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Catalogue of Radiation-Induced Damage of Rock Aggregates Identified by RBSM Analysis

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Abstract

This paper presents a catalogue of possible radiation-induced damage of rock aggregates, which was compiled using the relevant literature and confirmed numerically. The catalogue describes two common and six specific cases of rock aggregate damage. Additionally, the catalogue is supported by a validated numerical model, which is based on Rigid-Body Spring Model. The detailed numerical analysis and the result description of all the cases shown in the catalogue are presented in this paper. The main dependencies are also discussed and the related conclusions are drawn, of which the most important are that the damage of the rock can be delayed and increased rapidly after the delay; even a small amount (1%) of a highly expansive mineral leads to a significant reduction of mechanical properties of the rock; and a partial recovery of the elastic modulus of the rocks is possible even for significantly damaged rocks. It is believed that this paper will help to predict the radiation-induced degradation of rock aggregates as well as support the future development of related analytical models.

1. Introduction

Concrete structures in nuclear power plants (NPPs), such as concrete biological shields (CBSs), may be subjected to ionizing radiation. Based on the relevant research (Hilsdorf et al. 1978; Field et al. 2015; Le Pape et al. 2016), the radiation-induced volumetric expansion (RIVE) of rock aggregates in concrete composition is considered to be the principal cause of concrete deterioration under ionizing conditions. Then, the RIVE of rock aggregates may lead to significant concrete damage which may result in cracks in CBS which consequently compromise its shielding and load-bearing properties (Le Pape 2015; Khmurovska et al. 2019; Bruck et al. 2019; Kambayashi et al. 2020). Due to this fact, the RIVE and the related deterioration of rock aggregate mechanical properties need to be predicted. The problem of the prediction of RIVE of aggregates is hindered by the fact that volume of the related experimental investigations is very limited (Elleuch et al. 1972; Hilsdorf et al. 1978; Denisov et al. 2012; Maruyama et al. 2017) due to the extremely high cost of the irradiation itself, the complexity of post-irradiation examination and the subsequent storage of the irradiated aggregates, which effectively become radioactive waste.

The RIVE of rock aggregates is caused by RIVE of the constitutive minerals, which depends on the neutron fluence and its energy spectrum and the irradiation temperature (Dubrovskii et al. 2010; Denisov et al. 2012; Rosseel et al. 2016; Khmurovska and Štemberk 2019). Therefore, in order to predict the RIVE of aggregates, the RIVE of the constitutive minerals needs to be known. There are few numerical models of RIVE of minerals which can be used for this purpose (Le Pape et al. 2018; Denisov et al. 2012).

In this study, the numerical model of RIVE of minerals published by Denisov et al. (2012) was analyzed in detail, which revealed common trends in the development of the rock RIVE in accordance with the nature of the constitutive minerals and their volume fraction in the rock. The basic knowledge of the fracture mechanics and geology in combination with a substantial amount of imagination helped to derive the corresponding implications in terms of the crack volume and the elastic modulus degradation. These findings were summarized in a catalogue of the possible radiation-induced damage of rock aggregates which was confirmed with a numerical model based on Rigid-Body Spring Model (RBSM) (Kawai 1978). The RBSM numerical model also helped to visualize the important stages of the progressing damage during irradiation.

2. Radiation-induced damage of rock aggregates

2.1 Catalogue

As it was noted in the previous section, the RIVE of rock aggregates depends on the RIVE of the constitutive minerals which are present in their composition. However, the sum of the RIVE of minerals is significantly lower than the RIVE of the rock (Le Pape et al. 2020;
Denisov et al. (2012). The reason of this phenomenon can be found in the additional expansion due to the intragrain and intracrystalline defects, including cracks, which are caused by their damage. In this study, the crystal RIVE anisotropy was considered as the driving mechanism of the intragrain and intracrystalline irradiation-induced defects, since, based on the analysis of the data published in Denisov et al. (2012), it was confirmed that the additional cracks do not occur in the minerals with isotropic expansion (Khmurovska and Štemberk 2021). Another reason of the rock RIVE increase is in the formation of additional grain boundary cracks due to the uneven expansion of the minerals (Denisov et al. 2012) and the distribution of the grains. Therefore, generally, the total RIVE of a rock is believed to consist of three components, namely, the RIVE of minerals, the additional expansion due to the intragrain and intracrystalline defects, and the expansion due to the grain boundary crack occurrence, which can be expressed as follows.

\[
\left( \frac{\Delta V}{V} \right)_{rock} = \sum_{i} \left( \frac{\Delta V}{V} \right)_{i} + \sum_{i} \left( \frac{\Delta V}{V} \right)_{\text{defect},i} + \left( \frac{\Delta V}{V} \right)_{\text{crack}},
\]

where \( n \) is the total number of minerals in the rock composition, \( \left( \frac{\Delta V}{V} \right)_{i} \) is the RIVE of the \( i \)-th mineral in the rock composition, \( V_{i} \) is the volume fraction of the \( i \)-th mineral in the rock composition, \( \left( \frac{\Delta V}{V} \right)_{\text{defect},i} \) is the volume of intragrain and intracrystalline defects in the \( i \)-th mineral in the rock composition and \( \left( \frac{\Delta V}{V} \right)_{\text{crack}} \) is the volume of grain boundary cracks.

In Eq. (1), it is assumed that the intragrain and intracrystalline defects, including cracks, do not cause damage by themselves due to the possible time-dependent microcrack recovery originated from the defect healing (Geng et al. 2017; Renard et al. 2000; Meredith 2013), but that they create an additional expansion, which may result in the damage. Moreover, phenomenologically, on the microscale level, the RIVE is similar to the thermal expansion (Zuo et al. 2010; Isaka et al. 2019; Peng et al. 2019). Thus, similarly as in the case of thermal damage, the cracks initiate within the weakest links, which are the grain boundaries (Isaka et al. 2019).

It was considered that the grain boundaries remain the weakest links despite an increase of the intragrain and intracrystalline defects (damage of grains and crystals). This assumption leads to the conclusion that the damage caused by the intragrain and intracrystalline defects may be neglected, while the additional expansion due to these defects is taken into account.

Additionally, it was assumed that the intragrain and intracrystalline defects are insignificant in homogeneous materials, when the volume fraction of the rock-forming minerals is higher or equal to 95%, and thus they can be neglected.

Based on these assumptions, the numerical model of RIVE of minerals published by by Denisov et al. (2012), which is described in the Appendix, was analyzed and a catalogue of possible damage of aggregates was summarized in Table 1.

In the catalogue, the fluence means the neutron fluence with the energy above 10 keV, the RIVE of minerals is \( \left( \frac{\Delta V}{V} \right)_{i} \) and the total RIVE of minerals is \( \sum_{i} \left( \frac{\Delta V}{V} \right)_{i} V_{i} \), the crack volume is \( \sum_{i} \left( \frac{\Delta V}{V} \right)_{\text{defect},i} V_{i} + \left( \frac{\Delta V}{V} \right)_{\text{crack}} \), the RIVE of the rock is \( \left( \frac{\Delta V}{V} \right)_{\text{total}} \) and the relative \( E \) is the relative elastic modulus of the rock (the elastic modulus after irradiation divided by the initial elastic modulus), which represents an isotropic damage.

The irradiation-induced damage cases were derived as follows: firstly, in the common case, it was assumed that the crack size increased linearly with the RIVE increase. This is confirmed by the data on crack volume published in the reference source (Denisov et al. 2012). Then, it is known from the fracture mechanics that the relation between the crack size and the damage-related residual stresses may be approximated by the power law (Anderson 2017), which leads to the first common case. The other cases were derived from the first one with the assumption of crack closing effect and with the assumption that the uniform RIVE does not cause any damage. As can be seen in Table 1, two common cases and six specific cases were defined. It is believed that the common cases are more frequent, while the specific cases are rather rare, since some specific conditions need to be met in order to achieve such a behavior.

In Case 1, there is always one mineral with dominant RIVE at every neutron fluence level. Such RIVE of minerals leads to linear increase in the crack volume with respect to the total RIVE of minerals and subsequently results in the elastic modulus degradation, which can be approximated as a power function. The difference in the RIVE of the minerals and their volume fraction defines the slope of the crack volume function and the rate of the elastic modulus degradation. This case is common for multicomponent rocks, for example, some types of granite with an average irradiation temperature of 40°C. It should be noted that the damage case of the rock may change with alteration of the irradiation temperature, due to the temperature-induced variation of the behavior of constitutive minerals. Thus, the irradiation temperature is always indicated together with the example of the rock type.
| No. | Fluence | Total RIVE of minerals vs. RIVE of minerals | Crack volume vs. Total RIVE of minerals | RIVE of rock vs. Relative $E$ |
|-----|---------|-------------------------------------------|----------------------------------------|-------------------------------|
| 1   | ![Graph 1](image1.png) | ![Graph 2](image2.png) | ![Graph 3](image3.png) | ![Graph 4](image4.png) |
| 2   | ![Graph 1](image5.png) | ![Graph 2](image6.png) | ![Graph 3](image7.png) | ![Graph 4](image8.png) |
| 3   | ![Graph 1](image9.png) | ![Graph 2](image10.png) | ![Graph 3](image11.png) | ![Graph 4](image12.png) |
| 4   | ![Graph 1](image13.png) | ![Graph 2](image14.png) | ![Graph 3](image15.png) | ![Graph 4](image16.png) |
| 5   | ![Graph 1](image17.png) | ![Graph 2](image18.png) | ![Graph 3](image19.png) | ![Graph 4](image20.png) |
| 6   | ![Graph 1](image21.png) | ![Graph 2](image22.png) | ![Graph 3](image23.png) | ![Graph 4](image24.png) |
| 7   | ![Graph 1](image25.png) | ![Graph 2](image26.png) | ![Graph 3](image27.png) | ![Graph 4](image28.png) |
| 8   | ![Graph 1](image29.png) | ![Graph 2](image30.png) | ![Graph 3](image31.png) | ![Graph 4](image32.png) |

Table 1: Catalogue of irradiation-induced damage cases of rock aggregates.
In Case 2, there is no single mineral with dominant RIVE over all fluence levels. At the beginning of irradiation, one mineral has dominant RIVE and, with the increasing neutron fluence, another mineral becomes dominant in terms of RIVE. Such a behavior leads to nonlinear increase in crack volume and subsequently results in very complex shape of the elastic modulus degradation function. Generally, at the beginning of irradiation, the relative elastic modulus decreases according to the power law. However, when RIVE of the mineral with lower RIVE approaches the RIVE of the mineral with the dominant RIVE, the rate of the elastic modulus degradation decreases. After the exchange of the minerals according to their RIVE dominance, the rate of the elastic modulus degradation again increases. The exchange of the minerals according to their RIVE dominance may repeat, which leads to even more complex shapes of the elastic modulus degradation function. This case is also common for multicomponent rocks, for example, some types of gabbro with an average irradiation temperature of 80°C.

Generally, Case 3 is similar to Case 2. However, in Case 3, there is a mineral with dominant negative RIVE at the beginning of irradiation, which can be, for example, represented by glass. In this case, the total RIVE of the minerals is negative at the very beginning. The contraction of one mineral in the rock composition and expansion of the other minerals leads to the rapid increase in crack volume and results in very fast degradation of the elastic modulus. When expansion becomes dominant over the contraction, the behavior of the rock becomes similar to Case 2. This case occurs among multicomponent rocks, for example, some types of basalt with an average irradiation temperature of 140°C.

Case 4 is common for homogeneous materials or for materials which consist of minerals with similar or identical RIVE. In this case, the expansion is homogeneous, therefore, cracks or degradation of the elastic modulus do not occur. It should be noted that if the rock consists of several constitutive minerals with identical RIVE, the intragrain and intracrystalline defects may occur depending on the volume fraction of the minerals and their anisotropic behavior, therefore, the crack volume would not be equal to zero. However, based on the assumption that such defects do not cause damage, the degradation of the elastic modulus would not occur. This case is rather common for one- or two-component rocks, for example, obsidian with an arbitrary irradiation temperature (RIVE does not depend on the irradiation temperature here).

Case 5 is typical for those rocks which consist of the constitutive minerals with similar or identical of RIVE at the beginning of irradiation. However, the RIVE of the minerals differs at higher fluence levels. Such a rock composition leads to zero crack volume and results in zero damage at the beginning of the irradiation. However, when the RIVE of each of the constitutive minerals becomes different, the cracks start to propagate, which leads to the reduction of the elastic modulus. Generally, this case can be considered as the combination of Case 1 and Case 4. Similarly as in Case 4, the occurrence of the intragrain and intracrystalline defects is also possible. This case is rather common for two-component rocks, for example, some types of limestone with an arbitrary irradiation temperature (RIVE does not depend on the irradiation temperature here).

Case 6 may occur in two-component rocks with a small volume fraction (about 1%) of a highly expansive constitutive mineral and a large volume fraction (about 99%) of constitutive mineral with low RIVE, for example, hornblende, which contains impurity, with an average irradiation temperature of 90°C. Another necessary condition for this case is that the low-RIVE mineral is dominant in terms of RIVE at the beginning of irradiation. Such a composition leads to the occurrence of small crack volume and slight reduction of the elastic modulus at the beginning of irradiation. When the RIVE of the constitutive minerals becomes equal, the cracks close, which may lead to the recovery of the elastic modulus. However, when the highly expansive mineral becomes dominant, new cracks occur, which leads to the significant reduction of the elastic modulus.

Case 7 is similar to Case 6 with the only difference that the RIVE of the constitutive minerals differs significantly at the beginning of irradiation, but the ultimate RIVE of the rock constituting minerals is rather similar. Such a composition leads to the gradual crack volume increase at the beginning of irradiation and to the crack closing when the RIVEs of the minerals become similar. Since the ultimate RIVEs of the constitutive minerals do not differ much, no additional cracks occur when the minerals exchange according to their RIVE dominance. Similarly, at the beginning of irradiation, the elastic modulus reduces and recovers when the RIVEs of the minerals become similar. This case is common for olivinite, which contains impurity, with an average irradiation temperature of 80°C.

Case 8 is a combination of Cases 6 and 7. This case is also valid for two-component rocks, for example, serpentinitized dunite with an average irradiation temperature of 230°C. However, no limitation regarding the volume fraction is considered in this case. Therefore, the crack volume increases gradually at the beginning of irradiation, then the cracks are partially closed when the RIVEs of constitutive minerals become similar, which is followed by occurrence of new cracks. The relative elastic modulus decreases at the beginning of irradiation, then partially recovers and then decreases again. It should be noted that the rock may be totally damaged even before the recovery of the elastic modulus. This depends on the difference of the RIVE of each constitutive mineral and their volume fraction.

In order to confirm the individual cases of the proposed catalogue of radiation-induced damage cases of aggregates, an already developed RBSM-based numerical model was utilized.
2.2 Numerical interpretation and validation
The grain-based models (GBMs) are widely used in order to model behavior of crystalline rocks based on their composition (Wang et al. 2021; Zhou et al. 2019; Wong et al. 2018; Hofmann et al. 2015). The GBMs show precise results and good correlation with the experimental data. However, the high number of material constants and tuning of the internal model constants are needed in order to simulate each individual rock (Wang et al. 2021; Hofmann et al. 2015). This complicates significantly the use of GBMs especially when the amount of experimental data is limited. Therefore, the proposed numerical modeling utilizes an improved Voronoi-based 2D RBSM (Bolander and Saito 1998; Gu et al. 2013; Rasmussen et al. 2018; Wang et al. 2008), where a square domain was discretized into a set of Voronoi cells. In the model, the Voronoi cells represent rigid bodies which are interconnected by sets of zero-length springs. Each set of springs consists of one normal spring with the stiffness of $k_n$ and one shear spring with the stiffness of $k_s$. Three sets of springs were placed along each Voronoi cell boundary, so that the acting length for each spring set is equal to 1/3 of the boundary length, $l$, as shown in Fig. 1. Therefore, each rigid body has two translational and one rotational degrees of freedom (Meng et al. 2018; Yao et al. 2015).

The numerical model was tuned for the numerical samples with the dimensions of $1 \times 1 \times 1$ cm and with 1000 elements (Voronoi cells). The detailed information regarding the utilized modified 2D RBSM model was published in (Khmurovska and Štemberk 2021). Thus, only a brief explanation is given below.

Two fracture criteria were adopted (Yao et al. 2015) in order to describe the normal and the shear spring failures. The stress-strain relation for the normal springs is linear elastic with the slope which corresponds to the elastic modulus of the spring, $E_{int}$, up to the tensile-strength dependent value, $\sigma_t$, where the brittle failure occurs [Fig. 2a]). The behavior of the normal spring in compression is considered as perfectly elastic without failure or softening (Yao et al. 2015). The Mohr-Coulomb type criterion is assumed as the shear spring failure criterion, which is defined by the angle of internal friction, $\phi$, cohesion coefficient, $C$, normal stresses, $\sigma_n$, and critical normal stresses, $\sigma_{cr}$. If the shear stresses, $|\sigma_s|$, exceed the maximum shear stresses, which are defined by the Mohr-Coulomb type criterion, or the normal tensile stresses exceed $\sigma_t$, the cohesion component is eliminated and the criterion reduces to the residual form [Fig. 2b), from Yao et al. (2015)]. In the model, the angle of internal friction, $\phi$, remains constant even after the damage. This feature was adopted from (Yao et al. 2015) for the sake of simplicity in order to reduce the amount of calibrated parameters. However, the model allows for the use of different friction angle for pristine and damage states. All the necessary relations for the numerical simulation are published in (Khmurovska and Štemberk 2021).

As was shown above, the total RIVE of a rock defined by the Eq. (1) consists of three components. The first component of the total RIVE of a rock is the RIVE of minerals, which was obtained using the model of RIVE of minerals (Denisov et al. 2012), which is briefly de-
scribed in the Appendix.

The second component of the total RIVE of a rock is the expansion due to the intragrain and intracrystalline defects, which were calculated as follows.

$$\gamma \frac{\Delta V}{V} \text{defect} = \gamma \frac{\Delta V}{V}, \quad (2)$$

where $\gamma$ is the model parameter which depends on the anisotropy coefficient, $K_a$, as follows.

$$V_{\text{max}} < 95\% \quad \begin{cases} 
\gamma = 0 & \text{if } K_a = 1, \\
\gamma = 0.5 & \text{if } 1 < K_a \leq 2, \\
\gamma = 1 & \text{if } K_a > 2,
\end{cases}$$

$$V_{\text{max}} < 95\% \quad \text{for any mineral} \quad \gamma = 0 \quad \text{for any } K_a, \quad (3)$$

where $K_a$ is the anisotropy coefficient, which can be calculated according to Denisov et al. (2012) and which was introduced in (Khmurovska and Štemberk 2021), and $V_{\text{max}}$ is the volume fraction of the rock-forming mineral.

The third component of the total RIVE of a rock is the additional expansion due to the formation of additional grain boundary cracks, which is the direct output of the RBSM analysis, while the first and the second components of the total RIVE are inputs for the RBSM analysis.

The above described model was implemented in an in-house 2D RBSM code which was developed in MATLAB. The modified Newton-Raphson method was used to solve the system of nonlinear equations.

In the numerical simulation, each element (Voronoi cell) of the 2D numerical samples of rocks was loaded with the linear strain in the in-plane directions. The applied linear strain of the $i$-th mineral, $\varepsilon_{\text{lin},i}$, was calculated as follows.

$$\varepsilon_{\text{lin},i} = \left( \frac{\Delta V}{V} \right)_i + \left( \frac{\Delta V}{V} \right)_{\text{defect},i} \frac{1}{3}, \quad (5)$$

The boundary conditions for the numerical simulation were chosen to allow for free expansion of the numerical rock sample.

In the developed RBSM code, the minerals are assigned randomly to the elements (Voronoi cells) with respect to their volume fraction in the rock composition. The permitted deviation in volume fraction assignment is ±1%.

The total RIVE of the rock was obtained with the assumption that the linear strain in the out-of-plane direction of the 2D model is equal to the average strain in the in-plane directions. Therefore, in order to obtain the total RIVE of the rock, the average linear strain in the in-plane directions was calculated and multiplied by 3. The self-weight of the rocks was neglected in the numerical simulations.

The validation of the numerical model was performed on the data published in Denisov et al. (2012), where 130 experimental data points of RIVE and 112 experimental data points for elastic modulus degradation for 35 different rocks at different temperatures are presented.

The validation in terms of RIVE was published in (Khmurovska and Štemberk 2021), where it was shown that the numerical model is capable of estimating the RIVE of rocks accurately (the coefficient of determination $R^2=0.960$ and the residual standard error $RSE=0.961\%$). However, it should be noted that the model fails to predict the RIVE of the rocks with pronounced volume change due to the gas release, such as serpentinized dunites and sedimentary carbonates with the irradiation temperature close to the dissociation temperature of minerals, which is due to the fact that the numerical model does not take into account the radiation-induced gas release and the related volume change.

Regarding the validation of the model in terms of the elastic modulus degradation, it should be noted that the standard procedure of validation with calculation of the coefficient of determination and residuals is not applicable here, since the results in terms of the elastic modulus degradation published by Denisov et al. (2012) are split in groups and there is no chance to distinguish the results for the individual rocks. Therefore, the validation is performed by visual comparison of the group of experimental results with the group of numerical results, similarly as in Sasano et al. (2020). The groups presented in Denisov et al. (2012) and the corresponding rock numbers are listed below.

**Igneous rocks**

1. Granites, granodiorites, diorites, quartz andesite (1, 2, 3, 4, 5, 6, 9, 10)
2. Liparite (7)
3. Obsidian (8)
4. Albitite, labradorite, urtite, gabbro, gabbro-porphyr (11, 12, 13, 14, 15, 16)
5. Diabases, basalts (17, 18, 19, 20)
6. Pyroxenite, peridotite, olivine, dunite (21, 22 23, 24)
7. Serpentinized dunites (25, 26)
8. Hornblende (27)

**Sedimentary silicate rocks**

1. Aleurolite (28)
2. Sandstone (29)

**Sedimentary carbonate rocks**

1. Limestones, dolomite rock (30, 31, 32, 35)
2. Magnesite rock (33)
3. Siderite rock (34)

It should be noted that 1% of quartz was assumed in the rock 27 (medium-grain hornblende) and 1% of oligoclase was considered in the rocks 30, 31 and 33 (fine-grain limestone, small-grain limestone and fine-grain porous magnesite), since the incomplete composition (99% or about 100%) is indicated in the reference source (Denisov et al. 2012) for these rocks.
The rock composition is shown in the Appendix under the corresponding numbers.

The validation of the model in terms of degradation of the elastic modulus of the rocks is shown in Figs 3 to 5, where the experimental results from Denisov et al. (2012) are indicated with markers and the calculated results for different rocks are represented by curves. The number, which is shown in the legend, corresponds to the rock identifying number.

Figure 3 shows the radiation-induced elastic modulus degradation vs. the total RIVE of igneous rocks in accordance to the groups specified by Denisov et al. (2012). As can be seen, generally, the model shows good correlation with the experimental results with a slight overes-

Fig. 3 Radiation-induced elastic modulus degradation of igneous rocks: a) granites, granodiorites, diorites, quartz andesite; b) liparite; c) obsidian; d) albite, labradorite, urtite, gabbro, gabbro-porphyry; e) diabases, basalts; f) pyroxenite, peridotite, olivine, dunite; g) serpentinized dunites; h) hornblendite.
timation of the elastic modulus degradation, which means that the prediction is on the safe side. Only the radiation-induced degradation of rocks 25 and 26 (fine-grain serpentinitized dunites) does not correlate well [Fig. 3g]. The reason for this is that the model fails to predict the RIVE of the rocks with pronounced volume change due to the gas release.

Figure 4 shows the radiation-induced elastic modulus degradation vs. total RIVE of sedimentary silicate rocks. As can be seen, the model overestimates the degradation of the elastic modulus of sedimentary silicate rocks 28 and 29 (fine-grain laminated aleurolite and small-grain sandstone). This may be related to the fact that the creep of sedimentary silicate rocks has high temperature sensitivity in comparison to that of the igneous rocks (Brantut et al. 2013). For example, the creep strain rate in sandstone increases by approximately three orders of magnitude when the temperature is increased from 25°C to 75°C (Brantut et al. 2013). The creep of the rocks was not taken into account in the proposed numerical model. However, this neglected effect may be significant for sedimentary silicate rocks due to the fact that the experimental data were obtained at the average irradiation temperature from 85°C to 215°C (Denisov et al. 2012) and since it is known that the creep can reduce the rate of radiation-induced damage (Giorla et al. 2017; Khmurovska et al. 2019; Kambayashi et al. 2020).

Figure 5 shows the radiation-induced elastic modulus degradation vs. the total RIVE of sedimentary carbonate rocks in accordance to the groups specified by Denisov et al. (2012). As can be seen, the model shows good correlation with the experimental data. Only the uncertain
results related to the rock 35 (medium-grain dolomite), seen in Fig. 5a), do not show any significant damage according to the numerical simulation. However, due to the fact that there is no chance to distinguish the results for the individual rocks in the group according to Denisov et al. (2012), it is difficult to estimate the level of inaccuracy.

From all the above, it is possible to conclude that the numerical model shows good correlation with the experimental data for all the rocks except the rocks with significant radiation-induced gas release (fine-grain serpentinized dunites) and the rocks with significant creep (fine-grain laminated aleurolite and small-grain sandstone).

3. Results and discussion

The numerical analysis of all the 35 rocks, which were examined in Denisov et al. (2012), was performed at different temperatures in order to confirm the possible irradiation-induced damage cases which are catalogized in Table 1. The results in terms of the RIVE of minerals, the RIVE of rocks, the crack volume (the grain boundary cracks and the intragrain and intracrystalline defects) and the degradation of the elastic modulus were then analyzed. The examples of all of the described cases are discussed in detail below.

3.1 Case 1

As an example of Case 1, the rock 1 (large-grain granite) calculated at the temperature of 40°C was considered. The detailed results of these calculations are shown in Fig. 6, with the input expansion of minerals (neutron fluence vs. RIVE of minerals) is given in Fig. 6a). In this graph, the dotted curves denote the expansion of the minerals without defects, \( \Delta V \), and the solid curves denote the expansion of the minerals with defects, \( \Delta V + \Delta V_{\text{defects}} \). Also, the average irradiation temperature and the volume fraction of the individual minerals are given in the graph as inputs [Fig. 6a)]. Moreover, the color of the curves in this graph represents different minerals and also corresponds to the color of Voronoi cells within the changing crack pattern [Fig. 6b)]. The graphs of total RIVE of minerals vs. crack volume and total RIVE of rock vs. relative elastic modulus, which are shown in Figs. 6c) and 6e), respectively, are similar to those in the catalogue (see Table 1). Additionally, in Fig. 6d), the evolution of the RIVE in terms of neutron fluence is shown. Also, it should be noted that only the grain boundary cracks, excluding the intragrain and intracrystalline defects, are shown in the crack pattern.

It can be seen from the Fig. 6 that the increase of crack volume is linear with respect to the total RIVE of the constitutive minerals and the elastic modulus degradation can be approximated by the power law with respect the total RIVE of the rock.

![Fig. 6 Example of Case 1 (rock 1 at 40°C) of radiation-induced damage of rocks: a) graph of neutron fluence vs. RIVE of minerals; b) change in crack pattern; c) graph of total RIVE of minerals vs. crack volume; d) graph of neutron fluence vs. total RIVE of rock; e) graph of total RIVE of rock vs. relative elastic modulus.](image-url)
The crack patterns in Fig. 6 are shown at two stages, which are the initial stage (green color) and the ultimate stage of the maximum RIVE (orange color). The cracks are growing gradually due to the dominant expansion of quartz (dark red color), while the greatest cracks are observed in the microcline (light blue color), which has the lowest RIVE, and while no unusual change in the crack pattern could be observed.

3.2 Case 2
As an example of Case 2, the rock 15 (small-grain gabbro) calculated at the temperature of 80°C was considered. The detailed results of these calculations are shown in Fig. 7 in the similar way as it is explained in Section 3.1. It should be noted that Case 2 is the most frequent case of the radiation-induced damage of the analyzed rocks.

As can be seen, the shape of the crack volume evolution curve and the shape of the elastic modulus degradation curve are very complex (Fig. 7), and thus they cannot be approximated by simple functions.

At the beginning of irradiation, hornblende (dark blue color) has the highest RIVE rate and therefore, the cracks occur predominantly in oligoclase (light blue color), which has a much smaller RIVE rate. However, at the moment when the RIVE of quartz (dark red color) approaches the RIVE of hornblendite, the rock RIVE rate decreases. With the decreasing of the rock RIVE, the crack occurrence is also slowing down and even some cracks may be closed by the quartz expansion. This also leads to the reduction of the elastic modulus degradation rate. The stage, when the RIVE of quartz is equal to the RIVE of hornblende, is shown in Fig. 7 in green color. After this point, the RIVE of quartz becomes dominant and the highest number of new cracks occur in hornblende, while the existing cracks in oligoclase grow. This leads to the second wave of the rock RIVE, which results in the rapid elastic modulus degradation. The rock at the highest RIVE stage is depicted in Fig. 7 in orange color, where the significant change in crack pattern can be seen.

3.3 Case 3
As an example of Case 3, the rock 20 (fine-grain basalt) calculated at the temperature of 140°C was considered. The detailed results of these calculations are shown in Fig. 8 in the similar way as it is explained in Section 3.1. Ore and glass are considered only as free of defects, because these constitutive minerals have isotropic expansion (Denisov et al. 2012) and, thus, it was considered that the intragrain and intracrystalline defects would not occur inside them.

As can be seen in Fig. 8, at the beginning of irradiation, the dominant RIVE is negative due to the contraction of glass (light red color). This leads to the significant crack formation (predominantly around glass) and a very rapid reduction of the elastic modulus up to the point when the positive RIVE becomes dominant. This stage is indicated with green color in Fig. 8. After that, the small cracks in labradorite (light blue color) occur predominantly due to the expansion of olivine (dark red color) up to the instant when the RIVE of labradorite only still increases. This stage is indicated with orange color in Fig. 8. During this...
period, the crack volume increases steadily and the elastic modulus reduces although at a smaller rate. Next, due to the expansion of labradorite, the cracks in and around labradorite are closing. This reduces the evolution of crack volume rate even more as well as the elastic modulus degradation rate. After that, the RIVE of labradorite becomes dominant, which leads to the gradual increase in the total RIVE of the rock, the crack volume and the reduction of the elastic modulus. This stage is indicated with purple color in Fig. 8. The main conclusion which can be drawn from this case is that the fastest increase in the crack volume and the fastest degradation of the elastic modulus are associated with the contraction of one constitutive mineral and the expansion of the other constitutive minerals or, in other words, they are associated with the greatest difference in the RIVE of the rock constituting minerals.

3.4 Case 4

As an example of Case 4, the rock 8 (obsidian) calculated at arbitrary temperature, as RIVE does not depend on the irradiation temperature here, was considered. The detailed results of the calculations are shown in Fig. 9 in the similar way as it is explained in Section 3.1. Glass is considered only as free of defects because these constitutive minerals show isotropic expansion (Denisov et al. 2012) and, thus, it was considered that the intragrain and intracrystalline defects would not occur inside them. Also, the defects were neglected in homogeneous materials when the volume fraction of the rock-forming minerals is higher or equal to 95%.

Figure 9 evidences no cracks and no degradation of the elastic modulus due to the expansion of the homogeneous rock. Two-component rocks with the identical RIVE of the constitutive minerals would show the same response. If the volume fraction of the rock-forming mineral is lower than 95% and the RIVE of the constitutive minerals is anisotropic, the intragrain and intracrystalline defects would occur in the graph showing the total RIVE of minerals vs. the crack volume. However, since these defects are also homogeneously distributed within the rock, this would only increase the RIVE of the rock and would not lead to the elastic modulus degradation. The main conclusion which can be drawn from this case is that the RIVE does not always result in crack formation and reduction of the elastic modulus.

3.5 Case 5

It should be noted that there is no rock within 35 rocks examined by Denisov et al. (2012) with the response exactly corresponding to Case 5 shown in the catalogue in Table 1, however, such a response is possible. As an example of Case 5, the rock 32 (fine-grain limestone) calculated at arbitrary temperature, as RIVE does not depend on the irradiation temperature here, was considered as the nearest approximation. The detailed results of these calculations are shown in Fig. 10 in the similar way as it is explained in Section 3.1.

As can be seen in Fig. 10, this rock does not correspond exactly to Case 5 given in the catalogue in Table 1 in terms of the crack volume. The reason for this is that...
the volume fraction of the rock-forming mineral is lower than 95% and that the RIVE of the constitutive minerals is anisotropic. In the model, the homogeneous intragrain and intracrystalline defects, which do not affect the
elastic modulus, then occur. This is the reason why this rock defers slightly from Case 5 presented in the catalogue in Table 1. However, if the volume fraction of the rock-forming minerals of this rock would be different, e.g. 95% and 5% instead of 80% and 20%, the rock response would correspond exactly to Case 5 given in catalogue.

In Case 5, the expansion of the rock constituting minerals is similar at the beginning of irradiation (Fig. 10). Similarly as in Case 4, it does not lead to the crack formation (except the intragrain and intracrystalline defects) and the elastic modulus degradation. This stage is indicated with green color in Fig. 10. After that, the RIVE of the rock constituting minerals becomes different which leads to the gradual increase of the grain boundary crack volume. This results in a significant reduction of the elastic modulus. This stage is indicated with orange color in Fig. 10. The main conclusion which can be drawn from this case is that the damage of the rock can be delayed and subsequently increased rapidly after the delay.

3.6 Case 6

As an example of Case 6, the rock 27 (medium-grain hornblendite) calculated at the temperature of 90°C was considered. The detailed results of the calculations are shown in Fig. 11 in the similar way as it is explained in Section 3.1. In this case, the defects were neglected because the rock was considered to be homogeneous, when the volume fraction of rock-forming minerals is higher than 95%.

As can be seen in Fig. 11, at the beginning of irradiation, hornblende (light blue color) has a higher RIVE than quartz (dark red color). This leads to the occurrence of small cracks around the quartz and to the slight reduction of the elastic modulus. This stage is indicated with green color in Fig. 11. After that, the RIVE of quartz becomes equal to the RIVE of hornblende and the cracks are closing. This leads to the recovery of the elastic modulus. This stage is indicated with orange color in Fig. 11. Next, the RIVE of quartz becomes dominant. This leads to the significant increase in crack volume and the degradation of the elastic modulus. This stage is indicated with purple color in Fig. 11. From this analysis, it can be concluded that even a small amount of a highly expansive mineral may lead to a significant reduction of mechanical properties of the rock.

3.7 Case 7

As an example of Case 7, the rock 23 (medium-grain olivinite) calculated at the temperature of 80°C was considered. The detailed results of the calculations are shown in Fig. 12 in the similar way as it is explained in Section 3.1. In this case, the defects were neglected because the rock was considered to be homogeneous, when the volume fraction of rock-forming minerals is equal to 95%.

At the beginning of irradiation, due to the higher RIVE of olivine (dark red color), the cracks in and around ore (light blue color) occur. This results in reduction of the
elastic modulus. This stage is indicated with green color in Fig. 12. After that, the cracks in and around ore continue growing, however, new cracks do not occur. This leads to the increase in RIVE and the crack volume without any reduction of the elastic modulus. This stage is indicated with orange color in Fig. 12. Next, the RIVE of the rock constituting minerals becomes equal, which leads to the crack closing and the recovery of the elastic modulus. This stage is indicated with purple color in Fig. 12. After that, ore continues to expand, however, as the difference in the RIVE of ore and olivine is insignificant and the volume fraction of ore is low, no additional cracks occur and no reduction in the elastic modulus can be observed. The main conclusion which can be drawn from this case is that the increase in crack volume does not always lead to the reduction of mechanical properties of the rock.

3.8 Case 8

As an example of Case 8, the rock 26 (fine-grain serpentinitized dunite) calculated at the temperature of 230°C was considered. Despite the fact that the numerical model shows poor validation for this rock, because the model fails to predict the RIVE of the rocks with pronounced volume change due to the gas release, such response is possible. The detailed results of the calculations are shown in Fig. 13 in the similar way as it is explained in Section 3.1. Serpentine, as an assumption, is considered without any defects, because these constitutive minerals have isotropic expansion (Khmurovska and Štemberk, 2021) and, thus, it was considered that the intragrain and intracrystalline defects would not occur inside them.

At the beginning of irradiation, due to the higher RIVE of olivine (dark red color), the cracks in and around serpentine (light blue color) occur. This results in reduction of the elastic modulus. This stage is indicated with green color in Fig. 13. After that, the RIVE of serpentine becomes significant and the cracks start closing. In the model, all the grain boundary cracks can be closed, even though there is no possibility of defect healing, however, the total model response correlates with real observation on damaged rocks, when the cracks can be closed only partially (Zhang et al. 2020; Peng et al. 2015; Zhu and Arson 2014). Such crack closing results in a significant, but not complete, recovery of the elastic modulus. The stage where all the boundary cracks are closed is indicated with orange color in Fig. 13. Next, the RIVE of serpentine becomes dominant and new cracks in and around olivine occur. This results in the second wave of the elastic modulus degradation. This stage is indicated with purple color in Fig. 13. The main conclusion which can be drawn from this case is that the partial recovery of the elastic modulus is possible even for significantly damaged rocks. However, it should be noted that the rock can be fully damaged before the crack closing starts, and thus, there would not be a chance for mechanical properties to recover.

4. Conclusions

The paper presents a catalogue of the possible damage
(elastic modulus degradation) of rock aggregates, which was validated with an RBSM numerical model.

The numerical model showed good correlation with the experimental data in terms of the elastic modulus degradation for all the rocks except the rocks with significant radiation-induced gas release, such as fine-grain serpentinized dunites, and the rocks with significant creep, such as fine-grain laminated aleurolite and small-grain sandstone.

The paper presents detailed description of two common and six specific cases of rock aggregate damage, which can be used as a reference when simple analytical models will be developed.

Considering the specific cases of the radiation-induced damage of rock aggregates, it was demonstrated numerically that:

1. The steepest increase in crack volume and the fastest degradation of the elastic modulus are associated with the contraction of one constitutive mineral and the concurrent expansion of the other constitutive minerals or, in other words, it is associated with the greatest difference in the RIVE of the rock constituting minerals.
2. The RIVE does not always result in crack formation and reduction of the elastic modulus.
3. The damage of the rock can be delayed and then increased rapidly after the delay.
4. Even a small amount (1%) of a highly expansive mineral leads to a significant reduction of mechanical properties of the rock.
5. The increase in crack volume does not always lead to the reduction of mechanical properties.
6. Partial recovery of the elastic modulus of the rocks is possible even for significantly RIVE-damaged rocks.

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Appendix

According to Denisov et al. (2012), the RIVE of minerals can be defined as follows.

\[
\frac{\Delta V}{V} = \begin{cases} 
\left( \frac{\Delta V}{V} \right)_{\text{max}} \cdot \left( e^{a(T) \cdot dpa} - 1 \right) & \text{if } a(T) \neq \infty \text{ and } \beta(T) \neq 0, \\
\left( \frac{\Delta V}{V} \right)_{\text{max}} + a(T) \cdot e^{b(T) \cdot dpa} & \text{if } a(T) = \infty \text{ and } \beta(T) = 0,
\end{cases}
\]  
(A1)

where \( \frac{\Delta V}{V} \) is the RIVE of the mineral in \%, \( \left( \frac{\Delta V}{V} \right)_{\text{max}} \) is the maximum RIVE of the mineral in \%, which is shown in Table A1, \( dpa \) is the displacement per atom, \( a(T) \), \( b(T) \), \( \alpha(T) \) and \( \beta(T) \) are the temperature dependent model parameters, \( T \) is the average irradiation temperature in K and \( A, B, A_B, B_B \) and \( \beta_0 \) are the model coefficients which are shown in Table A1.

The composition of the rocks, which were examined by Denisov et al. (2012) are shown in Tables A2 to A4.

\[
\beta(T) = (A_B + B_B \cdot T)^{-1} + \beta_0, 
\]  
(A5)
Table A1 Constants of model of RIVE of minerals according to Denisov et al. (2012).

| Mineral       | $\Delta V/V_{\max}$ [%] | $-\ln A_\alpha$ [-] | $B_\alpha$ $10^3$ [K] | $-\Delta \beta_\theta$ [% K$^{-1}$] | $B_\beta$ $10^2$ [% K$^{-1}$] | $\beta_0$ [% -1] |
|---------------|-------------------------|---------------------|------------------------|--------------------------------------|-------------------------------|-----------------|
| Quartz        | 17.90                   | 13.12               | 3.96                   | 10.80                                | 3.65                          | 0.40            |
| Potassium Feldspar | 7.70                 | 15.58               | 4.74                   | 6.30                                 | 2.11                          | 1.10            |
| Albite        | 9.70                    | 15.10               | 4.41                   | 8.20                                 | 2.76                          | 0.74            |
| Oligoclase    | 7.00                    | 14.80               | 4.41                   | 5.90                                 | 1.99                          | 1.03            |
| Labradorite   | 4.50                    | 13.80               | 4.20                   | 3.20                                 | 1.10                          | 1.60            |
| Nepheline     | 10.00                   | 14.00               | 4.20                   | 6.10                                 | 2.10                          | 0.70            |
| Analcime      | 4.00                    | 13.60               | 4.20                   | 4.30                                 | 1.40                          | 1.00            |
| Muscovite     | 15.00                   | 12.70               | 4.20                   | 20.00                                | 7.60                          | 0.19            |
| Biotite       | 15.00                   | 14.40               | 4.20                   | 9.10                                 | 3.10                          | 0.46            |
| Chlorite      | 1.50                    | 12.30               | 4.20                   | 0.61                                 | 0.21                          | 7.00            |
| Serpentine    | 2.00                    | 11.38               | 4.20                   | 1.20                                 | 0.40                          | 3.5             |
| Enstatite     | 2.75                    | 10.30               | 3.92                   | 2.40                                 | 0.79                          | 3.25            |
| Diopside      | 2.75                    | 10.30               | 3.92                   | 2.40                                 | 0.79                          | 3.25            |
| Augite        | 2.80                    | 10.30               | 3.92                   | 2.40                                 | 0.79                          | 3.25            |
| Hornblende    | 1.70                    | 9.70                | 3.80                   | 0.45                                 | 0.15                          | 3.00            |
| Olivine       | 0.75                    | 8.90                | 4.20                   | 0.46                                 | 0.16                          | 9.30            |
| Calcite       | 0.40                    | -3.70               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Magnesite     | 0.70                    | -3.00               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Siderite      | 0.70                    | -3.00               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Dolomite      | 0.80                    | -3.00               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Chromite      | 1.00                    | -0.69               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Magnetite     | 0.60                    | -1.20               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |
| Quartz glass  | -3.00                   | -3.60               | 0.00                   | $\infty$                              | 0.00                          | 0.00            |

Table A2 Composition of examined igneous rocks according to Denisov et al. (2012).

| No. | Name                          | Composition                  |
|-----|-------------------------------|------------------------------|
| 1   | Large-grain granite           | Quartz: 25%, Microcline: 50%, Oligoclase: 24%, Biotite: 1% |
| 2   | Medium-grain granite          | Quartz: 30%, Microcline: 35%, Oligoclase: 20%, Biotite: 5%, Hornblende: 10% |
| 3   | Medium-grain granite          | Quartz: 30%, Microcline: 40%, Oligoclase: 25%, Ore: 5% |
| 4   | Small-grain porphyry granite  | Quartz: 40%, Microcline: 30%, Oligoclase: 15%, Muscovite: 13% |
| 5   | Medium-grain granodiorite     | Quartz: 20%, Microcline: 10%, Oligoclase: 40%, Hornblende: 13%, Biotite: 15% |
| 6   | Medium-grain granodiorite     | Quartz: 20%, Microcline: 20%, Oligoclase: 40%, Biotite: 10%, Hornblende: 10% |
| 7   | Aphanite liparite             | Quartz: 35%, Microcline: 33%, Oligoclase: 32% |
| 8   | Obsidian                      | Glass: 100% |
| 9   | Fine-grain quartz andesite    | Quartz: 30%, Oligoclase: 60%, Diopside: 10% |
| 10  | Small-grain diorite           | Quartz: 5%, Oligoclase: 55%, Mica: 30%, Ore: 10% |
| 11  | Small-grain albite            | Albite: 70%, Analcime: 30% |
| 12  | Large-grain labradorite       | Labradorite: 75%, Diopside: 15%, Biotite: 5%, Magnetite: 5% |
| 13  | Large-grain urtite           | Nephelin: 85%, Diopside: 10%, Apatite: 5% |
| 14  | Medium-grain gabbro           | Labradorite: 60%, Diopside: 40% |
| 15  | Small-grain gabbro            | Oligoclase: 53%, Quartz: 2%, Hornblende: 45% |
| 16  | Gabbro-porphyr                   | Oligoclase: 50%, Diopside: 50% |
| 17  | Fine-grain porous diabase     | Labradorite: 60%, Diopside: 39%, Ore: 1% |
| 18  | Fine-grain diabase            | Labradorite: 50%, Augite/Enstatite: 40%, Chlorite: 10% |
| 19  | Small-grain poikilitic diabase| Labradorite: 45%, Olivine: 30%, Hornblende: 20%, Magnetite: 5% |
| 20  | Fine-grain basalt             | Labradorite: 50%, Olivine: 30%, Ore: 10%, Glass: 10% |
| 21  | Small-grain pyroxenite        | Olivine: 50%, Plagioclase: 10%, Enstatite: 40% |
| 22  | Medium-grain peridotite       | Olivine: 80%, Diopside: 15%, Ore: 5% |
| 23  | Medium-grain olivinite        | Olivine: 95%, Ore: 5% |
| 24  | Small-grain dunite            | Olivine: 80%, Serpentine: 10%, Enstatite: 10% |
| 25  | Fine-grain serpentinized dunite| Olivine: 60%, Serpentine: 40% |
| 26  | Fine-grain serpentinized dunite| Olivine: 25%, Serpentine: 75% |
| 27  | Medium-grain hornblende       | Hornblende 100% approx. |

Table A3 Composition of examined sedimentary silicate rocks according to Denisov et al. (2012).

| No. | Name                          | Composition                  |
|-----|-------------------------------|------------------------------|
| 28  | Fine-grain laminated aleurolite| Quartz: 30%, Feldspar: 65%, Mica/Ore: 5% |
| 29  | Small-grain sandstone         | Quartz: 45%, Feldspar: 45%, Ore: 10% |
Table A4 Composition of examined sedimentary carbonate rocks according to Denisov et al. (2012).

| No. | Name                      | Composition            |
|-----|---------------------------|------------------------|
| 30  | Fine-grain limestone      | Calcite: 99%           |
| 31  | Small-grain limestone     | Calcite: 99%           |
| 32  | Fine-grain limestone      | Calcite: 80%, Dolomite: 20% |
| 33  | Fine-grain porous magnesite | Magnesite: 99%      |
| 34  | Fine-grain porous siderite | Siderite: 90%, Quartz: 10% |
| 35  | Medium-grain dolomite     | Dolomite: 95%, Siderite: 5% |