DEVELOPMENT OF WRAPPING PAPER WITH IMPROVED OPACITY, STRENGTH, AND WHITENESS

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1. Introduction

The studies belong to the field of pulp and paper industry. Results could be used in the production of paper for packing food products, which should comply with increased demands for the indicators of opacity, whiteness, and mechanical strength.

In Ukraine, acting standards GOST 1341-97 «Vegetable parchment. Specifications», GOST 1760-86 «Greaseproof paper. Specifications», GOST 8273-75 «Wrapping paper. Specifications» regulate the production of paper with a mass of 60 g, 80 g, 100 g, and higher, per 1 m². According to these standards, it is necessary to introduce up to 14% of the mineral filler of the mass of absolutely dry fiber [1] to its composition to provide the necessary level of indicators for whiteness, opacity, mechanical strength of the paper. However, high content of mineral filler leads to a significant reduction in the mechanical strength of paper — breaking length or destructive effort. This will not meet the requirements of sanitary norms and rules of its use for the manufacturing of wrapping products.

Wrapping paper should have matt surface with a whiteness of 70–88%, opacity of at least 90%, and high mechanical strength. It should not contain optical bleaches. Its manufacturability should ensure reprocessing with the use of modern high-speed polygraphic equipment and long-term exploitation of products made on its basis.

Wrapping paper must have certain properties, which are characterized by a set of indicators that ensure high quality of products manufactured on its basis. Homogeneity of paper structure, capability to perceive the printing ink, whiteness, surface smoothness, microgeometry of the surface, softness, resistance to dust, resistance of the surface to plucking exert influence on the printing properties of paper. Among the factors that determine the quality of printed products are also a degree of contrast between printed and unprinted areas, sharpness of the print, its translucency, that is, the indicators of paper opacity.

Opacity is the capability of paper to withstand penetration of light, a property that ensures the non-transparency of the back of a paper cloth and determines the ability to cover visually black prints on the laid-under paper or on the opposite side of the same paper. This indicator is especially important for double-sided copying when the print is applied on both sides of a sheet of paper.

The lower the mass of a paper per 1 m², the more difficult it is to provide the uniform transparency of its structure, opacity, whiteness, and mechanical strength. Exactly this complex of properties ensures high quality of printed materials, terms and conditions of use.
The strength, printing and deformation properties of the paper depend on the strength of the forces of fiber interconnection, the size, the degree of development and strength of the fibers themselves and their location in the structure of the paper. It is possible to achieve the required level of printing properties of the paper, uniformity and homogeneity along the length and width of the cloth using a fibrous mass with appropriate sizes of cellulose fibers or with an appropriate degree of their grinding.

It is almost impossible to arrange the production of fibrous matter with the same fiber size and create such conditions and perform grinding so that all the fibers are processed equally. This is particularly true for grinding in conical or disk mills of continuous action. Due to the unevenness of paper according to the indicators mentioned, the paint is fixed only on the protruding roughness of paper. As a result, the printed sample will be uneven (it will have spaces – unprinted areas), and this will lead to the rejection of product.

Under current conditions, more and more food manufacturers are trying to increase the popularity of their products at the expense of attractive wrapping. The high aesthetic properties of wrapping material should be accompanied by the ability to preserve the wrapped product. Therefore, the development of paper with high printing properties, increased opacity, strength, and whiteness for food products is a relevant task.

2. Literature review and problem statement

Scientists from many countries of the world are working to increase opacity, strength, and whiteness of paper.

A method for increasing the whiteness of paper on the basis of bleached sulfite cellulose ground to a grinding degree of 35 °SR was developed. Gluing compound, coagulant, and mineral filler – kaolin 8–23% of absolutely dry fiber in paper [2] is added. The achieved level of paper whiteness is 78%. However, this method cannot be widely used since the achieved level of the whiteness of paper is low. At the same time, high content of filler in the paper (23%) leads to a significant reduction in the mechanical strength of paper.

Increasing the whiteness of paper is achieved through filling with kaolin, modified cesium [3]. However, the use of such a filler in the production of wrapping paper for food products is dangerous for the health of consumers.

A process for manufacturing wrapping paper was proposed, the composition of which is added with, as a filler, in addition to kaolin, zeolite at the total content of the filler mixture in paper of 12.3% of the absolutely dry fiber [4]. However, zeolite has abrasive properties. Therefore, its use with a high content in paper can cause damage to nets, cloths, press rolls and a mirror polished surface of the drying cylinders of a paper converting machine. This will lead to additional costs for the elimination of these effects.

The process of offset paper manufacturing was developed, which implies grinding of hardwood and softwood bleached cellulose until reaching the average length of fibers at 2.2–2.5 mm and 0.9–1.1 mm, respectively. As a filler, kaolin is used. This makes it possible to provide the required level of opacity, but the paper obtained in this way has a high mass of 1 m² – 200 g/m² [5]. According to results of theoretical calculations based on the dependence of Gurevich-Kubelka-Munk, one should note that it is almost impossible to achieve the desired level of opacity of paper with a mass of 1 m² – 60–65 g using kaolin as a filler. To achieve this, it would be advisable to increase the paper’s ash content to 24–28% by increasing kaolin content in the paper composition to the same level. In this case, the strength of paper intended for the production of wrapping products reduces substantially.

Increasing the barrier properties of paper is also achieved through the use of biopolymers [6]. However, due to hydrophilicity, crystallization, fragility, and instability of the melts, there may occur difficulties in processing most biopolymers, which impedes full application at industrial scale [7].

Calcium carbonate is used to reduce crystallization of cellulose [8]. However, in this case, high values of quality indicators of the paper can only be achieved under condition of gluing with a large part of polymeric materials.

The use of nanotechnologies is developing in the manufacturing of wrapping materials for food products. However, high selectivity of nanostructures makes it possible to use them for a wide range of goods [9]. In particular, a method for the separation of crystalline cellulose is proposed, which makes it possible to increase strength and density of paper; however, its whiteness and stability of printing are lost in this case [10].

Another direction of the application of nanotechnologies in the production of paper is the regulation of temperature of cellulose agitation [11]. It increases the strength of paper only, but does not improve its aesthetic characteristics.

Significant increase in the strength and opacity of paper is achieved by gluing with melamine and urea resins [12]. However, the use of such fillers is not recommended in case of contact with food.

The methods of increasing barrier properties were developed, implying the saturation of paper with a composition based on polyvinyl alcohol and epichlorohydrin resin. Due to the application of this method, high value for the indicators of water and grease resistance is achieved [13], however, aesthetic properties were not investigated.

Thus, different ways, methods, approaches, and technological solutions are used in each case to ensure the necessary level of this or that quality indicator of the wrapping paper. However, none of these solutions yields a comprehensive result in increasing the values of quality indicators of wrapping paper. The increase in whiteness and opacity is achieved due to factors that reduce the strength of paper. Thus, the task of a simultaneous increase in the whiteness and opacity of paper while maintaining its strength is not resolved. Processes of achievement of uniformity of paper quality indicators over the entire width of cloth also require further investigation.

3. The aim and objectives of the study

The aim of present study is to increase the opacity, strength, and whiteness of wrapping paper by combining cellulose of various types, as well as methods of grinding, titanium dioxide and polyvinyl alcohol.

To achieve this objective the following tasks were set:

– an increase in opacity and mechanical strength of food wrapping paper;

– maintaining the whiteness of paper without use of optical bleaches;

– improvement in color acceptance and printing stability by utilizing properties of the microstructure of cellulose fibers, mineral fillers and chemical gluing and binding agents.
4. Developing a method for the production of wrapping paper with enhanced opacity, strength, and whiteness

The required quality level of paper for food wrapping is ensured by the following indicators: whiteness 70–88 % (without the use of optical bleachers), smooth surface without gloss, opacity (not less than 90 %), mechanical strength. Such paper should also comply with the requirements of SanPiN42-123-4240-86 «Permissible quantities of migration (PQM) of chemical substances released from polymeric and other materials in contact with food products, and methods for their determination». The achievement of such indicators requires an integrated approach to defining requirements for cellulose fibers, gluing, binding, and strengthening agents, their correlation and interconnection in the structure of paper.

The structure of paper for food wrapping is a set of fibers chaotically oriented in space, axes of which are mainly orientated parallel to the plane of formation of the cloth. In this case, strength is provided only by direct fiber bonds. That is why the particles of mineral filler, which are introduced into the composition of such paper, break these connections due to their location between the fibers of cellulose in the structure of paper. This reduces the strength of paper and products made on its basis. Therefore, to reduce the negative effect, the filler must be a finely dispersed (with the smallest diameter of the particles) material. This provides a high unrolled surface and porosity and satisfactory sorption capacity of paper, which are important when applying paint on its surface. Application of mineral filler of high dispersion degree contributes to obtaining paper with the required indicators of mechanical strength, softness, and elasticity. High levels of opacity and whiteness are important for paper used for food wrapping.

Thus, by using mineral highly-dispersed fillers, gluing and binding chemicals with prepared cellulose fibers, it is possible to obtain paper with a new purpose, to predict and ensure achievement of specialized quality indicators.

For the typography method of printing, the most widespread process of paper manufacturing from bleached sulphate cellulose of softwood and bleached sulphate cellulose of hardwood, which is chosen as an analogue, is the closest to its intended purpose and technical essence [14].

In the specified process of paper production, grinding of sulphate softwood cellulose is carried out to a degree of grinding of 18–20 °SR. Further grinding is carried out in a conical mill to a grinding degree of 37–40 °SR; grinding of hardwood cellulose is carried out to a grinding degree of 21 °SR. Grinded fiber of softwood and hardwood cellulose is mixed in the pool; the gluing agent and kaolin filler are added in a mass fraction of 10 % from absolutely dry fiber. For the manufacturing of paper, a mixture of ground fibers of softwood and hardwood cellulose is used at the ratio of % by weight, 80:20; 70:30; 60:40; 50:50.

The basis of present study is the solution to the problem on creating a paper cloth with a high degree of opacity and convenience of printing of the required information, which is applied on both sides of the sheet. The paint should not be reflected on the reverse side and migrate directly into the product. The problem is solved by choosing and combining fibers of cellulose on the basis of softwood and hardwood, methods and quality of cellulose fibers grinding, fraction particle sizes of mineral filler, preparation of paper pulp for the formation (pouring out) of structure.

Different printing methods put forward different demands for paper quality indicators. However, the main and important factor, besides opacity, whiteness and mechanical strength, which affects quality of the printed production, regardless of the type of printing, is still the heterogeneity of the structure, and hence the properties of the paper cloth.

Paper heterogeneity is the main reason for uneven ink absorption in the printing process, which results in a decrease in the range of image grading, not printing of its parts and receiving a spotted imprint.

The sulphate cellulose is most suited to such requirements. Its fibers are lighter than those of the sulfate, easier to bleach and grind. It is used traditionally for the production of various types of paper for printing, it is flexible and elastic. This contributes to maintaining and binding the filler in the structure of paper.

Fibers on the basis of sulfate bleached cellulose of softwood and sulfate bleached cellulose of hardwood are evenly distributed throughout the matrix. This creates a three-dimensional basis with the effect of grid structures and contributes to maintaining a mineral filler inside this system (Fig. 1).

![Fig. 1. Microstructure of paper with a filler (x640): a — based on sulfate bleached cellulose of softwood, b — based on sulfate cellulose of softwood and hardwood](image)

The desired high homogenization (alignment) of the structure of paper is achieved as a result. In general, due to the uniform distribution of all the components involved in the pouring of paper cloth, the formation and complex of its properties improve, which in turn leads to an improvement in the quality of paper products in general.

For the production of paper, specially designed fine-ground pulp fibers (hereinafter FGPFs) are used — natural fibers of softwood and hardwood pulp the particle size from 20 to 2000 μm. They play the role of a fibrous amorphous additive to the paper pulp. This contributes to an increase in the opacity of paper and maintenance of mechanical strength (Fig. 2), uniformity of structural and physical properties of paper along and across the paper cloth (Fig. 3).

The studies we conducted showed that for the production of paper for wrapping the food products, the best results were obtained when using cellulose with a mass share of fractions of 650–680 μm at 94–96 %. For other ratios, either the opacity or the mechanical strength of the paper is lost.

Ground mixture of FGPF at a mass fraction from 0.8 to 2.2 % of absolutely dry fiber has optimum properties as an additive to the paper pulp. Such ratio makes it possible to stabilize the pulp and, accordingly, the properties of paper over the entire width of the cloth. The technology of paper production involved grinding and preparation of a paper pulp composition, introduction of filler, gluing and binding agents,
dilution with water, pouring, pressing, and drying of a paper cloth.

Achieving the result of improved indicators of opacity and whiteness of paper for wrapping the food products is fulfilled by using sulfate bleached cellulose of softwood (SBCS) and sulfate bleached cellulose of hardwood (SBCH) in the composition. It is important to keep the appropriate degree of grinding of fibers and the ratio in paper pulp. This is achieved by controlling the raw material arriving directly into the paper-making machine. As an additive, natural FGFPF, with a whiteness of 88%, with a mass share of fractions with a fiber size of 640–680 μm at 90–98%, at a mass share of 0.8–2.2% of absolutely dry fiber. Titanium dioxide in different proportions and mass fraction of absolutely dry fiber is introduced to paper pulp as a filler. The mass fraction of the filler is 2–6% of absolutely dry fiber. The introduction of gluing substances and the manufacturing of paper was carried out using a paper-making machine with flat sieves in line with technological procedure shown in Fig. 4, but, as a binder, polyvinyl alcohol-fiber (PVA) was introduced at a mass fraction of 0.7–1.0% of absolutely dry fiber. The specified range is based on the results of previous studies [13].

The use of titanium dioxide as a filler is explained as follows: as a mineral filler for paper, kaolin is commonly used, which contributes to the growth of the paper’s initial opacity. However, in the presence of gluing and binding agents, kaolin has relatively little effect on opacity and may even strengthen transparency of paper [15].

Titanium dioxide, in comparison with other mineral fillers found in paper composition (for example, kaolin), has a high degree of dispersion and refractive index, with whiteness exceeding 96%. Therefore, the best consumer properties are achieved when creating mixtures of fillers based on titanium dioxide and swollen perlite: opacity, mechanical strength, and whiteness of paper for wrapping the food products. In this case, consumption of the mixed filler in comparison with kaolin is negligible. After all, in paper for the offset printing method, the content of kaolin in accordance with the requirements of GOST 9094-89 «Paper offset printing. Specifications» reaches 14%. Opacity of paper depends on the scattering coefficients of paper components. This, in turn, depends on size of the filler particles: if the size of filler particles reduces to the corresponding optimum, the scattering coefficient and paper opacity increase.

Fig. 2. Opacity and discontinuous paper length depending on the size of cellulose particles and mass fraction particle

Fig. 3. Difference in the mass of paper with an area of 1 m² in 8 points depending on the FGFPF mass share

Fig. 4. Technological procedure of paper production

Fig. 5. Opacity of paper depending on the type and mass fraction of the filler
The production of paper samples was carried out in a paper-making machine KM-1 in the following way: sulfate bleached cellulose of softwood and sulfate bleached cellulose of hardwood were milled after separate spreading in a hydraulic separator (1.5 m³) to achieve a degree of grinding of 42–44 °SR and 24–26 °SR, respectively. Ground fibers of two types of cellulose were mixed together in water to create a fibrous composition in different ratios. A titanium dioxide filler, FGPF, and a polyvinyl alcohol fiber at a different mass fraction of absolutely dry fiber were added to the resulting suspension of the fibrous composition. Samples of paper with a mass of 60 g per 1 m² were made from the paper pulp obtained according to the proposed variants and the analogue (Table 1).

### Table 1

Formulation of compositions of the examined paper samples

<table>
<thead>
<tr>
<th>Composition components</th>
<th>Samples</th>
<th>Proposed variants</th>
<th>Analogues [14]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Sulfated bleached cellulose of hardwood: degree of grinding, °SR</td>
<td>42</td>
<td>42</td>
<td>42</td>
</tr>
<tr>
<td>ratio, % by weight</td>
<td>78</td>
<td>86</td>
<td>90</td>
</tr>
<tr>
<td>Sulfated bleached cellulose of hardwood: degree of grinding, °SR</td>
<td>24</td>
<td>24</td>
<td>24</td>
</tr>
<tr>
<td>ratio, % by weight</td>
<td>22</td>
<td>14</td>
<td>10</td>
</tr>
<tr>
<td>FGPF, % of absolutely dry fiber</td>
<td>0.8</td>
<td>1.2</td>
<td>1.8</td>
</tr>
<tr>
<td>Titanium dioxide</td>
<td>0.65</td>
<td>0.75</td>
<td>0.85</td>
</tr>
<tr>
<td>Polyvinyl alcohol fiber, % of absolutely dry fiber</td>
<td>0.65</td>
<td>0.75</td>
<td>0.85</td>
</tr>
</tbody>
</table>

Thus, when comparing properties of the proposed composition and the analogue-paper, it is possible to determine the effect of FGPF, titanium dioxide, and polyvinyl alcohol fibers on them.

5. Results of examining wrapping paper with improved opacity, strength, and whiteness

The obtained samples of paper were tested for the indicators of opacity, whiteness, and discontinuous length in accordance with GOST 8273-75 «Wrapping paper. Specifications», as well as the uniformity in the indicators of mass of 1 m² and humidity in 8 points along width of the sheet. Results of testing the samples are given in Table 2, 3.

The proposed technology makes it possible to obtain paper pulp, cellulose fibers of which are characterized by high specific surface of the dispersed phase. In this case, in the course of the proposed method of cellulose grinding and as a result of its swelling, there is a loosening of the structure of the fiber. This contributes to its longitudinal and transverse splitting into flexible anisometric particles and fibrillation (combining into thin fibers – whiskers). A significant number of reactive hydroxyl OH groups of cellulose is also released when this method is applied. They participate in the formation of intermolecular bonds between elements of the suspension of paper pulp (cellulose fiber, binders and gluing compounds, filler) in the process of pouring and forming a paper cloth.

To increase mechanical strength and resistance of paper to abrasion, to reduce linear deformation of paper for writing and printing under high humidity conditions, and also to bind the components of paper pulp, a binder component from the polymer dispersion – polyvinyl alcohol fiber was introduced into its composition. It is characterized by the capability to soften partially in the wet state under the action of elevated temperature and to bind (glue) the base cellulose fibers firmly. In turn, fine-dispersed particles of mineral filler are arranged between them and on their surfaces (Fig. 6).

### Table 2

Results of examining paper for the indicators of strength, opacity, and whiteness

<table>
<thead>
<tr>
<th>Indicator name</th>
<th>Indicator value, samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proposed variants</td>
<td>Analogues [14]</td>
</tr>
<tr>
<td>Mass fraction of filler, % (titanium dioxide)</td>
<td>2 4 5 6 7 Kaolin 10 Kaolin 14</td>
</tr>
<tr>
<td>Mass fraction of FGPF, %</td>
<td>0.8 1.2 1.8 2.0 2.2 – –</td>
</tr>
<tr>
<td>Mass fraction of PVA binding agent, % by weight</td>
<td>0.65 0.75 0.85 0.95 1.0 – –</td>
</tr>
<tr>
<td>Mass of paper with an area of 1 m², g</td>
<td>60.2 60.2 60.2 60.3 60.2 60.3 60.3</td>
</tr>
<tr>
<td>Discontinuous lengths, m</td>
<td>5480 5480 5460 5380 5320 3500 3000</td>
</tr>
<tr>
<td>Opacity, %</td>
<td>86 88 88 90 92 90 90 90</td>
</tr>
<tr>
<td>Whiteness, %</td>
<td>82 84 86 88 88 82 84 84</td>
</tr>
<tr>
<td>Linear deformation of paper, %</td>
<td>1.4 1.4 1.2 1.2 1.0 2.6 2.2</td>
</tr>
</tbody>
</table>

### Table 3

Results of examining paper for the indicators of mass and humidity in 8 points

<table>
<thead>
<tr>
<th>Indicator name</th>
<th>Indicator value, samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proposed variants</td>
<td>Analogues [14]</td>
</tr>
<tr>
<td>Mass of paper with an area of 1 m², range in 8 points, g</td>
<td>60.2–60.3 60.2–60.4 60.2–60.3 60.3–60.5 60.3–60.5 60.1–60.9 60.2–61.1</td>
</tr>
<tr>
<td>Humidity, range in 8 points, %</td>
<td>5.5–5.8 5.6–5.8 5.6–5.8 5.8–5.8 5.7–5.8 5.2–5.8 5.4–5.8</td>
</tr>
</tbody>
</table>
A comparative analysis of the research results shows that high mechanical indicators of paper strength are achieved not only by mechanical interweaving of elements of the fiber composition but also by partial fusion (gluing) of the particles of the binder – polyvinyl alcohol fiber, fibers of cellulose, and particles of the filler in paper.

Polyvinyl alcohol fibers as binder and ground cellulose fibers enhance the adhesion of fibers in the paper structure and contribute to the growth of mechanical and adsorption holding of the filler particles by cellulose fibers. This provides paper with a uniform structure, increases its strength, smoothness, and resistance to plucking the fibers and particles of the filler during ink printing and friction in comparison with the paper-analogue.

As can be seen from analysis of the research results, the introduction into the composition of paper up to 2–5% of the combined filler based on titanium dioxide and FGPF did not practically reduce its mechanical strength and whiteness compared to the paper without a mineral filler. However, the degree of opacity increased to a large extent due to the insignificant consumption of filler compared to the use of traditional mineral fillers and paper-making methods.

The paper-analogue is characterized by uneven mass indicators per 1 m² and humidity in 8 points, which reduces quality of color acceptance by the surface of paper during printing (Fig. 7).

The increased value of deformation strength indicator results in the incompatibility of paints during multi-color printing. This is confirmed by results of comparison of the proposed samples with analogues in terms of print sharpness and color acceptance (Fig. 8).

The designed samples of paper, compared to analogues, differ by the uniformity of quality indicators along width of the sheet, by improved indicators of opacity, strength, and whiteness, as well as by a decreased indicator of the linear deformation of paper cloth when wetted. This indicates maintaining high quality of printed products and uniformity in the application of developed paper, which makes it possible to utilize it for the production of wrapping materials for food products.

By employing results of the study conducted, it was possible to design a paper wrapping material, which possesses simultaneously high functional (strength and opacity), aesthetic (whiteness), and technological properties (color acceptance and print sharpness). The latter is a particularly important element in the marketing strategy of food manufacturers because it enables creation of packaging that is attractive to consumer and may increase sales.

The above results were achieved by introducing into the composition of paper up to 2–5% of the combined filler based on titanium dioxide, FGPF, and polyvinyl alcohol fibre.

Results of the study conducted could be applied when manufacturing wrapping paper, as well as while choosing a packing material for one or another food product. This is especially important at present – under conditions of European integration and production of goods that meet international standards.

In the future, it is expedient to carry out studies into changes in the quality of products packed in the designed wrapping material.

7. Conclusions

1. The method we developed for the production of wrapping paper with improved opacity...
and strength implies the following: fibers of sulfate bleached cellulose of hardwood and softwood are exposed to the processes of breakdown in a hydraulic separator, to grinding and purification of the fiber suspension with the introduction of mineral fillers, gluing agents, pouring (forming) of a paper cloth. Grinding of bleached sulfate cellulose of softwood leads to a degree of grinding of 42–44 μm; the bleached sulfate cellulose of hardwood – to a grinding degree of 24–26 μm. Fine-ground cellulose fibers at a mass share of fraction of 640–680 μm at 93–95 % are used. The obtained fractions of fibers are mixed before the formation (pouring) of paper, % by weight: softwood cellulose – 78–92, hardwood cellulose – 21.2–5.8, FGPF – 0.8–2.2. As a binder agent, polyvinyl alcohol fiber is introduced at a mass fraction of 0.7–1.0 % of absolutely dry fiber. The set of such activities makes it possible to increase discontinuous paper length to over 5000 m and opacity to 92 %, while the linear deformation decreases by 2–2.5 times in comparison with the analogue.

2. Achieving the level of whiteness of 88 % is ensured by the introduction of titanium dioxide to paper pulp as a filler, with a mass share of fraction of 2–3 μm at 95–96 % and whiteness (reflection coefficient) of 94 %, at a mass fraction of 2–6 % of absolutely dry fiber.

3. The use of polyvinyl alcohol fiber also makes it possible to improve color acceptance and stability of printing. This is due to the creation of resistance to the plucking of fibers and particles of the filler by paint during printing and friction. The results are confirmed by the uniform mass indicators per 1 m² and humidity in 8 points.

References