STUDY OF THE STABILITY OF FOAM AND VISCOELASTIC PROPERTIES OF MARSHMALLOW WITHOUT GELATIN

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Abstract: Marshmallow is a special kind of chewing souffle, prepared on the basis of sugar and gelatin, belongs to popular confectionery products. The study aims at developing a marshmallow technology with the replacement of gelatin (taking into account religious and ethnic restrictions in human nutrition) with various non-starch polysaccharides. Taking into account the data on the synergism of polysaccharides, some pairs of non-starch polysaccharides (xanthan gum, guar gum and locust bean gum) with the total concentration of 1–2%, based on which eight marshmallow samples had been produced, were experimentally selected. The organoleptic quality of these samples was estimated using the subjective estimation of shape retention, elasticity, and an increase in volume. The marshmallow texture indicators were analyzed using a tool-software complex "TA.XT plus Texture Analyzer". The shelf life was estimated by measuring the moisture content and water activity using a water activity analyzer "HygroPalmAw" (Rotronic, Switzerland), which is equipped with a dielectric moisture sensor. The study of the moisture content and water activity of eight selected samples with different concentrations of xanthan gum and vegetable gums allows to refer the corresponding samples to a class of products with a mid moisture content. Based on the study, the organoleptically acceptable and economically viable marshmallow samples were selected, each of which successfully reproduces the main attributes of the well-proven and widely consumed traditional marshmallow.

Keywords: Food hydrocolloids, synergism, organoleptic estimation, moisture, water activity, textural characteristics, surface tension

INTRODUCTION

In the American and European confectionery markets, a special kind of chewing souffle prepared on the basis of sugar and gelatin – marshmallow – is widely known. The popularity of marshmallow is explained by its specific structural and mechanical properties against the background of other confectionery products (marshmallow and pastille): elasticity and its shape-retaining ability [1]. Due to its unique texture, marshmallow is also currently in high demand in eastern countries.

Gelatin, being a protein capable of forming elastic jelly, simultaneously performs two functions during the preparation of marshmallow – a foaming agent and an effective densifier [2]. With the addition of sugar and the subsequent intensive shaking, gelatin forms a white plastic air mass, which begins to show distinct elastic properties after hardening.

Along with these advantages, gelatin is characterized by a serious disadvantage – its animal origin. It is because of this that confectionery gelatin-based products cannot be included in the category of Halal food products allowed in Islam, which is the main obstacle to the wide spread of classic marshmallow in eastern countries [3]. The use of gelatin is also problematic and unsafe because of the restrictions imposed on its use by people with disturbed water-salt metabolism and urolithiasis. In addition, in view of its origin, gelatin may contain the spores of pathogenic microorganisms.

This problem can be solved by replacing gelatin with various polysaccharides – vegetable, algal or microbial gelling agents, a lot of which are widely used in the confectionery industry [4]. The task in this case is reduced to the qualitative and quantitative selection of one or several structure-forming polysaccharides that show synergistic properties, the products on the basis of which, in its texture, could be comparable to original marshmallow. The development of this type of products will help us to avoid the restrictions of their further distribution on religious and medical principles, thereby increasing sales volumes.

Thus, the search, development and testing of an alternative marshmallow production technology without using gelatin is an urgent scientific and practical task.

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STUDY OBJECTS AND METHODS

Technology. The study was carried out on the basis of the chair "Food Technology" of Saratov State Agrarian University named after N.I. Vavilov (Saratov, Russia) and the Food industry laboratory of the Research Institute of Food Science and Technology (Mashhad, Iran).

The basis for sample preparation is a typical marshmallow formulation that includes the stages of the swelling and dissolution of gelatin and egg albumin and dissolution followed by boiling sugar, glucose syrup and water, adding inverted sugar to the boiled syrup and shaking the mixture [5]. The product obtained is characterized by the following ratio of its components: gelatin – 2.08%, water – 25.11%, albumin – 0.69%, sugar – 38.91%, the composition of syrups – 33.21%. These values were accepted as basic in the development of alternative marshmallow formulations without gelatin.

The gelling agents were selected taking into account both their individual effect and the synergistic effect exhibited when interacting with another polysaccharide [6]. The concentrations of the studied polysaccharides were taken within the range from 0.2% – the minimum value required for the exhibition of a gelling effect, up to 1% – the value above which the use of a polysaccharide is generally unprofitable.

Objects. The commercial samples of the following materials were used in the studies:

1. Food Gelatin, State Standard Specification № 11293–89, Russia;
2. Agar, Fujian Quanzhou Quangang Chemical Plant, China;
3. Iota Carrageenan, Sarda Starch Pvd. Ltd. India;
4. Kappa Carrageenan, Sarda Starch Pvd. Ltd. India;
5. Guar Gum, Sarda Bio Polymers Pvt. Ltd., India;
6. Locust bean gum from Ceratonia siliqua seeds, Sigma-Aldrich Co. LLC, USA;
7. Sodium Alginate, DuPont Nutrition & Health, France;
8. HMP, Genu Pectin Factory, Copenhagen, Denmark;
9. LMP (apple and citrus origin), ZPOW Pektowin, Poland;
10. Xanthan Gum, Danisko, Spain;
11. Pea Protein Isolate, Shandong Jindu Talin Foods Co., Ltd., China;
12. Dry Albumen «Albumix», Eurosab LLC, Russia;
13. Organic Brown Rice Protein, Jiangxi Golden Agriculture Biotech Co., Ltd., China;
14. Whey Protein Concentrate, Belorussia;
15. High Fructose Syrup F55, Shandong Scents Jianyuan Bio-Tech Co., Ltd., China;
16. Refined Milled-Sugar, State Standard Specification № 21–94, Russia;
17. Food Salt, State Standard Specification № R 51574–2000, Russia.

Quality evaluation methods. For the preliminary characterization of structural and mechanical properties of the marshmallow samples, the point system of organoleptic estimation was developed reflecting the elasticity of the material, its ability to retain its shape and increase in volume. The following estimates are used in the system: "+/-" – indicates that the studied quality is exhibited in the best way in the sample and remains unchanged over time; "+" – the sample characterizes itself satisfactorily; "+/-" – the initially satisfactory quality of the sample gets lost with time; "-" – the material shows itself unsatisfactorily.

For the deeper study of the samples obtained by means of technical means, the following types of analysis were carried out: a water activity and moisture test, a texture analysis, the measurement of surface tension and the microphotography of sections.

Water activity is one of the most important physical characteristics that determines the texture properties of the product, as well as the rate of chemical and biological reactions therein [7]. Water activity is usually understood as the ratio of the pressure of water vapor above the studied material to that above pure water at the same temperature. To find this parameter, a "HygroPalmAw" (Rotronic, Switzerland) instrument equipped with a dielectric moisture sensor was used in our studies [8]. The working principle of the sensor is based on a change in the conductivity of the hygroscopic polymer depending on the relative humidity of the chamber [9]. The measurements were carried out in accordance with the generally accepted procedure [10]. Subsequently, for the reliable interpretation of the obtained data, the moisture content of the samples was additionally established using a method for drying to a constant mass in a drying cabinet [11].

Later on, to study the structural properties of control and experimental marshmallow samples, a textural analysis method was used, implemented in the hardware and software complex "TA.XT plus Texture Analyzer" manufactured by "Stable Micro Systems". The method aims at carrying out the overall estimation of the samples from the point of view of their perception by man [12].

During the study, the samples were subjected to the controlled compression forces using a specially selected probe. The resistance of materials was controlled using a calibrated load cell. The loads determined as a result of the experiment are the functions of the properties and modes of testing the samples [13].

The surface tension value in the liquid medium that forms this foam has a higher value for the stability of the foam that is the basis of aerated confectionery products [14]. Thus, this parameter had been determined for the systems before they were shaken. The measurement was carried out using the high-precision surface tension meter K100 manufactured by "Kruss". This device implements a direct measurement method, continuously recording the resistance force tested by a specially selected probe when immersed in the studied medium [15].

RESULTS AND DISCUSSION

Table 1 presents the results of organoleptic estimation of the obtained marshmallow samples according to the described point system. The samples with the best indicators are highlighted in color.
Table 1. Results of the organoleptic estimation of the obtained marshmallow samples with the addition of egg albumin

| Item No. | Hydrocolloid | Concentration, % | Parameters | Shape safety | Increase in volume | Elasticity |
|---------|--------------|------------------|------------|--------------|-------------------|------------|
| 1       | Gelatin (water swelling) | 2.0 | ++ | ++ | ++ |
| 2       | κ-carrageenan (water swelling) | 1.0 | – | – | – |
| 3       | Agar (water swelling) | 1.5 | ++ | + | – |
| 4       | HMP (water swelling) with citric acid (40%) | 1.0 / 1.2 | + | – | – |
| 5       | HMP (dry mixing with sugar) with citric acid (40%) | 2.5 / 1.2 | – | – | – |
| 6       | LMP (dry mixing with sugar) | 1.5 | + | + | + |
| 7       | Sodium Alginate (water swelling) | 1.0 | – | – | – |
| 8       | Xanthan Gum & Guar Gum (separate water swelling) | 1.0 : 1.0 | + | – | – |
| 9       | Xanthan Gum & Guar Gum (combined water swelling) | 0.5 : 0.5 | – | + | + |
| 10      | Xanthan Gum & κ-carrageenan (water swelling) | 0.5 : 1.0 | + | – | + |
| 11      | Xanthan Gum & κ-carrageenan (water swelling) | 1.0 : 0.5 | ++ | + | ++ |
| 12      | Xanthan Gum & κ-carrageenan (water swelling) | 0.8 : 0.7 | ++ | + | ++ |
| 13      | Xanthan Gum & κ-carrageenan (water swelling) | 0.6 : 0.9 | ++ | + | ++ |
| 14      | Xanthan Gum & κ-carrageenan (water swelling) | 0.4 : 1.1 | – | – | – |
| 15      | Xanthan Gum & κ-carrageenan (water swelling) | 0.2 : 1.3 | – | – | – |
| 16      | Xanthan Gum & κ-carrageenan (water swelling) | 0.7 : 0.8 | ++ | + | ++ |
| 17      | Xanthan Gum & κ-carrageenan (water swelling) | 0.9 : 0.6 | ++ | + | ++ |
| 18      | Xanthan Gum & κ-carrageenan (water swelling) | 1.1 : 0.4 | +/– | + | + |
| 19      | Xanthan Gum & κ-carrageenan (water swelling) | 1.3 : 0.2 | +/– | + | + |
| 20      | Xanthan Gum & κ-carrageenan (water swelling) | 1.0 : 1.0 | +/– | + | + |
| 21      | Xanthan Gum & κ-carrageenan (water swelling) | 1.0 : 0.5 | +/– | – | + |
| 22      | Xanthan Gum & κ-carrageenan (water swelling) | 0.5 : 0.5 | – | – | – |
| 23      | Xanthan Gum & Carob (water swelling) | 1.0 : 1.0 | + | + | + |
| 24      | Sodium Alginate & HMP (dry mixing with sugar) | 1.0 : 1.0 | – | – | – |
| 25      | Sodium Alginate & Guar Gum (dry mixing with sugar) | 1.5 : 1.0 | + | + | + |
| 26      | Sodium Alginate & Guar Gum (dry mixing with sugar) | 1.0 : 1.0 | + | + | + |

Table 2. Composition of the studied marshmallow formulations, %

| Formulation number | Guar gum | Carob bean gum | Xanthan | Water | Whey protein | Egg albumin | Sugar | Invert syrup |
|--------------------|----------|----------------|---------|-------|--------------|-------------|-------|--------------|
| 1                  | 0.80     | –              | 0.70    | 31.56 | 0.64         | –           | 35.77 | 30.53        |
| 2                  | 0.80     | –              | 0.70    | 31.56 | –            | 0.64        | 35.77 | 30.53        |
| 3                  | 0.70     | –              | 0.80    | 31.56 | 0.64         | –           | 35.77 | 30.53        |
| 4                  | 0.70     | –              | 0.80    | 31.56 | –            | 0.64        | 35.77 | 30.53        |
| 5                  | 0.90     | –              | 0.60    | 31.56 | 0.64         | –           | 35.77 | 30.53        |
| 6                  | 0.90     | –              | 0.60    | 31.56 | –            | 0.64        | 35.77 | 30.53        |
| 7                  | –        | 1.00           | 1.00    | 35.47 | 0.60         | –           | 33.41 | 28.52        |
| 8                  | –        | 1.00           | 1.00    | 35.47 | 0.60         | –           | 33.41 | 28.52        |

As can be seen from Table 1, those samples have the best texture indicators which are prepared on the basis of agar and low-etherized pectin, the combinations of xanthan gum with guar gum and locust bean gum, and also the combination of sodium alginate and guar gum. The results obtained are in good agreement with the known data on the degree of synergism between hydrocolloids [16, 23]. For further studies, 8 formulations based on xanthan, guar and locust bean gum were selected as the cheapest options, Table 2.

Table 3. Water activity and the humidity of the studied samples

| Sample | a_w | W, % |
|--------|-----|------|
| 1      | 0.6741 ± 0.0040 | 20.42 ± 0.0018 |
| 2      | 0.6813 ± 0.0080 | 21.14 ± 0.0040 |
| 3      | 0.6985 ± 0.0102 | 22.85 ± 0.0087 |
| 4      | 0.6893 ± 0.0022 | 21.91 ± 0.0110 |
| 5      | 0.7218 ± 0.0017 | 25.16 ± 0.0133 |
| 6      | 0.7221 ± 0.0067 | 25.20 ± 0.0026 |
| 7      | 0.7725 ± 0.0050 | 29.27 ± 0.0081 |
| 8      | 0.7773 ± 0.0017 | 29.73 ± 0.0183 |
and disaccharides the highest decrease in water activity is provided by fructose. Glucose and lactulose have a less pronounced effect, and sucrose rounds out the given sequence [18]. It should be noted that to some extent the mass ratio of sucrose and invert sugar also affects the nature of the dependence of water activity on moisture. This is consistent with the known data on the highest efficiency of invert sugar compared with sucrose as a component that reduces water activity [19]. In this case, the combination of invert sugar and sucrose has a synergistic effect and provides a decrease in water activity by about 85%. The reverse ratio of the above components at the same concentration makes it possible to reduce the water activity by about 35%. The application of various volumes of polysaccharides does not practically affect a change in the water activity in the system [20].

The data obtained in the course of the experiment, as well as the rheological curves, Fig. 2a, based on them, describe one compression cycle and, in accordance with Fig. 2b, allow to determine the following characteristics of the product:
(1) the work spent on deformation and relaxation, as well as the overall work;
(2) sample hardness;
(3) the initial and final relaxation rates;
(4) Young's modulus;
(5) elasticity [21].

The force needed to chew the product is characterized by its rigidity and is equal to the peak value of the curve. The cohesiveness of the product determines the energy of molecular bonds therein, can be estimated from the energy necessary for product deformation and can geometrically be defined as the sum of the areas formed by the curve and the abscissa before and after reaching the rigidity peak [22]. In this case, the former area characterizes the adhesive force relative to the degree of density of the material being studied, and the inverse relation of the areas is elasticity, i.e., the ability of the product to undergo significant elastic deformations without destruction. More precise information about the elasticity of the product is provided using Young's modulus calculated as the ratio of the pressure on the sample to a change in its linear size during the test. Having constructed tangents to the initial and final points of the section that describes the relaxation time, it is possible to determine the angle of their slope and calculate the corresponding rates at which the sample is reduced. Table 4 presents the study results.

It follows from the obtained data that the properties of the samples vary significantly depending on the composition. Thus, Samples 3 and 8 significantly exceed the control in all the parameters, while samples 2 and 4 only slightly exceed the corresponding characteristics. In terms of total of its properties, sample 1 is the closest to the control and is only inferior to it in the final reduction rate. The behavior pattern of Sample 7 differs from that of the above being inferior to the control in a number of properties (work, hardness, the initial relaxation rate, Young's modulus) and in a number of properties – superior to it. Samples 5 and 6 surpass the control in terms of performance and rigidity, are approximately equal to it in the initial reduction and elasticity rate, but significantly surpass the control in terms of the final reduction rate and Young's modulus.
Table 4. Textural characteristics of the studied product

| Parameter                        | Sample 1 | Sample 2 | Sample 3 | Sample 4 | Sample 5 | Sample 6 | Sample 7 | Sample 8 | Control (C) |
|----------------------------------|----------|----------|----------|----------|----------|----------|----------|----------|-------------|
| Hardness work done, J            | 0.11     | 0.18     | 0.32     | 0.18     | 0.14     | 0.13     | 0.06     | 0.21     | 0.11        |
| Relaxation work done, J          | 1.95     | 3.18     | 5.58     | 3.27     | 2.59     | 2.30     | 1.23     | 3.75     | 1.96        |
| Total work done, J               | 2.06     | 3.35     | 5.90     | 3.45     | 2.73     | 2.43     | 1.29     | 3.96     | 2.08        |
| Hardness, g                      | 214.68   | 353.43   | 637.40   | 352.12   | 274.44   | 265.61   | 125.49   | 425.74   | 226.16      |
| Initial resilience rate, g/s     | 24.47    | 43.40    | 51.15    | 42.43    | 25.80    | 27.40    | 10.74    | 48.96    | 26.27       |
| Final resilience rate, g/s       | 3.65     | 1.78     | 3.71     | 0.78     | 2.03     | 2.03     | 1.00     | 4.24     | 0.80        |
| Young's modulus, Pa              | 2406.05  | 3961.10  | 7143.72  | 3946.42  | 3075.81  | 2976.85  | 1406.44  | 4771.52  | 2534.71     |
| Resilience                       | 18.04    | 17.94    | 17.47    | 18.49    | 17.79    | 17.22    | 19.48    | 17.59    | 17.14       |

Fig. 3. Surface tension forces in the samples before shaking.

Since it is known that gums used in the present paper refer to neutrally charged polysaccharides, the strong ionic and electrostatic types of interaction between their molecules are excluded [23]. This assumption is confirmed by the low energy of intermolecular bonds, which in turn clearly demonstrates the amount of energy consumed for the compression and relaxation of the sample. It follows therefrom that molecules interact mainly at the level of non-covalent associations with the formation of weak hydrogen bonds. At the same time, the rigidity of the obtained polymer systems reflects the quality of this interaction, when various amounts of these bonds cause changes in the rigidity of separate samples.

The reduction rate, different at the initial and final stages of relaxation, reflects the pattern of the highly elastic deformation of the sample as a result of a change in the shape of flexible polymer molecules that are curved or rolled up [24]. Accordingly, the material with a high relaxation rate has a more ordered structure. The latter indicates the degree of synergy between the components of the polymer system. Indeed, there is good correlation in the experimental results between the parameters of the relaxation rate and rigidity, which, as noted above, indicates the amount of stable bonds between the molecules of xanthan and locust bean gum.

As can be seen from Table 4, another consequence of a more dense and homogeneous texture is the high value of Young's modulus: the polymer networks with a higher concentration of knots are characterized by a higher resistance to compression under elastic deformation. At the same time, the elasticity of the material established as a result of the experiment gives a notion of the distance between the knots of the polymer network, which is approximately the same in all the analyzed samples.

Thus, in terms of total of properties, the samples the rheological curves of which are at the same level or higher than the curve for the control gelatin-based sample should be considered the most advanced from a technological point of view.

Figure 3 shows the curves obtained using a Krüss K100 apparatus for the surface tension forces acting on the probe, when immersed in the experimental system.

It can be seen from Fig. 3 that the surface tension forces decrease rather significantly over time, moreover, curve 1 decreases below such a low value as 10 mN/m within 35 s, and curve 7 becomes so close to it that brings these systems closer to classical gelatin-based ones. The time dependence is usually explained by the adsorption of one or more surface active components on the surface of the platinum plate hampered by the high viscosity of the system. However, the minimum value of $\sigma$ is unusual.

To interpret these curves, let us consider the components of the mixture. After water, invert syrup based on sucrose (35.77%), glucose and fructose (10.38%) takes the first place. It is known that the 70% syrups of the mentioned sugars have a value of surface tension of 46, 55 and 43 mN/cm, respectively [25]. According to other data [26, 27], the addition of sucrose only increases the surface tension of solutions (the article [26] provides a linear nomogram, and the article [27] shows the effect of inorganic substances). This cannot explain the noted decrease in surface tension, but once again indicates the significant effect of the method for its measuring on the result.
The surface tension of egg protein is estimated as 46.1–50.3 mN/m [28], moreover, the equilibrium value was established after 600 s (in our case, \( \sigma \) stops to change after 40 s). It should be noted that compositions 1, 7 and 3, the kinetic curves \( \sigma \) of which are below the others, contain whey protein, but not egg protein. Even the small amounts of whey protein sharply reduce \( \sigma \) of the aqueous solution [29]. Thus, 1% yields \(~46\) mN/m. The minimum value of \( \sigma \) is \(~43\) mN/m at 5%, then it begins to grow. The measurement procedure is based on using a tensiometer with a platinum ring manufactured by the same company (Krüss). However, composition 5 is another one that includes whey protein that differs little from composition 1 in the content of guar and xanthan gums and is identical to it in the rest.

The third group of ingredients is gums. The book [30] provides surface tension values for guar gum (55 mN/m) and locust bean gum (50 mN/m) for 0.7%. These values cannot explain a decrease in \( \sigma \) to 10 mN/m, in addition, various gums are used in compositions 1 and 7.

In the paper [31], the value \( \sigma \) of xanthanum was measured using Wilhelmi method with a platinum plate, depending on time. It decreased with time, which is explained by the diffusion and adsorption of polymer macromolecules on the platinum surface. The addition of low molecular salt (0.1 M NaCl) accelerated the equilibrium and lowered the equilibrium value of \( \sigma \). However, this time continued to be measured in hours, and the minimum value of the surface tension did not decrease below 45 mN/m.

Everything taken together makes it possible to propose that the noted decrease in surface tension to 10 mN/m is due to the rather rapid diffusion of a rather low-molecular component to the platinum plate and adsorption thereon. It is presumably a component of whey protein.

Figure 4 shows the photos of the cuts of the samples under study, on the basis of which it is possible to judge the quality of their texture. The following can be distinguished among the significant parameters: the thickness of the sample, the cleanliness of the cut, the size and distribution of gas cavities. Thus, the thickness of the sample indicates the ability of plastic mass for efficient aeration, the presence of various sags, burrs and torn edges on the cut – the adhesive properties of the product, and the size and uniformity of distribution of air bubbles in its volume – the consistency of the texture properties of the product.

In accordance with the above, the samples with the numbers 3 and 4 are characterized by the highest thickness, relatively clean cuts and the uniform distribution of the air bubbles that are approximately equal in size, which indicates quality texture. The samples with the numbers 1 and 2 are also quite large, they are easy to cut and they are evenly saturated with gas. In contrast to the former two couples, samples 5 through 8 exhibit high adhesion properties and, as can be seen from the figure, are less amenable to aeration.

Figure 5 presents a marshmallow preparation flow sheet.

![Fig. 4. Sections of experimental marshmallow samples.](image-url)
Fig. 5. Marshmallow preparation flow sheet

The RF patent No. 2626580 and the Iran patent No. 93555 "Protein and carbohydrate confectionery base and the method of its preparation" (developed by Klyukina O.N., Ptichkina N.M., Kodatsky Yu.A., Nepovinnykh N.V., S. Yeganehzad and R. Kadkhodaei) have been received for the developed marshmallow formulations and technology.

CONCLUSIONS

The organoleptic estimation, taking into account the elasticity of the system, its ability to retain its shape and the fact that it beats well, confirmed the presence of the developed synergy in hydrocolloid agar: low-etherified pectin and guar gum: xanthan gum / carob bean gum / sodium alginate couples. The studies of moisture and water activity in eight selected formulations with various ratios of xanthan and vegetable gums make it possible to classify the corresponding samples as a class of products with intermediate moisture. In addition, a close correlation was found between the parameters studied, and a synergistic effect was found between sucrose and invert sugar that leads to a sharp decrease in water activity. On the other hand, there is no noticeable effect of different concentrations of polysaccharides on water activity in the final product.

The obtained profile of the texture analysis gives an idea of the degree of effect of the product composition on its viscoelastic properties that vary widely. As follows from the obtained data, Sample 3 has the most quality and ordered structure, which makes it the most promising alternative to gelatin-based marshmallow.

It is obvious that the graphs for surface tension indicate the prolonged adsorption, the pattern of which is still unclear and needs a further study. At the same time, it was found that the pattern of the gelatin curve...
is completely opposite to the curves that correspond to all other polysaccharides and this is of interest, too. In all other respects, the experimental data indicate that the liquid preparations under study have a surface tension sufficient to provide stable foaming in the final product, which is not inferior to the classical marshmallow formulation from this point of view.

Finally, the photos of the cuts clearly demonstrate the quality of the texture of the produced samples. In fact, the samples with the numbers 3 and 4 are characterized by the best structure: they are large, the air bubbles are distributed evenly inside them. Together with the obtained graphs, these images indicate that the claimed samples are the best in this experiment and can be recommended for production.

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