The structure formation of composite materials based on latex-impregnated non-woven needle-punched sheets

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The structure formation of a composite material produced by latex impregnation of porous materials of different nature is determined by the impregnation regimes, which should ensure that the pores are filled with latex, and also by squeeze rolling of the impregnated material. Filling of the porous material with latex depends on the structure of the pores and the dry residue of the latex. Squeeze rolling of the impregnated sheet is carried out with a roll gap smaller are larger than the thickness of the porous material. With a roll gap greater than the thickness of the porous material, a composite material with a surface film of rubber is obtained. With a roll gap smaller than the thickness of the porous material, the rubber is distributed in the bulk of the material.

Needle-punched non-woven sheets comprise a highly porous fibrous material with a coefficient of porosity of 0.8–0.9 and a system of communicating pores. The porous structure of the sheet is conducive to complete filling with latex, and the degree of filling is determined by the dry residue of the latex. With a squeeze roll gap smaller than the thickness of the sheets, porous composite materials with increased mechanical properties [1, 2] and controllable permeability [3] are obtained.

By comparison with the impregnation of low-porosity materials, the main problem of which is filling of the pores with latex, the structure formation of a composite material based on latex-impregnated sheet is determined by the processes occurring during drying. In the process of drying of the impregnated material, structure formation of the composite material occurs with a constant volume of the sheet, or the sheet volume changes, generally through shrinkage. With an equal degree of impregnation, composite materials produced with a constant volume of the sheet or with shrinkage have different physicomechanical properties [4]. Methods for monitoring the process of structure formation of composite materials and the establishment of the dependence of the structure of the composite materials on the structure of the non-woven sheet used for impregnation are of practical importance, and are the focus of this work.

To produce composite materials, use was made of non-woven sheets of different structure (Table 1), manufactured from polyester fibre (TU 6-13-0204077-95-91) of 20 μm diameter. Fibrous mats were obtained by the mechanical forming method with subsequent punching.

<table>
<thead>
<tr>
<th>Specimen number</th>
<th>F × 10³, kg/m²</th>
<th>d × 10³, m</th>
<th>ρₛ, kg/m³</th>
<th>k/ρₛ, rel. units</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>220</td>
<td>1.6</td>
<td>131</td>
<td>1.02</td>
</tr>
<tr>
<td>2</td>
<td>260</td>
<td>2.8</td>
<td>90</td>
<td>1.24</td>
</tr>
<tr>
<td>3</td>
<td>373</td>
<td>3.3</td>
<td>115</td>
<td>0.98</td>
</tr>
</tbody>
</table>

Note. F – surface density of sheet; d – thickness of sheet; ρₛ – density of sheet

Table 1. The structural characteristics of non-woven sheets and the parameter of formation of composite materials based on them

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with a density of treatment of 180–320 cm\(^{-2}\) [5], which ensured variation of the structure of the sheets (Table 1).

The sheets were impregnated with latex DVKhB-70 with a dry residue of 26% (TU 38.303-04-03-90). Variation in the degree of impregnation was achieved by diluting the latex with distilled water. For complete filling of cavities with latex, the specimen immersed in latex was rolled with a roller. After impregnation, the specimen was passed through squeeze rolls with a controllable roll gap, which amounted to 0.6 of the thickness of the sheet used for impregnation. Drying of the specimen to constant weight was done by air blowing at a temperature of 60°C. The degree of impregnation of the sheet (\(C_m\)) was calculated from the expression:

\[
C_m = \frac{m_1 - m}{m}
\]

(1)

where \(m_1\) and \(m\) are respectively the mass of the impregnated material and sheet, kg.

The formation of a composite material with a constant volume of the sheet or with change in its volume is reflected by the different influence on density. The density of the composite material produced with a constant volume of the sheet depends only on the degree of impregnation and is directly proportional to the content of rubber. Shrinkage of the sheet occurs with a simultaneous increase in mass and reduction in volume, which limits assessment of the form of the dependence of density on degree of impregnation. Equations describing linear dependences of density on degree of impregnation with a constant volume of the sheet have the general form:

\[
\rho = \rho_n + kC_m
\]

(2)

where \(\rho\) and \(\rho_n\) are respectively the density of the composite material and non-woven sheet, kg/m\(^3\), and \(k\) is the coefficient of dimensionality, kg/m\(^3\).

The coefficient \(k\) reflects the change in density of the composite material with unit change in the degree of impregnation.

Equation (2) can be written in the form of a more convenient equation for analysis of the mechanism of structure formation of the composite material:

\[
\rho = \rho_n \left(1 + \frac{k}{\rho_n}C_m\right)
\]

(3)

The dependence of the density of the composite material only on the degree of impregnation is reflected by fulfillment of the condition \(k/\rho_n = 1\). Reduction in the volume of the non-woven sheet is reflected by fulfillment of the condition \(k/\rho_n > 1\). It should be pointed out that fulfillment of the condition \(k/\rho_n > 1\) reflects the process of structure formation of the composite material that occurs with increase in the volume of the sheet.

Thus, the magnitude of the proposed parameter \(k/\rho_n\), which is determined from the dependence of density on degree of impregnation, reflects the mechanism of structure formation of the composite material and its dependence on the structure of the sheet. The dependences of the density of the composite materials on the degree of impregnation of sheets of different structure are presented in Figure 1, and the values of the parameters \(k/\rho_n\), obtained from these dependences are presented in Table 1.

As can be seen from Table 1, the formation of composite materials produced by impregnating non-woven sheets of >115 kg/m\(^3\) density occurs with retention of the volume of the sheet (the values of \(k/\rho_n\) for these materials approach unity). For a composite material produced by impregnating sheet of 131 kg/m\(^3\) density, shrinkage is absent up to the maximum degree of impregnation achieved in the work, equal to 0.8. With reduction in the density of the sheet to 115 kg/m\(^3\), shrinkage is absent with a degree of impregnation of less than 0.3. With a degree of impregnation greater than this critical value, the density indices of the composite material deviate from a linear dependence, become lower than the theoretical density obtained by extrapolation of the dependence of density on degree of impregnation with a degree of impregnation greater than the critical value (the broken line in Figure 1), and approach a constant magnitude (see Figure 1).

When non-woven sheet of minimum density is used, the formation of the composite material is associated with shrinkage, which occurs with a degree of impregnation of less than 0.2. The linear section of the dependence of density on degree of impregnation (Figure 1) reflects constant shrinkage with a unit increase in degree of impregnation. With a degree of impregnation of over 0.2, the density of the composite material, like the density of material produced using sheet of 115 kg/m\(^3\)
density, becomes lower than the theoretical density and approaches a constant magnitude (Figure 1).

The dependence of the structure formation of the composite materials on the density of the sheets is determined by the specific nature of the technology used for their impregnation. The industrial impregnation process includes the following stages: saturation of the sheet with latex in a dipping bath, transfer of the impregnated sheet in the free state to squeeze rolls, pressing in the gap of the squeeze rolls to remove excess latex, recovery of the volume of the sheet after leaving the squeeze roll gap, and drying.

The laboratory procedure for producing specimens ruled out any dependence of the structure formation of the composite material on the stage of latex saturation of the fibrous matrix. Prolonged and intense rolling with a roller of a specimen immersed in latex ensured complete filling of its volume with latex when latex with a larger value of dry residue was used. The remaining stages of industrial impregnation were simulated by using strips of sheet, the free end of which was fed into the squeeze roll gap, which ensured continuous movement and a constant feeding of impregnated strip into the squeeze rolls. Drying of the impregnated specimen was carried out by blowing with heated air, which is characteristic of an industrial unit.

Another factor of structure formation of the composite material is the specific nature of the structure of the non-woven sheet. Non-woven sheet has sections that differ in fibre packing density and orientation (Figure 2). Some fibres form bunches, which consist of through-thickness-oriented fibres with a relatively high packing density. Bunch formation is a consequence of the capture of fibres by the needle barbs [5]. Between the bunches, the fibres are oriented in the plane of the sheet, and their packing density is lower than the packing density in the bunches. Increase in the density of the sheet leads to an increase in the number of bunches, their thickness, or the number of fibres in their composition, and the length of the bunches approaches the thickness of the sheet (Figure 2).

During drying there is a reduction in the length of the ‘bridges’ of rubber particles that are formed between fibres in the process of impregnation. A consequence of the reduction in the length of these bridges is the movement of fibres and a reduction in the distance between them, which causes shrinkage of the sheet. The proposed mechanism of shrinkage explains the dependence of its occurrence on the density of the non-woven sheet. For sheets of relatively high density, which reflects the process of effective formation of bunches, a considerable proportion of the fibres enter the composition of bunches and have limited mobility. The limited mobility of the fibres is reflected by low shrinkage of the sheet in the process of drying of the impregnated material. Fibres in sheet of minimum density do not enter the composition of bunches and therefore possess high mobility, which leads to shrinkage during drying.

The non-uniform distribution of fibre packing density and orientation leads to a different capacity of individual sections of the sheet to hold sorbed latex. Therefore, at different stages of impregnation, the redistribution of latex occurs in the sheet. When the sheet leaves the dipping bath and moves in a free state towards the squeeze rolls, latex flows out of sections of fibres of low packing density and remains in the bunches (Figure 3). A local distribution of latex is most characteristic of sheets of high density or high efficiency of bunch formation. Filling of the bunches with latex is reflected by the formation of drops of complex shape on the surface of the sheet (Figure 3). A small part of the latex is retained in the form of shells around fibres positioned between the bunches. For sheets of low density, the distribution of latex in shell form is predominant, and here its relatively uniform distribution over the surface of the sheet is observed (Figure 3).

With an impregnation density greater than the critical magnitude, reduction in the experimental density of the composite materials relative to the theoretical magnitude (Figure 1) reflects an increase in the volume of the sheet during drying. Increase in the volume of the sheet is a consequence of increase in the rigidity of the fibres as a result of the formation on their surface of adsorbed shells of rubber. The rigidity of the fibres increases the rate of relaxation, a result of which is disentanglement of fibres and their straightening out.

Figure 2. Transverse sections of non-woven sheet of (a) 90 kg/m³ and (b) 131 kg/m³ density.

Figure 3. The distribution of latex through the thickness of sheet of (a) 90 kg/m³ and (b) 131 kg/m³ density at the stage of flow of latex from the sheet in the free state.
During impregnation with diluted latex, the latex is held in the bunches and remains within them after squeezing by rolls. Therefore, with a degree of impregnation below the critical magnitude, the density of the composite material is directly proportional to the content of rubber, which is reflected by a linear section of the dependence of density on degree of impregnation. A considerable number of bunches of sheet of maximum density ensures retention of latex up to the maximum degree of impregnation.

When sheets of low density are impregnated with latex with high dry residue, which is a condition of producing composite materials with a high degree of impregnation, some of the latex is located in bunches, and some between them. The latex between the bunches, which remains after squeezing in the rolls, is adsorbed on the surface of fibres and on drying forms a solid shell of rubber. Increase in the volume of the sheet is also promoted by the relatively high mobility of the fibres.

Thus, the parameter obtained from the dependence of density on degree of impregnation determines the structure formation of the impregnated material with retention of the volume of the fibrous material or with change in its volume. Shrinkage of the composite material depends on the density of the sheet, which reflects the effectiveness of formation of bunches and their structure. The production of composite materials with a high content of rubber and high density is achieved by using non-woven sheet of over 115 kg/m³ density. The limiting density of the impregnated materials depends on the capacity of the sheets to hold the latex at the stage of movement of the impregnated sheet towards the squeeze rolls.

REFERENCES


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