3.4 FBRM Results

FBRM was used to monitor real-time changes in the count rates of various chord length fractions and to collect the kinetic trends corresponding to nucleation and crystal growth rate. In addition, the impact of the incubation time was analyzed with this technique. Fig. 8 illustrates typical FBRM results of CCSTD.

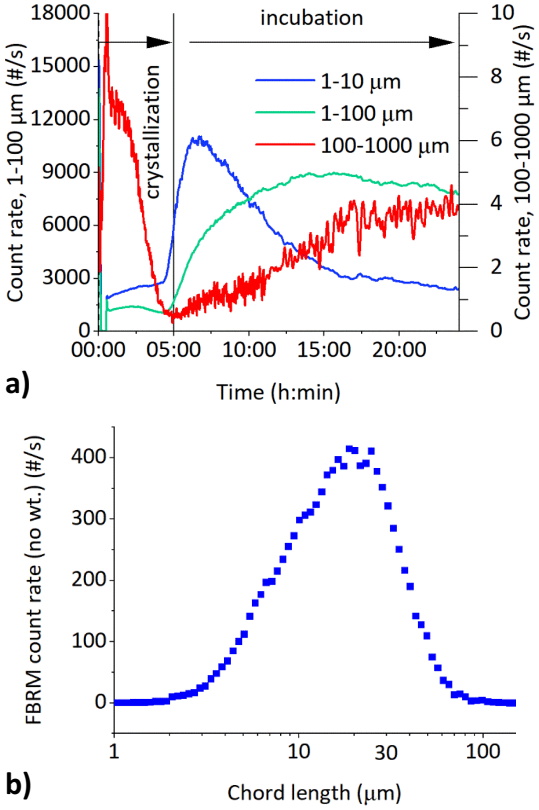

Figure 8. FBRM result of CCSTD with a two-stage cooling program and 20 h of incubation. (a) FBRM graph and (b) final unweighted chord length distribution.

As can be seen from Fig. 8, the incubation time increased the count rate of the largest chord length fraction. However, based on the results obtained, the incubation period can be shortened to 10 h since there is no significant effect of incubation beyond this time.

3.4 Separation Results and Crystal Purity Determination

Usually, a basket centrifugal filter is preferable for crystal separation compared with a Büchner funnel and benchtop centrifuge since it usually provides a final product of lower moisture content. However, due to the low quantities, small crystal size, and high viscosity of the mother liquor, a basket centrifuge was not used much in the present work. In addition, in most cases, a Büchner funnel could not be used due to the low crystallization temperature and high viscosity. In this case, since the filtration rate was low, the crystals dissolved before they had time to be filtered. Thus, a benchtop refrigerated centrifuge was chosen as the main separation method. The results of crystal purity, analyzed by HPAE chromatography, are summarized in Tab. 1.

Table 1. Results of crystal purity studies.

Exp. no.	Separation method	g-force	Glucose [wt %]	Galactose [wt %]
3	Basket centrifugal filter, 6000 rpm	2866×*g*	15	85
5	Basket centrifugal filter, 6000 rpm	2866×*g*	12	88
7	Benchtop centrifuge, 3500 rpm	1424×*g*	27	73
8	Benchtop centrifuge, 3500 rpm	1424×*g*	25	75
10	Benchtop centrifuge, 13 500 rpm + washing	21 187×*g*	1	99
11	Benchtop centrifuge, 13 500 rpm + washing	21 187×*g*	4	96

The results of the HPAE chromatography analysis showed that the crystal separation method and washing can ensure high crystal purity (up to 75–99 %). The target level for galactose purity was 90 %.

4 Conclusions

According to the XRD and HPAE chromatography analyses, crystallization of a glucose-galactose industrial side stream yielded α-D-galactose. The highest m_{obt}/m_{th} of 88 % and crystal purity of 99 % were achieved by CCSTD and by adjusting the filtration and cake washing steps. Thus, the resulting crystal purity and yield were higher than the purity and yield reported previously in patents and scientific publications, while the size of the crystals was smaller (about 60 μm).

Moreover, it was proved that galactose crystallization from a highly viscous process solution is a slow process that proceeds successfully at a cooling rate of less than 5 K min^{-1}. The viscosity played a key role in the crystallization kinetics. The viscosity measurements showed that the viscosity of the concentrated industrial side stream was significantly higher than the viscosity of pure glucose and galactose solutions. In addition, the concentrated industrial side stream demonstrated thixotropic and pseudoplastic behavior.

It should be mentioned here that FBRM analysis showed that sufficient incubation duration increased the crystal yield. Nevertheless, based on the results obtained, the incubation period can be shortened to 10 h since there is no significant effect of incubation beyond this time.

Preliminary studies of glucose-galactose side stream crystallization have shown promising results and the need for further investigation.

Supporting Information

Supporting Information for this article can be found under DOI: https://doi.org/10.1002/ceat.202300067.

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The authors have declared no conflict of interest.

Symbols used

CR	[K h^{-1}]	cooling rate
d_{50}	[μm]	median crystal size
m_{obt}	[g]	obtained crystal mass
m_{th}	[g]	theoretical crystal mass

Abbreviations

CC	cooling crystallization
CCSTD	cooling crystallization by stepwise temperature decrease
CSD	crystal size distribution
EC	evaporative crystallization
FBRM	focused beam reflectance measurement
HPAE	high-performance anion exchange
SEM	scanning electron microscopy
XRD	X-ray powder diffraction

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