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Review

A Review of Concrete Carbonation and Approaches to Its Research under Irradiation

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Abstract: The current state of knowledge on concrete carbonation has proven that this phenomenon is one of the key factors influencing the reinforced concrete durability reduction during the operational period. To date, the carbonation process has been researched quite deeply; however, the dependence of its course on a variety of external and internal factors poses a significant problem in service life predictions for concrete constructions. The development of nuclear infrastructure around the world in recent years has set scientists the task of investigating such processes in conditions different from those usual for industrial and civil construction. In particular, information in open sources on the course of the carbonation process under irradiation conditions is insufficient. The manuscript analyzes the existing data on concrete carbonation, including a review of the main methods for studying the carbonation process, key factors influencing the course of this process, applied methods of mathematical analysis, predictive models of service life, dynamics of carbonation development, and the application of such analytical models in practice. The available information about the carbonation process under various types of irradiations on the causes, dynamics, and mechanisms of carbonation and corrosion processes occurring in reinforced concrete during operation is also considered. Based on the results of the analysis carried out in the study, recommendations are given for further development in the research field of carbonation process in concrete structures of nuclear power plants in order to comprehensively predict their service life.

Keywords: carbonation; cement-containing materials; durability; irradiation; nuclear power plants; reinforced concrete; service life of reinforced concrete structures

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1. Introduction

Concrete carbonation is a considerably popular topic in the field of construction, material sciences, and production of concrete structures. Due to the presence of carbon dioxide in the air, carbonation may occur in any cement-containing materials [1]. Typically, the carbonation reaction is considered an unfavorable phenomenon since it contributes to a durability decrease in concrete. The most common and greatest negative factor occurring due to concrete carbonation is the corrosion of the reinforcement bars as a consequence of the alkalinity reduction [2].

The process of carbonation occurring in concrete is greatly influenced by the operating conditions of concrete structures and cement-containing materials (e.g., temperature, relative humidity, CO_2 concentration, load, etc.). The main air component that is aggressive towards concrete is carbon dioxide, which neutralizes the cement stone, and other acidic gases interact with the already carbonized concrete. The global volume of CO_2 concentration in the atmosphere is increasing every year due to the rapidly developing industry [2]. Thereby, carbon dioxide affects all reinforced concrete structures in direct contact with the air atmosphere, causing the process of concrete carbonation.

In some studies [1–3], it is noted that the concrete carbonation influence sharply decreases the service life of reinforced concrete structures 20 to 30 years after the operation.

In some cases, the changes in durability of concrete structures as a result of corrosion process was observed after 3–5 years of operation due to the high concentration of CO_2 in the atmosphere and certain environmental conditions, like high temperature and humidity [2–5]. These damages are caused by the steel reinforcement's depassivation, corrosion due to the neutralization of the protective layer of concrete (carbonation), and subsequent destruction by steel corrosion products (rust) that increase in volume. In addition, various technological factors influence the carbonation process's course. Among those factors, the cement phase composition, water–cement ratio, structure durability, and thickness are the most notable [4].

However, it is still rather problematic to predict the dynamics of this process under normal conditions due to the influence of external factors. This issue is especially acute for nuclear power plants (NPP) since these structures are subject to unique external influences, and there are currently very few studies in this area.

The analysis of the technical documents on the design of cement-based reinforced structures showed the lack of a methodology that allows predicting their durability with sufficient accuracy. This is due to the heterogeneity of concrete, the complexity of the physical and chemical processes of concrete carbonation, and the difficulty in predicting the influence of external factors and technological features in the prefabrication process of structures.

2. The Carbonation Process and Methods for Its Research

Due to the low concentration of carbon dioxide in the air (about 0.04%), the carbonation process in concretes proceeds slowly [1,3]. The researchers in the works [1–4] consider this to be the key factor for the research of reinforced concrete durability dynamics. According to the existing data, when conducting a research of this process in a laboratory, various methods of accelerated carbonation of concrete are used [1].

Determining the service life of concrete structures is associated with a number of difficulties in the calculations. This is usually associated with a strong spread of statistical data on the mineralogical composition of concrete and the influence of external factors.

2.1. Chemical and Physical Meanings of the Carbonation Process

Technical documents define carbonation of concrete as a chemical process that reduces the pH from about 12.5 to less than 9 by absorbing carbon dioxide. The chemical reaction (mineralization) between carbon dioxide and the hydration products of cement-containing materials occurs either naturally due to the sorption of carbon dioxide from the air or under some external influence (for example, under the influence of groundwater). It has been scientifically proven that the concrete carbonation dynamics depends on many different factors, such as the phase composition of the cement used, the water–cement ratio, the hardening conditions, and the exposure time [3]. In addition, it was noted that this process is significantly affected by the operating conditions of the structure, such as temperature, humidity, and carbon dioxide content in the surrounding atmosphere.

An important factor is the ability of concrete to sequester carbon from the air. Usually, cement-containing materials, when cured, have a porous structure with open capillary pores on the surface, capable of passing various substances (gaseous and liquid). As a result, when the structure is exposed to open air, CO_2 interacts with cement hydration products, and thus the carbonation process occurs. In the absence of other factors, it proceeds rather slowly, but the influence of temperature and humidity can significantly accelerate it. In the hydrated Portland cement, during the carbonation process, a number of certain stable compounds are formed in accordance with Table 1.

Table 1. Carbonation products of the main hydrates of Ordinary Portland cement [2].

Hydrates	Carbonation Products
$Ca(OH)_2$	$CaCO_3$, H_2O
C-S-H	$CaCO_3$, H_2O, silica gel
Calcium aluminate hydrate	$CaCO_3$, H_2O, aluminate gel
Hydrated ferrite phases	$CaCO_3$, H_2O, iron oxides, aluminate gel
Calcium monosulfoaluminate, ettringite	Gypsum, H_2O, aluminate gel

As it was noted in [3], the concrete is subject to carbonation during the operation of the building due to exposure to the natural environment at a rate of approximately one millimeter per year.

The authors also note that the carbonation process is significantly accelerated with the concentration increase in the CO_2 in the air near concrete structures (and other types of cement-containing products).

As the carbonation rate of the cement-containing materials in the natural environment is considered to be very low, in many studies of the carbonation process, an accelerated test method is used to study this phenomenon, in which the test samples are placed in an environment with a high content of CO_2 [4–8]. As noted by the authors of [4–8], the usage of accelerated methods for laboratory testing is an important tool for researching the dynamics of carbonation of various cement-containing materials, including concretes.

Many studies [2–11] have been carried out on concrete carbonation in a laboratory environment with various conditions. However, such an important point as the simulation of underground construction conditions and the impact of groundwater on the structure were rarely taken into account. The researchers in [9] considered the course of the carbonation process in two variations: "dry" (in the above-ground part of the structure) and "wet" (in the underground part).

The "dry" carbonation method involves the following steps: (1) interaction of CO_2 with water, resulting in the formation of carbonic acid; (2) the resulting carbonic acid interact with calcium hydroxide to form calcium carbonate. In parallel, the reaction of the interaction of C-S-H gel with carbonic acid proceeds, resulting in the formation of amorphous silica gel and calcium carbonate. The calcium carbonate formed at this stage enhances the durability characteristics of concrete, whereas amorphous silica gel reduces the permeability of the material [10,11].

During "wet" carbonation, the brine interacts with the components of concrete, as a result of which the amount of calcium carbonate decreases, and the strength of concrete also decreases.

There is no experimentally confirmed laboratory method for modeling "wet" carbonation that adequately simulates the conditions of the underground construction. Therefore, it is necessary to build a systematic theoretical model to characterize the carbonation reaction processes in terms of the associated phase transformation, reaction rate, and their effects on the mechanical properties of well cement.

According to existing studies [1,3,6–12], the most destructive effect of the carbonation process in concrete is the one on reinforcement. The carbon dioxide penetrates deeply into the material due to the porous structure of the hardened cement paste (HCP). The main product of concrete carbonation is calcium carbonate ($CaCO_3$) [12], which is formed as a result of the interaction of calcium hydroxide ($Ca(OH)_2$) with carbon dioxide (CO_2). The chemical reaction, in this case, takes the following form: $Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$. In addition, calcium carbonate is formed in lesser quantities due to the interactions of other hydrated phases of cement with carbon dioxide (in particular, with calcium silicate hydrates, C-S-H) [12]. In the case of cements with a high content of gypsum and tricalcium aluminate (C_3A), the formation of alumina ferrite tri-sulfate (ettringite, AFt) and monosubstituted alumina ferrite (AFm) also occurs [13].

As a product of cement hydration, calcium carbonate provides the durability characteristics of concrete [14]. However, during the operation of a formed concrete structure,

the carbonation process mostly has a negative effect. Firstly, this is due to a change in the alkaline environment inside the structure [15–17]. When using reinforcement, concrete poses as a protective layer against external influences, and an oxide layer is formed on the surface of the steel reinforcement bars, which prevents corrosion [15]. However, as a result of carbonation, the alkaline environment inside the concrete changes, and the oxide layer depassivates when the pH drops to a level of 9.5 and below [18–23]. That process leads to corrosion of the reinforcement and (in some cases) to the destruction of concrete.

Reinforcement corrosion caused by concrete carbonation causes great damage to the national economy of any country. A study conducted in 1975 showed that the annual losses from various types of corrosion in the United States amounted to about 70 billion US dollars [24]. Reinforcement corrosion losses caused by concrete carbonation accounted for 40% of the total loss [25]. In total, 36% of buildings in the UK need renovation or replacement due to steel corrosion caused by concrete carbonation. In the industrial areas of the former Soviet Union, corrosion of steel was a frequent phenomenon. This happened at most factories of heavy and light industries. This phenomenon has led to colossal losses (approximately 40 billion rubles)—16% of the fixed assets of the industry [25,26].

The researchers in [27] note that carbonation leads to an increase in most of the mechanical characteristics of concrete, including shrinkage, a change in the modulus of elasticity and durability.

One of the ways to improve the operational characteristics of concrete is a dispersed reinforcement with fibers [28–30]. However, the change in the carbonation process, in this case, has been researched insufficiently (especially in the field of NPP construction).

The porosity of concrete is reduced because the formed carbonates occupy a larger volume than the hydroxides. The formation of carbonates within the structure leads to an increase in the density of the material. The water released as a result of the carbonation reaction can react with non-hydrated cement and lead to its delayed hydration. As a result of this mechanism, the pores are closed, and the air permeability of concrete is significantly reduced, which leads to the development of corrosion processes within the material [14]. In particular, reinforcement corrosion occurs, leading to the development of stresses in concrete and subsequent crack opening. Clogging of capillary pores leads to an increase in the strength of the surface layer of concrete [31] and an increase in resistance to those types of external influences that are controlled by the porous structure of concrete [32].

The researchers in [33] were the first to confirm that the alkali–silica reaction in concrete (ASR) is the cause of concrete destruction. ASR occurs in concrete by reacting potassium and/or sodium hydroxide and calcium hydroxide in the solid state with silica in the presence of water. The reaction results in the formation of calcium silicate hydrate (or calcium/sodium silicate hydrate).

It should be noted that alkalis do not participate in the reaction. Initially, from the alkaline phases, anhydrous phases of sodium oxide (NaO_2) and potassium oxide (K_2O) are present in the cement. In the process of hydration, they are converted into sodium/potassium hydroxide ions, whereas they themselves, as ions, do not affect hydration and accumulate in the pore solution. A number of studies [33–35] note that alkalis are contained in some aggregates, and thus the carbonation process is not affected by their presence as long as the pH remains below a certain critical value.

2.2. Accelerated Laboratory Testing for Carbonation Research

The most popular method used in the research of the carbonation process is accelerated carbonation. In practice, various methods of accelerated carbonation of cement-containing materials are used, among which the most common is the aging of the researched samples in a corrosive atmosphere containing high concentrations of CO_2. The most widely used method of carbon dioxide mineralization in cement materials is their early age hardening during carbonation. At an early age, concrete hardening during the carbonation process causes reactions between hydrates and CO_2, resulting in formations of calcium carbonate and silica gel. This method involves the use of special corrosion chambers with CO_2 supply.

A number of studies [5,6,8,26–39] used CO_2 concentrations in the range from 0.15% to 100% as part of the accelerated carbonation method.

In reinforced concrete structures, concrete plays the role of a protective layer that prevents the development of corrosion processes. At the same time, as shown by a number of studies [6,8,15–21], carbonation is a dynamic process that develops on the concrete surface and penetrates deeply into the structure.

The researchers in [6] focused on comparing the carbonation dynamics in natural conditions and in the laboratory when simulating the carbonation process by an accelerated method. In the case of laboratory tests in an environment with the content of calcium carbonate in the chamber at a concentration of up to 100%, a relative humidity of 60%, and a temperature of 22 ± 2 °C, the authors noted that the dynamics of the carbonation process were about 40 times higher compared to the naturally carbonized concrete. To simulate natural carbonation, the authors exposed the concrete test samples to an environment with a relative humidity of 50 ± 5% and a temperature of 22 ± 2 °C.

As a result of the experiments, the researchers in [6] also reported a five-fold increase in the depth of carbonation when conducting an accelerated test with a CO_2 concentration of 5% compared to the natural environment with low carbon dioxide concentration (at around 0.03%). This fact proves the impact of carbon dioxide concentration on the carbonation depth. Further analysis indicated that the higher concentrations of carbon dioxide (more than 5%) significantly influenced the microstructure formation compared to the natural carbonation process.

The researchers in [5] studied the changes in the chemical and phase composition of the HCP at different concentrations of CO_2. The concentrations they chose were as follows: natural environment simulation (0.03%), 3%, 10%, and 100%. As a conclusion, they noted that the microstructures of HCP contained in an environment with a CO_2 concentration of up to 3% and in a natural environment (i.e., 0.03% CO_2) were practically no different. In addition, they found significant differences in the microstructures of HCP carbonized at higher CO_2 concentrations (between 10 and 100%).

The researchers in [37] proposed the following test sequence of the accelerated carbonation method. Concrete samples were manufactured in the form of cylinders with a diameter of 150 mm or 100 mm cubes. After demolding, the samples were moved to the drying chamber and kept to the state of 60% of internal relative humidity. Then, the test samples were placed in a chamber with a CO_2 concentration of 100% under a pressure of 1.5 MPa. According to the researchers, this approach makes it possible to achieve complete saturation of the samples with gas in two weeks and to speed up the carbonation process as much as possible. At the same time, it was noted that the use of cements with a higher water–cement ratio increased the rate of CO_2 diffusion during laboratory tests.

In a number of studies [37–43], it was noted that early carbonation could have a positive effect on the production of precast concrete structures. This phenomenon can be explained by the fact that carbonation accelerates durability development and does not have significant impact on the formation of the microstructure in the early stages of concrete hardening. As a result, a controlled carbonation process can accelerate the production of reinforced concrete structures. According to a number of researchers, the durability of ordinary concrete is not worsened by carbonation but is rather positive [44].

The existence of two main useful features associated with the carbonation of cement systems has been experimentally proven. The first is the accelerated durability gain of the cement matrix during curing process due to the reaction of carbonation [45]. The second is the ability of concrete to capture CO_2. The issue of CO_2 absorption by concrete (sequestration) has been raised in the scientific community for a long time. The volume of concrete use in the world today is second only to the volume of water consumption [46,47].

The researchers in [48] focus on the environmental component of cement-containing materials and the possibility of reducing harmful carbon dioxide emissions by using the carbonation reaction. However, for this, it is necessary to adapt the composition of such materials for targeted CO_2 absorption. They concluded that the diffusion rate

of carbon dioxide has a significant effect on the carbonation process, on the formation of the microstructure and volume of the pore space of concrete during hardening in particular [49–51].

In a number of literary sources [3,5–7,14,36], to determine the effect of carbonation reaction on the changes in physical and mechanical properties of concrete and the research of the corrosion process, attempts were made to carry out a complete carbonation of the studied samples. The researchers in [37] prepared a test facility that was capable of carrying out the maximum possible carbonation of concrete samples (up to 150 mm in diameter or up to 100 mm × 100 mm in cross section) for two weeks, using not only high CO_2 content but also maintaining high pressure (about 1500 kPa).

The correlation of data obtained as a result of natural and accelerated carbonation of various cement-containing materials is still a significant problem [6,52,53]. It is related to the difference in carbonate phases [54] caused by the carbon dioxide concentration differences. In addition, the CO_2 concentration affects the carbonation rate of calcium silicate hydrates (C-S-H) [36], pH of the solution, pore volume, mineral composition, and pore microstructure of carbonized samples [55].

The researchers in [56] noted that the products of the carbonation reaction could differ significantly from those obtained in the natural environment with a high concentration of CO_2 during accelerated tests. This was due to a change in the reactivity of some phases with a change in the alkalinity of the environment.

Given these results, the accelerated carbonation test data's applicability highly depends on the initial choice of appropriate exposure conditions for concrete. For Portland cement-containing materials, the similarity in microstructure changes caused by carbonation at low CO_2 concentrations (0.03 to 3%) was observed [52–56]. Therefore, it is possible to simulate the natural carbonation of concrete by an accelerated testing procedure using CO_2 concentrations up to 3% [36].

2.3. Possible Research Methods for Studying Carbonation

The use of phenolphthalein solution (as a pH indicator) is a widely applied test method that allows determining the depth of carbonation in cement-containing materials. When the pH level exceeds 9.0, the color of the solution changes from colorless to red (or pink). During the carbonation of the concrete system, the pH level decreases. As a result, the phenolphthalein solution is colorless on the carbonized surface and red (or pink) on the non-carbonized surface.

The red (pink) color on the surface indicates a pH level above 9.0, but the sample surface may still be partially carbonized. Consequently, this test method allows only an approximate assessment of the carbonation depth.

Thermal gravimetric analysis (TGA) makes it possible to quantify the degree of carbonation of cement-containing materials. This test method allows determining the relative changes in some components of the phase composition of cement, e.g., $Ca(OH)_2$ and $CaCO_3$. By studying the changes in the concentrations of these components, it is possible to understand the degree of carbonation in the samples under research. Sometimes TGA in combination with mass spectrometry (TGA-MS) [32] or infrared spectrometry (TGA-IR) is used to analyze carbonized samples. These test methods (TGA-MS and TGA-IR) make it possible to carry out a comprehensive analysis of gases released when carbonized samples are heated. Therefore, decomposing phases can be identified.

The application of Fourier transform infrared spectroscopy (FTIR) is widely used to obtain qualitative [57–59] or semi-quantitative [60] analysis of carbonation of cement-containing materials. This method is used to study the formation of carbonate phases based on the spectrum vibrations in the mid-infrared region. At the same time, the capabilities of this method make it possible to identify various phases of anhydrous calcium carbonate (calcite, vaterite, and aragonite). Unlike other methods, FTIR allows studying the dynamics of the process since the method relies on the registration of the vibrations of characteristic groups and does not depend on the crystallinity of the sample. At the same time, Raman

spectroscopy and scanning electron microscopy can be used in addition to the FTIR method to bring more detailed information about the carbonation process [60].

Modern research methods of materials also use Raman spectroscopy as a means of obtaining qualitative information on changes in phase composition of cement-containing materials occurring during carbonation. For example, the most common things registered by Raman spectroscopy in this area of science include the research of the formation of $CaCO_3$, polymerization of C-S-H, decomposition of ettringite, etc. [61,62].

The scanning electron microscopy (SEM) method is mainly used to obtain a qualitative assessment of the carbonation effect on the cement-containing material's microstructure. SEM is multifunctional. It can be used to assess changes in the chemical composition of Portland cement stone when used in conjunction with energy dispersion spectrometers (EMF) [63,64]. By using SEM, it is also possible to obtain quantitative information, for example, porosity (from SEM image analysis [63]).

Nuclear magnetic resonance (NMR) spectrometers 29Si (also 27Al) are used to assess the silicate (and aluminate) particles' polymerization under the effect of carbonation [65–67]. In the study in [65], the possibility of identifying the presence of carbonate particles presence in an aqueous solution using the cross-polarized 13C {1 H} NMR was shown.

One of the analytical methods for quantitative assessment of changes in the phase composition of cement-containing materials is the X-ray diffraction (XRD) method. This research makes it possible to identify the processes developing as a result of carbonation of samples.

Existing studies have confirmed the possibility of using gamma rays to monitor the dynamics of the density of the cement matrix as a result of carbonation [32,68,69].

In the study in [70], the researchers used the distribution function of X-ray pairs to explore the particle size changes in C-S-H phase during the carbonation reaction.

For a visual assessment of structural changes in concrete, an advanced modern method of micro-computed tomography (CT) can be used. In the process of examining samples, micro/meso scale models are created. Based on images, realistic sizes, shapes, volume fractions, and distributions of several phases are taken into account [71–73]. Thus, this method makes it possible to evaluate the pore space (as well as its changes) and the transformation of phases during the carbonation of concrete [74].

3. Existing Mathematical Models and Computer/Numerical Methods for the Calculation of Concrete Carbonation

3.1. Fundamentals of Carbonation Modeling

At present moment, there are a number of different mathematical predictive models for the carbonation process, compiled by the researchers based on the results of the studies, taking into account various factors. Among the most notable are the early theoretical model [22], the experimental model [75–78] based on the combination and interrelation of theoretical and experimental data [79–83], etc. This happens for several reasons, including a wide range of influencing factors, the complexity of selecting and taking into account environmental conditions, the discreteness of material properties, and the uncertainty of a number of other factors. The combination of all these factors directly affects the course and rate of concrete carbonation.

According to the results of numerous studies [1–3,5,6,46–48,82–85], by the year 2000, the global concentration of CO_2 in the atmosphere increased to about 365 parts per million. Based on the available data, the researchers extended the available statistics and concluded that the concentration of CO_2 in the atmosphere will exceed 1000 parts per million at the end of 2100 [85].

The possible influence of global climatic changes on concrete carbonation has been considered in some studies [81–83,86]. The researchers in [81] established the correlation between the concentration of CO_2 in the atmosphere and the concrete carbonation degree.

In 2007, a model for predicting carbonation was proposed based on the data obtained in [81] and Fick's first law [74].

Based on the Yun carbonation reaction [81], the researchers [87] proposed a model that could take into account the variable concentration of carbon dioxide in the environment as the main model for calculating carbonation. In addition, in further studies, the researchers explored the impact on concrete carbonation of various scenarios of CO_2 emissions into the environment with high, medium, and low carbon emissions in accordance with the IPCC report data on climate research in 2010 [82]. The researchers in [83] improved Yun's model [81] by proposing to carry out an integral correction of the volume of CO_2 reacting during carbonation taking into account climate change. They also studied the influence of the environment on concrete carbonation in Australia. In the research, they determined the residual service life of reinforced concrete structures with an increase in temperature and CO_2 concentration (based on climate forecasting data in 2011) [83].

3.2. Predicting the Service Life of Concrete Structures Using the Existing Mathematical Models

At the moment, numerous studies on concrete carbonation have been carried out. However, it should be taken into account that there are countless differences in the experimental conditions, as well as in the compositions of the tested concrete. Therefore, generalization of observations by one mathematical model is not possible. Each of the factors is regulated by its own laws, interconnected with each other. As an example, the CO_2 diffusion depends on the concrete structure transport properties. These properties directly depend on the type of cement, moisture content, and the temperature of the environment in which the concrete is located. During the carbonation of concrete or cement-containing materials, the decomposition of portlandite occurs with the formation of water. This fact will inevitably affect CO_2 and water diffusion [88]. Any analytical model for predicting carbonation is, by its very nature, already quite complex to calculate because of all conjugated (or synergistic) effects of various factors involved in the carbonation process.

For reinforced concrete structures, it is important to prevent complete carbonation of the protective layer of concrete. Under the condition of reinforcement corrosion, it becomes difficult to determine the concrete structure's service life [44]. The carbonation process begins with the concrete's surface. Due to the direct contact of the surface with carbon dioxide and water, it will carbonize rapidly. However, it was noted that a layer of carbonized concrete slows down the carbonation process of the inner layers. Consequently, the carbonation rate is not constant. Over time, the changes in concrete microstructure cause the carbonation process to slow down, as the rate at which the carbon dioxide passes through a thickening layer of modified concrete components lowers [88].

There are numerous studies in the literature based on existing experimental data, aimed at improving the understanding of the kinetics of carbonation of concrete [44,54,85,89–104]. A number of early experimental studies regarding concrete carbonation, both in vivo [89,96,105–108] and controlled indoor conditions [107,109–111], have led the researchers to put forward hypotheses. The approximated data show that the concrete carbonation depth (the "carbonation boundary") is directly proportional to the square root of the concrete age [19].

Random field modelling [112] is a recently developed and refined method for modelling the spatial randomness of carbonation properties in concrete structures. Random field modelling of heterogeneity is used to eliminate the dependence of numerical results on the geometric representation. To model the spatial variability of material properties, the theories of stochastic process are applied. Random fields of material properties are regarded as abstractions of microstructures and adhere to specific statistical criteria, including the mean, standard derivation, and correlation length, which are used in engineering applications [112].

Various predictive models based on the stoichiometry reaction $[Ca(OH)_2] = [Ca^{2+}]$ have also been developed. The researchers in [113] proposed a predictive model for the porosity variation during carbonation based on diffusion processes occurring in the pore

space of concrete. The mathematical algorithm proposed by the studies makes it possible to describe the process of formation of the pore space as a result of the interaction of concrete with silicon dioxide. The researchers in [9] proposed a predictive model for the change in volume fractions due to the carbonation reactions. The stoichiometric model was used to analyze phase transformations occurring in well cement during carbonation.

Recently, the researchers in [92] developed the finite elemental framework software (based on the ANSYS 14.0 software). The use of this software allows us to analyze the influence of the most important factors on the process of concrete carbonation. The analysis of the used software parameters allows us to set analogies of concrete carbonation processes, including complex thermal analysis with certain experimental conditions. The originality of the software is the consideration of nonlinear time-dependent factors affecting the carbonation process, as well as their interactions. Most of the researchers who have developed practical models for the analysis of concrete carbonation have made a general conclusion about the difficulty in creating a universal carbonation model that is able to describe all possible factors affecting the carbonation process, due to their large numbers and the large discretization of data in laboratory tests or in real life conditions.

Currently, in this area, the application of various machine-learning methods (including artificial neural networks application) to create mathematical models in solving this issue are known [111,114–116].

According to the study in [116], artificial neural networks proved to be effective in carbonation analysis. A model based on a neural network with a radial basis function (RBF) was created by some researchers [116]. The ratios between gel phases, cement content, and concrete hardening time were chosen as input parameters. The output data were only the depth of concrete carbonation. A total of 72 concrete carbonation datasets were used to build the RBF neural network and carbonation model, of which 60 datasets were applied for training, and another 12 datasets were applied for model validation. The results of the prediction model showed that the obtained data correlated well enough with the test results.

Artificial neural networks have proven to be very useful in determining a number of other parameters, such as the diffusion coefficient, etc. To receive the comparative data set on the diffusion coefficient of CO_2, the researchers in [114] analyzed various experimental results in their research. Several components of the mixture design were chosen as neurons for subsequent neural network training using a back-propagation algorithm. These parameters were the cement content, the ratio of water to cement, and the volume of the aggregate, including the conditions of exposure to relative humidity.

4. The State of Research of Concrete Carbonation Process under Radiation

Concrete structures of NPP operate not only in conditions of designed physical and mechanical loads and environmental influences, but these structures are also affected by radioactive radiation.

Many literary sources have researched environmental factors and operating conditions affecting the carbonation process in concrete. However, the NPP-specific factor., i.e., the presence of radiation and its effect on concrete carbonation, depending on the type and power, was not considered. Also, the existing mathematical models for carbonation calculation do not take that factor into account.

4.1. The Influence of Radiation on the Concrete Structures of NPP

The research of the radiation effect on concrete is directly related to the possibility of extending the NPP service life. Of particular interest to researchers is the effect of radiation exposure (primarily the irradiation fluence and the received dose) on the changes in the physical and mechanical properties of concrete structures of NPPs with light water reactors (LWR). In addition, there is great interest in the issue of changing the dynamics of corrosion processes in reinforced concrete during irradiation.

Studies on the effects of irradiation in concrete were carried out by Clark (1958) [117], Komarovsky (1961) [118], Dubrovsky (2012) [119], and Fillmore (2004) [120]. The most widely cited review is Hilsdorff's (1978) [121]. The review shows that the loss of concrete compressive strength begins at the neutron fluence of about 1.0×10^{19} n/cm^2. In [122], a similar result was observed for the concrete tensile strength at the same neutron fluence.

According to the data from these sources, based on the results of their research, the researchers made the following general findings on the changes in the concrete mechanical properties during irradiation:

- Gamma radiations have no effect on the concrete properties up to 10^{10} Gy;
- At fluences of fast neutrons more than $\sim 10^{19}$ neutron/cm^2, a significant degradation of concrete occurs, and the volume of the aggregates increases with such irradiation, which causes shrinkage of the HCP, resulting in intense cracking;
- The defined value of concrete shrinkage strain at a dose of 5×10^{19} neutron/cm^2 is about 2% for Portland cement and 0.3% for aluminous cement;
- Volume increase in the aggregate changes depending on the type of aggregate: at a fluence of 5×10^{19} neutron/cm^2, it is approximately 1.0% for flint, 0.9% for limestone, and 0.1% for serpentinite used as a filler in the concrete of the biological protection of the reactor;
- Based on the obtained data on the volumetric expansion of the aggregate and the deformation of concrete during irradiation, it is noted that the actual values of these parameters are greater than the calculated ones;
- The calculated and actual value of the concrete expansion ratio differ significantly during irradiation, and the researchers in [122] explained this by the aggregate mineralogical composition impact on the carbonation process;
- In the case of a high content of silicon oxide in the aggregate, the actual value of the concrete expansion rate is greater, e.g., the concrete expansion rate for a sample with siliceous aggregate is about five times higher than for a sample with limestone;
- When irradiated, only a slight expansion of crystalline quartz is observed, whereas the usage of quartz as an aggregate for concrete causes a significant concrete expansion at a fluence of 3×10^{19} neutrons/cm^2, and, at the same time, the deterioration of the concrete mechanical properties due to irradiation with fast neutrons is greater for concretes with a higher deformation degree;
- The negative effect of irradiation on the concrete tensile strength is more significant than on the compressive strength, and this statement suggests that the degradation of concrete under the influence of fast neutrons occurs not simply due to a violation of the structure of the cement paste and aggregates but rather due to complex internal processes inside the irradiated cement and aggregates.

In a study [123], researchers explored the effect of the neutron fluence increase on the compressive strength of concrete and confirmed the results of the research [121]. It is noted in some studies [117–123] that the effect of concrete irradiation depends on a large quantity of variables, including aggregate type, cement mineral composition, mixture proportions, curing conditions, environmental exposure, and internal moisture content. In addition, it was noted that many key factors influencing the carbonation process were not sufficiently researched in existing reviews on this issue.

The researchers in [124] considered the effect of radiation on concrete, and data on irradiated concrete are systematized. Along with the changes in the strength indicators, the researchers also specified the appearance of volumetric expansion and a change in porosity under the action of neutron irradiation. It should be noted that the volumetric expansion and change in porosity have a significant effect on the rate of the concrete carbonation process. The carbonation rate increase is also significantly affected by elevated temperatures and a decrease in pH.

Further research is needed to fill the knowledge gaps on corrosion process development dynamics change due to the irradiation effect. This will allow a better understanding

of the changes in the physical and mechanical properties that occur in the concrete structures of NPPs, and the course of the carbonation process under radiation exposure.

4.2. Extending the Service Life of NPP Concrete Structures Based on Changes in the Concrete Properties

In some studies [117–123], structural, physical, and mechanical changes were noted during the irradiation of concrete. In irradiated concrete, the pH of the cement paste decreases, the durability changes, and the linear dimensions of the components of the mortar matrix and concrete aggregates change as well.

It should be noted that the given fluence values at which the harmful effects of radiation on concrete were observed were not considered in most of the existing studies on the long service life expected for concrete structures (>30–40 years) since the possibility of NPP lifetime extension was not considered at the time of publication [117,121].

The nuclear industry needs to research the influence of radiation exposure on biological protection for up to 80 years. This will provide a justification for assessing the possibility of extending the life of the NPP. In addition, an explanation of the effect of the general fluence of neutrons on the properties of concrete and on the rate of corrosion in concrete is required.

The Atomic Energy Act of 1954 [125] and the rules of the Nuclear Regulatory Commission (NRC) regulate the operation of commercial power reactors. Primary licenses are issued for 40 years and can be extended for another 20 years without restrictions on the number of renewals. Primary license period (40 years) is proved on the basis of economic and antimonopoly considerations. To date, the service life of many NPPs has been extended based on their technical conditions by 20 years.

The average service life of concrete structures in the USA is 32 years [126]. The lifetime of most NPPs in the United States has already been extended from 40 to 60 years, and a further prolongation to 80 years is currently being considered. In the Russian Federation, the standard operating life of a concrete structure was 30 years. However, taking into account the technical state at present and the extension period, the operating life for individual NPP units is 50 years or more.

Extending the lifetime of NPPs has initiated the research of potential factors affecting the wear of concrete structures [127]. In the research in [127], the main emphasis is placed on the level of technical and operational experience available, the predicted decrease in the operational properties of concrete, and the probability of concrete deformation under the radiation influence. In the same work, a detailed analysis of degradation under the influence of radiation of cement-containing materials and degradation mechanisms, including concrete corrosion, was carried out. The researchers concluded in their study that most of the factors with negative impact on the operational characteristics of concrete structures could potentially affect the containment concrete structure, which is of primary interest in relation to NPP safety.

A similar research [128] was conducted in France, where NPPs were the basis of the country's energy supply. At the time of the research, nuclear power plants in the country had an average operating life of 27 years (from 13 to 34 years). In this work, the researchers also paid special attention to the issue of extending the life of NPPs after 40 years or more.

Many researchers in their studies [129–141] noted that various types of radiation had different mechanisms of influencing the changes in concretes, but everyone agreed that there were significant decreases in its physical and mechanical properties as a result of internal processes in concrete, initiated by irradiation.

4.3. Influence of Various Factors on Concrete Carbonation under Irradiation

4.3.1. Effect of Gamma Radiation on Concrete Carbonation

The authors of the work [129] investigated the changes in the properties of concrete under the influence of γ-radiation. In their research, they found that a variety of physico-

chemical processes took place in the structure of concrete cement stone during operation. These processes directly affect the carbonation of concrete:

- Hydrolysis of molecular water occurs, and, at this time, H_2O_2 peroxide is formed;
- H_2O_2 reacts with calcium (contained in cement), and, as a result of this reaction, $CaO_2 \cdot 8H_2O$ peroxide octahydrate is formed;
- This substance is metastable and decomposes as a result of subsequent carbonation reactions, and, eventually, calcium peroxide and water are formed;
- CaO_2 reacts with H_2O, and the formation of portlandite $Ca(OH)_2$ and oxygen occurs;
- Portlandite and CO_2 react, carbon dioxide enters the concrete through the pores, and, as a result of the reaction, calcite $CaCO_3$ and water are formed.

From this follows the conclusion that the absorbed radiation dose affects the calcite content. The irradiation of concrete by gamma-radiation causes a sequence of chemical reactions in the material. This is accompanied by radiolysis of water, as well as the formation of calcite. Calcite crystals are known to reduce the size of the pore space and the material's durability [130].

4.3.2. Effect of Neutron Radiation on Concrete Carbonation

Modern concepts about the changes in the concrete mechanical properties under the influence of neutron radiation are based on the Hilsdorff curves [131]. The data for those curves were initially obtained from the studies in the 1970s. In studies of 2013 [131], the Hilsdorf compressive strength curve was revised based on the recently occurred data.

An inherent problem observed in existing studies is the lack of systematic studies covering the entire range of energy densities that can be expected in LWR biological protection. Only a few researchers [132–141] have carried out studies with fluences above 1×10^{19} neutron/cm^2.

The changes in concrete structures under the effect of neutron irradiation (typical for LWR power plants) have been considered from the point of their performance during the long-term operation period. In [124], the researchers made several important conclusions based on the analysis in their research:

- The neutron irradiation of concrete at energy density levels of 1.0×10^{19} neutron/cm^2 and above can significantly reduce the compressive strength (with the lower limits of concrete durability at 50% of the estimated value);
- The tensile strength of concrete is more susceptible to neutron radiation than the compressive strength;
- At a density levels of 1.0×10^{19} neutron/cm^2 and above, a gradual decrease in the elasticity modulus was observed, and, in this case, it should be taken into account that the effect of elevated temperature is also observed in this range of irradiation density, which should also be taken into account in the calculations;
- Data indicate that silica aggregates pose the highest risk of adverse effects on concrete due to its tendency to amorphize under the influence of neutron irradiation, the increase in volume, and the higher thermal expansion coefficient;
- It is expected that the calculated fluence of fast neutrons (with an energy of more than 0.1 MeV) in the LWR biological shield will reach levels over a long period of operation (more than 40 years) at which negative changes in the concrete's physical and mechanical properties will be significant.

In the considered studies, the neutron radiation influence on concrete carbonation was not studied. However, taking into account the results in a detailed study of the mechanisms of destruction of concrete [130], as a result of carbonation of concrete, a decrease in pH and a loss of passivating properties should be expected.

4.3.3. Effect of Other Notable Factors on Concrete Carbonation under Irradiation

There are very few open data on changes in the properties of NPP concrete structures under the influence of radiation. Despite this, the researchers in [142,143] found that there

are cases of corrosion of NPP concrete under the action of ASR. For example, in the case of the reinforced concrete foundation of the turbine generator at Ikata NPP No. 1 (Ikata, Japan, Shihoku Electric Power), there are signs of corrosion due to ASR [142].

To assess the state of NPP concrete structures, the researchers in [144] conducted a study based on the following data: structure location, the degree of concrete cracking, concrete deformation, changes in concrete compressive strength, and the elasticity modulus.

The research of the ASR effect on the changes in the physical and mechanical properties of reinforced concrete structures of NPPs was carried out in [145]. In [145], the compressive strength and tensile strength were determined on the prism-shaped concrete samples subjected to ASR. To simulate the ASR effect, the samples were kept for about six weeks in an environment of 100% relative humidity and a temperature of 40 °C. At the same time, only the early age of concrete was covered by the data obtained in the research. Thus, it did not allow assessing the effect of ASR on NPP concretes whose ages exceed 30 years.

In the research in [146], the question was first raised about the possibility of the occurrence of ASR in concrete during irradiation since it increases the reactivity of the alkaline phases contained in some aggregates.

The researchers in [147] suggested that a decrease in the resistance of a concrete structure to radiation occurs due to the significant content of silicon oxide phases in the aggregates used.

Based on this statement, they made the following conclusions:

- The silicate aggregates' reactivity significantly increases under the influence of irradiation, although the occurrence of ASR is not guaranteed due to the different initial conditions necessary for this;
- The concrete of the reactor containment does not receive the highest dose of radiation;
- The calculated absorbed dose of gamma radiation for NPP concrete with a service life of 60 years is about 109 Gy, and this value is below the critical level at which significant physical and mechanical changes in concrete occur.

To assess the "theoretical" risk of ASR, the electric utility EdF developed a methodology in the 1990s based on the following parameters:

- Calculation of the active alkali content in the concrete mixture in accordance with the French recommendations LCPC 1994, and active alkali in concrete includes active alkali from all concrete components (cement, aggregates, water, etc.);
- The validation of the aggregate in accordance with the French LCPC 1994 guidelines (non-reactive, potentially reactive, or potentially reactive with passivating effect);
- Characterization of the concrete structure from an environmental point of view (elevated temperature, humidity, or normal operating conditions).

In the field of nuclear energy, there are strict requirements for ensuring the safety of nuclear power plants. For this industry, the study of reinforcement corrosion caused by concrete carbonation under the influence of radioactive radiation is relevant.

The researchers in [148] concluded that understanding the process of interaction of ionizing radiation with the mineral constituents of concrete for protective structures of nuclear reactors and subsequent corrosion is important for developing measures for the maintenance of NPPs, as well as determining the levels of safety in NPPs and the possibility of extending their service lives. In addition, they noted that water under the influence of ionizing radiation decomposes into oxidizing and reducing particles (OH, H_2O_2, O_2, etc.). This interaction of particles with steel reinforcement has not been researched well enough. Radiolysis products are chemically active. Their concentrations in the phase compositions of homogeneous solutions can be determined by a number of factors (fluctuations in pH and temperature and the presence of other dissolved substances).

The researchers in [149,150] noted that the formations of H_2 and H_2O_2 increased by more than two orders of magnitude during radiolysis, with an increase in pH from 6.0 to 10.6. Observations in [129] demonstrated the dependence of a number of characteristics on pH and potential. The factors of formation, conversion of the oxide film on carbon

steel, and the rate of corrosion directly depend on the pH and potential. This leads to the conclusion that the effect of radiolysis of water on metal corrosion and the transfer of corrosion products is often difficult to predict.

It should be taken into account that metal structures are used in conditions of ionizing radiation. The mechanism of the effect of the radiation on the corrosion of metals (including carbon steel) is insufficiently studied [146]. Many of the observed effects of ionizing radiation are contradictory. Many key questions remain unanswered. There is no reliable information that the radiation energy initially absorbed in the main metallic or aqueous phase is a key factor in the corrosion of materials.

Studies on the influence of γ-radiation, carried out by some researchers [148–151], showed that corrosion of carbon steel increases under the conditions of radiation exposure.

5. Conclusions

The analysis of the results of the literary sources shows that the carbonation of concrete is an important indicator that determines the durability of reinforced concrete structures of NPPs.

The researchers of the publications studied the influences of various factors on carbonation, including radiation. However, in the published materials, there is a lack of experimental data due to the long experimental period under conditions of exposure to radiation and of theoretical studies allowing the development of a methodology for assessing the effect of radiation on concrete carbonation.

At the moment, there is no publicly available information in the literature on the process of concrete carbonation of NPP-reinforced concrete structures in the conditions of operation periods of power units. However, an analysis of the state of research allows us to conclude that the irradiation influence on the concrete corrosion process and, namely, concrete carbonation is a popular scientific direction that makes it possible to increase the accuracy of predicting the service lives of NPP-reinforced concrete structures. Furthermore, very little research has been performed in this area.

Carbonation process dynamics prediction is a scientifically complex problem mainly because there are many different production and operational factors that must be considered. For NPPs, many of those factors are unique. Therefore, most of the existing carbonation prediction models for civil engineering are inapplicable to concrete constructions of NPPs.

6. Future Directions

Existing methods for the analysis of concrete carbonation are generally applicable to NPP structures exposed to radioactive radiation. However, the absence of an accelerated carbonation technique that simulates the effect of radiation on concrete in laboratory conditions greatly complicates the research process in this area. Based on the current analysis, for further development of a methodology for assessing the concrete carbonation under irradiation conditions, it is recommended to consider the effect of concrete components used in the protective structures of NPPs with VVER and PWR reactors on the microstructure and the concrete carbonized zone formation during accelerated testing. Also, it is suggested to consider the effect of a combination of factors operating environment and radiation on the carbonized zone and its microstructure during accelerated carbonation.

The predictive models of concrete carbonation considered above in most cases are based on many external factors. However, the influence of radiation exposure is not taken into account in any of them. It should be noted that the "multi-factorial nature" of such models makes it possible to make appropriate adjustments and adapt them to various conditions. In the case of nuclear power plants, for example, there are studies [118,119,133–135] confirming the possibility of simulating concrete radiation exposure in laboratory conditions using a special temperature and humidity regime. By taking this approach as a basis, it is possible to create a predictive model for assessing the durability of NPP concrete structures, subject to full-scale laboratory research.

To develop carbonation depth predictive models for NPP-reinforced concrete constructions with service lives up to 100 years, it is necessary to conduct experimental studies on the carbonation process, taking into account such factors as the type and degree of radiation influence, CO_2 content, relative humidity, temperature, water–cement ratio, types of filler, types and amounts of mineral additives and admixtures, the amounts of cement used, and the brand of concrete.

Such an approach to further research in this area will make it possible to progress the development of a universal prediction model for concrete carbonation and the comprehensive methodology that allows us (with sufficient accuracy) to predict the reinforced concrete structures' durability of both NPP power units and the building structures of general-purpose buildings, taking into account the concrete carbonation mechanism.

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