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# Influence of dispersion of a new type whole crystals seed on crystallization process and sugar quality

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## ABSTRACT

The aim of the work was to study effectiveness of the influence of a new type crystal seed with different microcrystals dispersion on the process of sucrose crystallization and sugar quality. The dispersion of microcrystals of the crystal-forming agent with whole crystals was determined and curves of their differential distribution were presented, which showed the average particle sizes of the "Ester K 01" are 18.2 µm and 46.4 µm. The morphological characteristics were determined of particles of the whole crystals seed and the seed slurry produced at a sugar factory. The duration of massecuite boiling was investigated and a comparative characterization of the quality indicators of the resulting massecuite, mother liquor, and sugar was given depending on the use of the whole crystals seed with different dispersion of microcrystals in comparison with the factory slurry. Microphotographs of sugar crystals in the massecuite were obtained with their size recorded 5, 10 and 20 min after the input of various seeds. The morphology of sugar crystals was studied in the finished massecuite. A comparison is given of the granulometric composition of sugar crystals obtained with crystal-forming agents with different dispersion. It was found based on the results of the study that when using the "Ester K 01" (46.4 µm) in thick juice with high turbidity the corresponding crystals are formed faster in the total amount of the massecuite. It was also found that there is a kind of blocking of the process of adjacent growth of crystals from the particles of impurities of thick juice with high turbidity. This contributes to the formation of more uniform, homogeneous crystals of the correct shape. The results are presented in the article of tests of the whole crystal seed "Ester K 01" (18.2 µm) and "Ester K 01" (46.4 µm) at sugar factories using the method of working on the seed magma and the method of shock crystallization respectively. The dependences were obtained of the average linear size, the amount of sugar of the 0.65 mm fraction, the coefficient of variation and the coefficient of uniformity of sugar crystals on the consumption of the whole crystals seed "Ester K 01" (crystal size 46.4 µm). The regression equation was developed in natural form for the process of sucrose shock crystallization. It was established the rational consumption of the crystal-forming agent "Ester K 01" (crystal size 46.4 µm), which ensures the effective crystallization of sucrose for the process of crystallization by the shock induced method.

## 1. Introduction

The timely introduction of crystals and the cessation of their formation have the vital importance for the process of massecuite (boiled syrup consisting of sugar crystals and inter-crystalline solution) boiling and obtaining high-quality finished massecuite in the sugar industry. The more crystallization centers are formed at the time of introduction, the smaller the sugar crystals will be, and vice versa, the fewer crystallization centers are introduced, the larger are the crystals in the finished massecuite [1-11]. The crystallization centers are created by bringing the sugar solution to a labile state (degree of supersaturation 1.25...1.3), followed by the introduction of a crystal-forming agent

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Received 17 June 2024; Received in revised form 27 September 2024; Accepted 30 September 2024 Available online 2 October 2024 0022-0248/© 2024 Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies. (seed), due to which new crystallization centers are formed. Then the solution is transferred to the second stage, for which the degree of supersaturation is reduced to metastability (1.1...1.2). At this stage, only crystal growth occurs without the formation of new crystallization centers. This is designed to produce a high quality massecuite with homogeneous crystals.

High-quality crystal forming agent is required to ensure a uniform granulometric composition of sugar, reduce the content of conglomerates and minimize the formation of secondary crystallization centers throughout the entire massecuite boiling cycle [1,5,7,9,10,12].

Crystallizing agents are recognized as a crystal particles or even small crystals of the same substance that are added to a supersaturated solution. Each of the crystal forming particles is a crystallization center.

Crushed powdered sugar, slurries and pastes are used as a crystal seeds [13–22]. Many factories use suspensions that are prepared independently by grinding sugar in a laboratory ball mill in a liquid medium that does not dissolve sugar. Alcohol is most often used as a dispersing medium, and the size of sugar microcrystals in 1 ml is 5...50  $\mu$ m [13–21], which leads to their rapid stratification.

The experience of sugar factories has shown that the content of conglomerates in white sugar can be reduced by 40–50 % by using highquality crystal-forming agents with high crystal uniformity, surfactants and mechanical circulation in vacuum pans [12].

It is extremely important to use the seeds with properly formed sugar crystals, without twin crystals and clusters [9,7,12]. Heterogeneity of crushed crystals leads only to uneven formation of crystallization centers in the vacuum pans, that is extremely undesirable. Also it is necessary to work with the lowest possible concentration of the crystal seed in a properly organized crystallization process in order to form homogeneous sugar crystals without conglomerates [9].

Earlier a team of authors [23] has already proved the effectiveness of using the whole crystals seed "Ester K 01" (SPE "Electrogazokhim", Ukraine) to obtain uniform, homogeneous sugar crystals by the method of shock crystallization (the formation of sugar crystals. This occurs when the solution reaches a certain degree of supersaturation, by the introduction of crystallization centers (sugar crystals)). This crystal forming agent is a suspension of homogeneous sugar crystals with an average size of about 10  $\mu$ m, obtained by precipitation from a supersaturated sugar solution and stabilized with a specially selected food surfactant. It's use reduces the duration of massecuite boiling by 15–20 min, increases the volume of crystals in the massecuite that allows to obtain sugar with a high percentage of uniform, homogeneous crystals with a minimum content of fines and powder [23].

Recently factories have been using seed magma (syrup containing microcrystals of sugar and used to form the bulk of sugar crystals) for crystallization during the boiling of the A product massecuite [18,19,24–28]. The purpose of seed magma use is to create a specially prepared charge with seed crystals of uniform shape without conglomerates, using auxiliary equipment. This is difficult to achieve under normal conditions of boiling the seed magma on powdered sugar. For example, when fine crystallization centers are less than 10 µm, they go through a critical growth phase, so they can either dissolve or cause spontaneous growth and formation of new crystallization centers [1,9,18,25,28]. With the size of the crystals of the starting material from 20 µm, the process of spontaneous dissolution of crystals can be completely excluded, since the crystallization centers introduced into the boiled syrup already have a sufficiently large surface area, which significantly intensifies the process of growing the resulting crystals [9,18,25,28-30].

The occurrence of additional crystallization centers (loss of "flour") and the formation of conglomerates can be avoided only by introducing a seed with a uniform crystal size and precise dosing of thick juice (syrup used to make massecuite) input into the metastable boiling zone [19]. That is why the use of the crystal forming agent "Ester K 01" will be very appropriate in this process.

Specialists of SPE "Electrogasokhim" provide the possibility of

manufacturing a whole crystals crystal-forming agent with different microcrystal sizes. Therefore, it is interesting to investigate the effect of different microcrystal sizes of whole crystals seed on the process of crystal formation and the quality of finished products during boiling on the seed magma.

## 2. Materials and methods

Experimental studies were carried out in the technical laboratory of the Research and Production Enterprise Electrogazokhim (Kyiv, Ukraine) and in the laboratory of the Department of Sugar Technology, Sugar-containing Products and Ingredients of the Institute of Food Resources of the National Academy of Agrarien Sciences of Ukraine (Kyiv, Ukraine).

Production tests were conducted in the conditions of sugar factories: LLC "Narkevytskyi Sugar Factory", Ukraine; PJSC "Salyvonkivskyi Sugar Factory ", Ukraine.

Crystallization process at Narkevytsky Sugar Factory LLC is fully automated and is carried out using the method of boiling the seed magma (Braunschweig method). One uses in the process filtered thick juice, the initial characteristics of which are shown in Table 1. The consumption of the factory slurry for boiling the seed magma is 1.7 dm<sup>3</sup> per vacuum pan with a capacity of 50 tons, and the amount of crystal-forming agent "Ester K 01" No.1 was taken 10 % less than the amount of factory slurry that is 1.5 dm<sup>3</sup>. The reduced amount of the crystal-forming agent is because the average size of its particles is more than 10  $\mu$ m and thus the dissolution and loss of active crystallization centers can be excluded [1,9,18,25,28].

Massecuite boiling was carried out according to the current settings of the control program under the same conditions for the studied ingredients. The thick juice for the seed magma was thickened in a separate vacuum pan to Brix = 75 %, then it was sent to a crystallizer-cooler, where the degree of cooling can be adjusted. When the degree of supersaturation reaches 1.1 at a temperature of 50 °C, the thick juice was added to the test samples of the seed slurries and the crystal base was built up to an approximate particle size of 100...130  $\mu$ m. The resulting crystal base is again sent to the vacuum pan, where the crystal size increases to 200  $\mu$ m. The total duration of the seed magma boiling is about 3.0 h.

The finished seed magma in the amount of 4.5 tons is fed through closed pipelines into a 60-ton vacuum pan to produce the main massecuite. The duration of massecuite boiling in the main vacuum pan is 2.5 h.

The process of seeding start at PJSC "Salyvonkivsky Sugar Plant" was carried out by the method of shock crystallization (degree of supersaturation 1.2...1.3). Different amounts of crystal-forming agent were used for the process in order to establish the rational amount:  $10 \text{ cm}^3$ ,  $20 \text{ cm}^3$ ,  $30 \text{ cm}^3$  per vacuum pan with a capacity of 60 tons that corresponds to an amount of 0.2 cm<sup>3</sup>,  $0.3 \text{ cm}^3$ ,  $0.5 \text{ cm}^3$  per 1 t of finished massecuite.

Standard procedures were followed in all investigations of the qualitative and quantitative indices of products and semi-products [31]. The following штієктьутеі were used in the laboratory research: refractometer Abbemat (Co. Anton Paar, Austria) was used to determine the solids content; saccharimeter MPP 5300/5500 Sucromat (Co. Anton

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The sugar thick	i juice's quality	parameters.
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Parameter	Value
Brix, %	$64.4\pm0.1$
Sucrose content, %	$61.9\pm0.1$
Purity, %	$96.00\pm0.01$
Colour, ICUMSA units	$968\pm3$
Turbidity, mg/dm <sup>3</sup>	$84.6 \pm 0.5$
Calcium salts, %	$0.036\pm0.003$
Ash content, %	$0.141\pm0.002$
Concentration of hydrogen ions (pH <sub>20</sub> )	$\textbf{7.6} \pm \textbf{0.1}$

Paar, Austria) was used to determine the sucrose content; colorimeter KFK-3–01 (Co. ZOMZ, Eastern Europe) was used to determine color; turbidity meter AQ4500 (Co. Orion, USA) was used to measure turbidity; laboratory ionometer Temp AD8000 (Co. ADWA, Hungary) was used to determine pH value; conductivity meter OK 102/1 (Co. Radelkis, Hungary) was used to determine the ash content; and analytical balance AS 220.R2 (Co. RADWAG, Poland) was used to weigh the analysis samples. Determination of the granulometric composition of the obtained sugar samples was carried out according to the Butler method described in [31].

## 2.1. Objects of research

The objects of research were the following: crystal-forming agents intended for use in sugar production; semi-products and products of sugar production: thick juice, massecuite, mother liquor (run-off syrup remaining after the sugar crystals are separated from the massecuite), sugar crystals in massecuite, finished sugar obtained in laboratory and production conditions.

For the research two prototypes of crystal forming agents "Ester K 01" No. 1 and "Ester K 01" No. 2 were manufactured at production facilities of SPE "Electrogasokhim", using a special technology for precipitation of microcrystals from supersaturated solution and their stabilization with a specially selected food surfactant. We used as a control sample factory slurry (Narkevytsky Sugar Factory LLC), which was obtained at sugar factory by grinding sugar in a ball mill in the presence of isopropyl alcohol as a stabilizer.

## 2.2. Investigation of the dispersion of the seed particles

The crystal-forming agents' particle size (dispersion) was ascertained by means of laser diffractometry. Diffraction laser analyzer SALD-201 V (Co. SHIMADZU, Japan) was used for the measurements. The light source was a semiconductor laser with a wavelength of 670 nm. Wing SALD II-201 V V3.3 software (Co. SHIMADZU, Japan) was used to process correlation functions.

At first crystallizing agent samples were manually shaken, then mixed with a magnetic stirrer for ten minutes and then placed in an ultrasonic stirrer for ten minutes in order to prepare them for measurement. A thoroughly blended sample was used for the measurements. One hundred milliliters of isopropyl alcohol were employed as a solvent.

## 2.3. Microscopy of seed particles and sugar crystals in massecuite

The GENETIC PRO MONO (A) microscope (Co. Delta Optical, Poland) was used to examine the samples under the microscope in compliance with the operating instructions [3,32]. A digital camera MCMOS 5100 5.1MP USB2.0 (Co. SIGETA, Ukraine) was used to record micrographs. ToupView V3.7 was the program used for handling microphotography. (ToupTek Photonics Co., Ltd., Hangzhou, China).

## 2.4. Production of thick juice

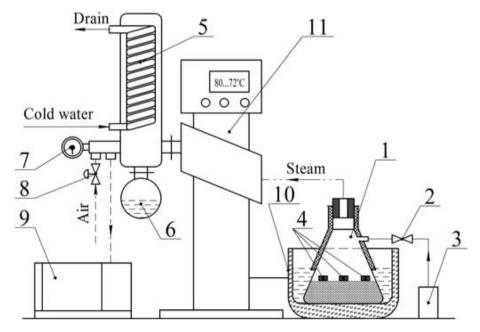
Thick juice from Ukrainian sugar factory (LLC Narkevytsky Sugar Factory) was used for all laboratory tests involving crystal-forming agents.

In Table 1 are shown quality values of the factory thick juice samples.

## 2.5. Process of massecuite boiling and sugar obtaining

Prepared thick juice samples in the amount of 1 kg were used to obtain massecuite. It was boiled according to the method of preparation of seed magma (Regensburg method) in a pilot plant [23], which was developed on the basis of the RE-2000A vacuum rotary evaporator (Shanghai ya rond biochemistry instrument factory, China) (Fig. 1).

Vacuum boiling vessel 1 is equipped with thermal insulation to avoid condensation of water vapor. There is a need in the process of massecuite boiling sometimes for the so-called "pumping" of fresh thick juice, i.e. the addition of fresh portions of the thick juice to the container using for this purpose crane 2. The pumping of the thick juice occurs under the action of vacuum from the container 3. The porous elements 4 are used to intensify the boiling process of the thick juice under vacuum. Water vapor from a vacuum tank enters the vacuum cooler 5 where it is condensed and condensate is collected in a flask 6. Vacuum is created by a vacuum pump 9. The depth of vacuum is regulated with the help of a reducer 8. Vacuum meter 7 is used for indication. Constant required



**Fig. 1.** Schematic of a pilot plant for studying the effect of seeding materials on the crystal formation process: 1 - vacuum boiling vessel with thermal insulation; <math>2 - tap for "pumping" thick juice; 3 - vessel for "pumping" thick juice; 4 - porous boiling intensifiers; <math>5 - vacuum cooler; 6 - vacuum receiving flask for condensate; <math>7 - vacuum meter; 8 - reducer for adjusting the vacuum depth; 9 - vacuum pump; 10 - water bath with automatic temperature control; <math>11 - housing with control unit.

temperature in a container with a solution of thick juice is mainteined with the help of water bath 10 with thermostat in automatic mode and the control unit 11.

Bunzen flask 1 is used as a container for crystallization of massecuite because it is made of vacuum glass and has a pipe for connecting the tap 2 and a system for pumping fresh thick juice. It is provided operation of the installation with Bunzen flasks with a volume of 0.5 dm<sup>3</sup> to 3 dm<sup>3</sup>.

## 2.5.1. Massecuite boiling

Tested massecuite samples were boiled under a vacuum. Firstly, 20–30 min the thick juice was evaporated under pressure P = -60 kPa, then the pressure was decreased up to -90 kPa. The temperature of the boiling process was maintained within 75 ... 80 °C. To reduce foaming and improve the process of boiling antifoaming agent "Esterin A 08" (SPE "Electrogasokhim", Ukraine) was added to massecuite in the recommended by manufacturer concentration (5 mg per 1 kg of massecuite).

A degree of supersaturation  $\text{Co} \ge 1,2-1,3$  is indicated by the thick juice content of dry matter reaching up to 80–82 % during the first 20 min of boiling at above mentioned conditions.

The crystallizing agents were added after reaching the degree of supersaturation in the following ratio: factory suspension (at the rate of 1,5 dm<sup>3</sup> per 50 tons of thick juice), «Ester K 01» crystal former No.1, No.2 (at the rate of 0,5 dm<sup>3</sup> per 50 tons of thick juice). Per 1 kg of thick juice such concentration will correspond to: factory suspension  $-30 \mu$ l; "Ester K 01″ No.1, No.2—10  $\mu$ l. The introduction of slurries in the form of suspensions was performed by a special micropipette.

Then crystallization was carried out by intensive cooling of the thick juice from 80 to  $72^{\circ}$  C at a cooling rate of  $0.25^{\circ}$  C/min using the appropriate program on the pilot unit (duration of about 30 min). After cooling crystallization the boiling of the massecuite and the increase of sugar crystals with two-time input of thick juice, which is required to reduce the degree of supersaturation and avoid the formation of a large number of new centers of crystallization and to promote the growth of existing crystals.

The end of crystallization was determined by the number of crystal seed. Periodically samples of massecuite were taken on glass and the number of crystals formed was observed. To obtain crystals of medium size (it is necessary that 1 mm of glass length has 5–6 crystal seeds).

## 2.5.2. Separation of massecuite

Massecuite was separated to extract sugar using a laboratory vacuum filtering apparatus that included a Bunsen flask, a Buchner nozzle with a filtration baffle, and a membrane vacuum pump N035AN.18 (Co. KNF, Germany). After that sugar samples were taken and cleaned using a 70 % ethyl alcohol solution.

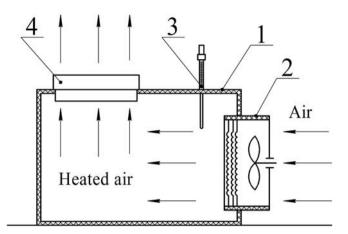
## 2.5.3. Sugar drying

The resulting sugar samples were dried at temperatures of 45–50  $^{\circ}$ C on a laboratory sieve in a specially designed installation, the scheme of which is shown in Fig. 2.

The calorifer with the fan heats the air entering the housing 1 to a set temperature. The heated air passes through a layer of sugar, which is placed on a sieve 4. The constant temperature in the installation is maintained with the help of a thermal contact thermometer 3. Sugar was stirred manually during drying. The flow rate is regulated by the speed of the fan, which is selected visually starting from the minimum, so that the sugar crystals on the sieve begin to rise slightly but do not blow away.

## 2.6. Statistical analysis

The results were expressed as mean  $\pm$  standard error. All experimental measurements were performed in triplicate. The optimization of the experimental data was carried out using the Harrington method, the purpose of which is to combine local criteria into a generalized



**Fig. 2.** Scheme of sugar drying installation: 1 - installation body; 2 - calorifer with a fan; 3 - thermal contact thermometer; 4 - laboratory sieve.

optimization criterion. The data were analyzed using the statistical software Mathcad Professional 2007. V. 14.0 (PTC, USA).

### 3. Results and discussion

## 3.1. Diagnosis of crystal-forming agents

Crystal-forming agents were evaluated to assess the preliminary qualitative properties. It was discovered that the regularity and production rate of sugar crystals are influenced by the size of their crystals [15,33,34]. The outcomes are shown in Fig. 3.

The results of the analysis of dispersed composition of the particles of the factory suspension (Fig. 3, A) showed that the average size of the particles is 4.1  $\mu$ m and distribution is uneven, bimodal with modes 0.7  $\mu$ m and 8.7  $\mu$ m. Also there are particles in the suspension with a size of less than 0.5  $\mu$ m and a maximum size of 14.4  $\mu$ m.

From the analysis of the particles size distribution of the crystalforming agent "Ester K 01" No.1 (Fig. 3, B) we can say that the distribution is in fact homogeneous, the average size of particles is 18.2  $\mu$ m with the mode of the peak 17.9  $\mu$ m. There are also particles with the smallest size of 4.2  $\mu$ m in the amount of 0.123 % by volume and a maximum size of 53.2  $\mu$ m in the amount of 0.333 % by volume.

The dispersive distribution of particles of the crystalline "Ester K 01" No.2 (Fig. 3, C) showed that the average size of the particles in the crystal-forming agent is 46.4  $\mu$ m. The distribution is uniform, with a peak mode in 66.1  $\mu$ m. The crystal-forming agent contains particles with the smallest diameter of 14.4  $\mu$ m in the amount of 0.125 % by volume and the highest 82.2  $\mu$ m in the amount of 5.1 % by volume.

Also very important is the morphology (form) of crystals, they should be transparent, have the right syngonia (a set of elements of crystal symmetry) inherent for sucrose, clear faces without damage and foreign inclusions [1,3,9,13,15,18,33,35,36]. The results of morphological studies of crystalline particles are presented in Fig. 4.

Factory suspension (Fig. 4, A) has a destroyed symmetry of crystals, which causes irregular shape particles. The particle of the seed is very small, which is a consequence of repeated long-term grinding of sugar crystals in the ball mill for the preparation of the suspension. Due to the very small size of particles they form associations that include more than a dozen particles even in the presence of a special stabilizer.

The "Ester K 01" No.1 (Fig. 4, B) contains solid crystals of the correct form inherent for sucrose. The technology of obtaining them is associated with the crystallization of microcrystals from the supersaturated sugar solution of high purity, which promotes the formation of uniform, homogeneous crystals without signs of surface destruction.

Particles of "Ester K 01'' No.1 (Fig. 4, C) are also obtained by crystallizing them from a supersaturated sugar solution of high purity, so

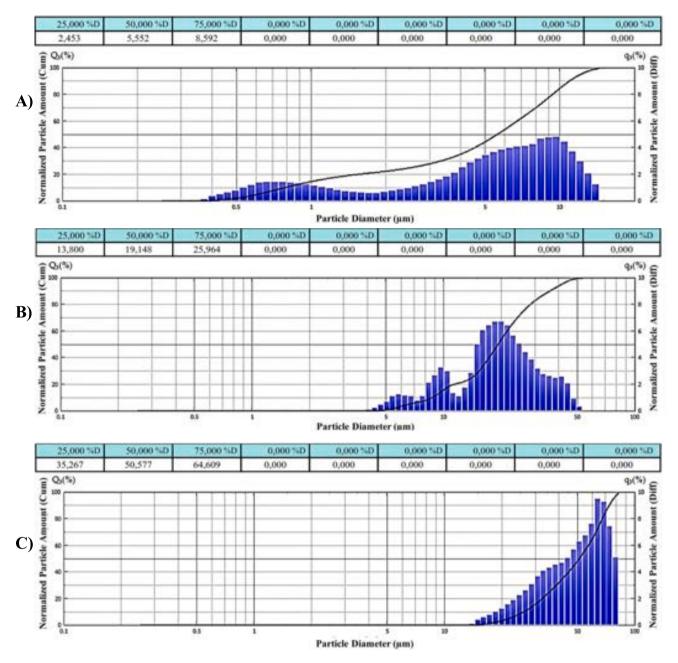


Fig. 3. Differential distribution of crystallizing agent particle sizes: A) standard factory slurry; B) crystal-forming agent «Ester K 01» No.1; C) crystal-forming agent «Ester K 01» No.2.

they are uniform, homogeneous and preserve the integrity of the sucrose crystal.

Thus, it is possible to conclude from the analysis of microphotos that, unlike the factory suspension, the particles of the crystal-forming agents "Ester K 01" No.1, No.2 are whole uniform crystals of the correct shape.

## 3.2. The duration of massecuite boiling

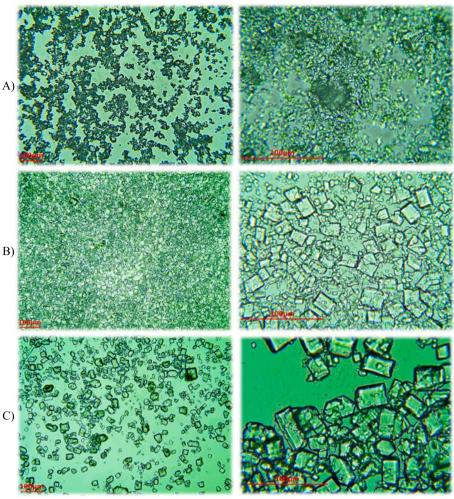
In Table 2 are presented the massecuite boiling duration data for various crystal-forming agents.

The analysis of the experimental (Table 2) showed that the duration of boiling of the massecuite on the crystal seed "Ester K 01" No.1 and "Ester K 01" No.2 is slightly lower than the duration of boiling on the factory suspension but close to it.

## 3.3. Morphological studies of sugar crystals

In order to evaluate the process of growth of crystals, morphological studies of sugar crystals in massecuite were conducted at different intervals after the beginning of the phase of their growth. Samples of massecuite for research were taken 5 min, 10 min and 20 min after the crystal-forming agent input. The finished massecuite was analyzed also. It is shown in Fig. 5 the morphological analysis of sugar crystals in massecuite that were obtained with different crystal-forming agents along with their corresponding sizes.

In the first five minutes after the beginning of the build-up, we observe the formation of uniform crystals of sugar on the crystal-forming agent "Ester K 01" No.2, the size of which is mainly 0.10-0.12 mm (37 crystals) (Fig. 5, C). "Ester K 01" No.1 crystal-forming agent provides the formation of sugar crystals with slightly worse uniformity. In the system and the main number of crystals is with a size of 0.06-0.10 mm (48



10x magnification

40x magnification

Fig. 4. Microphotos of seed particles: A) Factory suspension in its own stabilizer: A) "Ester K 01" No.1 in its own stabilizer; C) "Ester K 01" No.2 in its own stabilizer.

Table 2The duration of the massecuite boiling.

Crystallizing agent	Duration of massecuite boiling, min
Factory suspension	65 75
«Ester K 01» No1 crystal-forming agent	60 65
«Ester K 01» No.2 crystal-forming agent	60 65

crystals) (Fig. 5, B). Multiple sugar crystals are formed on the factory suspension. Crystals are uneven in size, the main number of crystals has size from 0.04–0.14 mm (101 crystals), conglomerates are presented (Fig. 5, A).

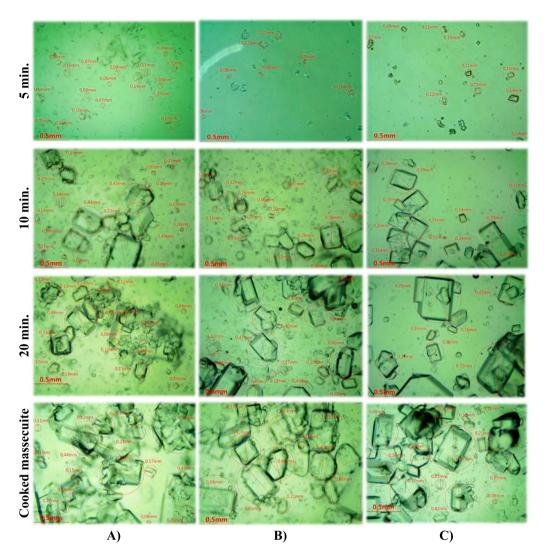
Ten minutes after the start of the crystals growth phase they are uniform, homogeneous in case of using crystal-forming agent "Ester K  $01^{\prime\prime}$  No. (Fig. 5, C). The main number of crystals has size 0.31-0.39 mm (23 crystals), as well as there are single crystals with a size of 0.14 and 0.24 mm. Less uniform crystals grow on the crystal-forming agent "Ester K  $01^{\prime\prime}$  No.1. The size of the bulk of crystals is 0.20-0.40 mm (21 crystals), there is also a number of small crystals with a size of 0.06-0.14 mm (Fig. 5, B). Factory suspension gives sugar crystals uneven in size Fig. 5, A), some individual crystals reach 0.5–0.65 mm in the presence of the main number of crystals from 0.07-0.43 mm (29 crystals). There are single conglomerates with size 0.27 and 0.33 mm.

In twenty minutes the bulk of uniform sugar crystals with a size of 0.5-0.72 mm (6 crystals) were obtained using the crystal-forming agent "Ester K  $01^{"}$  No.2 (Fig. 5, C)., with the single crystals of 0.16 and 0.3 mm.

Most sugar crystals obtained with "Ester K  $01^{"}$  No.1 are slightly smaller in size 0.3–0.5 mm (14 crystals), but also uniform, although small crystals with a size of 0.11 to 0.23 mm are also presented in the massecuite (Fig. 5, B). Sugar crystals obtained on factory suspensions have (Fig. 5, A) uneven dimensions of 0.08–0.5 mm (122 crystals); a significant number of small crystals 0.08–0.12 mm, which are also combined in conglomerates with a size of 0.33–0.40 mm. There are also single crystals of 0.9 mm.

In the finished massecuite the most uniform crystals with a size of 0.5-0.7 mm (9 crystals) and 0.2-0.3 mm (17 crystals) are formed by the use of the crystal-forming agent "Ester K 01'' No.2 (Fig. 5, C). Sugar crystals obtained with "Ester K 01'' No.1 (Fig. 5, B) also have uniform dimensions -0.4-0.6 mm (12 crystals), there are single crystals of 0.7 and 0.9 mm and small crystals 0.11 mm. Although the crystals obtained with Ester K 01 No.1 are uniform in size, most of them have irregular shape. False crystals are present also [9]. Massecuite obtained on the factory suspension contains uneven crystals with a size of 0.20-0.60 mm (21 crystals) and conglomerates 0.3-0.8 mm (9 crystals), formed from crystals of 0.1-0.20 mm in size (Fig. 5, A).

The formation of crystals of irregular shape and false crystals on the crystal-forming agent "Ester K 01" No.1 (Fig. 5, B) and conglomerates on the factory suspension (Fig. 5, A) can be associated with significant turbidity of the original thick juice  $-84.6 \text{ mg/dm}^3$  (Table 1), which is 2.8–3.4 times higher than the recommended normative parameter (25–30 mg/dm<sup>3</sup>) [37]. Such thick juice may contain a significant amount of impurities [3,39–44]. In particular, calcium-containing



**Fig. 5.** Microphotographs of sugar crystals in the massecuite after 5, 10, 20 min of input of crystal-forming agents and microphotograph of boiled massecuite (10x magnification): A) a standard factory suspension; B) the crystal-forming agent "Ester K 01" No.1; C) the crystal-forming agent "Ester K 01" No.2.

compounds with sucrose (calcium sugars, etc.), which according to [38] have a ratio of 1:4 in the structure of the "calcium: sucrose" complex. These compounds can also be nuclei for crystallization or be included in the structure of a newly formed sugar crystal [3,5,9,16,24,29,39–44]. The high turbidity of the thick juice is usually caused by poor main and after filtration of the juice of the second carbonization and filtration of thick juice after the evaporation station. The size of the particles of impurities of non-filtrated juice of the second carbonization is  $5...16 \,\mu\text{m}$  [45] and will actually correspond to the size of the particles of impurities in unsatisfactory filtered thick juice.

So the use of crystalline centers of agent "Ester K 01" No.1 with an average particle size of 18.2  $\mu m$  (Fig. 3, B), which is somewhat adjacent to the size of impurities in thick juice [45] will lead to almost simultaneous nucleation of crystals, both from the impurities of the thick juice and from microcrystals. It is obvious that such adjacent crystal formation can lead to irregular shape crystals with foreign inclusions (Fig. 5, B).

The factory suspension has a particle size in the range of 0.3...14.4 µm, the average size of 4.1 µm (Fig. 3, A), which is also adjacent to the size of the particles of thick juice impurities [45]. With the use of such a suspension for the crystalline process, part of the crystallization centers of less than 10 µm will dissolve [1,9,18,25,28] that reduce the number of active crystallization centers, which will contribute to the formation of larger crystals [1–11]. Spontaneous growth and formation of multiple

crystallization centers could be induced by some quantity of particles with a size of less than 10  $\mu$ m [1,9,18,25,28] (Fig. 5, A, 5 min.). All this will contribute to the formation of a large number of conglomerates in the finished massecuite taking into account the adjacent crystal formation with particles of thick juice impurities (Fig. 5, A).

To the contrary the size of the crystal-forming agent particles in "Ester K 01" No.2 is already 46.4  $\mu$ m, which is almost 3.0 times higher than the maximum size of particles of impurities of thick juice [45]. Obviously, if they are used for the crystal formation process, such crystallization centers will grow faster to the appropriate size and faster form the corresponding crystals in the total amount of massecuite than the particles of impurities of thick juice. Consequently there is a kind of blocking the process of adjacent growth of crystals from the particles of impurities of thick juice with high turbidity, which will contribute to the formation of more uniform, homogeneous crystals of the correct shape (Fig. 5, C).

## 3.4. Qualitative sharacteristics of semi-products and sugar

The following products were examined: massecuite, intercrystal solution and sugar in order to evaluate the impact of different crystalforming agents on the quality of intermediate products. In Table 3 are presented the acquired results.

Since the same initial syrup was used in all experiments during

#### Table 3

Analyses of intermediate products obtained with different crystal-forming agents during massecuite boiling.

Intermediate product	Crystal-forming agents	pH <sub>20</sub>	Brix,%	Sucrose content, %	Purity quotient, %	Color, ICUMSA units	Ash content, %
Massecuite	Factory suspension	$6.9\pm0.1$	$90.6\pm0.1$	$84.20\pm0.1$	$92.90\pm0.02$	906 ± 4	$0.145\pm0.002$
	«Ester K 01»	$\textbf{6.9} \pm \textbf{0.1}$	$\textbf{90.2} \pm \textbf{0.1}$	$83.80\pm0.1$	$92.90\pm0.02$	$928\pm3$	$0.143\pm0.002$
	No.1						
	«Ester K 01»	$\textbf{6.9} \pm \textbf{0.1}$	$\textbf{90.4} \pm \textbf{0.1}$	$84.00 \pm 0.1$	$92.90\pm0.02$	$918\pm3$	$0.144 \pm 0.002$
	No.2						
Mother liquor	Factory suspension	$\textbf{6.7} \pm \textbf{0.1}$	$\textbf{58.8} \pm \textbf{0.1}$	$53.40 \pm 0.1$	$90.81 \pm 0.03$	$841\pm 5$	$0.086\pm0.004$
	«Ester K 01»	$\textbf{6.9} \pm \textbf{0.1}$	$\textbf{58.8} \pm \textbf{0.1}$	$53.20\pm0.1$	$90.47 \pm 0.04$	$858\pm5$	$0.088 \pm 0.004$
	No.1						
	«Ester K 01»	$\textbf{6.9} \pm \textbf{0.1}$	$58.6 \pm 0.1$	$52.80\pm0.1$	$90.10\pm0.04$	$862\pm4$	$0.090\pm0.003$
	No.2						
Sugar obtained	Factory suspension	_	-	$97.75 \pm 0.05$	_	_	$0.058\pm0.002$
	«Ester K 01»	-	-	$97.80 \pm 0.05$	-	_	$0.055\pm0.002$
	No.1						
	«Ester K 01»	-	-	$97.75\pm0.05$	-		$0.054\pm0.002$
	No.2						

laboratory crystallization, the differences in purity, color and ash for the samples of molasses obtained with different crystal formers are almost insignificant (Table 3).

As can be seen from Table 4, the lowest sugar content, purity and the highest ash content in the mother liquor is in case of use of the crystal-forming agent "Ester K 01" No.2. Somewhat worse characteristics were obtained by the use of "Ester K 01" No.1. The mother liquor obtained in case of use of the factory suspension has the worst characteristics among the studied samples.

The highest content of sucrose is observed in the sugar sample with the use of the crystal-forming agent "Ester K 01" No.1 (Table 3), but the difference between the values is insignificant because of the same sugar content in all samples. The ash content is almost 7.0 % lower in the sugar samples obtained with the "Ester K 01" crystal-forming agents No.1, No.2 in comparison with the sugar obtained on the factory suspension. The low ash content in sugar obtained with crystal formers"Ester K 01" No. 1, No. 2 is confirmed by its higher content in the corresponding samples of intercrystalline solution. This indicates a good correlation with the results of study of the particles morphology. Since we have a good separation of the intercrystalline solution and sugar crystals, more homogeneous sugar crystals (fewer agglomerates) with a minimum number of inclusions are formed when using whole crystals of "Ester K 01" No. 1, No. 2.

## Table 4

Screen, diameter	The average size	Sugar mass on screen, g				
	of crystal, d",	Factory	"Ester	"Ester		
	mm	suspension	К01"	К01"		
			No.1	No.2		
2.50	2.70	0	0	0		
1.25	1.35	$6.54 \pm 0.03$	0.94 $\pm$	0.76 $\pm$		
			0.05	0.05		
1.00	1.10	$20.22 \pm 0.02$	18.64 $\pm$	14.16 $\pm$		
			0.02	0.02		
0.80	0.90	$24.06\pm0.02$	$21.46~\pm$	$\textbf{24.82} \pm$		
			0.02	0.02		
0.50	0.65	$31.82 \pm 0.02$	42.52 $\pm$	43.38 $\pm$		
			0.02	0.02		
0.20	0.35	$16.78\pm0.02$	16.18 $\pm$	16.66 $\pm$		
			0.02	0.02		
Bottom	0.10	$\textbf{0.58} \pm \textbf{0.06}$	0.26 $\pm$	0.22 $\pm$		
			0.05	0.05		
Mean aperture	-	$0.800~\pm$	0.746 $\pm$	$0.731~\pm$		
MA, mm		0.009	0.005	0.006		
Coefficient of	-	$35.90\pm0.01$	$\textbf{33.80} \pm$	$32.50~\pm$		
variation CV, %			0.02	0.02		

## 3.5. Granulometric composition of the resulting experimental sugar samples

High-quality sugar should be homogeneous in size, should not contain clusters (conglomerates), that is crystals that stick together and "flour" – too small crystals that are capable of passing through a sieve of 0.25 mm [9,30,46].

Analysis of granulometric composition was carried out to assess crystals quality of the obtained sugar samples. The findings are presented in Table 4.

The analysis of the data of Table 4 shows that the highest average linear size (0.800 mm) is observed in the sugar sample obtained with the factory suspension. The use of crystal "Ester K 01" No.1, No.2 for seeding of crystallization centers gives almost the same average linear size of crystal sugar, but slightly smaller with "Ester K 01" No.2.

The largest average linear size of sugar crystals for the use of factory suspension can be explained by a decrease in its concentration by dissolving particles less than 10  $\mu$ m, and therefore a decrease in number of active crystallization centers [1,9,18,25,28]. It is because of larger crystals formation at low seed concentration [1–11].

The highest ratio (Table 4) of sugar crystals – 35.9 % we have in case of use factory suspension, which is obviously associated with the significant unevenness of the particles from 0.3 to 14.4 µm (Fig. 3, A). During nucleation such crystallization centers smaller than 10 µm pass through the critical phase of growth, so they can both dissolve and cause spontaneous growth and formation of multiple small dispersed centers of crystallization ("flour") [1,9,18,25,28]. All this will cause the formation of more uneven sugar crystals. Such heterogeneity ratio is unsatisfactory according to the recommendations [28,30].

"Ester K 01" No.1 and No.2 crystal agents assure smaller unevenness ratios - 33.8 % and 32.5 %, which is 5.8 % respectively and 9.5 % less than for the factory suspension and is a satisfactory characteristic according to the recommendations [28,30]. The lower coefficient of variation for "Ester K 01" No.2 can be explained by the less influence of the related growth of crystals from the particles of impurities of thick juice. In addition, it is known [47] that the number of active growth centers on the surface of a solid crystal is smaller by two orders than the total surface of a sucrose molecule. That is, only a part of the areas of solid crystals of the "Ester K 01" No.1 and No.2 actively participates in the process of new crystal formation, and most of their areas do not contribute to the process of nucleation, since they are located on a flat crystal surface [47]. Therefore, due to this, we also have a minimal content of fines and conglomerates in the maturing massecuite and sugar, unlike the seed from crushed sucrose crystals, which enhance nucleation due to the larger specific area surface and the actual absence of flat crystal areas.

The analysis of the presented sugar fractions (Table 4) shows that the

most uniform crystals of sugar were obtained for the use of the crystalforming agent "Ester K 01" No.1 and No.2. The number of crystals with a fraction of 0.65 mm is 42.52 % and 43.38 % respectively, that is 25.18 % and 26.65 % higher than the sugar obtained on the factory suspension. The number of crystals with a fraction of 1.35 mm is only 0.94 % and 0.76 %, respectively, which is 85.63 % and 88.38 % less than the sugar obtained from the factory suspension.

## 3.6. Industrial tests

## 3.6.1. Testing of crystal-forming agent "Ester K 01" No. 1

Industrial tests were carried out to check the effectiveness of the crystal-forming agent "Ester K 01" No. 1 at Narkevytsky Sugar Factory LLC. Factory suspension was used as a control sample (Fig. 3, A, Fig. 4, A). Comparative results are presented in Table 5 of the granulometric composition of sugar obtained in industrial conditions using the factory slurry and the crystal-forming agent "Ester K 01" No.1.

The analysis of the data in Table 5 shows that the average linear size of the sample of sugar produced with the crystal-forming agent "Ester K  $01^{"}$  No.1 is 5.3 % higher compared to the sugar produced with the factory slurry. The coefficient of variation is 36.58 %, which is 1.77 % lower compared to sugar boiled using the factory slurry. The larger average linear size of sugar crystals can be explained by the larger average size of its microcrystals that contributes to the faster formation of larger sugar crystals compared to the factory slurry. Therefore, it is necessary to reduce the duration of the basic massecuite boiling using crystal-forming agent "Ester K  $01^{"}$  No.1 by adjusting the control program. The lower coefficient of variation for sugar obtained on the crystaforming agent "Ester K  $01^{"}$  No.1 is due to a more uniform dispersed composition of its particles.

The analysis of sugar fractions (Table 5) showed that the use of crystal-forming agent "Ester K 01" No.1 in industrial conditions ensures the production of larger sugar crystals. The number of crystals fraction of 0.90 mm is 15.38 %, which is 55.0 % more compared to the use of the factory suspension. At the same time the number of crystals with an average size of 0.65 mm and 0.35 mm is 50.83 % and 14.32 % that is 14.23 % and 11.88 % less respectively, compared to sugar produced with the factory slurry. Crystals of small fraction - 0.10 mm make up only 0.11 %, which is 88.9 % less compared to sugar boiled on the factory slurry.

It was also performed microscopy of samples of finished sugar obtained using the factory suspension and crystal forming agent "Ester K 01'' No.1 in order to assess the crystal structure. The results of the study are shown in Fig. 7.

Sugar obtained from the factory slurry (Fig. 6, A) contains mainly

## Table 5

Granulometric composition of sugar obtained in the industrial conditions at Narkevytsky Sugar Factory LLC.

1 1 0			
Screen, diameter	Average size of crystal, d", mm	Sugar mass on so Factory suspension	rreen, g "Ester K01" No.1
2.50	2.70	$1.86\pm0.03$	$2.41\pm0.03$
1.25	1.35	$5.40\pm0.03$	$\textbf{6.44} \pm \textbf{0.03}$
1.00	1.10	$9.32\pm0.03$	10.51 $\pm$
			0.03
0.80	0.90	$6.92\pm0.03$	15.38 $\pm$
			0.02
0.50	0.65	$59.26\pm0.02$	50.83 $\pm$
			0.02
0.20	0.35	$16.25\pm0.02$	$14.32~\pm$
			0.02
Bottom	0.10	$0.99\pm0.05$	$0.11 \pm 0.05$
Mean aperture MA,	-	$0.710\pm0.005$	$0.750~\pm$
mm			0.005
Coefficient of	-	$37.24\pm0.01$	$36.58~\pm$
variation CV, %			0.01
Bottom Mean aperture MA, mm Coefficient of		$\begin{array}{c} 0.99 \pm 0.05 \\ 0.710 \pm 0.005 \end{array}$	$\begin{array}{c} 0.02 \\ 0.11 \pm 0.05 \\ 0.750 \pm \\ 0.005 \\ 36.58 \pm \end{array}$

conglomerates, which are a combination of two or more crystals and false crystals. Such crystals have significant damage that indicates their low strength and the presence of foreign inclusions in the crystal structure [9,28].

At the same time analysis of micrographs of sugar obtained on the crystal-forming agent "Ester K 01" No.1 (Fig. 6, B) showed that the obtained crystals are transparent, have the correct shape characteristic of sucrose and clear faces without damage. This is evidence of high crystal strength. The correctness of the outer crystal faceting and its internal structure that corresponds to the strict laws of symmetry denotes the absence of foreign inclusions [9].

Thus it can be concluded that good physicochemical and quality indicators of sugar are confirmed when using the crystal forming agent "Ester K 01" No.1 in comparison with the factory suspension. It is necessary to reduce the duration of main massecuite boiling by adjusting the automatic process control program in order to obtain a larger number of smaller crystals using the crystal-forming agent "Ester K 01" No.1.

## 3.6.2. Testing of crystal-forming agent "Ester K 01" No. 2

Production tests of the crystal-forming agent "Ester K  $01^{"}$  No.2 were carried out at PJSC "Salyvonkivskyi Sugar Factory". Comparative results of the granulometric composition of sugar obtained in production conditions on the crystallizing agent "Ester K  $01^{"}$  No.2 are presented in Table 6.

Analysis of the data in Table 6 shows that the largest average linear size of sugar (0.766 mm) is obtained with a crystal-forming agent consumption of  $0.5 \text{ cm}^3$ , a slightly smaller size (0.615 mm) is obtained at a consumption of  $0.3 \text{ cm}^3$  and the smallest is 0.565 mm at a consumption of  $0.2 \text{ cm}^3$ . We obtained the lowest coefficient of variation 30.67 % at a flow rate of  $0.5 \text{ cm}^3$  which is 12.30 % and 10.82 % lower compared to sugar obtained at a flow rate of  $0.2 \text{ cm}^3$  and  $0.3 \text{ cm}^3$  respectively. It is a good indicator according to recommendations [28,30].

The results of the analysis of sugar fractions (Table 6) show that the largest number of sugar crystals with a fraction of 0.65 mm – 52.67 % was obtained using an amount of 0.3 cm<sup>3</sup> of "Ester K 01" No.2 that is 3.78 % and 5.64 % lower compared to sugar obtained at a consumption of 0.2 cm<sup>3</sup> and 0.5 cm<sup>3</sup> respectively. The amount of sugar crystals of 0.90 mm fraction is the largest at a flow rate of 0.5 cm<sup>3</sup> and is 26.50 % that is 70.72 % and 51.21 % more compared to the flow rates of 0.2 cm<sup>3</sup> and 0.3 cm<sup>3</sup> respectively. Also the largest amount of sugar is observed with an average fraction size of 1.10 mm and 1.35 mm (9.50 % and 5.00 %, respectively) at a flow rate of 0.5 cm<sup>3</sup>.

At the same time number of crystals with an average size of 0.35 mm, at a consumption of crystal-forming agent "Ester K 01" No.2—0.5 cm<sup>3</sup>, is 9.30 % that is 76.18 % and 68.46 % less, compared to sugar obtained at a consumption of 0.2 cm<sup>3</sup> and 0.3 cm<sup>3</sup> respectively. At the same time crystals of small fraction - 0.10 mm make up only 0.10 %, that is 94.22 % and 81.13 % less compared to sugar boiled at a flow rate of 0.2 cm<sup>3</sup> and 0.3 cm<sup>3</sup> respectively.

Thus the consumption of 0.5 cm<sup>3</sup> per 1 ton of massecuite of the crystal-forming agent "Ester K 01" No. 2 ensures the formation of a larger number of large sugar crystals, which is obviously due to the average particle size of the seed  $-46.4 \mu m$ , This size eliminates spontaneous dissolution and loss of active crystallization centers and chaotic formation of multiple fine crystallization centers ("flour"), since the crystallization centers introduced into the boiled syrup already have a sufficiently large surface area that significantly intensifies the process of growing the resulting crystals [1,9,18,25,28–30].

The use of crystal-forming agent "Ester K 01'' No.2 with a consumption of 0.3 cm<sup>3</sup> per 1 ton of massecuite leads to formation of a slightly smaller number of large crystals compared to the use of 0.5 cm<sup>3</sup> but allows to obtain the largest amount of sugar with a fraction of 0.65 mm and a significant amount of crystals with a fraction of 0.35 mm. The consumption of 0.2 cm<sup>3</sup> per 1 ton of massecuite provides the largest amount of sugar of fine fraction – 0.35 mm with a significant content of

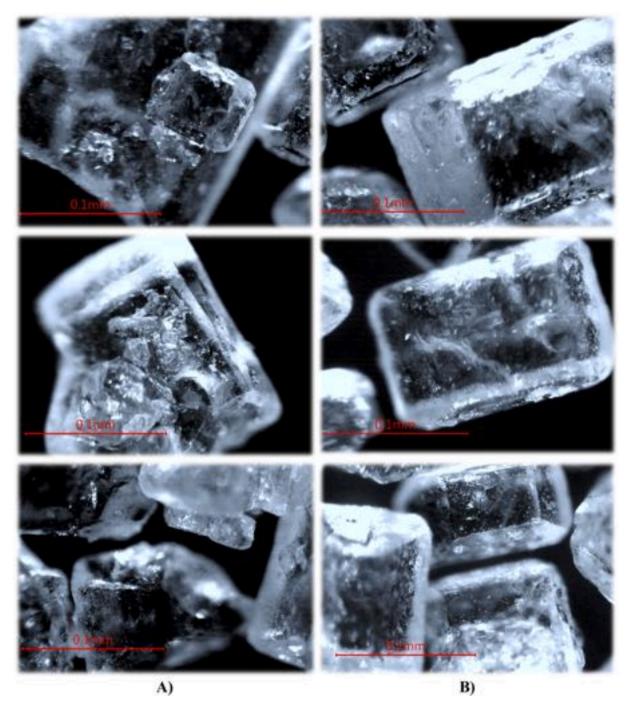


Fig. 6. Micrographs of sugar crystals obtained in industrial conditions (40x magnification): A) standard factory suspension; B) crystal-forming agent "Ester K 01".

## 0.65 mm fraction.

Thus it is possible by using different amounts of crystal-forming agent "Ester K 01" No.2 for the shock crystallization process with an average particle size of 46.4  $\mu m$  to adjust significantly the average size of the commercial sugar crystal fraction to meet the needs of the consumer market.

The authors carried out a mathematical analysis and optimization of the consumption of the crystal-forming agent "Ester K 01" No.2 for the shock crystallization process. The optimization of the rational consumption was performed based on the desired average sugar fraction size of 0.65 mm. In the calculations we used: average linear size of sugar crystals (Mean aperture) MA, mm; number of sugar crystals of 0.65 mm fraction C, %; coefficient of variation of sugar crystals CV, %; coefficient of homogeneity of sugar crystals H, %. The homogeneity coefficient is characterized by the ratio of the mass of the two largest adjacent fractions by sieve analysis to the mass of the sample under analysis and is expressed as a percentage [28,30]. The calculated homogeneity coefficients for sugar obtained at different consumption rates of crystal-forming agent "Ester K 01" No. 2 are presented in Table 6.

The dependences are presented in Fig. 7 and Fig. 8 of the average linear size, the amount of sugar of the 0.65 mm fraction, the coefficient of variation and the coefficient of homogeneity of sugar crystals depending on the consumption of the crystal-forming agent. Correspondent regression equations were obtained in natural form.

The dependences were obtained of sugar crystallization efficiency on the consumption of crystal-forming agent "Ester K 01'' No.2 (Fig. 9) using these regression equations and the generalized optimization

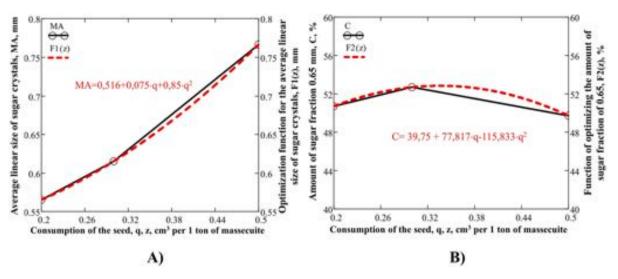


Fig. 7. Dependences of the average linear size, the amount of sugar of the 0.65 mm fraction on the consumption of crystal-forming agent and their regression equations in natural form: A) mean aperture, MA; B) amount of sugar of 0.65 mm fraction, C.

## **Table 6** Granulometric composition of sugar obtained with the crystallizing agent "Ester K 01" No.2 at different concentrations.

Screen diameter	The average	Sugar mass on screen, g			
	size of crystal,	$0.2 \text{ cm}^{3}$	0.3 cm <sup>3</sup>	0.5 cm <sup>3</sup>	
	d", mm	per 1 ton	per 1 ton	per 1 ton	
		of	of	of	
		massecuite	massecuite	massecuite	
2.50	2.70	0	0	0	
1.25	1.35	$\textbf{0.41} \pm \textbf{0.06}$	$\textbf{0.78} \pm \textbf{0.06}$	$\textbf{5.00} \pm \textbf{0.03}$	
1.00	1.10	$\textbf{2.11} \pm \textbf{0.03}$	$\textbf{3.60} \pm \textbf{0.03}$	$\textbf{9.50} \pm \textbf{0.03}$	
0.80	0.90	$\textbf{7.76} \pm \textbf{0.03}$	12.93 $\pm$	$26.50~\pm$	
			0.02	0.02	
0.50	0.65	50.68 $\pm$	52.67 $\pm$	49.70 $\pm$	
		0.02	0.02	0.02	
0.20	0.35	39.04 $\pm$	$\textbf{29.49} \pm$	$\textbf{9.30} \pm \textbf{0.02}$	
		0.02	0.02		
Bottom	0.10	$1.73\pm0.04$	$\textbf{0.53} \pm \textbf{0.06}$	$\textbf{0.10} \pm \textbf{0.07}$	
Mean aperture	_	$0.565 \pm$	0.615 $\pm$	$0.766~\pm$	
MA, mm		0.005	0.005	0.007	
Coefficient of	-	$34.97~\pm$	$34.39~\pm$	$30.67~\pm$	
variation CV, %		0.01	0.01	0.02	
Homogeneity	_	89.70 $\pm$	82.20 $\pm$	$\textbf{76.20} \pm$	
coefficient H, %		0.02	0.02	0.05	

criterion according to the Harrington method, which essentially characterizes the crystallization efficiency.

The weighting coefficients for the generalized optimization criterion were chosen taking into account the importance of local optimization criteria (MA, C, CV, H). The values of the weighting coefficients were taken in three variants. Thus in the first variant (I), all the weighting factors were the same, in the second variant (II), the values of the weighting factors for C and H were chosen to be larger and in the third variant (III) even larger. Since the amount of sugar in the 0.65 mm fraction and the coefficient of uniformity of sugar crystals in this case play a more important role in choosing a rational concentration of the seed for the crystal formation process.

Analysis of the dependencies shown in Fig. 9 showed that the rational consumption of "Ester K 01" No.2, which ensures effective sugar crystallization, is  $0.30...0.31 \text{ cm}^3$  per 1 ton of massecuite that corresponds to a consumption of 18.0...18.6 cm<sup>3</sup> for a vacuum pan with a capacity of 60 tons of massecuite.

## 4. Conclusions

Summarizing the results we can say that the duration of massecuite boiling with the crystal-forming agent "Ester K 01" (18.2  $\mu m$ ) and "Ester K 01" (46.4  $\mu m$ ) is 5–10 min shorter than the duration of boiling with the factory slurry.

It was found from the analysis of morphological, dimensional and quantitative features of sugar crystals in the massecuite at different stages of its production that more uniform in size crystals of regular shape were obtained using the crystal-forming agent "Ester K 01" (46, 4  $\mu$ m), slightly worse in terms of the corresponding characteristics but also uniform are the crystals obtained with "Ester K 01" (18.2  $\mu$ m). Sugar crystals obtained from the factory slurry are uneven and contain a significant number of clusters (conglomerates).

It has been established that the use of the factory suspension and crystal-forming agent "Ester K 01" (18.2  $\mu$ m) for the seeding of crystallization centers in thick juice with turbidity 2.8–3.4 times higher than the recommended standard value, leads to almost simultaneous nucleation of crystals from both thick juice impurities and microcrystals of the seed. Such adjacent crystallization causes the formation of conglomerates and irregularly shaped crystals with foreign inclusions.

It is obvious based on the analysis of microphotographs of sugar crystals in the massecuite for different time intervals that the crystallization centers of "Ester K 01" (46.4  $\mu$ m), when used for the process of crystal formation in high turbidity thick juice, grow faster to a given size and form the corresponding crystals in the total amount of massecuite faster than the particles of thick juice impurities. Consequently there is a kind of blocking of the process of adjacent growth of crystals from the particles of high turbidity thick juice that contributes to the formation of more uniform, homogeneous crystals with correct shape.

It was proved based on the analysis of the crystal size distributions of sugar obtained with different crystallizers in the laboratory that the most uniform sugar crystals were obtained using the crystallizing agents "Ester K 01" (18.2  $\mu$ m) and "Ester K 01" (46.4  $\mu$ m).

Industrial tests in the conditions of Narkevytsky Sugar Factory LLC confirmed that the use of crystal-forming agent "Ester K 01" (18.2  $\mu$ m) in the method of boiling on the seed magma provides more uniform and larger sugar crystals. It was found from the analysis of microphotographs of sugar obtained with the crystal-forming agent "Ester K 01" (18.2  $\mu$ m) under production conditions that the obtained crystals are transparent, have the correct shape characteristic of sucrose, clear edges without damage. That is evidence of high crystal strength. The absence of foreign inclusions is indicated by the correctness of the crystal face and its

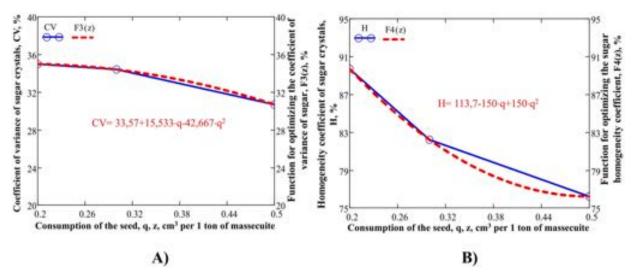
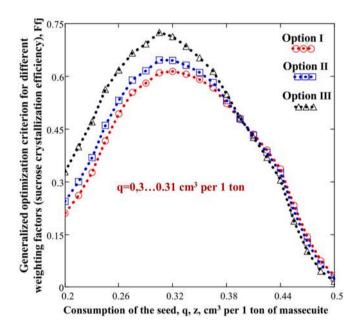


Fig. 8. Dependences of the coefficient of variation and the coefficient of homogeneity of sugar crystals on the consumption of crystal-forming agent and their regression equation in natural form: A) coefficient of variation, CV; B) homogeneity coefficient, H.



**Fig. 9.** Dependences of sugar crystallization efficiency (optimization function (Ffj)) on the consumption of crystal-forming agent "Ester K 01" No.2.

internal structure that corresponds to the strict laws of symmetry. In contrast sugar produced from factory slurry contains mainly conglomerates and false crystals with significant damage. This indicates their low strength and the presence of foreign inclusions in the crystal structure.

It was found in the process of seeding with the crystal-forming agent Ester K 01" (46.4  $\mu m$ ) by the method of shock crystallization under the production conditions of PJSC "Salyvonkivskyi Sugar Factory" that at a consumption of 0.5 cm<sup>3</sup> per 1 t of massecuite we have the lowest coefficient of variation and the largest number of sugar crystals of large fraction (0.90, 1.10 mm and 1.35 mm) compared to the consumption of 0.2 cm<sup>3</sup> and 0.3 cm<sup>3</sup> per 1 t of massecuite. At the same time the largest amount of sugar crystals with a fraction of 0.65 mm and 0.35 mm was obtained at the consumption of Ester K 01" (46.4  $\mu m$ ) – 0.3 cm<sup>3</sup> and 0.2 cm<sup>3</sup> per 1 ton of massecuite respectively. It was determined, based on the data of industrial tests in the conditions of PJSC "Salyvonkivskyi Sugar Factory" using the generalized optimization criterion, that for the process of crystal formation by the method of shock crystallization the

rational consumption of "Ester K 01'' (46.4 µm) is 0.30...0.31 cm<sup>3</sup> per 1 t of massecuite. This corresponds to a consumption of 18.0...18.6 cm<sup>3</sup> for a vacuum pan with a capacity of 60 tons of massecuite.

The perspective of our further work is to develop and work out a methodology for determining the rational amount of whole crystal crystallizing agents for operation with the seed magma.

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## CRediT authorship contribution statement

Serhii Volodymyrovych Tkachenko: Funding acquisition, Formal analysis, Data curation, Conceptualization. Tamila Volodymyrivna Sheiko: Resources, Project administration, Methodology, Investigation. Olena Mykhailivna Anisimova: Supervision, Software, Resources. Vasyl Viktorovych Petrenko: Writing – review & editing, Methodology, Investigation. Kostiantyn Dmytrovych Skoryk: Supervision, Funding acquisition, Formal analysis. Olha Illivna Dzhohan: Writing – original draft, Software, Conceptualization. Liubomyr Mykhailovych Khomichak: Visualization, Validation. Inha Vadymyrivna Kuznietsova: Writing – original draft, Investigation, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

Data will be made available on request.

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