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SELECTION OF GELLING AGENTS TO ACHIEVE HIGH HOMOGENEITY OF A CONCENTRATED PRODUCT FROM QUINCE (CYDONIA OBLONGA) FRUITS USING GRAPH THEORY

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This study investigates the quality of a concentrated quince fruit product. Given the widespread use of concentrated fruit products, particularly quince jelly, in various climatic zones, as well as their potential as a medicinal and dietary product, assessing product quality using mathematical methods, particularly graph theory, is of interest in processing technology.

Quince fruits, with their rich chemical composition, stand out among other fruits with their pleasant aroma. These volatile components migrate into the finished product, even after the raw materials are processed. Quince jelly is obtained by concentrating the juice, resulting in the formation of a colloidal system. However, manufacturing the product using gel technology, or more accurately, sol-gel technology, is determined by the added ingredients and environmental parameters. The advantage of this technology is that the resulting product has a more homogeneous appearance and a pleasant taste. The viscosity of the resulting product varies little across grades, averaging 2.17·10^4 mPa·s, and its Valent strength is 400. In terms of material flows, the consumption per ton of finished product was 1,328 kg.

The structure of fruit jelly is formed by the addition of gelling agents to the juice. Therefore, this product is not considered the result of a strict sol-gel processing method, where the transition from sol to gel structure occurs through chemical reactions. In fruit jelly, the ingredients themselves create a three-dimensional network structure, but not a solid crystalline one. This network structure is formed by hydrolysis of pectin substances and the polycondensation of polygalacturonic acids, resulting in the formation of salt bridges

Keywords: sol, gel, thixotropy, syneresis, xerogel, colloid, graph, vertices, edge, incidence, adjacency

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1. Introduction

Since sol-gel technology enables nanoscale production, its application range is vast. The substance used in this technology has the ability to form polymolecules, polysolvates, and micelles depending on conditions, which subsequently enables the formation of sol nanoparticles [1]. Although fruit juices acquire new physicochemical properties during thermal thickening, they have comparatively low chemical indices. The use of sol-gel technology in processing not only creates conditions for the formation of gelatin and similar products at lower temperatures but also improves their aggregation properties. This makes it easier to impart a structural form to the product, eliminate thixotropy, and increase its viscosity.

It should be noted that in the traditional method of preparing fruit jelly, sugar is dissolved in the juice and heated by adding albumin. Gelatin is used to form jelly. Sol-gel technology, however, utilizes special polysaccharides that float in the product for a period of time without settling. Subsequently, under the influence of external factors, these particles aggregate to form a gel structure. This process occurs by increasing the concentration of the colloidal solution. Evaporation, extraction, chemical condensation, hydrolysis, and polymerizing agents are used for this purpose. Depending on the method used, aerogel, xerogel, cryogel, and other gels can be obtained.

Colloidal solutions are known to occupy a middle place between coarsely dispersed and true solutions. In sol-gel technology, a sol refers to a dispersed system in which the dispersed medium is a liquid and the dispersed phase consists of solid nanoparticles. Depending on the degree of sol maturation, particle aggregation begins, gradually leading to the formation of clusters with a specific structure. Sol-gel technology is widely used as a method for the liquid-phase synthesis of nanomaterials. One of the characteristics of the nanoworld is the absence of the concept of "phase" and the ability to self-organize. Products obtained using gel technology are primarily considered objects with fractal (fragmented) properties. The formation of this structure is caused by the aggregation of nanoparticles [1–3].

In general, gel production technology consists of standard operations, such as preparing raw materials for processing, converting the product into a gel, molding the gel, cooling, and packaging. Since jelly is a gel formation, the dispersed medium, that is, fruit juice, is dispersed in the gelling agent.

Indeed, since gel technology is aimed at forming porous structures, the formation of nanostructures in the product is completed by drying. However, for fruit jelly, this form is formed during the resting phase during storage after cooling. This stepwise sol-gel transformation occurs somewhat differently for other colloidal solutions. For example, in the first stage, the chemical composition of the product is formed, that is, a sol is formed. The particle size in this dispersed phase is 10^{-9}–10^{-6} m. The second stage is characterized by an increase in the density of the dispersed phase, that is, the formation of coagulation bonds between the particles. This can also be called the initial stage of gelation. In such a colloidal system, the distance between particles is 10^{-8}–10^{-7} m, and such systems exhibit thixotropy. In the third stage, separation of the dispersion medium occurs. As a result, phase bonds are formed between the particles, thixotropy is lost, and mechanical destruction of the structure becomes irreversible. The gel, which undergoes the subsequent drying stage, transforms into a finely porous, solid-structured material – a xerogel [2, 3]. According to [3], the rigidity of the gel depends on the spatial network of the colloidal structure. Understanding the origin of this rigidity is key to developing gels with the desired properties.

Graph theory is a branch of discrete mathematics. Graphs are essentially represented as geometric diagrams, but the points (vertices) of this diagram and the lines (edges or arcs) between them have specific meanings for the process under consideration. All graph theory problems can be solved in matrix and graphical form. Research conducted to study changes in the dispersed state of quince fruits during processing led to a mathematical approach to such transformations. Since the sol-gel transition is a chemical transformation, the goal is to find this transition point in order to obtain a high-quality finished product. Therefore, the application of graph theory to gel technology is highly relevant for solving this problem.

2. Literature review and problem statement

Only one species of quince is known to exist – *Cydonia oblonga L.*, all its cultivars being derived from this species. The fruits are large, round, pear-shaped, and wrinkled. The flesh can be white, yellow, firm, or soft. The fossilized portion of the quince fruit, primarily the core, consists of lignin and cellulose, which creates certain difficulties in processing, although these compounds are harmless to humans.

In total, quince is grown on 69,600 hectares of land worldwide. Quince fruits are characterized by their aroma and astringency. The aroma of these fruits is due to the esters of enanthic and pelargonic acids they contain. The astringency of quince fruits is due to the polyphenols they contain, primarily tannins. Chemically, quince contains 8–11% sugar, 0.85–2% total acids, 4.7–9.6% pectin, 0.42–0.68% tannins, and numerous vitamins and minerals. Methods for preparing concentrated products such as jam, preserves, jellies, candied fruit, etc., from quince fruit are described in [4, 5]. However, issues related to determining the transition point of the product from one state to another remain unresolved. A way to overcome these difficulties may be to apply gel technology not only to the preparation of jelly but also to other canned and confectionery products.

As is well known, fruit jelly is concentrated fruit juice mixed at high temperature with sugar, acid, and a gelling agent. Both fresh and sulphited fruit juices or syrups may be used as raw materials, provided that the content of the main juice in the mixture is at least 65%.

The paper demonstrates that, in general, the process of gel formation is a type of chemical condensation. The initially formed molecularly dispersed substance tends to precipitate. Therefore, for molecular aggregation, external factors (active acidity of the medium, temperature, added ingredient, concentration, etc.) must be selected so that aggregate growth is completed at a certain stage. Furthermore, the use of chemical condensation in the production of concentrated fruit products creates conditions for the subsequent imparting of the required structural form to the resulting freely dispersed particles [5]. However, issues related to determining the sol-to-gel transition point remain unresolved. A solution to this problem is the application of discrete mathematics methods, particularly graph theory. This approach to the process allows for the elimination of some process steps and the simplification of the flowchart. As a result of the cumbersome data, a more compact functional diagram emerges, making it easier to control and monitor the sol-gel transition. When making quince jelly, three components were used as gelling agents: pectin, agar-agar, and carrageenan. These components share a common chemical composition, as their structure consists primarily of polysaccharides, specifically galacturonic acid derivatives. Therefore, they all exhibit gelling properties because they can form a three-dimensional gel structure. However, these gelling agents differ in the degree of methoxylation and the presence or absence of salt bridges.

Highly esterified apple pectin, with an esterification degree greater than 50%, was selected for quince jelly. This pectin gels the product with a low active acidity and high dry matter content. This is a distinguishing feature of pectin compared to other gelling agents. Highly methoxylated pectin molecules form a gel structure by interacting with each other through hydrogen bonds, which retain water. Gelation occurs due to hydrogen bonds between the hydroxyl groups of carbons C_2 and C_3 in the galacturonic acid molecule and the carboxyl groups. A high degree of esterification of pectin also accelerates setting time. Such studies were conducted in [6]. However, both sugar and acid (citric acid) play important roles in gelling. If the solution is saturated with sugar, less acid is required for jelly formation. It has been established that, for example, the optimal recipe for jam is 281.79 g of sugar, 4.13 g of pectin, and 264.66 cm^3 of citric acid [7]. However, the pulp content of the jam must be taken into account. The presence of acid in this environment prevents the dissociation of free carboxyl groups, resulting in a reduced likelihood of forming negatively charged particles, thereby reducing their repulsion from each other. This explanation for gelation is also confirmed in [8]. Citric acid was chosen as an ingredient in the study because it promotes the formation of a more stable pectin gel. The positive role of tartaric acid in the formation of such gels was also demonstrated in [8].

Gelatin is commonly used in traditional fruit jelly preparation. However, being an animal product, this substance increases the caloric content of the jelly, and gelatin is easily denatured by heat. Therefore, choosing plant-based gelling agents prevents the aforementioned drawbacks. It is known that polygalacturonic acid chains are formed by salt bridges (Ca^{2+} or Mg^{2+} ions). Analysis of study results [9] showed that large amounts of metal ions reduce the organoleptic appeal of the product. Therefore, highly esterified pectin was chosen, in which gelling occurs without the formation of salt bridges.

Another gelling agent used in the study is agar-agar, as this polyol also plays an important role in gel formation. Agar-agar is a sulfated galactan, or more precisely, a mixture of two polysaccharides: agarose (β-D-galactopyranose and 3,6-anhydro-α-L-galactopyranose) and agaropectin (D-galactose and L-galactose with acidic side groups such as sulfates, galcturonases, and pyruvates). Therefore, the mechanism of its gel formation is considered to be a double bond model. It has been established that structural heterogeneity is, to a certain extent, necessary for the proper functional efficiency of red algal galactan. Agarose is responsible for the gel strength (agar-agar Bloom strength is 600–1200), and agaropectin is responsible for the solution viscosity. The fine organization of the polysaccharide chains makes these polymer structures effective molecular sieves. As noted in [10, 11], a flowing liquid is a sol, and a critical concentration of substances transforms it into a gel. Before use, it is kept in a cold liquid for 15 minutes, then heated for 5 minutes with stirring.

The use of agar-agar in a gelatin-free mixture is due to the fact that gelatin added to the agar-agar solution leads to a decrease in the shear stress of the system at a given velocity, that is, this composition makes the system more fluid [12].

Compared to other gelling agents, agar-agar does not require either sugar or acid for gelling. This is due to its high hydrating capacity and easy hydrolyzability in an acidic environment, resulting in a loss of gelling properties.

Carrageenan was selected as another gelling agent in the study. This substance forms a slightly turbid gel, has an elastic or brittle structure (depending on the type), and is thermoreversible. Carrageenan has a higher sulfate content (24–53%) compared to agar-agar. Chemically, it is a mixture of β-D-galactose-4-sulfate and 3,6-anhydro-α-D-galactose monomers with esterified sulfate residues. These residues are bonded to various cations (potassium, sodium, calcium). The most powerful gelling agent among the three types (kappa, lambda, and iota) of carrageenan is kappa($\varkappa$)-carrageenan, which was used in studies [13, 14]. A distinctive property of carrageenan is its interaction with proteins [15].

All of the above points to the feasibility of conducting research on the sol-gel transition of dispersed systems using food products as an example. When quince jelly is traditionally prepared, the product is not very thick, and the liquid separates from the main structure, giving the product an unmarketable appearance. This phenomenon is due to changes in the chemical composition of the raw materials, so careful selection of gelling agents is essential to prevent this problem.

3. The aim and objectives of the study

The aim of our study is to identify a mechanism for achieving high homogeneity in a concentrated quince product. This will, in practice, improve the product stability required for finished jelly.

To achieve this goal, the following objectives were set:

– to study the effect of gel technology on the quality of quince jelly;

– to conduct a comparative analysis of the effects of various gelling agents (pectin, agar-agar, carrageenan) on the physicochemical properties of quince jelly;

– to construct a functional flow chart for quince jelly production, including the key stages of the sol-gel transition.

4. The study materials and methods

The subject of our study is the quality of a concentrated quince fruit product.

The use of chemical condensation in the production of fruit jelly can affect the structure and, consequently, the appearance of the finished product. Since quince jelly is highly liquefied to achieve the desired consistency, this phenomenon can be approached from the perspective of the sol-gel transition. The influence of carbohydrate gelling agents must be taken into account.

Making products using gel technology, including jelly production, is a complex technological process. It can be assumed that this process involves a number of physical, mechanical, and chemical transformations in the raw materials. Adding auxiliary materials to this, one can imagine the complexity of this process. It could be expected that the effect of gelling agents on the sol-gel transition would improve the structure and appearance of the finished product. It was assumed that the application of graph theory to our technology would be successful, as it would shorten and simplify the raw material processing flow chart.

The study's materials were quince fruits (Jardam, Sary Quince, and Velechin varieties).

To improve the technology, discrete mathematics methods, specifically graph theory, were applied.

As is well known, each object and transformation in graph theory is represented by vertices and edges, which correspond to specific changes. Therefore, the technological process was formalized by the following formula (1), where the set of graph vertices is defined as V, and the set of edges is defined as E

$$G=(V,E), \tag{1}$$

where $E \subseteq V \times V$ because V^2 is the set of all two-element subsets.

In material flow graphs, vertices can be sources or sinks, and edges (arcs) represent flows. In such flows, technological operations act as converters (operators) of physical flows, that is, these operators translate the parameters of input flows to the parameters of output flows [16, 17]. The functional dependence of these operators can be expressed as follows

$$y = f(x, z, k), \quad (2)$$

where y is the output flow vector; x is the input flow vector; z is the control parameter vector; and k is a parameter accounting for the element design.

Statistical analysis of the quantities of the main ingredients of quince jelly was performed. The arithmetic mean of n determinations was found using formula (3)

$$\bar{x} = \frac{\sum_{i=1}^{n} x_i}{n}, \quad (3)$$

where x_i is an individual result of multiple measurements; n is the number of parallel measurements.

The standard deviation was estimated using formula (4)

$$S = \sqrt{\sum_{i=1}^{n} (x_i - \bar{x})^2 / (n-1)}. \quad (4)$$

The relative standard deviation was calculated using formula (5)

$$b = \frac{S}{\bar{x}} \cdot 100. \quad (5)$$

Random errors of the analysis results for the confidence probability ($\alpha = 95\%$) were estimated using formula (6)

$$\Delta = \pm \frac{t_\alpha S}{\sqrt{n}}, \quad (6)$$

where t_α is the distribution or Student's t-test.

The confidence limits for both ingredients will be as follows

$$\mu = \bar{x} \pm \Delta. \quad (7)$$

Quince juice is considered the primary raw material for jelly. The process flow for quince jelly is as follows: inspection, washing, crushing, heating, and enzymatic treatment, juice extraction, straining, clarification, decantation and filtration, addition of ingredients (creation of gel), and cooking of the mixture until the dry matter content reaches 65% for the pasteurized product and 68% for the unpasteurized product. Extracting juice from quince fruit is challenging due to its biological structure. Therefore, the pulp is heated to 40–45°C and a pectolytic enzyme suspension is added at a concentration of 0.03%. The resulting mass is pressed, filtered, and transferred to a clarifier. The heated juice is left to ferment for 3–4 hours. The resulting suspended particles are removed by decantation, and the juice is filtered. The filtered juice with added sugar is boiled down in a double-boiler or vacuum apparatus until the dry matter content reaches 65%. The boiling process should last no more than 30 minutes (Fig. 1).

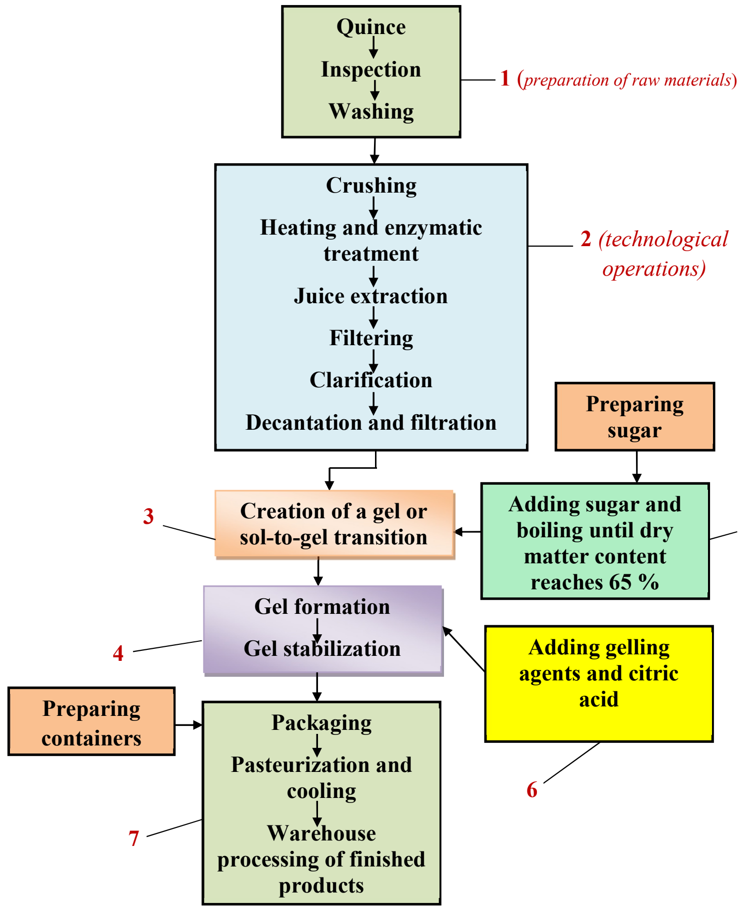


Fig. 1. Technological flow chart of quince jelly production

According to the recipe, acids and gelling agents were added to the mixture at the end of the boiling process (gel formation). After this procedure, the mixture was boiled for another 5–6 minutes. The active acidity of the medium reaches 3.2. The finished boiled jelly was filtered hot through cheesecloth or nylon mesh. Then, without allowing it to cool, the jelly was packaged. Glass, tin, aluminum, or thermoplastic polymer jars (to stabilize the jelly) can be used as containers. The filled container was immediately sealed. The jelly was pasteurized at 95°C for 10 minutes, then cooled and sent to warehouse operations. For comparative analysis, the necessary ingredients were taken in equal quantities. The yield of finished jelly was found using the following formula (8)

$$B = \frac{M_1 c_1 + M_2 c_2 + M_3 c_3}{C}, \quad (8)$$

where M_1, M_2, M_3 are the proportions of the respective ingredients, that is, quince, sugar, and gelling agent, kg; c_1, c_2, c_3 are the dry matter of the respective ingredients, %; C is the dry matter of the jelly, %.

Table 1 gives calculation results using formula (8) per 1000 kg of jelly.

Table 1

Recipe for ingredients for 1 ton of jelly

Product	Ingredients, kg			Output of finished jelly, kg	Juice weight, kg
	Juice	Sugar	Gelling agent		
Jelly	597.3	543.5	54.3	184	64.1

This means that the mass of quince juice per 100 g of jelly is 64.1 kg, which meets the standard requirements.

Gelling occurs precisely after the addition of gelling agents to the mixture. As a result, the Brownian motion of the particles slows down, and viscosity increases. Product processing conditions must be taken into account. Since the main component of all gelling agents is polygacturonic acids, prolonged heat treatment can destroy them by cleaving off the methoxyl groups of these acids. As a result, the gelling capacity of the ingredient decreases. By-product carbohydrates of gelling agents, such as rhamnose, arabinose, galactose, acetic acid, etc., do not affect the degree of gelling [5].

Sugar also positively affects the gelling capacity of gelling agents. This ingredient, by absorbing water from the hydrophilic parts of galacturonic acid molecules, promotes their aggregation. The sugar solution must be saturated. Replacing sucrose with glucose accelerates the gelling process, resulting in a softer and more flexible product. Glucose even prevents the formation of sugar crystals in the gel.

Gelling is also influenced by the structural components of the ingredients. For example, pectin contains methoxyl groups, while agar-agar does not, making pectin more suitable for gelling. Agar-agar and carrageenan are also considered acidic polysaccharides because they contain sulfuric acid residues.

As mentioned, to transform traditional jelly production into an effective sol-gel process, it is necessary to select the right gelling agent. Gel formation promotes a phase transition of the dispersed medium. If the resulting jelly needs to be preserved at the nanoscale, it is dried, and the product acquires a porous structure [16, 17].

The main reason for gel formation is that pectin and agar-agar react with Ca^{2+} ions, while carrageenan reacts with both Ca^{2+} and K^+ ions, forming a hard and brittle structure. Carrageenan and agar-agar gels are thermoreversible and exhibit hysteresis. Furthermore, carrageenan gels degrade more quickly and are not very strong. The presence of potassium ions shifted the formation and melting temperatures of κ-carrageenan hydrogel to higher temperatures, and the formation and melting temperatures of the gel increased with increasing potassium ion content [18].

Physicochemical analyses of the finished product were conducted. Gel strength was determined using a VC-1 instrument, and product viscosity was measured using an Ostwald viscometer (Table 2).

5. Results of a homogeneity study of a concentrated quince product

5. 1. Quality study of the finished product obtained using gel technology

Comparative analysis of quince jelly with various gelling agents revealed that the sample treated with pectin was elastic, soft, and transparent, with good light refraction and high solubility. The gel formed with kappa-carrageenan, although fragile, was transparent, while the gel formed solely by K^+ ions was prone to syneresis. The jelly prepared with agar-agar was dense and elastic.

The process flow chart (Fig. 1) shows the transformations the gel undergoes before it stabilizes. As mentioned above, gel formation begins with the addition of sugar to the product. This is because sugar absorbs the liquid phase and, as a result, increases viscosity. After gelling agents are added to the mixture, jelly formation occurs during cooling of the packaged finished product. The use of highly methoxylated pectin allows the gelling process to occur even in the absence of an acidic environment [5, 7].

Physicochemical analyses of the jelly revealed slight differences in these parameters (Table 2). As can be seen from Table 2, a comparison of all the physicochemical properties of the gelling agents yielded nearly identical values, but agar-agar exhibits a higher viscosity than the other ingredients.

5. 2. Comparative analysis of the effect of various gelling agents on the physicochemical properties of the finished product

Graphs of the relationship between viscosity and active acidity of quince jelly with different gelling agents (Fig. 2) show that with pectin and carrageenan, viscosity initially increases and then drops sharply.

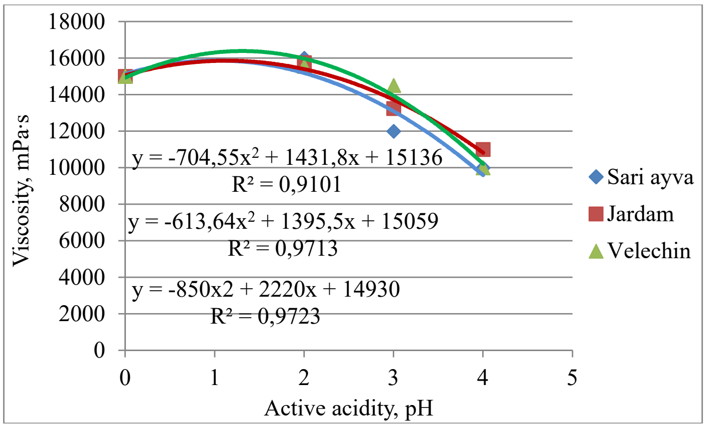
Quince fruit is known to contain pectin, so jelly made with pectin performed better in the sol-gel transition than those made with agar-agar and carrageenan. This is likely due to the similar origin (seaweed) of both gelling agents and their nearly identical chemical composition (the difference being the Ca^{2+} and K^+ ions in kappa carrageenan). The highly esterified pectin used was apple pectin, and in apple pectin, calcium ions do not participate in gelation [5, 6].

It has been established that the sol-gel transition in hydrocolloids is a first-order phase transition. It has been shown that under certain conditions, the sol-gel transition can acquire features characteristic of a percolation transition [17, 19]. The sol-gel transition in jelly preparation can be controlled by adjusting the chemical composition of the mixture. To clarify this, studies were conducted with different amounts of ingredients to determine the sol-gel transition point. The recipe for 1 ton of jelly was changed, except for the juice content. The research results are given in Table 3.

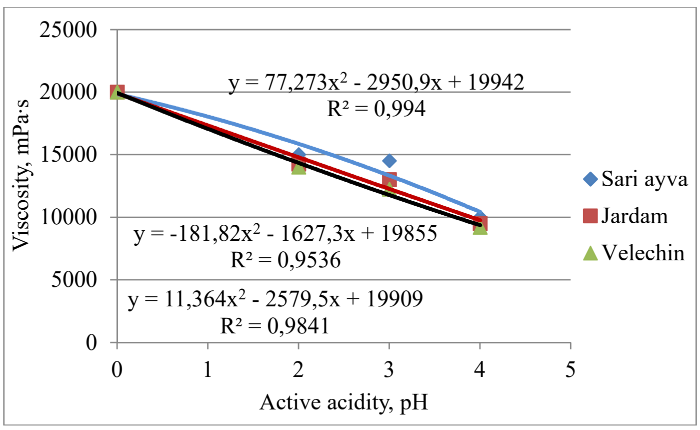
Table 2

Physicochemical properties of quince jelly

Indicator	Variety								
	Sary quince			Jardam			Velechyn		
	Pectin	Agar-agar	Carrageenan	Pectin	Agar-agar	Carrageenan	Pectin	Agar-agar	Carrageenan
Viscosity, mPa·s	1.5×10^4	3×10^4	2×10^4	1.5×10^4	3×10^4	2×10^4	1.5×10^4	3×10^4	2×10^4
Strength, Valent-based	300	400	500	300	300	500	400	400	500
Active acidity, pH	3.2	3.4	3.9	3.2	3.4	3.9	3.2	3.4	4.0



a



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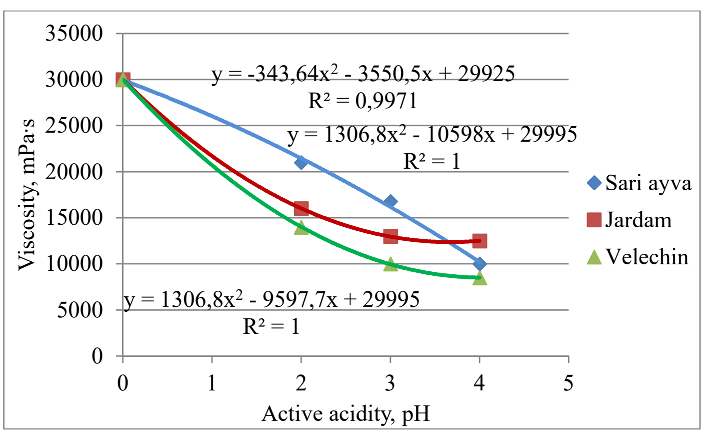

c

Fig. 2. Dependence of viscosity on active acidity of quince jelly with different gelling agents: *a* — with pectin; *b* — with agar-agar; *c* — with carrageenan

Table 3

Various recipe compositions for 1 ton of jelly

Recipe	Temperature-time	Gelling agents, g			Sugar, g	pH
		Pectin	Agar-agar	Carrageenan		
I	85°C–15 min	7.5	7.5	7.5	120	4.2
II	90°C–5 min	8.0	8.5	8.5	115	4.0
III	95°C–0 min	9.0	9.0	9.0	110	3.5

5. 3. Constructing a functional diagram of quince jelly based on the stages of sol-gel transition

Based on the data in Table 3, statistical analysis of the quantities of the main ingredients (pectin and sugar) of quince jelly was performed. The arithmetic mean, standard deviation, and relative standard deviation of three determinations were found (x_1 = 7.5 kg, x_2 = 8.0 kg, x_3 = 9.0 kg).

Then, using formula (7), we obtain the confidence limits for both ingredients:

$$6.28\ \text{kg} \le \mu_{\text{pectin}} \le 10.06\ \text{kg},$$

$$115\ \text{kg} \le \mu_{\text{sugar}} \le 127.4\ \text{kg}.$$

All obtained results are given in Table 4.

Table 4

Results of statistical processing of quince jelly recipe composition

Ingredient	The meanings of three parallel measurements, x_i			
	Arithmetic mean, $\bar{x}$	Root mean square value, *S*	Relative standard deviation, *b*	Random errors, *Δ*
Pectin	8.17 kg	0.76 kg	9.3%	1.89 kg
Sugar	115 kg	5 kg	4.3%	12.4 kg

It should be noted that the statistical data results (Table 4) show small deviations from the obtained standard data.

In the process flow chart for preparing quince jelly (Fig. 1), all operations are aimed at gel formation and its final stabilization. If we consider the process in terms of graphs, it can be represented as in Fig. 3.

The process flow chart in graph language will be more compact. In Fig. 3, the vertices (V = {1, 2, 3, 4, 5, 6, 7}) represent objects, and the edges (E = {*a*, *b*, *c*, *d*, *e*, *g*, *f*, *h*}) express transformation processes. Writing the numbers in functional form corresponding to each vertex allows for a more visual representation of the graphs, that is, some process operations are abbreviated.

For example, 1 is the preparation of raw materials (sum of three operations); 2 is the process operations (sum of six operations); 3 is the sol-gel transition; 4 – gel formation and jelly stabilization (sum of two operations); 5 – sugar addition and boiling; 6 – gelling agent and acid addition; 7 – container preparation and finished product storage (sum of three operations). Numerical conclusions can be drawn from this flow chart structure.

Fig. 3, *a* shows that the resulting graph consists of seven vertices and eight edges. The contour rank or cyclomatic number of this graph ($\nu(G)$) is 2. This number indicates that removing two edges from the graph yields a new type of graph – a tree. This value is found as the difference between the number of edges (E = 8) and vertices (V = 7), plus the connectivity component (9)

$$\nu(G) = |E - V| + 1 = 8 - 7 + 1 = 2. \qquad (9)$$

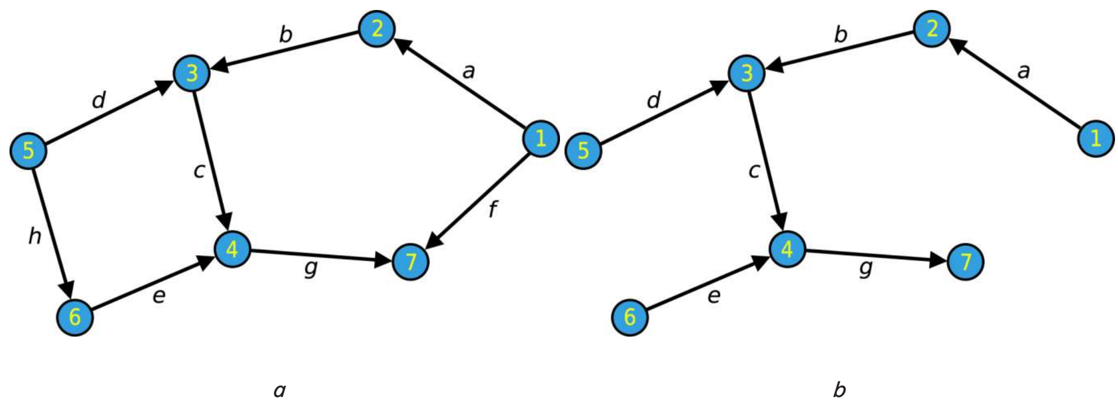

Fig. 3. Technological scheme of quince jelly: *a* – full graph; *b* – tree

This value also indicates the number of independent cycles in the graph.

Qualitative and quantitative transitions of physical flows allow us to express the symbolic mathematical model of the operator as a functional relationship. The properties of the *i*-th physical flow are characterized by the following parameters (Fig. 3, *b*)

$$Q_i = (m_i; x_i; t_i; T_i), \tag{10}$$

where m_i is the flow rate; x_i is the ingredient proportion; t_i is the temperature; T_i is the time.

Clearly, the graph tree can be used to identify flow graph vertices corresponding to process elements. These elements reflect changes in the flow rates of physical parameters. The graph arcs represent generalized flows. In Fig. 3, *a*, the arcs a, b, d, f, g, h can be replaced with the corresponding flow rates. Generally, these flow rates contain source vertices and sink vertices. In our tree, vertex 7 is the sink (*S*), that is, the finished product or output element. All other elements, that is, the input components, serve vertex 7.

Compared to the general jelly-making process, the functional diagram is more compact. The technical devices, that is, the process equipment, remain the same; only the approach to the processing changes. In the tree graph model, there are no connections between nodes 1 and 7, or 5 and 6. Unlike the cumbersome, conventional large-scale diagram, this clearly depicts the interrelationships between raw material processing operations.

Process graphs express the interrelationships, dependences, and influences of processes on each other. Representing graphs in matrix form better captures these interrelationships. The incidence matrix describes the topological properties of the graph, displaying the relationships between nodes and edges. The adjacency matrix shows the degree of adjacency between nodes (Fig. 4).

For each technological process, flow graphs can be constructed. Such graphs are homomorphic to the system under consideration and represent topological models of physical or material flows (2).

In material flow graphs, vertices correspond to elements of the technological system, which are distributed among sources and consumption channels. The edges of the graphs represent the overall transformations of materials [15, 16, 20, 21]. According to the law of conservation of mass and energy, the equation for the vertex flows along the edges is as follows:

$$\sum_{i=1}^{e} W_{ik} = 0, \tag{11}$$

where W_{ik} is the flow along the *i*-th edge or arc incident to the *k*-th vertex.

1	*aa*	*bb*	*cc*	*dd*	*te*	*ff*	*gg*	*hh*
11	-– 1	00	00	00	00	-– 1	00	00
22	11	-– 1	00	00	00	00	00	00
33	00	11	-– 1	11	00	00	00	00
44	00	00	11	00	11	00	=– 1	00
55	00	00	00	-– 1	00	00	00	-– 1
66	00	00	00	00	-– 1	00	00	11
77	00	00	00	00	00	11	11	00

a

	11	22	33	44	55	66	77
11	00	11	00	00	00	00	11
22	00	00	11	00	00	00	00
23	00	00	00	01	00	00	00
44	00	00	00	00	00	00	11
55	00	00	11	00	00	11	00
66	00	00	00	11	00	00	00
77	00	00	00	00	00	00	00

b

Fig. 4. Graph matrices: *a* – incidence; *b* – adjacency

The edge symbols on the material flow graph indicate consumption. The calculations for quince jelly determined raw material consumption (1,328 kg) per 1,000 kg of product, including waste (12)

$$T = \frac{B_{\text{juice}} \cdot 100}{100 - x}, \tag{12}$$

where B_{juice} is the juice requirement for 1 ton of jelly; *x* is waste, %.

Dividing the resulting value by the edges (1328 : 8 = 166 kg) yields the consumption rate for the entire process. Although this number is common to all edges, each edge still has its own numerical value depending on the stages of the process operations. If we distribute this number by the edges (Fig. 3), for example, for *a* it will be 55.3 kg; for *b* – 27.7 kg; for *c*, *h*, and *e*, this number remains constant; for *d* – 83 kg; for *g* – 55.5 kg.

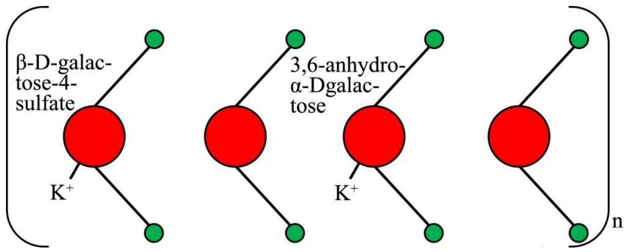

Fig. 5. The proposed molecular forest graph of kappa-carrageenan

The structures of gelling agents can also be viewed from a graph perspective. For example, in pectin, one molecule is the source, or rather the vertex, of its entire structure, while in agar-agar and kappa-carrageenan, two molecules act as vertices. A complex graph structure results in a forest (Fig. 5).

6. Results of applying gel technology for quince jelly: discussion and summary

Our results (Table 2) indicate that gel technology facilitates the formation of a product with a cross-linked structure, that is, at the nanoscale. This results in an even more homogeneous jelly, free of small particles. This can be explained by the fact that hydrolysis and polycondensation of the polymer structure of the gelling agents occurs during raw material processing. In this process, the dispersed medium is incorporated into the gelling agent. Furthermore, nucleation does not occur during gelling, and the resulting product is non-crystalline because this process creates a three-dimensional structure that prevents this formation. Despite the fact that the jelly acquires new physical and mechanical properties during the process, the quality of the finished product improves (Table 3).

Pectin was considered the most suitable of all three gelling agents used (Fig. 2, *a*) because pectin-based products have a pleasant taste, a good appearance, and a natural aroma, characteristic of the processed raw material. Fruit pectin's widespread availability and affordability should also be considered. Furthermore, the polygalacturonic acid bases of highly esterified pectin (Table 3) are highly susceptible to gelation [5].

In terms of physicochemical properties (Table 2), finished products from different varieties differ little. However, the Valent gel strength of carrageenan was 500, while the viscosity of agar-agar gel was $3{\cdot}10^4$ mPa. These differences, according to these data, are explained by the distinctive chemical structures of the gelling agents. Although galactan is the primary structural element for all of these ingredients, it differs in the residual molecule, the composition of functional groups, the degree of esterification, the presence of various salt bridges, etc.

A unique feature of the results is the use of graph theory to construct a functional diagram of the concentrated food product, taking into account the sol-gel transition (Fig. 3). There is data on the application of graph theory in the production of oilcake, semi-finished potato products, and canned goods, but, unlike [21], there is no detailed information on the reported studies. Therefore, taking this fact into account, a functional flow chart for jelly production was built, a process matrix was constructed, and calculations were performed for the general flow (10), (12).

Further studies are predetermined by the application of discrete mathematics methods, specifically graph theory. This approach to the process avoids the aforementioned difficulties. As an alternative to the traditional jelly production flow chart, the functional flow chart allows for a shorter raw material processing flow chart, giving the process a more compact form. This approach to the process will also improve the quality of the jelly production process, as this flow chart, by accelerating raw material processing, eliminates side processes (Fig. 1, 3, 4). Using the flow chart (11), it is possible to adjust ingredient feeds and control physical parameters at the sol-gel transition stage (Table 4). This result can be achieved by routing processes and eliminating individual process steps.

For clarity, a forest-like molecular graph of kappa-carrageenan was constructed (Fig. 5). The formation of this structure is explained by the fact that two kappa-carrageenan molecules act as vertices. Cyclization of this forest can reveal the underlying structure of the polymolecules.

A limitation of gel technology is the precise determination of the sol-to-gel transition parameters. As noted above, this transition is influenced by both external physical parameters and the chemical components of the raw materials. Therefore, practical application and further research, determining the sol-to-gel transition point is a complex task and represents an extension of this approach. It should also be taken into account that in the initial stages of the transition, the dispersed system differs little from the subsequent state. Only at the transition point does this difference disappear, and a different system – a gel – emerges.

7. Conclusions

1. The physicochemical properties of the resulting product showed little difference between varieties. These parameters are key during the sol-gel transition. The viscosity and strength of the product under study determine its appearance. The active acidity of the environment also influences the product's properties, as its value determines the final formation of the jelly structure.

A comparison of the physicochemical properties of quince jelly using different gelling agents revealed little difference. For example, the viscosity of pectin, agar-agar, and carrageenan in all varieties was 1.5×10^4 mPa·s, 3×10^4 mPa·s, and 2×10^4 mPa·s, respectively. The pectin strength of the Sary Quince and Jardam varieties was the same – 300, per Valent. Only the Velechin variety had a strength of 400 per Valent. It should be noted that all grades had the same carrageenan strength, that is, 500 per Valent. The active acidity of the gel was almost identical across all grades.

2. In an organoleptic comparison of gels with different gelling agents, pectin was preferred because the sample with this agent has a soft, transparent structure and dissolves well. Furthermore, for economic reasons, it has inexpensive processing sources and is therefore easily transported.

Based on recipe compositions, it can be noted that a temperature of 85–95°C and a cooking time of 10–15 minutes are suitable for the sol-gel transition, depending on the amount of gelling agents, sugar, and the active acidity of the medium. However, the final point of sol-gel transition occurs immediately after the addition of the gelling agent, and complete conversion occurs within 10 minutes. This time is required for microbiological requirements.

It can be concluded that fruits with a high pectin content more easily transition from sol to gel when pectin is added.

Furthermore, highly esterified pectin forms gels without the use of cations, has a regular molecular structure, and the resulting jelly exhibits greater stability.

3. To mathematically describe the raw material processing, the process flow diagram was represented in functional form. This diagram simplifies the approach to process engineering. The matrix representation of processes allows for a better understanding of the transformations occurring in the raw material. Constructing a complete process flow diagram provides greater clarity of these transformations. Using incidence and adjacency matrices, one can determine the degree of proximity and interaction of various process operations that result in various raw material transformations.

Conflicts of interest

The authors declare that they have no conflicts of interest in relation to the current study, including financial, personal, authorship, or any other, that could affect the study, as well as the results reported in this paper.

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Data availability

The manuscript has associated data in the data warehouse.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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