One of the complexities in the detecting of defects is the dependence of dielectric permeability on the concentration of CNT in the polymer material. It can vary considerably, up to the acquisition of the conductor properties. An electric capacitive method of non-destructive testing is capable of ensuring the detecting of defects in materials with different properties, from dielectrics to conductors [6]. Thus, the application of the electric capacitive method is the most suitable for detecting defects in NMPCM. The determination of the possibilities and limits of the application of the method in the testing of polymer nanocomposites allows the application of this method in the industrial production of critical parts and components.

2. Literature review and problem statement

In the manufacture of NMPCMs, the presence of defects in the product may be caused by two factors: the presence of agglomeration of CNT in the dispersion of the system and the unevenness of the temperature and mechanical fields in the processing equipment.

Ultrasonic homogenizing dispersants-cavitators are used for uniform distribution of CNT in a polymeric matrix [7].
When dispersing, it is important to effectively adjust the design and operating parameters of ultrasonic equipment to achieve maximum uniformity of the system [8].

Methods of mathematical modeling of the process are used when creating processing equipment for NMPCMs production [9]. The simulation allows predicting possible non-uniformity of temperature [10] and mechanical fields in the processing equipment at the design stage. Since the presence of CNT in the polymer matrix greatly increases the coefficient of wall slipping of the polymer, it is important to consider the wall effects [11–13] when modeling the production of NMPCMs, whose effect on the process may be significant. In addition, the presence of finely dispersed inclusions in the melt can significantly affect the behavior of the flow at the extruder exit, which also requires additional simulation [14]. The properties of polymeric materials with fibrous fillers can be taken into account using the kinetic equations described in [15].

The works [16, 17] consider the method of non-destructive testing that is capable of visualizing defects in a wide range of materials and structures, ranging from insulators to metal conductors. This method, with further modification, can be used to visualize defects in NMPCM.

In papers [18–20] the authors consider the non-destructive testing of non-metallic materials, in particular polymers, using the electric capacitive method and determine the possibility of finding defects in polymeric materials. However, the works are devoted only to the problem of finding defects in dielectric materials with homogeneous properties, and NMPCM can change their electrical properties depending on the concentration of CNT.

Inspection of products with NMPCM during operation may be carried out with the help of film multilayer sensors [21], which allow monitoring the fatigue durability and deformation in the material in real time.

The authors of [22, 23] developed a series of technical solutions aimed at improving the electric capacitive method of non-destructive testing, which allowed to increase sensitivity. The method is used for a wide range of materials and constructions, products made of dielectrics and conductive materials. The functional schemes of devices for realization of the given method were proposed. However, the issue of the application of the method for defects detecting in various materials was not considered.

Thus, the problem of defects detecting in materials with a wide range of properties variations by the electric capacitive method is not sufficiently studied.

### 3. The aim and objectives of the study

The aim of the study is to establish the limits of the application of the electric capacitive method of non-destructive testing of defects in NMPCM at different concentrations of CNT.

To achieve this aim, the following tasks were solved:

- numerical simulation was performed with different contents of CNT in NMPCM;
- electric field distributions and values of the relative electric capacity for different depths of defect and distance from the sensor to the surface were obtained;
- the limits of the method application were determined for NMPCM testing, namely: maximum defect depth and distance from the sensor to the surface.

### 4. Techniques of simulation of detecting defects in nanomodified polymer composite materials by electric capacity method

The issue of simulation of the electric capacity testing method for polymer materials was considered in [24]. Electromagnetic phenomena are described by Maxwell’s equations. In the general case, materials with both dielectric and conducting properties are described by the Maxwell-Ampere equations [25]

\[
\nabla \times \mathbf{H} = J + \frac{\partial \mathbf{D}}{\partial t},
\]

where \(H\) is the intensity of the magnetic field; \(J\) is the current density; \(D\) is the electric flux density.

To eliminate the intensity of the magnetic field, the equation (1) is transformed to the form

\[
\nabla \left( J + \frac{\partial \mathbf{D}}{\partial t} \right) = 0.
\]

Given that the derivative of the magnetic flux density \(B\) can be neglected in time, and according to the Faraday law, the electric field \(E\) is free to refer,

\[
\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} = 0.
\]

Thus, the electric field \(E\) can be described by means of an electric scalar distribution of the potential \(\phi(x, y, z)\)

\[
E = -\nabla \phi(x, y, z),
\]

using constitutional relations, we get

\[
J = \sigma(x, y, z)E,
\]
\[
D = \varepsilon(x, y, z)E.
\]

Thus, the expression (2) will look

\[
\nabla \left[ \sigma(x, y, z) \nabla \phi(x, y, z) \right] +
\n\nabla \left[ \frac{\partial}{\partial t} \varepsilon(x, y, z) \nabla \phi(x, y, z) \right] = 0,
\]

where \(\sigma(x, y, z)\) is the distribution of conductivity; \(\varepsilon(x, y, z)\) is the distribution of dielectric permittivity.

If electrical conductivity and dielectric permittivity are known, then the distribution of the electrical potential \(\phi(x, y, z)\) can be obtained by solving the equation (3). On the other hand, due to the time-derivative relation between dielectric and conducting properties, the equation (3) can not be solved. A practical way to solve this problem is to consider the system as “mostly dielectric” or “predominantly conductive”. In the first case, the equation (3) can be simplified to the Laplace equation

\[
\nabla \left[ \varepsilon(x, y, z) \nabla \phi(x, y, z) \right] = 0.
\]

In the latter case, the equation is written as

\[
\nabla \left[ \sigma(x, y, z) \nabla \phi(x, y, z) \right] = 0.
\]
Using the above quasistatic assumption, it is possible to apply the finite element method to solve the above equations and predict the potential distribution $\Phi(x, y, z)$, created by capacitive electrodes in a particular material and for a certain geometry. The numerical model satisfies the Dirichlet boundary condition

$$\Phi(x, y, z) = \begin{cases} V(x, y, z) & \text{if } (x, y, z) \in \Gamma_1, \\ 0 & \text{if } (x, y, z) \in \Gamma_2, \\ 0 & \text{if } (x, y, z) \in \Gamma_3, \end{cases}$$

where $\Gamma_1$ is the surface of the reference electrode, $\Gamma_2$ is the surface of the measuring electrode, $\Gamma_3$ is the outer surface of the computational regions, $V$ is the voltage applied to the reference electrode.

To determine the induced charge on the measuring electrode by the calculated distribution of the electric potential, the Gauss's law in the numerical form of the integral along the surface of this electrode is used. The Gauss's law can be written in the form

$$q = -\iiint_\Omega \mathbf{E}(x, y, z) \cdot d\mathbf{s},$$

where $s$ is the surrounding surface of a sensitive electrode.

To solve the problem, the COMSOL Multiphysics software was used and further analysis was carried out in the MATLAB system.

### 5. Results of simulation of the electric capacitive method of defect detecting in nanomodified polymer composite materials

Electrodes in the form of rectangles with a zero thickness were used in the simulation. It was assumed that the length of the rectangular electrode is much larger than the width, so the distribution of the electric field along the length can be considered constant. On this basis, the 3D geometry of the problem can be reduced to a 2D model, which can be used for solving this task. In this regard, the simulation was carried out in a two-dimensional planar formulation [24].

In order to unify the calculations, the relative value of “$H$” was introduced and all geometric sizes were given relative to this value. The geometry of the model is shown in Fig. 1.

![Fig. 1. Geometry of the material model with a defect](image)

The defect had the size of $1H$ to $2H$ and electrical properties of air at depths of $0.1H$ to $10H$. The distance from the sensor to the surface was simulated in the range from $0.03H$ to $0.5H$.

The convergence of the grid in order to determine the minimum required density of the calculation grid for obtaining a qualitative result of the calculation was studied. The characteristic size of the element was gradually diminished and the control parameter was compared with different values of the limiting dimensions of the element. As a control parameter, the value of the electric capacity between the electrodes was used. As a result, the finite-element grid was broken into 37300 elements.

The data obtained in [26] were used to determine the material parameters. The authors investigated the mechanical and electrical properties of NMPCMs on an example of low-density polyethylene (LDPE). The authors measured the electrical properties of the samples using an impedance analyzer (Agilent 4294) and a dielectric measurement system (Ando TRS-10T) according to ASTM D-149 and ASTM D-150 standards. The most useful data for this study are shown in Table 1. It demonstrates the values of dielectric permittivity and specific conductivity of the material at different concentrations of CNT.

<table>
<thead>
<tr>
<th>Concentration of CNT</th>
<th>Dielectric permeability</th>
<th>Conductivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 wt %</td>
<td>0.25</td>
<td>0 S/m</td>
</tr>
<tr>
<td>1 wt %</td>
<td>12</td>
<td>0.01 S/m</td>
</tr>
<tr>
<td>3 wt %</td>
<td>133</td>
<td>0.08 S/m</td>
</tr>
<tr>
<td>5 wt %</td>
<td>539</td>
<td>0.1 S/m</td>
</tr>
<tr>
<td>10 wt %</td>
<td>779</td>
<td>0.1 S/m</td>
</tr>
</tbody>
</table>

During simulation in COMSOL Multiphysics, the following settings were used:
- physical component – Electrostatic;
- task type – Stationary;
- material – LDPE (properties in Table 1);
- the initial value of the electric potential $V_i=0$ V;
- electric potential on the supporting electrode $V_s=1$ V;
- electric potential on the measuring electrode $V_s=0$ V.

Fig. 2 shows the distribution of electric potential on the model surface at different concentrations of CNT, which was obtained by simulation.

![Fig. 2. Distribution of electrical potential on the model surface at different concentrations of carbon nanotubes:](image)

$a$ – 0 wt %; $b$ – 1 wt %; $c$ – 3 wt %

From Fig. 2 it is seen that the concentration of CNT in the material affects the distribution of electrical potential along the model surface. Therefore, it is possible to determine the magnitude of the influence of the defect depth on this distribution.

To summarize the results of the study, the values of capacitances obtained during the simulation were reduced
to relative values. A number of simulations were conducted with a defect depth from $1H$ to $10H$ at different concentrations of CNT in LDPE.

Fig. 3 summarizes the simulation results in the form of dependencies of the relative electrical capacity between the electrodes and the defect depth at different concentrations of CNT in the material.

As can be seen from Fig. 3, the relative electrical capacity approaches to the constant value, depending on the concentration of CNT.

The Cochran statistical criterion was used to evaluate the homogeneity of the dispersion. In this case, the dispersion was calculated for each experiment with different concentrations of CNT

$$s^2 = \frac{1}{n-1} \sum_{i=1}^{n} (y_i - \bar{y})^2,$$

where $n$ is the number of experiments; $y_i$ are the results of a separate experiment.

The Cochran criterion is defined as the ratio of the maximum dispersion to the sum of the dispersions of all experiments

$$G = \frac{s_{max}^2}{\sum s_j^2}.$$  

The dependence of the relative electric capacity between the electrodes on the defect depth at different CNT concentrations in the material was studied. The value of the Cochran criterion was $G=0.594$, which does not exceed the critical value, which is $G_{cr}=0.602$ ($k=10$, $v=1$), with a confidence probability of 95 %.

In order to determine the limits of the possible application of the method for each of the investigated concentrations of CNT, a maximum defect depth was obtained. It is determined taking into account the threshold sensitivity of the device of 1 %, provided

$$\frac{dC}{dH} 100 \% \geq 1 \%.$$

The results are shown in Fig. 4.

This approximation of the dependence was obtained with the accuracy of approximation $R^2=0.99$ in the range of CNT concentrations from 0 wt % up to 10 wt %.

As can be seen from Fig. 4, at the CNT concentration of 1 wt % the sensitivity of the system is the largest and allows detection of defects at depths up to $5H$. The maximum defect depth decreases to $2H$ with further increase of the CNT concentration.

Inspection of products with NMPCM is not always possible with a minimum distance from the sensor to the product. To determine the influence of the distance from the sensor to the material surface, a number of simulations with a distance from $0.03H$ to $0.5H$ were conducted at different concentrations of CNT in LDPE.

Fig. 5 summarizes the simulation results in the form of dependencies of the relative electrical capacity between the electrodes on the distance between the sensor and the material surface at different concentrations of CNT in the material.

As can be seen from Fig. 5, the relative electric capacity approaches to the steady-state value, depending on the CNT concentration.

The dependence of the relative electric capacity between the electrodes on the distance from the sensor to the material surface at different CNT concentrations in the material was studied. The value of the Cochran criterion was $G=0.422$, which does not exceed the critical value, which is $G_{cr}=0.602$ ($k=10$, $v=1$), with a confidence probability of 95 %.

The limiting value of the distance from the sensor to the surface was obtained in order to determine the limits of the
The results of the research can be used in the design of technological regulations for the NMPCMs production, as well as in the development of complex devices for non-destructive testing.

Analyzing the plot of the relative capacity dependence on the distance between the sensor and the surface (Fig. 5), we can conclude that at a small distance there is a strong dependence both on the concentration and on the distance. This can be used to control the concentration of CNT in the testing point at a fixed distance. The prospect of further research is to apply the electric capacitive method for determining dielectric material permeability, which will allow inspection of the uniformity of the CNT distribution in the melt polymer under the continuous technological process of NMPCMs production. Furthermore, it is necessary to conduct a study of the method application in the industrial production of intelligent polymer systems [27].

### 7. Conclusions

1. A mathematical model based on Maxwell-Ampere, Faraday and Gauss's equations and satisfying the Dirichlet boundary condition was proposed. The simulation was carried out in a two-dimensional planar formulation and a minimum required density of the calculated grid was determined (37300 elements) to obtain a qualitative result of the calculation.

2. A number of numerical studies have been conducted with different contents of CNT in NMPCMs in the range of 0 wt % up to 10 wt %. The depth of the defect in the material was changed in the range of $1H$ to $10H$, and the distance between the sensor and the surface was varied in the range of $0.03H$ to $0.5H$.

3. The homogeneity of the dispersion is estimated using the Cochran statistical criterion. The value of the Cochren criterion did not exceed the critical one for all conducted experiments.

4. Approximation relations of the maximum defect depth and distance from the sensor to the surface were obtained depending on the content of CNT in NMPCM.

5. The limits of the method application in the testing of NMPCM are determined. The maximum defect depth was $5H$ at the CNT concentration of 1 wt % and with increasing the CNT concentration, the maximum defect depth decreases to $2H$. The maximum distance between the sensor and the surface was $0.33H$ at the CNT concentration of more than 5 wt %.

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### References


