Analytical description of the hydrogen evolution from concrete under the effect of gamma radiation

Aleksandr Denisov
Moscow State University of Civil Engineering, Yaroslavskoe shosse, 26, Moscow, Russia
E-mail: DenisovAV@mgsu.ru

Abstract. The formation of explosive gas mixtures due to the evolution of hydrogen from concrete structures for radiation protection of nuclear power buildings should be taken into account when designing and operating them. However, this requires methods for estimating and predicting the radiation evolution of hydrogen.

This paper was done due to the lack of analytical methods for determining the gas evolution from concrete and their components under the influence of gamma radiation. In the work, based on the available experimental data, the model was selected, checked, and adjusted for an analytical description of the process of hydrogen evolution from concrete on aggregates that do not contain water under the influence of gamma radiation.

When selecting the model, it was assumed that the evolution of hydrogen from concrete on aggregates that do not contain water under the effect of gamma radiation is mainly due to the radiation decomposition of evaporable water (free water and adsorbed water) from hardened cement paste (cement stone) and is determined by the amount of evaporable water in the material and the power of the absorbed dose of gamma radiation. It was taken into account that the amount of evaporable water during irradiation decreases with increasing time and temperature of irradiation.

Verification of the possibility of using the selected model, its necessary adjustment and determination of the main parameters of this model was performed on the basis of available experimental data on the evolution of hydrogen from Portland cement stone under the influence of gamma radiation. We used experimental data on the release of hydrogen and water from cement stone containing and practically not containing evaporable water when irradiated with gamma rays with an average energy of 1.25 MeV at the absorbed dose rate of 0.87 kGy/h, 3.84 kGy/h and 7.05 kGy/h at temperatures of 25, 40 and 60 °C for up to 2 months. The dependence of the model parameters on the absorbed dose rate of gamma radiation and the irradiation temperature is shown. For cement stone dried at 120 °C to remove evaporable water, the amount of residual evaporable water (residual evaporable water or decomposed chemically bound water) was studied and determined.

1. Introduction
Concrete structures for radiation shielding of nuclear power buildings are exposed to ionizing radiation. In this regard, issues of radiation stability and radiation changes of concrete are quite important.

At the present time, radiation changes in concrete have been sufficiently studied [1-24], methods for their analytical determination [9-12, 18] have been developed. But in addition to radiation changes as a result of radiolysis of water from concrete and their components, gases (mainly hydrogen) are
allocated [2-4, 18, 25-27], resulting in the formation of an explosive mixture. This should be taken into account in the design and operation of nuclear facilities. In this regard, the study of the radiation evolution of hydrogen from concrete and their components is relevant.

Radiation evolution of gases under the influence of ionizing radiation is the least studied. Data on the evolution of gases from concrete and their components during irradiation in nuclear reactors are given in [2-4, 18, 25, 26]. Moreover, the papers [18, 25] consider an analytical description of the process of gas evolution during irradiation in a nuclear reactor, as a function of neutron fluence, evaporable water content, irradiation temperature, and pressure. The evolution of gases from concretes and their components under the influence of gamma radiation is shown in [2, 3, 26, 27]. However, the issues of analytical description of the radiation evolution of gases under the effect of gamma radiation were not considered.

The purpose of this work is to substantiate the method of analytical description of the radiation evolution of hydrogen under the effect of gamma radiation from hardened cement paste (cement stone) and concrete on aggregates that do not contain water.

2. Research Methodology

To achieve the goal set in the work, the following main tasks were solved:

- Selection of model for analytical description of the process of hydrogen evolution under the action of gamma radiation from cement stone and concrete on aggregates that do not contain water.

- Checking on the basis of existing experimental data the possibility of using the selected model, making the necessary adjustments and determining the values of the parameters of this model for the conditions of existing experimental data.

When choosing a physical and mathematical model for analytical description of the process of radiation evolution of hydrogen from cement stone and concrete, the following regularities were taken into account, arising from existing experimental data [2-4, 18, 25-27]:

- According to [2-4, 18, 25, 26], radiation evolution of gases (mainly hydrogen) from concrete occurs due to radiolysis of water contained in the cement stone and in hydrate fillers. Radiation evolution of hydrogen from concrete on aggregates that do not contain water is caused by gas evolution from cement stone due to the presence of water in it.

- According to experimental data [26, 27], in most cases, the rate of hydrogen evolution from a cement stone under the effect of gamma radiation depends primarily on the amount of evaporable water (free water and adsorbed water), the absorbed dose rate, and the time of irradiation. The rate of gas evolution increases with increasing dose rate and the amount of evaporable water, decreases with increasing exposure time due to the release of water under the influence of heating and radiation. Although in experiments [26, 27] at the very beginning of irradiation, the rate of hydrogen evolution first increases, reaching a maximum. However, this is explained first by the accumulation of hydrogen in the structure (pores) of the material and only then by its evolution from the material [27].

In this regard, the choice of a model for analytical description of the process of radiation evolution of hydrogen under the effect of gamma radiation from cement stone and concrete on aggregates that do not contain water was carried out based on the following provisions:

- The evolution of hydrogen under the effect of gamma radiation from cement stone and concrete on aggregates that do not contain water is mainly caused by the radiation decomposition of evaporable water contained in the cement stone. In this regard, the radiation decomposition of chemically bound water in materials containing evaporable water can be neglected. Radiation decomposition of chemically bound water can only be noticeable if all the evaporable water is removed from the materials before irradiation. However, this situation is practically impossible under normal conditions without isolating the material from the environment. Even after removing all the evaporable water, the material absorbs water from the environment. And when exposed to high temperatures, chemically bound water becomes evaporable.
- The rate of radiation gas evolution of hydrogen from cement stone and concrete on aggregates that do not contain water can be assumed to be proportional to the amount of evaporable water in the material and the power of the absorbed dose of gamma radiation.

- The amount of evaporable water when exposed to gamma radiation with an increase in irradiation time decreases as a result of drying under the influence of heating, accompanying irradiation and as a result of radiation decomposition of this water under the effect of gamma radiation.

- Since the evaporable water in concrete on aggregates that do not contain water is located mainly in the cement stone of the layers between the aggregates, the radiation evolution of hydrogen can be described by the same model for cement stone and concrete. The differences are only related to the amount of evaporable water per unit mass.

In this regard, for the analytical description of the evolution of hydrogen under the action of gamma radiation from concrete on aggregates that do not contain water, a physical and mathematical model was adopted, described by the formulas:

\[
\begin{align*}
\left( \frac{dM_{H_2}}{dt} \right)_{t=0} &= a_H D_{rg} m_{awo}, \\
\frac{dM_{H_2}}{dt} &= a_H D_{rg} m_{awo}(t, T, D_{rg}),
\end{align*}
\]

where \( M_{H_2} \) is the relative amount of hydrogen evaporable from the material, mol/g;

\( t \) – the duration of irradiation, h;

\( \left( \frac{dM_{H_2}}{dt} \right)_{t=0} \) - the rate of hydrogen evolution at the initial irradiation period, including the hydrogen in the pores of the material, mol/(g-h);

\( \frac{dM_{H_2}}{dt} \) - the rate of hydrogen release at any time of irradiation \( t \), mol/(g·h);

\( a_H \) - the parameter of proportionality, mol/(Gy·g);

\( D_{rg} \) - the absorbed dose rate of gamma radiation, Gy/h;

\( T \) – the temperature of the irradiation, °C;

\( m_{awo} \) - the relative amount of evaporable water in the material at the initial moment of irradiation (before irradiation), g/g;

\( m_{awo}(t, T, D_{rg}) \) - the relative amount of evaporable water remaining in the cement stone or concrete at time \( t \) as a result of water release due to the temperature \( T \) accompanying the irradiation, as well as due to the effect of gamma radiation with the absorbed dose rate \( D_{rg} \), g/g.

Verification of the possibility of using the selected model, carrying out its necessary adjustments and determining the values of the parameters of this model was carried out on the basis of experimental data [26, 27] presented in figures 1-4.

These data were obtained in the works [26, 27] using samples of cement stone in the form of cylinders with a diameter of 50 mm and a height of 100 mm, made of a mixture of ordinary Portland cement without chemical additives and water with a water to cement ratio of 0.3. The samples were irradiated with gamma-rays with an average energy of 1.25 MeV at a facility with a gamma-radiation source of \(^{60}\)Co at an absorbed dose rate of 0.87 kGy/h, 3.84 kGy/h, and 7.05 kGy/h at temperatures of 25 °C, 40 °C, and 60 °C for up to 2 months. Irradiation was carried out in containers containing 3 samples. During irradiation, the selection and measurement of gases and water was carried out. Samples dried at 40 oC, weighing about 390 g, containing about 32 g (0.082 g/g) of evaporable water (samples D40), as well as samples dried at 120 °C, not containing evaporable water, were examined weighing about 358 g (D120 samples).

To test the possibility of using the formula (1) of the model and determining the parameter \( a_H \), the dependence was investigated \( \left( \frac{dM_{H_2}}{dt} \right)_{t=0} \) on \( D_{rg} m_{awo} \).
Figure 1. Change in the rate of hydrogen evolution from containers #1, 3, 5, 7, 9, 11 with three D40 samples upon exposure to gamma radiation according to [26]. In parentheses are the temperature and power of the absorbed dose of gamma radiation.

Figure 2. Change in the rate of hydrogen evolution from containers #2, 4, 6, 8, 10, 12 with three D120 samples upon exposure to gamma radiation according to [26]. In parentheses are the temperature and power of the absorbed dose of gamma radiation.

Figure 3. Release of water from containers #1, 3, 5, 7, 9, 11 with three D40 samples when irradiated with gamma radiation according to [26].

Figure 4. Release of water from containers #2, 4, 6, 8, 10, 12 with three D120 samples when irradiated with gamma radiation according to [26].

To test the possibility of using the formula (2) of the models the dependence of the ratio of the rate of hydrogen evolution at different irradiation times to the rate of hydrogen evolution at $t=0$ on the ratio of the evaporable water content at different irradiation times to the content of the evaporable water before irradiation was investigated:

$$\frac{dM_{H_2}}{dt} \left( \frac{dM_{H_2}}{dt} \right)_{t=0} \frac{m_{aw}(t,T,D_{rg})}{m_{awo}}.$$  (3)

When using the accepted model to test the results of the work [26, 27], the experimental data were taken according to the graphs shown in Figures 1-4, for 7-10 most typical values of the irradiation time. The rate of hydrogen evolution at the beginning of training was obtained by graphical extrapolation of experimental dependencies after reaching a maximum to $t=0$, since in the experiment at the beginning of irradiation, the rate was underestimated due to the accumulation of hydrogen in the pores of the cement stone.
3. The results of the research and their discussion

The results of the study of the dependence of the rate of hydrogen evolution from cement stone (D40) with evaporable water \( m_{awo} = 0.082 \text{ g/g} \) at the beginning of irradiation \( \left( dM_{H_2}/dt \right)_{t=0} \) on the value \( D_{rg}m_{awo} \) at the beginning of irradiation at different irradiation temperatures based on the results of processing experimental data [26, 27] are shown in figure 5. It can be seen that there is a linear relationship between the values \( \left( dM_{H_2}/dt \right)_{t=0} \) and \( D_{rg}m_{awo} \), described by expression (1). However, the angle of inclination of the approximation lines, the tangent of which characterizes the parameter \( a_H(T) \), turns out to be dependent on the irradiation temperature.

According to the graphs of Figure 5 it is established that the parameter \( a_H \) values are:

\[
a_H(T) = 69.8 \times 10^{-12} \text{ mol/(Gy-h)} \quad \text{at} \quad 25 \degree C;
\]

\[
a_H(T) = 92.0 \times 10^{-12} \text{ mol/(Gy-h)} \quad \text{at} \quad 40 \degree C;
\]

\[
a_H(T) = 102.6 \times 10^{-12} \text{ mol/(Gy-h)} \quad \text{at} \quad 60 \degree C.
\]

Parameter values increase with increasing irradiation temperature (figure 6). The temperature dependence of the coefficient can be approximated by the formula:

\[
a_H = a_0 T^{b_0},
\]

where \( a_0 = 17.08 \times 10^{-12} \text{ mol/(Gy-h)} \) \( b_0 = 0.444; \) \( T \) – the temperature of the irradiation, °C.

![Figure 5](image1)

**Figure 5.** Dependence of the rate of hydrogen evolution at the beginning of irradiation \( \left( dM_{H_2}/dt \right)_{t=0} \) from 1 g of cement stone D40 containing evaporable water on the value \( Dm = D_{rg}m_{awo} \) at the beginning of irradiation at different irradiation temperatures \( T \) (°C) based on the results of processing experimental data [26, 27].

![Figure 6](image2)

**Figure 6.** Dependence of the parameter \( a_H(T) \) of formula (5) obtained during processing of figure 5 on the irradiation temperature.

To use the formula (1) to describe the release of hydrogen from cement stone dried at 120 °C (theoretically without evaporable water), you can enter the concept of residual evaporable water. According to [26, 27] data, water and hydrogen are also released from D120 samples dried at 120 °C, although in much smaller amounts than from D40 samples with evaporable water. Although the D120 samples sorbed about \( 1 \times 10^{-4} \text{ g/g} \) of evaporable water before irradiation, the actual water release was up to \( 1.8 \times 10^{-3} \text{ g/g} \). This indicates the presence of residual evaporable water in the D120 samples. In fact, it may be part of the bound water, which is also subject to radiation decomposition.
Experimental data on the radiation evolution of hydrogen from cement stone samples dried at 120 °C based on the expression (1) and the obtained values allowed us to determine the content of this residual evaporable water. According to the results of calculations, it was found that the residual evaporable water in the samples of works [26, 27], dried at 120 °C, is:

- $m_{awo} = 0.0073 \, \text{g/g} - D_{rg} = 7.05 \, \text{kGy/h} \text{ and } T=60 \, ^\circ \text{C}$;
- $m_{awo} = 0.0071 \, \text{g/g} - D_{rg} = 3.84 \, \text{kGy/h} \text{ and } T=60 \, ^\circ \text{C}$;
- $m_{awo} = 0.0078 \, \text{g/g} - D_{rg} = 0.87 \, \text{kGy/h} \text{ and } T=60 \, ^\circ \text{C}$;
- $m_{awo} = 0.0074 \, \text{g/g} - D_{rg} = 3.84 \, \text{kGy/h} \text{ and } T=40 \, ^\circ \text{C}$;
- $m_{awo} = 0.0070 \, \text{g/g} - D_{rg} = 0.87 \, \text{kGy/h} \text{ and } T=40 \, ^\circ \text{C}$;
- $m_{awo} = 0.0061 \, \text{g/g} - D_{rg} = 0.87 \, \text{kGy/h} \text{ and } T=25 \, ^\circ \text{C}$.

It can be seen that $m_{awo}$ exceeds the amount of water released, practically does not depend on the dose rate of gamma radiation, and increases slightly with the increase in the irradiation temperature.

The dependences of the change in the ratio of the rate of hydrogen evolution from a cement stone containing evaporable water at time $t$ in relation to the rate in the initial irradiation period $K_{MHt=0}^{awo}$ from the remaining part of the evaporable water relative to its initial amount $K_{w/t=0}^{awo} = m_{awo}(t,T,D_{rg})/m_{awo}$ according to the results of processing experimental data [26, 27] are shown in figure 7(a)

![Diagram](a)

**Figure 7.** Dependences of the change in the irradiation process of the ratio of the rate of hydrogen evolution from a cement stone at time $t$ in relation to the rate in the initial irradiation period $K_{MHt=0}^{awo}$ from the remaining part of the evaporable water relative to its initial amount $K_{w/t=0}^{awo}$ according to the results of experimental data processing [26, 27].

(a) - from D40 cement stone containing evaporable water;
(b) - from D120 cement stone, dried at 120 °C, containing residual evaporable water.
Figure 7 (a) shows that with a decrease in the amount of the remaining part of the evaporable water during irradiation, the rate of radiation evolution of hydrogen relative to the rate at the beginning of irradiation decreases. However, the rate of hydrogen evolution decreases more significantly than the amount of remaining evaporable water. Moreover, the higher is the power of the absorbed dose of gamma radiation, the more significant is the difference between them. It follows that formula (2) does not correctly describe the process of radiation gas evolution from cement stone. Model adjustment is required.

A possible reason for the deviation is an additional decrease in the amount of water due to its radiation decomposition, since the higher is the power of the absorbed dose of gamma radiation, the greater is the rate of decomposition of water. However, analysis of the results showed that this is not the case. So even at a dose rate of 7.05 kGy/h and a temperature of 60°C after irradiation for 62 days, 0.056 g/g of evaporable water is released, and as a result of radiolysis, only 3.5 \times 10^{-4} g/g is decomposed, that is, 160 times less. In this regard, the effect can be neglected. It is possible that as the absorbed dose of gamma radiation increases, some of the evaporable water interacts with the cement stone and becomes bound.

Based on the processing of dependencies shown in Figure 7(a), it is established that the dependence of the value $\left(\frac{dM_{H_2}}{dt}\right)/\left(\frac{dM_{H_2}}{dt}\right)_{t=0}$ from the value $m_{aw}(t,T,D_{rg})/m_{aw0}$ can be described by the formula:

$$\left(\frac{dM_{H_2}}{dt}\right)/\left(\frac{dM_{H_2}}{dt}\right)_{t=0} = \left(\frac{m_{aw}(t,T,D_{rg})}{m_{aw0}}\right)^{b_w},$$

where $b_w$ - parameter equal to:

- $b_w=1.29$ – at the absorbed dose rate of gamma radiation $D_{rg}=0.87$ kGy/h and $T=25$ °C;
- $b_w=1.26$ – at the absorbed dose rate of gamma radiation $D_{rg}=0.87$ kGy/h and $T=40$ °C;
- $b_w=1.31$ – at the absorbed dose rate of gamma radiation $D_{rg}=0.87$ kGy/h and $T=60$ °C;
- $b_w=1.835$ – at the absorbed dose rate of gamma radiation $D_{rg}=3.84$ kGy/h and $T=40$ °C;
- $b_w=1.78$ – at the absorbed dose rate of gamma radiation $D_{rg}=3.84$ kGy/h and $T=60$ °C;
- $b_w=2.18$ – at the absorbed dose rate of gamma radiation $D_{rg}=7.05$ kGy/h and $T=60$ °C.

It can be seen that the values $b_w$ depend mainly on the power of the absorbed dose. This is clearly shown in Figure 8.

![Figure 8](image-url)

**Figure 8.** Dependence of the $b_w$ parameter of formula (5) on the absorbed dose rate of gamma radiation $D_{rg}$. The temperature of the radiation $T$ in the °C.

It is established that the dependence of the parameter on the absorbed dose rate of gamma radiation $D_{rg}$ (in Gy/h) can be approximated by the formula:
The dependences of the change in the ratio of the rate of hydrogen release from the cement stone dried at 120 °C at time \( t \) in relation to the rate in the initial period of irradiation \( \left(\frac{dM_{H_2}}{dt}\right)_{t=0} \) from the remaining part of the residual free water relative to its initial amount according \( m_{aw}(t,T,D_{rg})/m_{awo} \) to the results of processing experimental data of works [26, 27] are shown in figure 7 (b). It can be seen that the dependence of figure 7 (b) is similar to the dependence of figure 7 (a). However, due to the small values of the hydrogen evolution rate and therefore the greater influence of measurement errors, there are more significant variations and the unambiguous effect of the gamma radiation dose rate is not noticeable.

It turned out to be possible to calculate the values based on the degree of change of the hydrogen evolution rate only for the results obtained at the absorbed dose rate of gamma radiation 3.84 kGy/h and 7.05 kGy/h at temperatures of 40 °C and 60 °C.

The calculated values \( b_w \) for samples of cement stone dried at 120 °C are comparable to the values for cement stone with evaporable water and are:

- \( b_w = 3.12 \) – at the absorbed dose rate of gamma radiation \( D_{rg} = 3.84 \) kGy/h and \( T = 40 \) °C;
- \( b_w = 1.13 \) – at the absorbed dose rate of gamma radiation \( D_{rg} = 3.84 \) kGy/h and \( T = 60 \) °C;
- \( b_w = 2.20 \) – at the absorbed dose rate of gamma radiation \( D_{rg} = 7.05 \) kGy/h and \( T = 60 \) °C;
- \( b_w = 1.62 \) – for an averaged dependency across all values.

Approximately for cement stone, dried at 120 °C, can be assumed to be equal to \( b_w = 1.62 \) or to be equal to the values obtained with less error for cement stone with evaporable water.

Thus, for an analytical description of the process of radiation evolution of hydrogen, instead of the formula (2), you need to use the formula:

\[
dM_{H_2}/dt = \frac{a_H D_{rg} m_{awo}(m_{aw}(t,T,D_{rg})/m_{awo})^{\beta_w}}{b_w}
\]

4. Conclusion

1. Based on the experimental data available in the scientific and technical literature, the model was selected, verified, and adjusted for an analytical description of the process of hydrogen evolution from cement stone and concrete on aggregates that do not contain water under the influence of gamma radiation.

2. When choosing a model for the analytical description of the process of radiation evolution of hydrogen under the influence of gamma radiation from cement stone and concrete on aggregates that do not contain water, it was assumed that hydrogen evolution is mainly associated with the radiation decomposition of the evaporable water of the cement stone and is proportional to the amount of evaporable water in material and absorbed dose rate of gamma radiation. It was taken into account that the amount of evaporable water during irradiation decreases with an increase in the irradiation time as a result of drying under the action of heating, accompanying irradiation and as a result of radiation decomposition of this water under the influence of gamma radiation. Since evaporable water in concrete on aggregates that do not contain water is located mainly in the cement stone of the layers between the aggregates, it was believed that the radiation evolution of hydrogen can be described by the same model for cement stone and concrete.
3. The possibility of using the selected model, its necessary adjustment and determination of the main parameters of this model was performed on the basis of available experimental data on the evolution of hydrogen from Portland cement stone under the influence of gamma radiation. Experimental data were used on the evolution of hydrogen and water from cement stone containing and not containing evaporable water when irradiated with gamma rays with an average energy of 1.25 MeV at a plant with a gamma radiation source of $^{60}$Co at an absorbed dose rate of 0.87 kGy/h, 3.84 kGy/h and 7.05 kGy/h at temperatures of 25, 40 and 60 °C for up to 2 months.

4. The model was adjusted to take into account the change in the relationship between the amount of evaporable water and the rate of hydrogen evolution as the amount of this water decreases during irradiation due to the effects of heating that accompanies irradiation. For cement stone dried at 120 °C to remove evaporable water, the amount of residual evaporable water (residual evaporable water or decomposed chemically bound water) was studied and determined.

5. This model can be used for analytical description of hydrogen release from concrete on aggregates that do not contain water, under the action of gamma radiation with an average energy of 1-1.5 MeV at temperatures of 25 - 60 °C. This requires data on the irradiation temperature, the power of the absorbed dose of gamma radiation, the amount of evaporable water, and the change in its content during heating at the irradiation temperature. This model can be used as a basis for developing a method for analytical determination of the radiation release of hydrogen from concrete in a wide range of irradiation conditions.

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