It is impossible to effectively use water with a high salt content at car washes. In many places, access to water with a high salt content is almost unlimited but its utilization requires deionization. For this purpose, several methods are used, the main of which are reverse osmosis, electrodesorption, ion exchange methods, and distillation. However, they have significant drawbacks. Recently, the technology of capacitive deionization of water has been widely used, based on the removal of salt ions from the solution during the charge/discharge of "double" electric layers of carbon materials with a significant active surface (800–2,000 m²/g).

Theoretically, this process should be more energy efficient by using a low potential voltage (1–2 V). This paper considers the interrelation of physical parameters that affect the process of capacitive deionization of water. The dependences of voltage drop on the distance between electrodes for electrodes based on the material SAUT-1S (Belarus) have been investigated. It is shown that the main contribution to the sequential internal resistance is introduced by the resistance of the electrolyte. As the distance between the electrodes increases, the voltage drop on the serial internal resistance increases linearly. A decrease in the concentration of ions leads to a decrease in the conductivity of the solution, which causes an increase in energy consumption and a decrease in the efficiency of sorption. It has been demonstrated that the voltage drop at the serial internal resistance when the voltage on the electrodes is limited, which is set in order to avoid the transition of the electrode charging mode to the electrolysis of water, causes a significant drop in the efficiency of the capacitive deionization process.

Keywords: double electric layer, electrochemical impedance, capacitive deionization, water treatment for car washes

1. Introduction

There are no formal requirements for salt content or other parameters regarding the water that is used at car washes. However, a car cleanly washed with ordinary tap water will be covered with light strips of hardness salts after it dries. The high salt content in the water can lead to accelerated corrosion and subsequent breakdown of the car. Post-washing with demineralized water makes it possible to avoid the formation of plaque. This effect is achieved in water deionization devices.

Deionization is the process of extracting positive and negative ions from the occupied volume. Deionization can be carried out by various methods; for example, calcium ions can be extracted from the solution using ion-exchange systems for water treatment. However, more common in modern water treatment systems is the use of reverse osmosis. Reverse osmosis is a purification technique involving the "pushing" of water through a semi-permeable membrane. As a result, on the other side of the membrane, there is practically distilled water without salts and other contaminants but part of the water (up to 50 %) remains in the form of a concentrate and must be drained or directed for further use [1]. Reverse osmosis water is used at most self-service washers at the final washing stage. When drying, the drops do not leave traces of salts contained in them. However, these systems have some drawbacks. Due to the high sensitivity of membranes to contamination and a fairly high cost of replacement, high-quality mechanical cleaning is mandatory prior to this type of filter. If chlorinated water is supplied (from the city water supply), it is necessary to install a filter with activated coal – chlorinated water quickly spoils the reverse osmosis membrane. Softened water interacts better with car shampoos. In hard water, less active foam is formed, which leads to noticeable over-consumption of detergents.

In many places, there is virtually unlimited access to water with a high salt content but its use requires deionization.
The main methods for deionizing water are reverse osmosis, electrodialysis, ion exchange methods, and distillation. During distillation, water is removed from saline solutions by evaporation. Salts remain in a concentrated solution. Water that has evaporated is then condensed to produce clean water, and condensation energy can be reused to reduce overall energy consumption. Commercial technologies include multi-stage and multi-step distillation. Distillation is relatively old and proven technology and is widely used to treat water with high concentrations of salts [2]. Due to the high energy consumption for water evaporation, other deionization and desalination technologies such as reverse osmosis are becoming increasingly competitive. Reverse osmosis is a controlled process of membrane separation of solutions with different salt content under pressure. High pressure is used to cause the liquid to pass through a membrane that does not pass some of the ions of the dissolved salts. To separate the concentrated solution and the desalted water, the applied pressure must be higher than the osmotic pressure of the saline solution. Reverse osmosis is widely used to desalinate seawater and salt water [3]. Membranes used in reverse osmosis technologies require replacement every few years.

The electrodialysis method of deionization is based on the use of the effect of the passage of ions through ion-selective membranes under the action of an electric field. Two types of anion-exchange and cation-exchange membranes are used at the same time. These membranes are located on both sides of the main water flow, and, on the outside of the membranes, there are two electrodes. By applying the potential difference to these electrodes, the transfer of ions across the membranes is initiated. Since ion exchange membranes are semi-permeable, the transfer of charge through cation exchange membranes is associated with the movement of cations towards the cathode, and the transfer of charge through anion exchange membranes is associated with the movement of anions to the anode. At the end of the charge transfer, there is a splitting of water on the anode and cathode, which leads to the production of oxygen and H+ at the anode, and, at the cathode – hydrogen and OH-.

Due to the location of ion exchange membranes, there is a stream of highly concentrated salt brine and pure water at the outlet. The most effective is to use electrodialysis for desalination of solutions with low salt content, as desalination of brackish groundwater for use as drinking water [4].

Ion exchange is based on the interaction between ions of a certain type that are present in water and mobile ions of ion-exchange materials. Ion-exchange materials contain charged functional groups with a high affinity for some type of ion. Therefore, ions of this type are sorbed from the solution. Ion exchange is used to selectively remove a wide range of ions from the solution, where selectivity depends on the type of ion-exchange materials, as well as the operating mode of the plant. Ion exchange is mainly suitable for removing low concentrations of specific ions from the solution, and, therefore, used to remove metal ions, for example, in the mining industry [5].

It should be noted that existing methods of deionization and desalination are either energy-intensive, like evaporation, or imperfect, like osmosis technologies (membrane problem), electrodialysis, and ion exchange methods. That leads to a high cost of water, which is produced by these technologies [6]. One of the ways to resolve this issue is to devise and implement new methods of water desalination [7].

Given that the disadvantages of the above methods are not inherent in the systems of capacitive deionization of water, it is a relevant task to study their application in water treatment systems at car washes.

2. Literature review and problem statement

Paper [8] reports the results of designing and the possible introduction of new mesoporous materials in the most promising method of desalination of water, which is capacitive deionization (CDI). The mechanism of capacitive deionization of water is as follows: when voltage is applied to electrodes that are in the water with high salt content, solvated ions begin to move from the flow of pumped water to the surface of the electrodes. In this case, the ions are adsorbed at the surface, forming a double electric layer. At the same time, a stream of deionized water flows out of the plant. At the end of the sorption process, the polarity of the voltage applied to the electrodes changes to the opposite, and the solvated ions are desorbed. The water is enriched with ions and drained from the plant. In practice, electrochemical installations of capacitive deionization of water operate either under a cyclic or a two-stroke mode and produce both deionized water and water enriched with ions. The ratio between the flows of deionized water and water enriched with ions depends on the predefined level of deionization and the design of the plant.

The physical limitations faced by the development of capacitive water deionization systems are described in more detail in [9]. Thus, the specific capacity of the double layer for mesoporous carbons with a developed surface is about 20 μF/cm2. This means that when the potential falls on a double layer of 1 V, 20 μC of electric charge can be retained per square centimeter of the two-layer area. The ratio between the charge and the number of adsorbed ions is determined by the Faraday constant (F=96,485 C/mol), suggesting that all charges are involved exclusively in the formation of a double layer. In practice, not all electric charges are involved in the formation of a double layer and adsorption, some of the charges participate in other processes, for example, as a result of electrochemical reactions, as described in work [10].

For a long time, the development of the technology of capacitive deionization of water was constrained by the lack of materials with a sufficiently high developed inner surface. However, in recent decades (in the period from 1994 to 2021), with the development of technologies for the production of mesoporous carbons, materials with an active surface of 800–2,000 m2/g were created (for example, the design of aerogels at the Livermore National Laboratory), which are described in work [11], which led to the increased interest in the capacitive deionization of water.

It should be noted that for the implementation of the technology of capacitive deionization of water in an actual device, the potential of the double electric layer should not exceed the dissociation potential of water molecules of 1.23 V. The total potential difference between the electrodes that can be used depends on the redox chemical reactions occurring on the surface of the electrodes. For transition metal ions present in the solution, galvanic reactions, and metal isolation on one of the electrodes can occur. This process is commonly used to remove heavy metals, for example, in the treatment of wastewater in mining, described in work [12]. However, these galvanic reactions are
difficult to reverse for the desorption process, and, therefore, their presence is undesirable for the capacitive deionization process. Other possible sources of redox reactions include surface functional groups, mainly oxide, as shown in [13]. These surface functional groups can contribute to the splitting of water and, therefore, limit the use of voltage on the electrodes to about 1.5 V.

The voltage on the electrodes includes a voltage drop on the serial internal resistance and the capacitance of two double electric layers. The potential drop at the serial internal resistance is associated with the resistance of the electrodes, the solution in the interelectrode gap, the contact resistance between the current collector and the carbon electrodes, and determines the energy efficiency of the process of capacitive deionization of water.

Paper [14] describes the process of sorption/desorption, which occurs as follows. Initially, there is no potential difference on the electrodes, the ions are not absorbed. Consequently, the drop in potential on the double layers is zero. When the potential is applied to the electrodes as a result of a strong electric field, the ions move across the entire volume of the solution to the electrodes. Adsorption of ions leads to the shielding of the electric field. This means that the more ions are adsorbed in the double layers, the lower the absorption rate. When the potential drop on the double layer is equal to the potential on the electrodes, the electric field is shielded, and adsorption stops. In the opposite event, desorption reduces the drop in potential on the double layers, therefore, a decrease in the electric field. This means that the more ions are desorbed from the double layer, the lower the desorption rate. When the potential drop on the double layer becomes zero, the electric field becomes zero and the desorption process stops.

Capacitive deionization technology is associated with the technology of creating supercapacitors. Supercapacitors are based on the same physical principles as capacitive deionization. Two carbon electrodes with a highly developed surface are separated by a thin gasket and immersed in an electrolyte solution, as shown in work [15]. The difference between capacitive deionization technology and supercapacitors technology is that for supercapacitors, the amount of accumulated charge on the electrodes matters, not the number of ions removed. Therefore, stationary electrolytes are used in supercapacitors, while in capacitive deionization technology, the electrolyte flows between the electrodes. Supercapacitors are capable of storing large amounts of energy, which can be obtained at high speed during discharge. Supercapacitors are used in the electric part of cars in the systems for energy recovery spent on braking, for example, the kinetic energy recovery system (KERS) in Formula 1 cars. Other applications include mobile phones, toys, and flash devices in digital cameras.

There are many parallels between the development of supercapacitors and capacitive deionization technology. Paper [16] shows that the designs of the first supercapacitors and capacitive deionization systems are very similar. Subsequently, the structures began to differ greatly, since, in the technology of capacitive deionization, the electrolyte must pass between the electrodes.

Research continues into the use of electrodes made of carbon aerogel for capacitive deionization of water, which are described in work [17]. The use of carbon aerogels as electrodes resolved the issue of electrical contact due to their high strength and low internal resistance. The disadvantage of using carbon aerogel is its high cost compared to activated carbon, typically made from cheap materials such as coconut shells, and a significant drop in potential on consistent internal resistance. In addition, the titanium plates used are expensive. Work [18] shows that the development of new materials for use in supercapacitors continues. These materials were also used in the construction of capacitive water deionization systems. This is how materials based on carbon powder in the polymer matrix, carbon fibrous materials of carbon nanotubes and nanofibers, graphene electrodes are designed. In addition to the development of materials from which electrodes are made, much attention is paid to improving the design of capacitive deionization systems.

3. The aim and objectives of the study

The aim of this work is to determine the influence of various components of sequential internal resistance on the efficiency of the process of capacitive deionization of water to build promising water treatment systems. This will make it possible to construct commercially attractive water treatment systems for car washes.

To accomplish the aim, the following tasks have been set:
– to determine the effect of the electrolyte concentration on the charging-discharge parameters of the electrosorption module;
– to determine the dependence of the specific sorption capacity on the voltage supplied to the electrodes;
– to determine the dependence of the voltage drop on the serial internal resistance on the concentration of the electrolyte and the distance between the electrodes.

4. The study materials and methods

4.1. Preparation of carbon materials

Most electrode materials with a large surface area have a high resistivity, which is why the efficiency of using these materials for capacitive deionization of water is reduced. The carbon fabric SAUT-1S is one of those materials (its specific surface area is 2,000 m² per gram of material, its resistivity is 30–32 Ohm/cm). Although it has a large surface area, its resistivity is also high. To increase the electrical conductivity of sorption electrodes made of the carbon fabric SAUT-1S, a conductive layer is created directly on the surface of the electrode by applying materials that are insoluble in saline solutions under the action of the applied voltage and, at the same time, safe for human health. It’s titanium. Various techniques of applying titanium coatings to the surface of carbon materials are being actively studied. First of all, these are chemical methods [19]. The vacuum-arc method of applying a titanium coating on the surface of the carbon fiber fabric in order to improve its electrical conductivity was studied. Electric arc methods of vacuum spraying of coatings made of titanium [20], titanium oxide and nitride [21], allow them to be applied to materials of various structures, including carbon fibrous materials, such as the carbon fabric SAUT-1S.

The carbon fabric SAUT-1S the size of 30*30 cm was arranged on the holder in a vacuum chamber at a distance of 50 cm from the applied titanium cathode. After vacuuming the system to a pressure of 10⁻³ torr, an arc discharge (discharge current, 100 A) was lit between the carbon fiber fabric and the titanium cathode. Application of titanium coat-
ing on one side of the carbon fabric SAUT-1S is carried out for 10 minutes by electric arc method. The diameter of the sprayed electrode is d ≈ 30 cm. Pre-vacuuming to a pressure of \(10^4\) torr and the distance between the spray electrode and the carbon fiber fabric made it possible to evenly apply the titanium coating onto the surface of the carbon fabric. The deposition was carried out without the application of any additional stress to the carbon fiber fabric [22]. The thickness of the titanium coating layer is 10 μm. A sample of the carbon fabric with and without titanium coating is shown in Fig. 1.

![Fig. 1. The surface of carbon material: a — before application of titanium coating; b — after applying titanium coating](image1)

Fig. 1.

The resistivity of the fabric SAUT-1S before applying the titanium coating was 32 Ohm/cm. After applying a titanium coating on one side of the fabric of carbon fiber – 11 Ohm/cm. After application on both sides – 8 Ohm/cm. Photographs of the carbon fabric SAUT-1S, modified with titanium, acquired from an electron microscope, are shown in Fig. 2.

![Fig. 2. SEM photographs of the carbon fabric SAUT-1S modified by applying a layer of titanium with a resolution: a — 1 mm; b — 200 μm; c — 20 μm](image2)

Fig. 2. SEM photographs of the carbon fabric SAUT-1S modified by applying a layer of titanium with a resolution: a — 1 mm; b — 200 μm; c — 20 μm

Fig. 2 shows that the carbon fabric SAUT-1S consists of a large number of carbon threads with a thickness of about 10 μm. Fig. 2 clearly demonstrates white areas of the sprayed titanium/titanium nitride, which form additional contact pads between the individual filaments. An electrode was made of this fabric by fixing the fabric between two titanium frames made of titanium VT-1, which were welded together using spot welding.

4.2. Experimental equipment and measurement procedure

To power the electrochemical module, a programmable low-voltage power supply CDS1-5M10 (Ukraine) is used, which includes five separate modules, each of which has an operating voltage range of −1.5+1.5 V, and a current of 0–10 A. This unit makes it possible to charge the CDI module both under the mode of the current generator and the voltage generator mode. The electrodynamic parameters are controlled by the digital oscilloscope Tektronix 2024B (USA), which registers signals from a low-voltage voltage divider and a low-ohm current shunt. The conductivity and TDS of aqueous solutions are controlled in a buffer volume using the conductometer Bante 950 (Taiwan) with the function of temperature compensation. In the range of salt concentrations of 1,000–10,000 ppm, the measurement accuracy is 10 ppm. Aqueous solutions were prepared by dissolving a batch weighed to 0.1 g of chemically pure sodium chloride in distilled water. The volume of the solution in the system is 1 l. Circulation speed is 1 l/min. When studying the dependence of the relationship between the sorption capacity of the electrodes and the maximum voltage on the electrodes, an assembly of 8 permeable electrodes was used. The diagram is shown in Fig. 3.

![Fig. 3. Schematic of the experimental bench for estimating the sorption capacity of electrodes](image3)

Fig. 3.

To investigate the dependence of voltage drop at the serial internal resistance on the distance between the electrodes, an electrochemical module consisting of two electrodes was used. These electrodes were separated by insulating inserts of various thicknesses (3–15 mm) made of plexiglass. Insulating inserts occupied less than 10% of the volume between the electrodes.

Earlier, scientists from the National Scientific Center “Kharkiv Institute of Physics and Technology” determined specific sorption parameters of water capacitive deionization modules at the level of the best world samples. For the subsequent introduction of these devices into various practical applications, it is necessary to investigate the impact of various parameters on energy efficiency.

5. Results of studying the physical parameters that determine the efficiency of the modules of capacitive deionization of water

5.1. The dependence of CDI cell charging-discharge parameters on electrolyte concentration

Fig. 4 shows a typical oscillogram of the CDI cell charge/discharge with a low-voltage CDS1-5M10 power supply. At the first stage, DC charging is carried out in the mode of the current generator to the limiting voltage in the range of 1–1.5 V. After reaching the limiting voltage, the power supply switches to the voltage generator mode, while the cell is charged, an exponential voltage drop occurs, which is limited exclusively automatically.

The waveform shown in Fig. 4 demonstrates that under a DC mode, at the initial moment there is a jumping increase in voltage on the electrodes. This increase is associated with a voltage drop at constant internal resistance.

Fig. 5 shows the 20-minute CDI cell charge/discharge waveforms under voltage-limited modes of 2 V and 1.5 A current for different concentrations of sodium chloride in solutions. The discharge was carried out by maximum current through the internal resistance of the power supply.
The waveforms shown in Fig. 5 demonstrate that the charging modes differ at different concentrations of sodium chloride in the solution, due to the faster achievement of limiting nutrition parameters. Thus, with a decrease in concentration, there is a significant decrease in the charge transferred between the electrodes.

5.2. The dependence of specific sorption capacity on the voltage applied to the electrodes

Fig. 6 shows a plot of the dependence of changes in the concentration of the model solution and the total transferred electric charge on the concentration of the model solution in the interelectrode interval when the electrochemical module is charged for 20 minutes. The electric charge is calculated from the waveforms of current. Concentration changes are calculated as average over three measurements.

In addition, Fig. 7 shows the voltage drops on the serial internal resistance and the voltage drop in the capacitance of the electrochemical module. The voltage drops in capacitance are calculated as the difference between the voltage on the electrodes and the voltage drop on the serial internal resistance over three measurements.
5.3. The dependence of voltage drop in internal resistance on the concentration and distance between electrodes

A significant impact on the efficiency of the process of capacitive deionization of water is exerted by the amount of energy losses, which are due to the Joule heating of water and electrode elements during the flow of current. The characteristic that determines the magnitude of these losses is the sequential internal resistance.

Sequential internal resistance can be represented as follows:

\[ R_{ESR} = R_{cont} + R_{mat} + R_{sol} \]

where \( R_{cont} \) is the resistance on the contacts between the current collector and the carbon material, \( R_{mat} \) is the resistance of the carbon material, \( R_{sol} \) is the resistance of the solution between the electrodes. \( R_{sol} \) grows with increasing distance between the electrodes.

Fig. 8 shows a plot of the dependence of the voltage drop in the serial internal resistance on the distance between the electrodes for different salt concentrations. Voltage drops are calculated as average over three measurements.

The plots shown in Fig. 7 demonstrate that with an increase in the interelectrode distance, there is a linear increase in voltage in the serial internal resistance. For solutions with a conductivity of 10–20 mS/cm (TDS: 7,000–15,000 ppm), the conductivity of the solution has little effect on the overall resistance of the electrochemical system.

6. Discussion of results of studying the parameters of the module of capacitive deionization of water

The influence of electrolyte concentration on the charging-discharge parameters of the electrosorption module has been determined. Similar to [23], the voltage waveform in Fig. 4 has a “step” associated with a drop in potential on a sequential internal resistance. This voltage drop is associated with energy losses on the contacts of electrodes with current collectors, on the resistance of the carbon material and the solution between the electrodes, as described in work [24]. Since the voltage on the electrodes is limited to 1.5 V to prevent the onset of electrolysis, then with an
increase in a voltage drop at the serial internal resistance, there is a decrease in the voltage drop on the capacity of the electrochemical module; a decrease in the amount of charge transferred between the electrodes is observed.

Thus, with a decrease in concentration, there is an increase in voltage drop in the sequential internal resistance and a decrease in the change in the concentration of the model solution (Fig. 6, 7). With a decrease in the concentration of the solution, there is a decrease in the conductivity of the solution and an increase in the component of the sequential internal resistance associated with the resistance of the solution.

The dependence of the specific sorption capacity on the voltage supplied to the electrodes has been determined. Thus, when the voltage drop in the capacity of the double electric layers of the electrochemical module decreases from 1.3 V to 0.7 V (Fig. 7), there is a proportional decrease in the change in the concentration of the model solution, from 205 ppm to 120 ppm. Thus, the electro-sorption of ions associated with the formation of a double electric layer is directly proportional to the voltage drop in the capacitance of the electrochemical module.

The dependence of voltage drop in the serial internal resistance on the concentration of the electrolyte and the distance between the electrodes has been determined. It is worth noting that for concentrations of 10,000–15,000 ppm, the total voltage drop in the electrochemical impedance is less than 0.2 V (Fig. 8), that is, the voltage to which the reactive component of the electrochemical impedance can be charged exceeds 1.3 V. Thus, the resistance contribution of the carbon material and the contact resistance make a negligible contribution to the consistent internal resistance, relative to the contribution of the resistance of the electrolyte solution.

With a decrease in the resistance of the electrolyte (for example, with an increase in the concentration of salts), the contribution of the component associated with the resistance of the solution to the sequential internal resistance increases significantly, therefore, with a decrease in the concentration of ions, thermal energy losses increase during capacitive deionization of solutions.

Thus, in order to reduce energy losses, it is necessary to minimize all the components of the resistance included in the sequential internal resistance. Thus, the spraying of the conductive layer of titanium/titanium nitride can significantly reduce the resistance of the material, and reduce the distance between the electrodes of the electrolyte resistance. On the other hand, minimizing the distance between the electrodes leads to an increase in the hydrodynamic resistance of the flow electrode system. The increase in hydrodynamic resistance leads to an increase in the pressure necessary for pumping the solution through the electrodes and an increase in energy costs for the deionization process. Optimization of all parameters of this process requires additional research.

7. Conclusions

1. The influence of the concentration of salts in the solution on the parameters of charging-discharging the electrosorption module through a change in the voltage drop on the serial internal resistance has been determined. It is shown that with an increase in the concentration of salt ions by 7 times, the charge carried in the solution increased by 2 times. Thus, it has been demonstrated that with an increase in the concentration of salt ions in solution, the efficiency of capacitive deionization increases.

2. The dependence of specific sorption capacity under voltage restrictions on electrodes before the beginning of electrolysis processes has been determined. It is established that with an increase in voltage drop in the serial internal resistance, the charging voltage decreases proportionally, which leads to a proportional decrease in the specific sorption capacity. It has been demonstrated that with an increase in the concentration of salt ions by 13 times, the specific sorption capacity increases by times. Thus, it is shown that the specific sorption capacity of the process of capacitive deionization of water is directly proportional to the transferred charge.

3. It has been shown that the dependence of the voltage drop in the serial internal resistance on the distance between the electrodes depends on the range of electrolyte concentrations. Thus, for high concentrations of salts (more than 5,000 ppm), the distance between the electrodes has almost no effect on the voltage drop in the serial internal resistance. For concentrations exceeding 1,000 ppm, the resistance of the electrolyte is decisive for the drop in resistance on the successive internal resistance.

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