Measuring Setup for Investigation and Visualization of the Percolation Phenomenon in Non-Ordered Models of Metal-Dielectric Nanocomposites

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Abstract

The study uses the phenomenon of high voltage partial discharge to investigate the phenomenon of percolation and visualisation of the percolation channel. The phenomenon of partial discharges is very similar to the quantum tunneling phenomenon observed in metal-dielectric nanocomposites. In both cases the flow of alternating current occurs in the absence of direct contact between the metallic phase particles.

A measuring stand was developed and constructed to test models of metal dielectric nanocomposites using high voltage partial discharge. The stand consists of a 110 kV high voltage transformer, a voltage regulator protecting the constant rate of high voltage rise, a measuring system consisting of a measuring probe, voltmeters and a computer. The communication between the measuring probe and the voltmeter was made in digital technology with the use of fiber optic technology, which allowed the meter to communicate with the computer without any errors and eliminated the interference caused by a strong electromagnetic field resulting from the use of high voltage.

Systems modelling metal-dielectric composites were built, consisting of metallic elements in the form of disks, randomly distributed on the surface of the dielectric matrix. The number of disks was increased in series of 40 in each. The maximum number of disks was 1520. The dependence was determined of one of the important parameters characterising a partial discharge, i.e. the initial voltage, at which an electric current starts to flow between electrodes, on the concentration of the metallic phase. On the basis of these results, a percolation threshold was established for a matrix with a random distribution of metallic phase elements, the value of which is about 50%. Films and pictures of partial discharges with visible percolation channels were taken with the camera with which the stand was equipped.

Keywords: percolation, nanocomposites, partial discharge, high voltage.

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Измерительная установка для исследования и визуализации явления перколяции в неупорядоченных моделях нанокомпозитов металл-диэлектрик

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В статье для исследования явления перколяции и визуализации каналов перколяции использован высоковольтный частичный пробой. Частичный пробой очень похож на квантовое явление туннелирования, наблюдаемое в нанокомпозитах металл-диэлектрик. Как в первом, так и во втором случаях для протекания электрического тока не требуется контакт между частицами металлической фазы.

Разработана и изготовлена измерительная установка, предназначенная для измерений моделей нанокомпозитов металл-диэлектрик с использованием высоковольтного частичного пробоя. К сою став установки входят: высоковольтный трансформатор с максимальным напряжением до 110 кВ, регулятор напряжения первичной обмотки высоковольтного трансформатора, обеспечивающий постоянную скорость увеличения напряжения, измерительный зонд, вольтметры и компьютер. Сигналы от зонда и вольтметра с целью ограничения помех, связанных с сильным электрическим полем, передаются с использованием оптоэлектронного кабеля.

Разработаны и изготовлены двумерные модели нанокомпозитов, в которых частицы металлической фазы в форме металлических дисков размещены случайным образом на поверхности диэлектрической матрицы. Число металлических дисков во время измерений увеличивалось по 40 штук в серии до максимальной величины 1520.

Определена зависимость от концентрации металлической фазы одного из существенных параметров высоковольтного частичного пробоя – пороговое напряжение, выше которого между электродами начинает течь ток. На основании этих измерений определён порог перколяции для матрицы со случайным распределением элементов металлической фазы.

С помощью фотоаппарата, входящего в состав установки, зарегистрированы фильмы и фотографии явления частичного пробоя между элементами металлической фазы моделей, на которых видны каналы перколяции.

Ключевые слова: перколяция, нанокомпозиты, частичный пробой, высокие напряжения.

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Introduction

There are currently more and more studies and applications of nanocomposites and nanomaterials which are characterised by a number of interesting chemical, optical, mechanical and electrical properties [1]. These are materials that consist of at least two phases with at least one component less than 10^-7 m in size [2]. Their special properties are related to the influence of surface energy, much higher than that of solid materials, and to quantum phenomena that occur in nanocomposites. One of the types of nanocomposites is metal-dielectric nanocomposites. Such material has special electrical properties related to conductivity. Metallic nanoparticles are so small that they should be considered as wells of potential. Then all phenomena must be described on the principles of quantum physics.

In such materials, which are characterised by high resistivity, there may be a type of conductivity, based on step exchange (tunnelling) of electrons [3]. The condition of its observation is the weakening to the lowest possible level of ionic conductivity in dielectrics and electron or hole conductivity in semiconductors. The basis for the occurrence of the quantum effect of electron tunnelling between potential wells is that the wave function of the electron in a potential well of nanometre dimensions or smaller has a non-zero probability beyond the potential barrier. The probability of finding a valence electron at a distance r outside a three-dimensional potential well is given by the formula [3]:

\[ \Psi^2 = \exp(-2\alpha r), \]  

where: \( \alpha \) – inverse of the radius of the location of the electron; \( r \) – distance from the well of potential in which the electron is located.

\[ \alpha^{-1} \equiv R_B, \]  

where \( R_B \) – Bohr’s radius.

In the absence of an external electric field, the direction of electron tunnelling is random. After applying an electric field to the semiconductor, there is a partial ordering of electron jumps, related to the Debye factor [3]:

\[ \exp\left(\pm \frac{e \cdot r \cdot E}{k \cdot T}\right) = \sinh\left(\frac{e \cdot r \cdot E}{k \cdot T}\right), \]  

where: \( e \) – electron charge; \( r \) – distance between the well of potential from which the electron is tunnelling and the well to which it is jumping (tunnelling); \( E \) – electric field strength; \( k \) – Boltzman constant; \( T \) – temperature.

According to the precursor of the theory of electron tunnelling in disordered semiconductors and dielectrics [3] electron tunnelling between potential wells can take place in two ways. The first one occurs in the case of weak location of electrons and temperature, close to the temperature of liquid helium. The second mechanism occurs in the case of strong electron localisation and high temperatures. Then electron tunnelling occurs between the closest adjacent potential wells. The conductivity formula for the case of tunnelling between the closest neighbours [3]:

\[ \sigma = \sigma_0 \exp\left(-2\alpha r - \frac{\Delta W}{kT}\right), \]  

where: \( r \) – distance between wells between which the electron is tunnelled; \( \alpha \) – inverse of the radius of the location of the electron; \( \Delta W \) – activation energy.

As can be seen from formula (4), in the mechanism of electron tunnelling between the closest adjacent potential wells there is a strong temperature dependence of conductivity. The activation energy entering the formula is related to the fact that the tunnelling electron, as a rule, jumps from a lower energy level in the first well to a higher level in the second well. The distance between the potentials wells to be seen in formula (4) is a function of their concentration \( N \):

\[ r \approx N^{-\frac{1}{3}}. \]  

Formulas (4) and (5) demonstrate that conductivity increases faster than linearly as the concentration of potential wells increases. Mott’s calculations [3] show that when the distance between potentials wells decreases to a value two-and-a-half times greater than Bohr’s radius \( R_B \), as the concentration of potentials wells increases:

\[ r \equiv 2.5R_B, \]  

there will be a transition from dielectric type conduction with a positive temperature coefficient of conductivity \( d\sigma/dT > 0 \) to metallic type conduction with \( d\sigma/dT < 0 \).

The transition from dielectric type conductivity at low contents to metallic type conductivity in the area of high potential well contents is associated with percolation [4]. Below the percolation threshold there is a rapid increase in the conductivity of the nanocomposite with an increase in the metallic phase.
content, while above the percolation threshold the increase in conductivity significantly slows down and the transition to metallic type conductivity occurs.

Percolation phenomena cover a wide range of issues related to the flow of current in spatial networks, self-generated in metal-dielectric nanomaterials. Networks composed of resistive ($R$), resistive and capacitive ($RC$) and resistive, capacitive and inductive ($RLC$) elements are used to model such phenomena [5]. The network consists of nodes connected to each other by sections consisting of these elements [6]. When the network is continuous, it is a current flow channel from one side of the network (contact) to the opposite through the elements forming the network. After it is connected to a power source, current flows through the network. When more than one damage is introduced into the network, at some point the current stops flowing from one contact to another. It is said that the percolation threshold has been reached [7]. Such a state can be achieved in networks consisting of passive elements in two ways. In the first one, randomly determined nodes are eliminated. In the second one, the sections connecting the nodes are randomly cut. There are also other ways of modelling the percolation phenomenon, for example testing the current conduction by randomly mixed balls of identical dimensions, one of which is made of metal and the other of insulating material. Gradually increasing the content of metallic balls, we achieve a rapid increase in current intensity practically from zero, which corresponds to passing through the percolation threshold. Nowadays, the phenomenon of percolation has been analysed using the Monte Carlo method. The most common models studied were two-dimensional networks with translational symmetry. Such networks were damaged by eliminating nodes or crossing bonds with statistically independent probability $p$. Depending on the method of network damage, they formed models of bond percolation or node percolation. In the last few decades a huge amount of work has been put into finding precise and approximate ways to determine the values of percolation thresholds for different networks [8]. Exact thresholds are only known for some two-dimensional networks such as square or triangular.

Unfortunately, both in the grating, ball and Monte Carlo models below the percolation threshold, the conductivity of the investigated grating is zero, which is not observed in real nanocomposites. The models do not take into account the fact that in nanocomposites the current conductivity is also below the percolation threshold through electron tunnelling, and with the increase of metallic phase content the conductivity of the material increases [9]. Real nanocomposites are characterised, first of all, by random distribution of metallic phase elements in dielectric matrix, secondly, the flow of direct or alternating current occurs also in the absence of direct contact between adjacent nanoparticles. This is due to the quantum tunnelling phenomenon. Another problem related to both the network and ball models is that these models do not allow to determine the route of the percolation channel.

Comparing the voltage behaviour in the case of high voltage partial discharge (HVPD) and conductivity behaviour in the case of electron tunnelling in the nanocomposite we can state that they are similar. For both phenomena, HVPD and tunnelling, the resistance decreases with the decrease of distance. The application of a partial high voltage discharge does not require direct contact of the metallic phase elements to the current flow between the electrodes. Of course the scale of these phenomena is completely different. Tunnelling occurs on a nanometer scale, while the phenomenon of high voltage discharge – on a macroscopic scale. Due to the similarity of both phenomena, the phenomenon of high voltage discharge was used in the study to model the percolation phenomenon in metal-to-electric nanocomposites. The aim of this work was to develop and build a stand for research of the phenomenon of percolation and visualisation of percolation channels in disordered systems modelling metal dielectric nanocomposites, make models and define the percolation threshold for them and determine the route of percolation channels.

**Measuring stand**

In order to investigate the phenomenon of percolation in metal-to-dielectric nanocomposites based on an partial high voltage discharge, a measuring stand was developed and made, the block diagram of which is shown in Figure 1. A dielectric matrix 3 was used as a model of a two-dimensional metal-to-dielectric composite. In the model, the dielectric phase is modelled by air, while the metallic phase is represented by metal disks. The base of the matrix is XPS extruded polystyrene. The primary winding of the single-phase high voltage transformer 2, whose gearbox was 110 kV/220 V, was powered by autotransformer 1, which was used to control the voltage. It was a specially designed autotransformer.
with a stepper motor drive. Such a solution allowed precise regulation of the voltage increase at a constant speed. This is very important due to changes in the strength of the system depending on the rate of voltage increase. The stepper motor is controlled by an electronic system based on a sixteen-bit Atmega microcontroller. The program that has been written and imported to the microcontroller supports the motor controller in such a way that the value of voltage increase is digitally regulated in the range from 1 to 2 kV/s. On the surface of matrix 3 there are flat electrodes between which, as a result of the voltage increase, an partial discharge is created. The essence of the work was to register the changes in the HVPD current value along with the voltage increase for different concentration of metallic phase. Due to the very high values of the electromagnetic field it was significantly difficult.

Figure 1 – Block diagram of the measuring station: 1 – autotransformer; 2 – high voltage transformer; 3 – percolation model; 4 – measuring probe; 5, 6 – voltmeters; 7 – computer; 8 – stepper motor; 9 – stepper motor control electronics; 10 – camera

The first and fundamental problem was the measurement of the current intensity. Due to the use of high voltage it was not possible to make direct measurements. In order to measure the current in the grounding branch, resistance was incorporated into the system. Two resistors of 50 Ω each were used. The voltage on the resistor did not exceed 4 V. The tests showed the presence of residual inductance in the measuring circuit, which significantly deformed the course making it impossible to obtain reliable measurements. This problem was solved by using specialised non-inductive resistors. For smaller voltage values, i.e. up to 80 kV, the system met the requirements. For larger values, the electromagnetic field induced a voltage in the cables and resistors, which resulted in a very noisy signal. In order to limit the influence of the field, a Faraday cage was used, in which both resistors to which the test leads were connected were placed. It was a 1 mm thick copper cuboid, earthed, which allowed the wires to be earthed. Figure 2 shows a diagram of the measuring probe used in the model.

Figure 2 – Diagram of the measuring probe: 1 – BNC connector; 2 – resistors; 3 – measuring BNC output; 4 – copper casing

An additional element used to reduce electromagnetic field interference was the use of BNC RG213 50 Ω coaxial cables, double-insulated with braid. These cables were used in the branch between the model output and the probe in order not to interfere with the input current and in the branch that provided the measuring signal to the meter. Thanks to this solution, the measuring system was not exposed to a strong field and the received signal was free of noise resulting from its operation.

Then, a computer program with a graphical interface was created, communicating via a computer with two electronic meters 5 and 6, which had digital outputs. The communication was created in accordance with the RS-232 standard. This allowed the computer to receive 7 synchronised items of data from the two meters. One indicated the voltage value on non-inductive measuring resistors. The other was connected to an autotransformer supplying a high voltage transformer and was used to read the voltage given to the model. The program supported data recording at a frequency of 32 per second in a CSV format file. Such a resolution of measurements is sufficient to be able to plot characteristics such as the value of the voltage on the measuring resistors or changes in the current of the exhaust depending on the voltage supplied by the test transformer. As it turned out, such measurement method was effective only for HVPD registration. When the stand was applied in the full range, i.e. until the moment of a full discharge, the electromagnetic field of the resulting electric arc caused a disturbance of the digital signal between the meters and the computer,
which made it impossible to read the downloaded data. Therefore Universal Serial Bus cables were replaced by TOSLINK type optical fibre cables. This procedure reduced transmission errors to zero, thanks to which the created measuring station was used in the full measuring range.

Test results and their analysis

In order to measure the changes in the current intensity value as a function of the supply voltage depending on the concentration of the diffuse phase, it was necessary, in the first step, to place the metal disks in the dielectric matrix in strictly defined quantities depending on the series being tested. The matrices with the coordinates of points were generated by means of a computer program created. This program was based on a random number generator. The model of the matrix was generated for the smallest possible distances equal to 0 mm and the diameters of points were the same and amounted to 6 mm. The matrix generated in this way leaves the freedom to choose the number of disks in a series. The whole matrix was composed of 1520 points, randomly distributed on the matrix surface. Such a large number of disks was selected due to the fact that errors, resulting from the statistics, for sets over 1000 do not exceed 2 %. The matrix was divided into 38 series which contained 40 discs each [10]. It was not a continuum type because the program did not allow disk overlapping. The concentration of disks modelling metallic phase molecules is defined as the ratio of the metal surface area of the disks to the surface area of the matrix. Figure 3 shows a matrix for four concentration values of metallic phase as an example.

For testing, another series of metal discs, 40 in each, were placed on the matrix. When each series was mounted on the model, a program was run to connect to the meters and initiate the download of measurement data from them. A microcontroller-controlled stepper motor was started, driving an autotransformer to control the test transformer voltage. The rate of voltage increase was 2 kV/s in the range from 0 V to jump voltage. An electric field was generated between the electrodes on the matrix. Voltages on the test transformer and on the probe were read by electronic voltmeters and their values were stored in the computer memory. In this way, for a specific concentration value of the metallic phase, the course of voltage changes on the resistors in the probe depending on the model supply voltage was obtained. The voltage measured on the measuring probe was proportional to the current of the partial discharge, formed on the model of nanocomposite metal dielectric. The diagram for one of the series of measurements is shown in Figure 4.

![Figure 3](image1.png)

**Figure 3** – Coordinates of discs of computer-generated dielectric matrix for 12 %, 30 %, 42 %, 58 % concentration

One of the basic parameters of the partial discharge is the initial voltage. Only when this voltage is exceeded will an partial discharge be initiated and a current start to flow between the electrodes. From Figure 4 it can be seen that for a concentration of the metallic phase of 42 % the partial discharge starts at an initial voltage of about 35 kV.

![Figure 4](image2.png)

**Figure 4** – The course of the voltage value on the measuring probe depending on the model supply voltage for concentration of metallic phase 42 %
After exceeding this voltage, the current flowing through the model starts to rise faster and faster. After reaching the voltage value of about 45 kV, the increase of the current intensity is clearly slowing down. When a voltage of about 72 kV is reached, an electric arc is ignited and the overcurrent protection switches off the power supply. This can be seen on a vertical drop to zero current.

The most important feature of the partial discharge applied in the test is the flow of electric current through the matrix below the percolation threshold. This is due to the similarity of the partial discharge and quantum tunnelling of electrons. Metallic phase elements, exactly as in real nanocomposites, do not need any direct contact current for the flow of current and the flow of current between them depends on the value of probability of jumps as a function of distance. By comparing the waveforms for individual concentrations of the metallic phase shown in Figure 5, it is possible to determine the initial voltage at which an partially discharged current appears. Figure 5 shows that for the concentration of the metallic phase $X \leq 42\%$ the initial voltage of partial discharges appears at a voltage of about 36 kV. An increase in concentration up to 50% causes the initial voltage of partial discharges to decrease rapidly to about 15 kV.

Such a leap change can be interpreted as reaching the percolation threshold. Its value is $(46 \pm 4)\%$. The conductivity of the system suddenly changes, which means that a small change of concentration between 42 % and 50 % significantly affects the properties of the whole system.

![Figure 5 – Diagram of the voltage waveforms on the measuring probe depending on the supply voltage for different concentration of metallic phase](image)

During partial discharges radiation is generated in the areas of visible light and ultraviolet. One of the elements of the station equipment is a camera. Its application allows the visualisation of percolation channels which are recorded in the form of films and photographs. Figure 6 shows a picture of an partial discharge, on which percolation channels and disks modelling the metallic phase are visible. In the picture, the light is visible in places where there is no direct contact with the metallic elements of the model and an partial discharge occurs. The photos taken during the research show that in the matrix, similarly as in nanocomposites, a number of percolation channels may occur.

**Figure 6 – Example picture of percolation channel**

**Conclusion**

A measurement stand was developed and built to study the percolation phenomenon and visualise the percolation channel using a high voltage partial discharge. The phenomenon of partial discharge applied in the work is very similar to the quantum tunnelling phenomenon. In both cases the current flow occurs in the absence of direct contact between elements of the metallic phase.

The measuring equipment, i. e. the measuring probe and connecting cables, were made in such a way as to eliminate the interference caused by a strong alternating electromagnetic field resulting from the use of high voltage. The communication between the measuring probe and the voltmeter was made in digital technology with the use of optical fibre technology, which allowed the meter to communicate with a computer without errors.

Models of nanocomposites of metal dielectric type were used for the tests. Modelling systems
consisting of metallic elements in the form of disks with concentration varying from 0% to 72% randomly distributed in the dielectric matrix were generated by computer and built. During the research, the concentration of the metallic phase was gradually increased by placing successive series of discs, 40 in each. The maximum number of discs was 1520 at 72% concentration. The initial voltages of the exhaust were determined, at which the electric current starts to flow through the model, depending on the concentration of the metallic phase. Based on the obtained dependencies, a percolation threshold was determined, the value of which for the random distribution of the metallic phase elements is about \((46 \pm 4)\%\). The radiation generated during partial discharges in the visible light area was used to visualise the percolation channels. Photographs of partial discharges, on which percolation channels are visible, were taken during the tests. The photos taken during the study show that the matrix, similarly to nanocomposites, may contain a number of percolation channels.

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