

Increasing of drinking water quality by adding carbonate rocks to low mineralized groundwater: Case study from the Krupina district, Slovakia

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Abstract: The research has revealed that the increased mortality on cardiovascular and oncological diseases in the Krupina district, belonging among the most serious in Slovakia, is caused mainly by the low calcium and magnesium contents in the groundwater, being used for drinking by inhabitants of the Krupina district.

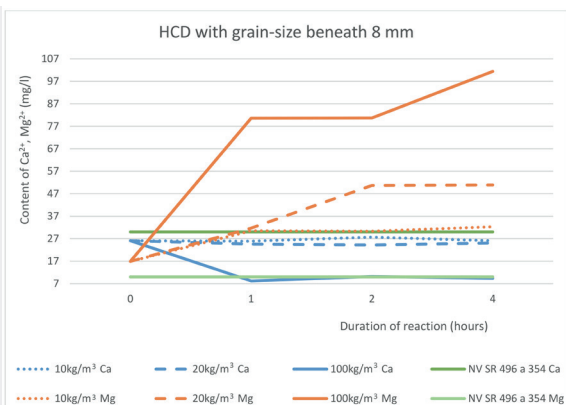
The paper provides the research results of artificial mineralization tests for one model (distilled) water and two waters from the representative home wells from the Krupina district at standard partial air pressure ($\text{CO}_2 \sim 0.03 \text{ vol.}\%$), as well as at increased CO_2 concentration in the water in laboratory static and dynamic conditions, applying carbonatic rocks (dolomite, magnesite, limestone), as well as the half-calcined dolomite (HCD) in amounts of 10, 20 and 100 kg/m^3 of the water and duration of mineralization process 1, 2, resp. 4 hours.

The laboratory tests of the mineralization of the water have manifested that there is possible significantly increase the content of Ca and Mg in the water to values reaching, or even overreaching, the requested values for drinking water. The highest enrichment was obtained using HCD and it related to magnesium increase. However, the HCD application caused the decrease of the Ca content. Therefore, the limestone or dolomite must be also used in the mixture, so for water mineralization in the home wells we recommend using a mixture of limestone and HCD, where the HCD content in the mixture should be about 30 %. No significant differences were found from the point of view of the grain-size of the carbonate rock used. The Ca and Mg contents increase with longer duration of mineralization process. The recommended amount of added rock is from 20 to 100 kg.m^{-3} of the water.

The controlled addition of carbon dioxide to waters with deeply deficient calcium and magnesium contents when using dolomite (or half-calcined dolomite) at the amount of 20 kg/m^3 causes a significant increase in the hardness and Ca and Mg concentrations in a relatively short time (up to 5 minutes) in mineralized water up to / above the recommended values according to the valid legislation.

Key words: Water mineralization, dolomite, half-calcined dolomite (HCD), magnesite, limestone, water hardness, calcium (Ca), magnesium (Mg)

Graphical abstract



Highlights

- One of reasons of increased mortality on cardiovascular and oncological diseases in regional scale can be the low Ca and Mg contents in the drinking water
- The simple way how to increase the low Ca and Mg contents in drinking water is to add a mixture of limestone (70 %) and half-calcined dolomite (30 %) into the home wells. The process of Ca and Mg deliberation can be even more accelerated by addition of CO_2 into the water.

Introduction

The research on the projects GEOHEALTH (www.geology.sk/geohealth) and LIFE FOR KRUPINA (www.geology.sk/lifeforkrupina) has revealed that the health state of the population in the Krupina district belongs among the worst in Slovakia. It is caused mainly by increased mortality on cardiovascular and oncological diseases mainly due to the low calcium and magnesium contents in the

groundwater, being used by inhabitants of the Krupina district (Rapant et al., 2015, 2017). The contents of these two important elements are mostly within the range of Slovak norms - Regulations of the Government of Slovak Republic Nos. 8/2016 Coll., 496/2010 Coll. and 354/2006 Coll. for the drinking water, but often their contents are lower than recommended values. The main objective of the laboratory research was to propose optimal composition of the additive carbonate rocks, their optimum grain-size and the

necessary quantities, which would be the most suitable for increasing the Ca and Mg contents in the low mineralized water in the individual home wells in the Krupina district.

Laboratory experimental work

Current general knowledge and basic concepts

Calcium (Ca) belongs among the principal macroelements forming the chemical composition of natural waters. It primarily enters into the groundwater by the dissolution of limestone, dolomite, gypsum, feldspar, less often by magnesite, amphibolite, pyroxene, a.o. In the groundwater there prevails a simple form of cation - Ca^{2+} . Depending on the total chemical composition of the water, various ionic associates may be formed, e.g. $[\text{CaCO}_3(\text{aq})]^0$, $[\text{CaHCO}_3]^+$, $[\text{CaOH}]^+$, etc. (cf. Rapant et al., 1996).

The natural source of magnesium (Mg) in the groundwater is represented mainly by the interaction products between water and rocks - dolomite, magnesite, amphibolite, resp. minerals - olivine, pyroxenes, etc. The Mg concentration in the water depends also on weathering conditions. In the groundwater the magnesium occurs prevalingly as cation - Mg^{2+} . Less often it forms complex compounds $[\text{MgSO}_4(\text{aq})]^0$, $[\text{MgCO}_3(\text{aq})]^0$, $[\text{MgHCO}_3]^+$ and $[\text{MgOH}]^+$. In relation on pH values Mg can precipitate from the moderately acid water as $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$, in neutral water as $3\text{MgCO}_3 \cdot \text{Mg}(\text{OH})_2 \cdot 3\text{H}_2\text{O}$ and at pH values higher than 9 as $\text{Mg}(\text{OH})_2$. At the presence of phosphates in water, magnesium produces a weakly soluble compound $\text{Mg}_3(\text{PO}_4)_2$ (cf. Rapant et al., 1996).

The term water hardness means the sum of Ca+Mg contents in mmol/l. We distinguish the temporary hardness (formed by calcium and magnesium carbonates and bicarbonates) and the constant (permanent) hardness formed by Ca and Mg salts, e.g. sulphates, chlorides, nitrates, etc. Of the poorly soluble calcium and magnesium compounds, the carbonates of these elements, $\text{CaMg}(\text{CO}_3)_2$, CaSO_4 , fluorides and phosphates are of utmost importance in hydrogeochemistry and technology. When heating the water, Ca (Mg) carbonates can be precipitated, forming a sludge to a solid crust, so-called incrustat ("waterstone") in the boilers. Magnesium is less damaging in this particular case because MgCO_3 is more soluble than CaCO_3 (Rapant et al., 1996). According to hardness, we divide the water into five groups (Tab. 1).

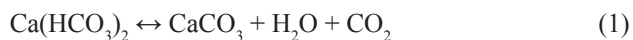
According to Regulation of the Government of Slovak Republic (RGoSR) No. 8/2016 Coll., establishing the requirements for the water for human consumption, and this water quality check as amended by the RGoSR No. 496/2010 Coll. in part B of Supplement 1 to RGoSR No. 354/2006 Coll. (Drinking water quality indicators and their limits) the part e) states the substances, which presence in drinking water is desirable and the recommended

Tab. 1
Water hardness scale.

Water designation	Degree of hardness (mmol.l ⁻¹)	Degree of hardness (°dH)
Very soft	< 0.69	< 3.89
Soft	0.7 – 1.25	3.9 – 7.0
Medium hard	1.26 – 2.5	7.01 – 14.0
Hard	2.51 – 3.75	14.01 – 21.0
Very hard	>3.76	> 21.01
Recalculation of the water hardness: 1 mmol/l = 5.6 °dH or 1 °dH = 0,1783 mmol/l		

value (RV) for the calcium and magnesium content (water hardness) should be within the range 1.1 to 5.0 mmol/l, RV of Ca^{2+} content has to be over 30 mg/l and RV of Mg^{2+} content in the range 10-30 mg/l. According to the above regulations, the limit value (LV) for the Mg^{2+} content is a value 125 mg/l (Mg content above this level causes the bitter taste of water) and for pH the LV is from 6.5 to 9.5. Optimum Ca and Mg concentration in drinking vode is not easy to determine, because medical requirements do not have to overlap with technical requirements (increased hardness - increased water precipitates formation).

The hydrogen carbonates are a common part of the chemical composition of the groundwater. Their content in natural waters is predominantly below 500 mg/l. In the case of groundwater saturation with carbon dioxide (CO_2), the HCO_3^- concentration may exceed the value 1000 mg/l. The main natural source of hydrogen carbonates in the groundwater is mainly the dissolution of limestone and dolomite, being accelerated by the saturation with CO_2 from different sources. Besides the simple HCO_3^- ions, the hydrogen carbonates are present at stronger mineralized waters, as well as ion accociates, mainly $[\text{CaHCO}_3]^+$, $[\text{MgHCO}_3]^+$, $[\text{MnHCO}_3]^+$, a.o. At heating they change to water vapour, CO_2 and carbonates (eq. 1). The carbonates of Ca and Mg form so called carbonatic incrust ("water stone"; Rapant et al., 1996).



Regarding the limited dissolution of CaCO_3 (resp. CaSO_4), the Ca concentration does not overreach the value 1000 mg/l in mineralized waters, while the high Ca concentration is bound on presence of sufficient amount of dissolved CO_2 . The magnesium in the natural waters is usually less abundant than calcium, though the weight ratio of Ca : Mg there varies as a rule within a range of 4 : 1 to 2 : 1. The magnesium content in the water above 250 mg/l is usually manifested by the bitter taste and the waters

with the high Mg concentration have laxative effects. Such groundwaters are not present in the territory of Slovak Republic (Rapant et al., 1996).

The Ca and Mg carbonates are solid, in the water (without the content of CO₂) insoluble, resp. very weakly soluble substances. The calcium carbonate CaCO₃ is present in the nature as limestone, calcite and aragonite. The magnesium carbonate MgCO₃ is known as dissoluble and anhydrous mineral magnesite. The carbonates of these elements react with CO₂ water solution, originating soluble hydrogen carbonates of Ca(HCO₃)₂, resp. Mg(HCO₃)₂ can exist only in the water solution in the presence of excess CO₂ (Gažo et al., 1981).

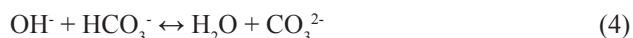
Because the CO₃²⁻ anion represents the strong alkaline substance, the carbonates in the water solution undergo strong hydrolysis according the equation (2), so they manifest alkaline reaction:



The hydrogen carbonates react weakly alkaline, because the HCO₃⁻ anion behaves weak alkaline (3):



Warming of the hydrogen carbonate solution causes the deliberation of CO₂, originating by the decomposition of the acid carbonate, which disbalances the equilibrium of the reaction (3) a new share of hydrogen carbonate ions and originating hydroxide ions enter immediately into reaction (4), so the hydrogen carbonate gradually changes to carbonate – it is the opposite to reaction 2:



The above stated reactions (2), (3) and (4) closely relate with the most important protolytic system in natural and supply waters: CO₂; - HCO₃⁻; - CO₃²⁻ (Gažo et al., 1981). Tab. 2 presents the dissolution values, resp. the multiple of dissolution of some minerals (compounds) according to Andrlik et al. (1967).

There is frequently suggested that weakly mineralized or demineralized waters should be mineralized by means of flow column, being filled by dolomite or limestone. The water flowing through the carbonate fill has to dissolve this fill and enrich by Ca and Mg in the case of dolomite, eventually by Ca in the case of limestone. The deliberation of Ca and Mg from dolomite and Mg from dolomite (event. from limestone and magnesite) into the water is influenced by numerous factors as the flow velocity of the water, resp. the ratio of solid phase : liquid phase (water), grain-size of the carbonate material, resp. its specific surface, concentration of dissolved CO₂, duration of the contact of solid

phase (dolomite, limestone, magnesite) with the water, etc. (Janda et al., 2010).

The carbonate ions delivered into the solution by the dissolution of dolomite or calcite react with the water (hydrolysis according to equation 2), producing the hydrogen carbonates or at least carbonic acid (eq. 3), which by this

Tab. 2
Dissolution values of some minerals (compounds) at various temperatures

Dissolution of anhydrous compound in g/100 g of solution at various temperatures			
Compound	t = 20°C	t = 50°C	t = 100°C
CaCO ₃	0.0014	0.0015	0.0017
CaSO ₄	0.204	0.210	0.065
CaSO ₄ ·2H ₂ O	0.250	0.240	0.200
Dissolution of compound in g/100 ml of the water			
MgSO ₄ ·7H ₂ O 117 240			490
MgCl ₂ ·6H ₂ O	305	425 (60°C)	910
K _s – dissolution coefficient			
CaCO ₃	0.87 · 10 ⁻⁸ mol/l (25°C)		
CaSO ₄	6.40 · 10 ⁻⁵ mol/l (18°C)		
MgCO ₃	2.60 · 10 ⁻⁵ mol/l (12°C)		

way deplete the system by part of carbonates. Higher concentrations of Ca and Mg ions will be present in the water at the contact with the air CO₂ (concentration of CO₂ in the air is in average around 0.03 vol.%), because its dotation into water will increase the dissolution of dolomite or calcite (Janda et al., 2010).

The research results have demonstrated that also in the most favourable case, i.e. at temperature of 10 °C, access of air and the use of the highest value of the multiple of dolomite dissolution K_s = 10exp(-17.09) by the calculation in the program PHREEQC, the equilibrium concentration of the Ca 16.8 mg/l and Mg 10.2 mg/l will be achieved, while the dissolution of CO₂ in the water at common partial pressure in the air is 0.7 mg/l at 10 °C and 0.44 mg/l at 25 °C (Janda et al., 2010).

In the case of weakly mineralized water and low concentration of “free” CO₂, the reaction speed is low and therefore the technology of water mineralization will become more efficient when using the guided dosing of CO₂ or e.g. by the control of dosing of diluted solution of the hydrochloric acid (HCl) into the column with carbonate fill or it is alternatively possible the direct dosing the dissolved compound of Ca and Mg as solution into the water with careful observation of pH of treated water. Table 3 states the dissolution categories of some calcareous, resp. magnesia compounds (Andrlik et al., 1967).

Tab. 3
Dissolution categories of some Ca and Mg compounds

Name of compound	Chemical formula	Dissolution
Magnesium hydroxide	$\text{Mg}(\text{OH})_2$	Non-soluble in the water, soluble in H^+
Calcium hydroxide	$\text{Ca}(\text{OH})_2$	Weakly soluble in the water
Calcium chloride anhydrous	CaCl_2	Soluble in the water
Calcium chloride hexahydrate	$\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$	Soluble in the water
Magnesium chloride anhydrous	MgCl_2	Soluble in the water
Magnesium chloride hexahydrate	$\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$	Soluble in the water
Calcium sulfate anhydrous	CaSO_4	Weakly soluble in the water
Calcium sulfate dihydrate	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	Weakly soluble in the water
Magnesium sulfate anhydrous	MgSO_4	Soluble in the water
Magnesium sulfate heptahydrate	$\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$	Soluble in the water

The water mineralization can be increased also by some products of thermic decomposition of Ca, resp. Mg carbonates (cf. Luptáková et al., 2013a, b, 2016; Vrabec et al., 2016), e.g. half-calcined dolomite - HCD ($\text{MgO} \cdot \text{CaCO}_3$), caustic lime (CaO), hydrate of lime $\text{Ca}(\text{OH})_2$ and magnesite clinker (MgO). In this case there is necessary to register the pH of mineralized water (for the pH of the water the limit value (LV) is from 6.5 to 9.5).

The division of carbonate rocks (according to recommended Slovak Technical Norm STN EN 72 1210) with the limestone and dolomite contents in various shares of calcite - CaCO_3 and dolomite - $\text{CaMg}(\text{CO}_3)_2$ is as follows (Tab. 4):

The half-calcined dolomite (HCD), we used in our laboratory experiments, is produced from dolomite raw mate-

rial in the Malé Kršteňany factory by the heat treatment at ca 720-950 °C in fractions 2-4, 4-8 and 8-11 mm. Besides the treatment of drinking water the HCD can be used also for the composite sorbents for the capture of pollutants and heavy metals from polluted waters.

Laboratory experimental part - mineralization of model (distilled) water

For the mineralization of *model (distilled) water* under the static (non-flowing) conditions, the carbonate rocks from localities Trebejov (dolomite), Košice-Bankov (magnesite) and Jaklovce (limestone) were used, all having the grain-size beneath 8 mm. Tab. 5 states the basic chemical and mineral composition of these samples and Tab. 6 their grain-size.

Tab. 4
Division of carbonatic rocks (Slovak Technical Norm STN EN 72 1210)

Name	Mineral content in %		Mineral composition in %	
	Calcite	Dolomite	CaCO_3	MgCO_3
Limestone	100 – 90	0 – 10	100 – 95.4	0 – 4.6
Dolomitic limestone	90 – 50	10 – 50	95.4 – 77.1	4.6 – 22.9
Calcareous dolomite	50 – 10	50 – 90	77.1 – 58.8	22.9 – 41.2
Dolomite	10 – 0	90 – 100	58.8 – 54.3	41.2 – 45.7

Tab. 5
Chemical and mineral composition of carbonate samples

Locality	Rock	Content of compounds (wt.%)			Content of minerals (wt.%)
		CaO	MgO	CO_2	
Trebejov	Dolomite	31.0	21.8	46.0	98.6 % dolomite
Stoichiometric dolomite		30.41	21.86	47.73	100 % dolomite (54.3 % CaCO_3 + 45.7 % MgCO_3)
Košice	Magnesite	2.8	43.5	49.7	86.8 % magnesite + 9.2 % dolomite
Stoichiometric magnesite		0	47.81	52.19	100 % magnesite
Jaklovce	Limestone	54.0	1.4	43.9	93.0 % calcite + 5.6 % dolomite
Stoichiometric limestone		56.03	0	43.97	100 % limestone

Table 5 manifests that the samples of carbonate rocks contain dominant share of dolomite - 98.6 % (Trebejov); magnesite - 86.8 % (Košice; besides 9.2 % of dolomite) and 93 % of calcite (Jaklovce; besides 5.6 % of dolomite). Table 6 shows that the rock grit with grain-size beneath 8 mm, used for mineralization of *model* (distilled) water contain only small amounts of the dust share (beneath 0.1 mm), being represented in the range from 5.25 to 8.12 %.

Tab. 6
Wet granulometric analysis of dolomite, magnesite and limestone samples

Grain-size class (mm)	Weight yield of the class (%)	Remaining over sieve (%)	Passing through the sieve (%)
Dolomite beneath 8 mm			
4 – 8	49.20	49.20	-
2 – 4	23.62	72.82	50.80
1 – 2	12.61	85.43	27.18
0.1 – 1	9.32	94.75	14.57
< 0.1	5.25	-	5.25
Together	100.00	-	-
Magnesite beneath 8 mm			
4 – 8	25.58	25.58	-
2 – 4	30.83	56.41	74.42
1 – 2	13.39	69.80	43.59
0.1 – 1	22.08	91.88	30.20
< 0.1	8.12	-	8.12
Together	100.00	-	-
Limestone beneath 8 mm			
4 – 8	49.95	49.95	-
2 – 4	19.42	69.37	50.05
1 – 2	11.51	80.88	30.63
0.1 – 1	12.19	93.07	19.12
< 0.1	6.93	-	6.93
Together	100.00	-	-

Experimental laboratory tests of mineralization of the *model* (distilled) water with the studied rock samples in static (non-flow) conditions were done in the stirrer reactor at various duration of reaction under atmospheric CO₂ (ca 0.03 vol.%) and shaking of the suspense, at free permeation of the suspense by CO₂ (8 litres of CO₂/1 minute) and stirring of suspense at input CO₂ pressure 0.3 MPa. The quantity of the solid phase to liquid phase was 10 grams of the sample per 500 ml of distilled water (20 kg/m³ of the water). The water hardness (mmol/l) of entering distilled water, water after mineralization, as well as the Ca and Mg

contents (mg/l) in all water samples after filtering of the solid phase were determined by the chelatometric method (Holzbecher, 1974). The results of the mineralization tests of *model* (distilled) water are stated in Table 7.

Table 7 demonstrates that the hardness of *model* (distilled) water is very low (very soft water) with the contents of Ca, resp. Mg only 2.21 mg/l, resp. 1.34 mg/l. After one hour lasting shaking of this water with the dolomite, magnesite, resp. limestone sample at atmospheric CO₂ the qualitative indicators of mineralized water have increased only slightly, and the reached values of water hardness were in the range of 0.315 to 0.405 mmol/l (very soft water) with the Ca content in the range of 6.62 to 10.22 mg/l, resp. Mg in the range of 1.94 to 5.84 mg/l. The recommended values according to valid legislative norms were not reached.

The most favourable results were reached after 5 minutes mineralization of the distilled water by dolomite at input CO₂ pressure 0.3 MPa and shaking of suspension, obtaining the water hardness of 3.51 mmol/l (hard water) with the contents of Ca 35.3 mg/l and Mg 63.9 mg/l. Tab. 7 shows that even after 5th minute of mineralization of distilled water by the magnesite sample at input CO₂ pressure 0.3 MPa, the Mg content was significantly increased (above the limit value 125 mg/l according to the RGoSR). The Ca content was increased only moderately (to 12.8 mg/l) from the present dolomite component of this sample. After mineralization of distilled water lasting 5 minutes by the limestone sample at the starting CO₂ pressure 0.3 MPa, the Ca content significantly increased (101 mg/l), similarly as the Mg content (68.4 mg/l) from the present dolomite component of this sample (5.6 %).

The results in Table 7 demonstrate that even the very soft water (distilled - demineralized water) can be effectively mineralized in relatively short time (up to 5 minutes), i.e. to reach the recommended values of Ca and Mg using the dolomite in the amount of 20 kg/m³ of the water with simultaneous controlled CO₂ dosing (increase of CO₂ concentration in the adopted water).

Laboratory experimental part – mineralization of the water from the wells

For laboratory tests of mineralization of waters from two wells from the Krupina district in static (non-flow) and dynamic (flow) conditions there were used carbonatic rocks from the Trebejov locality (dolomite with the grain-size beneath 4 mm - D) and half-calcined dolomite (HCD) from the locality Malé Kršteňany, having the grain-size beneath 8 mm, resp. their mixture in three total amounts 10, 20 and 100 kg/m³ water (non-flow conditions). For the mineralization tests of the well water with the carbonate crushed samples was taken water designated as *sample 6* (**Kňazová dolina - KD**) and *sample 11* (**Hontianske Nemce - HN**). Table 8 presents the selected qualitative parameters of these samples.

Tab. 7

Static tests of mineralization of the model (distilled) water at 20-22 °C and the carbonate amount of 20 kg/m³ of the water.

Water type and duration of mineralization reaction		Water hardness (mmol/l)	Ca ²⁺ (mg/l)	Mg ²⁺ (mg/l)
RGoSR Nos. 8/2016, 496/2010, 354/2006		1.1–5.0 (OH)	> 30 (OH)	10 - 30 (OH)
Distilled water (pH = 7.62 at 20.4 °C)		0.110	2.21	1.34
Dolomite (10 g + 500 ml distil. water)				
CO ₂ atmospheric (0.03 vol.%)	1 hour	0.405	6.62	5.84
CO ₂ bubbling (8 l/minute)	5 min.	3.36	25.7	66.1
CO ₂ at pressure (0.3 MPa)	5 min.	3.51	35.3	63.9
CO ₂ at pressure (0.3 MPa)	1 hour	5.55	70.5	92.1
Magnesite (10 g + 500 ml distil. water)				
CO ₂ atmospheric (0.03 vol.%)	1 hour	0.315	8.82	2.32
CO ₂ bubbling (8 l/minute)	5 min.	2.65	8.2	59.4
CO ₂ at pressure (0.3 MPa)	5 min.	5.63	12.8	129.1
CO ₂ at pressure (0.3 MPa)	1 hour	5.86	13.8	134.1
Limestone (10 g + 500 ml distil. water)				
CO ₂ atmospheric (0.03 vol.%)	1 hour	0.335	10.22	1.94
CO ₂ bubbling (8 l/minute)	5 min.	5.18	111.4	58.4
CO ₂ at pressure (0.3 MPa)	5 min.	5.355	101.0	68.4
CO ₂ at pressure (0.3 MPa)	1 hour	9.0	237.0	74.9

Note: **Green numbers** – fulfill recommended values (RVs), **red numbers** – do not comply with the recommended values. Designation is valid also for following tables.

Tab. 8

Measured and determined values of selected water quality indicators

Sample No.	Locality	Hardness category	pH at 21 °C	Ca (mg/l)	Mg (mg/l)	Ca+Mg (mmol/l)
6	KD	Soft water	6.99	33.7	9.4	1.225
11	HN	Medium hard water	6.32	26.1	16.9	1.345
RGoSR Nos. 8/2016, 496/2010, 354/2006			6.5-9.5 (LV)	>30 -RV	10-30 (RV)	1.1-5.0 (RV)
For comparison: Drinking water of the conduit in the ŠGÚDŠ Košice			7.42-21.5°C	48.1	19.5	2.0
For comparison: Well water - Košice surrounding (Sokol'			-	87.0	44.5	4.0

Note: RV - recommended value; LV - limit value.

Table 8 shows that water of the *sample 6* is soft with the Mg content beneath recommended value (RV), similarly as the water of the *sample 11* (medium hard water) has the Ca content beneath RV. For comparison with common drinking water we state also values of qualitative indicators for the town of Košice and its surrounding - Sokol' village.

Table 9 shows the chemical and mineral composition of dolomite and half-calcined dolomite (HCD) and Table 10 the grain-size characteristics of dolomite with the grain-size beneath 4 mm, resp. HCD with grain-size beneath 8 mm. The weight yields of individual classes, total remains

on sieves, resp. undersize products are related to dry state of the samples.

Table 9 shows that the dolomite grit with grain-size beneath 4 mm (D) contains 96.7 % of dolomite and small amounts of Si, Al and Fe oxides. The HCD grit with the grain-size beneath 8 mm contains 97.2 % MgO.CaCO₃ and minor amounts of Si and Al oxides.

Table 10 presents that dolomite fraction beneath 4 mm contains a small amount (less than 11 %) of dust fraction of the grain-size class beneath 0.1 mm and the fraction beneath 0.1 mm is represented in the HCD grit with grain-size beneath 8 mm only by negligible 0.29 %.

Tab. 9
Chemical and mineral composition of dolomite (D) and half-calcined dolomite (HCD)

Chemical compound	Chemical composition of the sample (wt.%)	
	Dolomite (D) beneath 4 mm	Half-calcined dolomite (HCD) beneath 8 mm
SiO ₂	1.56	0.43
TiO ₂	0.04	< 0.01
Al ₂ O ₃	0.80	0.19
Fe ₂ O ₃ total	0.35	0.05
CaO	29.5	37.9
MgO	21.4	26.6
MnO	0.03	< 0.01
Na ₂ O	< 0.2	< 0.2
K ₂ O	0.17	< 0.05
P ₂ O ₅	0.02	0.04
SO ₃	< 0.01	0.02
CO ₂	45.5	32.7
LOI	46.0	34.7
um	99.87	99.91
Mineral compound (wt.%)	96.7	97.2
Potential toxic elements	(mg/kg)	(mg/kg)
Pb	< 5	< 5
Hg	0.01	< 0.01
Cd	< 0.5	< 0.5
Ni	5	< 4
As	1.2	0.3
Sb	0.4	< 0.1
Cr	26	17

Table 9 states the contents of potential toxic elements (mg/kg) in grit of dolomite and half-calcined dolomite and Table 11 states the highest limit values (HLV) of these elements (µg/l) according to appropriate Regulations of the Government of Slovak Republic. Table 11 shows that the contents of these elements in leachates from these samples are distinctly lower than HLV stated in regulations.

Static (non-flow) mineralization tests of the water of sample 6 in laboratory conditions

The static mineralization tests of water *samples 6* (resp. *11*) (closed no-flow system) were realized in 1 liter vessels with the shaking of the water sample and dolomite with the

Tab. 10
Wet granulometric analysis of Trebejov dolomite and half-calcined dolomite from Malé Kršteňany

Grain-size class (mm)	Weight yield of the class (%)	Total remnant (%)	Total undersize product (%)
Dolomite beneath 4 mm (D)			
2 – 4	45.82	45.82	-
1 – 2	22.17	67.99	54.18
0.5 – 1	11.99	79.98	32.01
0.1 – 0.5	9.06	89.04	20.02
< 0.1	10.96	-	10.96
Together	100.00	-	-
Half- calcined dolomite beneath 8 mm (HCD)			
4 – 8	80.84	80.74	-
2 – 4	13.71	94.55	19.16
1 – 2	3.46	98.01	5.45
0.5 – 1	0.98	98.99	1.99
0.1 – 0.5	0.72	99.71	1.01
< 0.1	0.29	-	0.29
Together	100.00	-	-

grain-size beneath 4 mm (D), half-calcined dolomite with grain-size beneath 8 mm (HCD), resp. D : HCD = 1 : 1 in quantities 10, 20 and 100 kg/m³ of the water with duration of mineralization process 1, 2 and 4 hours at laboratory temperature (ca 20-22 °C). After each mineralization test, the pH of the water was measured (pH meter ProfiLine pH 3310 WTW GmbH), and the values of selected qualitative indicators of water samples were found by chelatometric titration method. Tables 12, 13 and 14 state the results of static tests of water mineralization in 6 laboratory conditions.

Table 12 shows that increased Ca contents, resp. recommended Mg contents in investigated *water 6* after static mineralization in laboratory conditions were reached in comparison to the input sample with all amounts of dolomite having the grain-size beneath 4 mm at all durations of the mineralization process (Ca contents were from 36.3 to 46.1 mg/l, Mg contents from 11.3 to 14.8 mg/l), except for the dolomite application in quantity 10 kg/m³ for one hour (Mg content is beneath RV) - Fig. 1.

Using the dolomite and HCD in the ratio of 1 : 1 in quantities of 10, 20 and 100 kg/m³ of water the Mg contents were significantly increased, i.e. 2.6 to 8 times (from 24.5 to 75.4 mg/l) generally during all the static mineralization durations,, though at the expense of Ca contents, which were below RV in the range of 8.0-26.5 mg/l, besides the application of this mixture in an amount of 10 kg/m³ in duration of 1 hour, where the Ca 30 mg/l content was on the RV boundary only. When applied D + HCD in

Tab. 11
Chemical composition of leachates from dolomite (D) and half-calcined dolomite (HCD)

Chemical compound	Content of potentially toxic elements		RGoSRNos. 8/2016, 496/2010 and 354/2006, App. 1, part B, a)
	in leachate from D	in leachate from HCD	
Pb (µg/l)	< 5	< 5	10 (HLV)
Hg (µg/l)	< 0.1	< 0.1	1 (HLV)
Cd (µg/l)	< 0.3	< 0.3	5 (HLV)
Ni (µg/l)	< 2	< 2	20 (HLV)
As (µg/l)	< 1	< 1	10 (HLV)
Sb (µg/l)	< 1	< 1	5 (HLV)
Cr (µg/l)	< 2	< 2	50 (HLV)

Note: HLV - the highest limit value

a ratio of 1 : 1 and quantity 100 kg/m³, the upper LV of the water pH was exceeded at all durations of mineralization (pH > 9.5) - Fig. 2.

When used the half-calcined dolomite with grain-size beneath 8 mm in amounts of 10, 20 and 100 kg/m³ of the water, the Mg contents were significantly increased, i.e. 3.6-10.9 times (from 34 to 102.5 mg/l), generally at all durations of static mineralization, though at the expense of Ca contents that were below RV in the range of 8.2-29.9 mg/l, besides the application of the HCD in amount of 20 kg/m³ at 2 hours duration, where the Ca content was 30.1 mg/l - Fig. 3.

The water *sample 6* with hardness 1.225 mmol/l (soft water) was mineralized using all amounts of dolomite during 1, 2 and 4 hours by the way that the water hardness has increased with increasing amount of dolomite and the

Tab. 12
Static tests of the mineralization of water - *sample 6* - at 20-22°C

Sample type	Quantity (kg/m ³)	Duration (hod.)	pH	Ca and Mg (mmol/l)	Ca (mg/l)	Mg (mg/l)
RGoSR Nos. 8/2016, 496/2010 and 354/2006	-	-	6.5-9.5 (LV)	1.1-5.0 (RV)	> 30 (RV)	10 – 30 (RV)
Water of the sample 6	CO ₂ 0.03 vol.%		6.99	1.225	33.7	9.4
Dolomite beneath 4 mm	10	1	7.40	1.285	36.3	9.2
	10	2	7.54	1.400	36.7	11.8
	10	4	7.65	1.460	37.9	12.5
	20	1	7.72	1.490	38.5	12.9
	20	2	7.69	1.500	38.1	13.4
	20	4	7.77	1.550	40.7	13.0
	100	1	7.87	1.615	46.1	11.3
	100	2	7.95	1.675	43.7	14.2
	100	4	7.90	1.725	44.7	14.8
Water of the sample 6	CO ₂ 0.03 vol.%		6.99	1.225	33.7	9.4
Dolomite + half-calcined dolomite (1 : 1)	10	1	8.47	1.755	30.0	24.5
	10	2	8.46	1.700	26.5	25.3
	10	4	9.07	1.750	25.1	27.4
	20	1	8.48	2.185	23.7	38.8
	20	2	8.41	2.240	22.7	40.7
	20	4	8.58	2.595	11.4	56.2
	100	1	9.52	2.670	13.6	56.7
	100	2	9.83	3.200	11.2	71.0
	100	4	10.16	3.300	8.0	75.4
Water of the sample 6	CO ₂ 0.03 % obj.		6.99	1.225	33.7	9.4
Half-calcined dolomite beneath 8 mm	10	1	8.54	2.120	25.9	35.9
	10	2	8.46	2.145	29.9	34.0
	10	4	8.47	2.165	10.0	46.6
	20	1	8.44	2.140	25.1	36.8
	20	2	8.40	2.180	30.1	34.8
	20	4	8.70	2.425	8.2	54.0
	100	1	8.95	4.215	15.4	93.1
	100	2	9.06	4.245	12.4	95.7
	100	4	9.17	4.505	11.6	102.5

Tab. 13
The static mineralization tests of water sample 6 at 20-22°C (differences)

Sample type	Quantity (kg/m ³)	Duration (hour)	pH	Ca and Mg (mmol/l)	Ca (mg/l)	Mg (mg/l)
Water sample No. 6	CO ₂ 0.03 vol.%		6.99	1.225	33.7	9.4
Dolomite with grain-size beneath 4 mm	10	1	+ 0.41	+ 0.060	+ 2.6	- 0.2
	10	2	+ 0.55	+ 0.175	+ 3.0	+ 2.4
	10	4	+ 0.66	+ 0.235	+ 4.2	+ 3.1
	20	1	+ 0.73	+ 0.265	+ 4.8	+ 3.5
	20	2	+ 0.70	+ 0.275	+ 4.4	+ 4.0
	20	4	+ 0.78	+ 0.325	+ 7.0	+ 3.6
	100	1	+ 0.88	+ 0.390	+ 12.4	+ 1.9
	100	2	+ 0.96	+ 0.450	+ 10.0	+ 4.8
	100	4	+ 0.91	+ 0.500	+ 11.0	+ 5.4
Water sample 6	CO ₂ 0.03 vol.%		6.99	1.225	33.7	9.4
Dolomite + half-calcined dolomite (1 : 1)	10	1	+ 1.48	+ 0.530	- 3.7	+ 15.1
	10	2	+ 1.47	+ 0.475	- 7.2	+ 15.9
	10	4	+ 2.08	+ 0.525	- 8.6	+ 18.0
	20	1	+ 1.49	+ 0.960	- 10.0	+ 29.4
	20	2	+ 1.42	+ 1.015	- 11.0	+ 31.3
	20	4	+ 1.59	+ 1.370	- 22.3	+ 46.8
	100	1	+ 2.53	+ 1.445	- 20.1	+ 47.3
	100	2	+ 2.84	+ 1.975	- 22.5	+ 61.6
	100	4	+ 3.17	+ 2.075	- 25.7	+ 66.0
Water sample 6	CO ₂ 0.03 vol.%		6.99	1.225	33.7	9.4
Half-calcined dolomite with the grain-size beneath 8 mm	10	1	+ 1.55	+ 0.895	- 7.8	+ 26.5
	10	2	+ 1.47	+ 0.920	- 3.8	+ 24.6
	10	4	+ 1.48	+ 0.940	- 23.7	+ 37.2
	20	1	+ 1.45	+ 0.915	- 8.6	+ 27.4
	20	2	+ 1.41	+ 0.955	- 3.6	+ 25.4
	20	4	+ 1.71	+ 1.200	- 25.5	+ 44.6
	100	1	+ 1.96	+ 2.990	- 18.3	+ 83.7
	100	2	+ 2.07	+ 3.020	- 21.3	+ 86.3
	100	4	+ 2.18	+ 3.280	- 22.1	+ 93.1

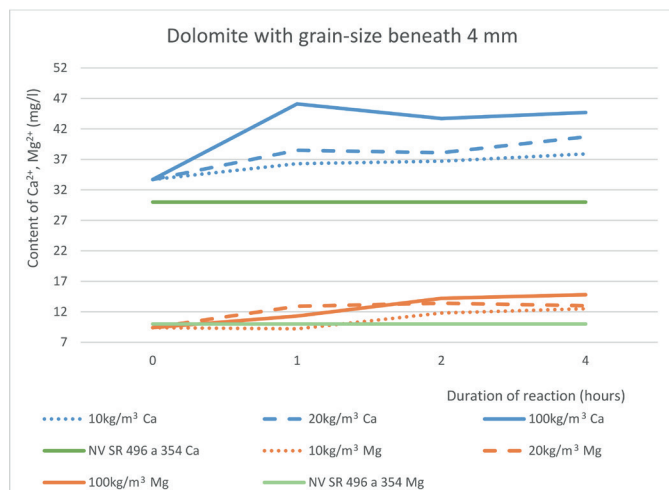


Fig. 1. The dependence of the release of Ca and Mg into the water of the *sample 6* from dolomite on its quantity and duration of the contact

duration of mineralization from 1.285 to 1.725 mmol/l (medium hard water).

After application of dolomite and HCD mixture in quantities of 10 kg/m³ (at duration of 1, 2 and 4 hours) and 20 kg/m³ (at duration of 1 and 2 hours), the water hardness was in the range of 1.700 to 2.240 mmol/l (medium hard water), at the amount of 20 kg/m³ (4 hours) and 100 kg/m³ (1, 2 and 4 hours), the water hardness varied from 2.595 to 3.300 mmol/l (hard water).

In the case of application of HCD in amounts of 10 and 20 kg/m³ of the water at all durations of mineralization, the water hardness was in the range of 2.120 to 2.425 mmol/l (medium hard water), at 100 kg/m³ of the water and all durations of the mineralization, the water hardness was 4.215-4.505 mmol/l (very hard water).

Tab. 13 shows that Ca and Mg contents - at the use of dolomite with the grain-size beneath 4 mm and quantities 10, 20 and 100 kg/m³ of the water - have increased in comparison with the values in the input *water 6* in the order: Ca increased in the range of 2.6 to 4.2 mg/l and Mg increased in the range of 0 to 3.1 mg/l at amount of 10 kg/m³; Ca increased from 4.4 to 7.0 mg/l and Mg increased from 3.5 to 4.0 mg/l at the amount of 20 kg/m³ and Ca increased from 10.0 to 12.4 mg/l, as well as Mg increased from 1.9 to 5.4 mg/l at the amount of 100 kg/m³ generally during all durations of static mineralization - Fig. 4.

Contents of Ca and Mg at application of dolomite and HCD in the ratio of 1 : 1 in the quantities of 10, 20 and 100 kg/m³ of the water were in comparison with the values in the input *water 6* in the order: Ca contents decreased at all doses from 3.7 to 25.7 mg/l, where the Mg contents increased significantly with an increasing amount, namely: Mg from 15.1 to 18.0 mg/l at the amount of 10 kg/m³; Mg from 29.4 to 46.8 mg/l at the amount of 20 kg/m³ and Mg from 47.3 to 66.0 mg/l at the amount of 100 kg/m³ generally during all static mineralization durations - Fig. 5.

The Ca and Mg contents at application of half-cal-cined dolomite with the grain-size beneath 8 mm in quantities 10, 20 and 100 kg/m³ of the water were in comparison with the values on input *water sample 6* in the order: Ca contents decreased at all doses of HCD in the range of 3.6-25.5 mg/l, while the magnesium contents radically increased with increasing amount of HCD: Mg was in the range of 24.6-37.2 mg/l at the amount of 10 kg/m³; Mg from 25.4 to 44.6 mg/l at the amount of 20 kg/m³ and Mg from 83.7 to 93.1 mg/l at the amount of 100 kg/m³ generally during all static mineralization periods - Fig. 6.

Table 14 reviews the differences in the values of selected qualitative indicators of water samples after 4 hour static water mineralization in comparison with values of the input (pre-mineralization) water of the *sample 6*.

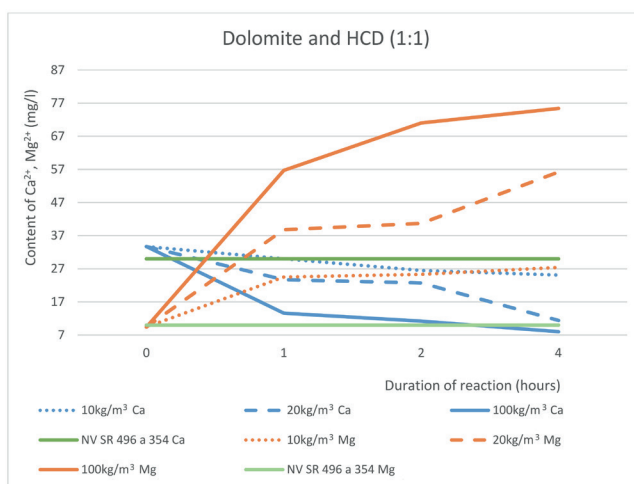


Fig. 2. Dependence of the release of Ca and Mg into water of *sample 6* from a mixture of dolomite and HCD on its quantity and contact time

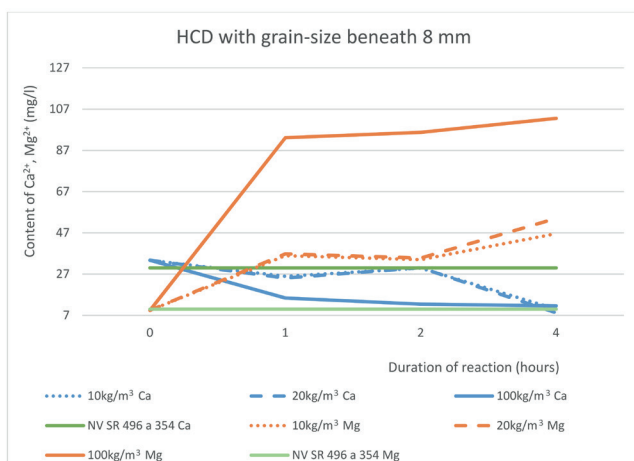


Fig. 3. Dependence of Ca and Mg release from HCD into the water of *sample 6* from its quantity and contact time.

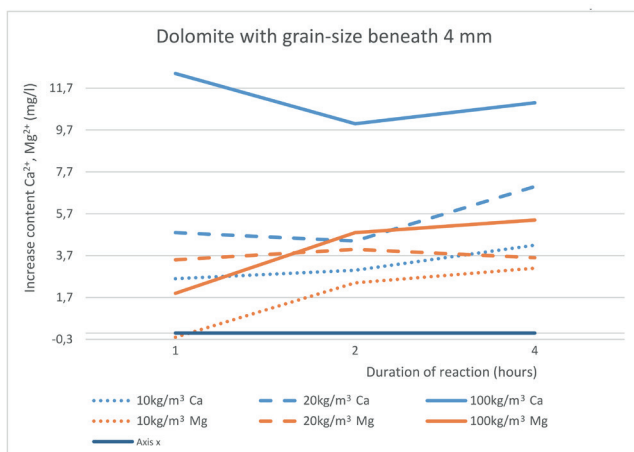


Fig. 4. The dependence of Ca and Mg increase in *water 6* from dolomite on its quantity and contact time.

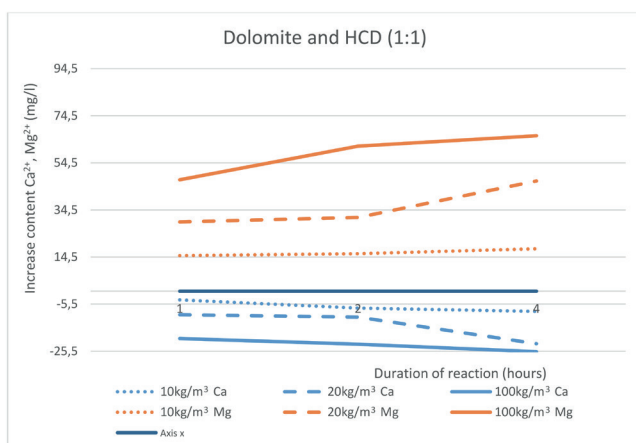


Fig. 5. The dependence of the increase of Ca and Mg in the water of *sample 6* on the quantity of the mixture of dolomite and HCD and on the contact time.

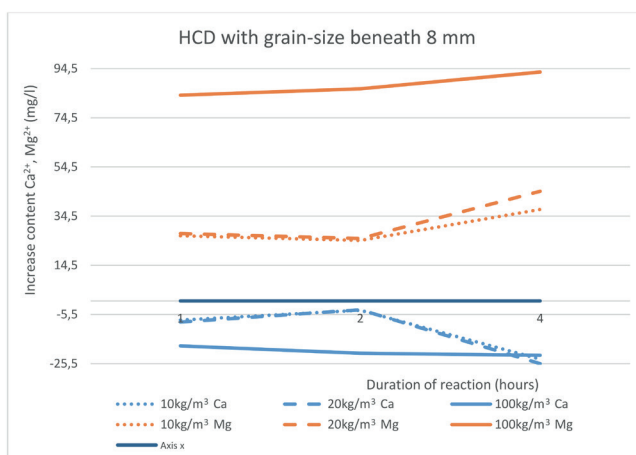


Fig. 6. Dependence of the increase of Ca and Mg in the water of *sample 6* on the quantity of the HCD and on the contact time.

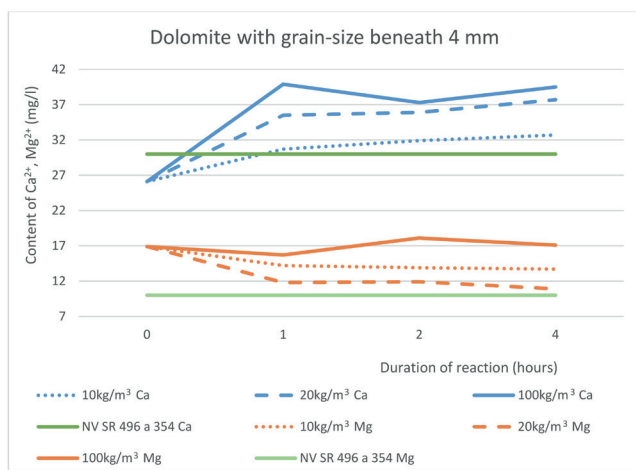


Fig. 7. Dependence of the release of Ca and Mg from the dolomite into *water 11* on its quantity and contact time

Table 14 shows that using the dolomite with the grain-size beneath 4 mm with its increasing quantity of 10, 20 and 100 kg/m³ after 4 hours lasting static mineralization of the *water 6*, the Ca and Mg contents increase, similarly at the water hardness in relation to the input *water 6* in the order for Ca: +4.2; +7.0 and +11.0 mg/l; for Mg: +3.1; +3.6 and +5.4 mg/l and for the water hardness: +0.235; +0.325 and +0.500 mmol/l.

When using a mixture of dolomite and HCD in the ratio of 1 : 1, with increasing quantity of 10, 20 and 100 kg/m³ after 4 hours of static mineralization of the *water 6* the content of Ca decreases in the following order: -8.6; -22.3 and -25.7 mg/l and significantly there increases the Mg content and water hardness in relation to input *water 6* in following order for Mg: +18.0; +46.8 and +66.0 mg/l and for water hardness: +0.525; +1.370 and +2.075 mmol/l.

When applying the HCD with the grain-size beneath 8 mm, its increasing amount of 10, 20 and 100 kg/m³ after 4 hour static *water 6* mineralization, the Ca content depletes in the order: -23.7; -25.5 and -22.1 mg/l, whereas the Mg content and water hardness distinctly increase in relation to input *water 6* in following order for Mg: +37.2; +44.6 and +93.1 mg/l, and for the water hardness: +0.94; +1.20 and +3.28 mmol/l.

Dynamic (flow) tests of water 6 mineralization in laboratory conditions

Table 15 states the results of dynamic mineralization of *water 6* in the flow regime in the glass column wide 4 cm in diameter and 13 cm high, having placed 150 g of appropriate type of carbonatic sample. This infill was penetrated in average with 1000 ml of the water during ca 2.5 hour during one test, which corresponds with the amount ~ 150 kg/m³ of the water.

Table 15 documents that at water flow of ca 7 ml/min., the application of dolomite has increased the hardness of mineralized water to 1.48 mmol/l (medium hard water) with increased contents of Ca (35.7 mg/l) and Mg (14.3 mg/l) with comparison to the input water. By increasing of the portion of HCD in the mixture with dolomite the water hardness usually increases, so in the ratio of dolomite and HCD of 1:2 the water is very hard (4.020 mmol/l), while the Ca content lowered to 20.6 mg/l and Mg content has substantially increased to a value of 85.2 mg/l in comparison with the values in the input water.

Using the HCD as well as the dolomite in the mixture with HCD, the Ca content lowered in comparison to the value in the input *water 6* to values within the range of 18.4-25.1 mg/l. On the other hand the Mg contents increased significantly after flow mineralization to the values within the range of 45.6 to 85.2 mg/l.

Tab. 14

Differences in the values of selected qualitative water sample indicators after 4 hours static mineralization of the water *sample 6* in comparison with values of the input water

Type of sample / sample quantity		pH	Hardness	Ca	Mg	Ca + Mg
(CO ₂ 0.03 vol.%)	kg/m ³	-	(mmol/l)	(mg/l)	(mg/l)	(mg/l)
Input water <i>sample 6</i>	-	6.99	1.225	33.7	9.4	43.1
Dolomite	10	+0.66	+0.235	+4.2	+3.1	+7.3
Dolomite + HCD – 1 : 1	10	+2.08	+0.525	-8.6	+18.0	+9.4
HCD	10	+1.48	+0.940	-23.7	+37.2	+13.5
Input water <i>sample 6</i>	-	6.99	1.225	33.7	9.4	43.1
Dolomite	20	+0.78	+0.325	+7.0	+3.6	+10.6
Dolomite + HCD – 1 : 1	20	+1.59	+1.370	-22.3	+46.8	+24.5
HCD	20	+1.71	+1.200	-25.5	+44.6	+19.1
Input water <i>sample 6</i>	-	6.99	1.225	33.7	9.4	43.1
Dolomite	100	+0.91	+0.500	+11.0	+5.4	+16.4
Dolomite + HCD – 1 : 1	100	+3.17	+2.075	-25.7	+66.0	+40.3
HCD	100	+2.18	+3.280	-22.1	+93.1	+71.0

Tab. 15

Dynamic (flow) tests of the water *6* mineralization at 20-22°C in the column.

Sample type	pH	Ca and Mg (mmol/l)	Type of water	Ca (mg/l)	Mg (mg/l)
Water <i>sample 6</i>	6.99	1.225	Soft	33.7	9.4
CO ₂ 0.03 vol.%	The column flow test with water sample No. 6				
6.667 ml/min; D; 150 g	8.42	1.480	Medium hard	35.7	14.3
6.850 ml/min; D : HCD = 2 : 1; 150 g	8.75	2.425	Medium hard	19.4	47.2
7.042 ml/min; D : HCD = 1 : 1; 150 g	8.86	2.500	Medium hard	25.1	45.6
6.579 ml/min; D : HCD = 1 : 2; 150 g	9.15	4.020	Very hard	20.6	85.2
7.042 ml/min; HCD; 150 g	9.20	3.270	Hard	18.4	68.3

Note: D – dolomite with grain-size beneath 4 mm; HCD – half-calcined dolomite beneath 8 mm

Static (non-flow) tests of water *sample 11* mineralization in laboratory conditions

Static tests of the water *11* mineralization in closed non-flow system were realized at the same laboratory conditions, as earlier with the water *6*. Their results are presented in tables 16, 17 and 18.

Table 16 manifests that the increased Ca contents (above RV), resp. recommended values of investigated water *11* after static mineralization in laboratory conditions were reached in comparison with the input sample with all amounts of dolomite with the grain-size beneath 4 mm for all durations of mineralization (contents of Ca 30.7-39.9 mg/l and contents of Mg 10.9-18.1 mg/l), while the Ca contents gradually increased with increasing amount of dolomite.

Using dolomite and HCD in the ratio 1 : 1 in amounts 10, 20 and 100 kg/m³ of the water, the Mg contents sig-

nificantly increased, i.e. 1.4-4.6 times (from 23.7 to 78.5 mg/l), generally at all durations of static mineralization, but at the expense of Ca contents, which varied beneath RV in the range of 8.8-27.7 mg/l. At application of D + HCD in the ratio of 1 : 1 and amount of 20 kg/m³ at the 4 hours duration of mineralization, resp. in amount of 100 kg/m³, there was at all durations of static mineralization exceeded the upper pH limit (pH nad 9.5) - Fig. 8.

At application of half-calcined dolomite with the grain-size beneath 8 mm in amounts of 10, 20 and 100 kg/m³ of the water, the Mg contents were significantly increased, i.e. 1.79 to 6 times (from 30.3 to 101.4 mg/l) generally at all durations of static mineralization, though at the expense of Ca contents, ranging beneath RV from 8.2 to 27.7 mg/l. At application of HCD during 2 and 4 hours and the HCD in amounts 20 and 100 kg/m³, the upper LV of water pH of the water was overreached (pH above 9.5) - Fig. 9.

Tab. 16
Static mineralization tests of the water 11 at 20-22°C

Sample type	Quantity (kg/m ³)	Duration (hod.)	pH	Ca and Mg (mmol/l)	Ca ²⁺ (mg/l)	Mg ²⁺ (mg/l)
RGoSR Nos. 8/2016, 496/2010 and 354/2006	-	-	6.5-9.5 (MH)	1.1-5.0 (OH)	> 30 (OH)	10 – 30 (OH)
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Dolomite with the grain-size beneath 4 mm	10	1	7.71	1.350	30.7	14.2
	10	2	7.40	1.365	31.9	13.9
	10	4	7.51	1.380	32.7	13.7
	20	1	7.83	1.370	35.5	11.8
	20	2	7.81	1.385	35.9	11.9
	20	4	7.63	1.390	37.7	10.9
	100	1	8.10	1.640	39.9	15.7
	100	2	8.00	1.675	37.3	18.1
	100	4	7.98	1.688	39.5	17.1
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Dolomite + half-calcined dolomite (1 : 1)	10	1	8.71	1.745	24.3	27.7
	10	2	8.47	1.800	25.9	28.1
	10	4	9.49	2.065	22.9	36.4
	20	1	8.94	1.660	27.5	23.7
	20	2	9.27	1.820	27.7	27.5
	20	4	9.64	2.250	25.1	39.5
	100	1	9.54	2.575	23.5	48.4
	100	2	9.64	3.365	10.0	75.7
	100	4	9.81	3.450	8.8	78.5
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Half-calcined dolomite with the grain-size beneath 8 mm	10	1	9.34	1.900	25.9	30.5
	10	2	8.92	1.935	27.7	30.3
	10	4	9.00	1.980	26.1	32.3
	20	1	9.42	1.920	24.6	31.7
	20	2	10.00	2.690	24.2	50.7
	20	4	10.10	2.720	25.1	50.9
	100	1	9.02	3.520	8.2	80.6
	100	2	9.90	3.575	10.2	80.7
	100	4	10.45	4.405	9.4	101.4

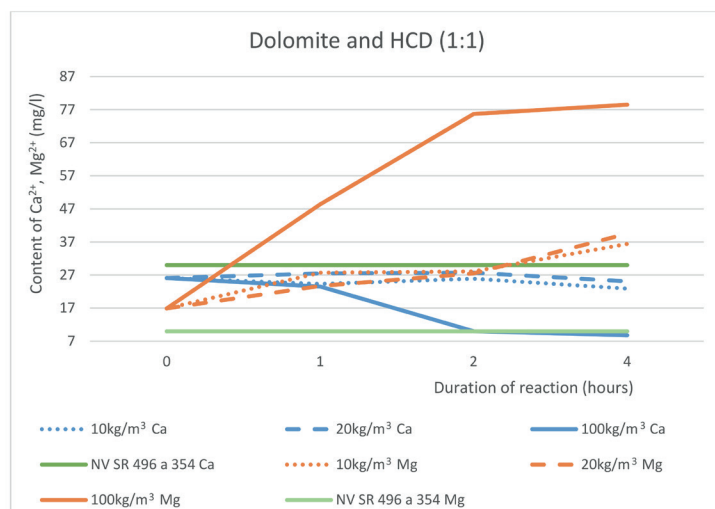


Fig. 8. Dependence of the release of Ca and Mg into water 11 from a mixture of dolomite and HCD on its amount and contact time.

The water *sample 11* with the hardness 1.345 mmol/l (value in the closeness of the lower boundary of medium hard water) was mineralized using all amounts of dolomite during 1, 2 and 4 hours by the way that the water hardness increased with increasing amount of dolomite and duration of mineralization process from 1.350 to 1.688 mmol/l (medium hard water).

At application of the mixture of dolomite and HCD in amounts of 10 and 20 kg/m³ (duration 1, 2 and 4 hours), the water hardness was in the range of 1.660-2.250 mmol/l (medium hard water), at the amount of 100 kg/m³ (1, 2 and 4 hours), the water hardness was from 2.575 do 3.450 mmol/l (hard water).

When applying the HCD in amounts of 10 kg/m³ (duration of mineralization process 1,

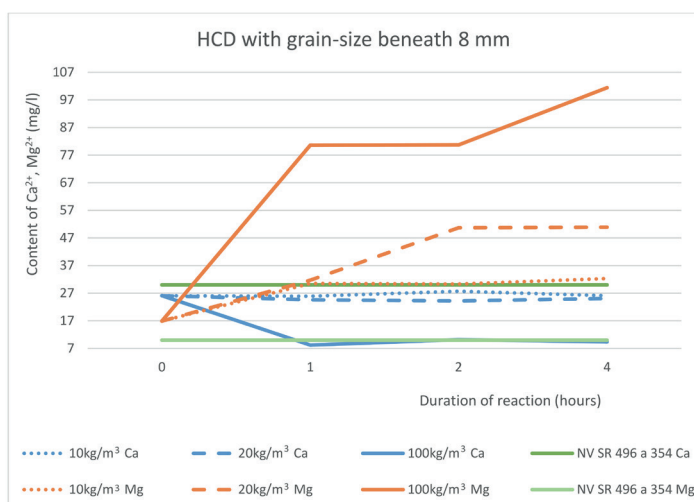


Fig. 9. The dependence of the release of Ca and Mg from HCD into the water *11* on its amount and contact time

Tab. 17
Static tests of mineralization of the water with designation 11 at 20-22°C (differences)

Sample type	Quantity (kg/m ³)	Duration (hour)	pH	Ca and Mg (mmol/l)	Ca (mg/l)	Mg (mg/l)
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Dolomite with grain-size beneath 4 mm	10	1	+ 1.39	+ 0.005	+ 4.6	- 2.7
	10	2	+ 1.08	+ 0.020	+ 5.8	- 3.0
	10	4	+ 1.19	+ 0.035	+ 6.6	- 3.2
	20	1	+ 1.51	+ 0.025	+ 9.4	- 5.1
	20	2	+ 1.49	+ 0.040	+ 9.8	- 5.0
	20	4	+ 1.31	+ 0.045	+ 11.6	- 6.0
	100	1	+ 1.78	+ 0.295	+ 13.8	- 1.2
	100	2	+ 1.68	+ 0.330	+ 11.2	+ 1.2
	100	4	+ 1.66	+ 0.343	+ 13.4	+ 0.2
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Dolomite + half-calcined dolomite (1 : 1)	10	1	+ 2.39	+ 0.400	- 1.8	+ 10.8
	10	2	+ 2.15	+ 0.455	- 0.2	+ 11.2
	10	4	+ 3.17	+ 0.720	- 3.2	+ 19.5
	20	1	+ 2.62	+ 0.315	+ 1.4	+ 6.8
	20	2	+ 2.95	+ 0.475	+ 1.6	+ 10.6
	20	4	+ 3.32	+ 0.905	- 1.0	+ 22.6
	100	1	+ 3.22	+ 1.230	- 2.6	+ 31.5
	100	2	+ 3.32	+ 2.020	- 16.1	+ 58.8
	100	4	+ 3.49	+ 2.105	- 17.3	+ 61.6
Water sample 11	CO₂ 0.03 vol.%		6.32	1.345	26.1	16.9
Half-calcined dolomite with the grain-size beneath 8 mm	10	1	+ 3.02	+ 0.555	- 0.2	+ 13.6
	10	2	+ 2.60	+ 0.590	+ 1.6	+ 13.4
	10	4	+ 2.68	+ 0.635	0.0	+ 15.4
	20	1	+ 3.10	+ 0.575	- 1.5	+ 14.8
	20	2	+ 3.68	+ 1.345	- 1.9	+ 33.8
	20	4	+ 3.78	+ 1.375	- 1.0	+ 34.0
	100	1	+ 2.70	+ 2.175	- 17.9	+ 63.7
	100	2	+ 3.58	+ 2.230	- 15.9	+ 63.8
	100	4	+ 4.13	+ 3.060	- 16.7	+ 84.5

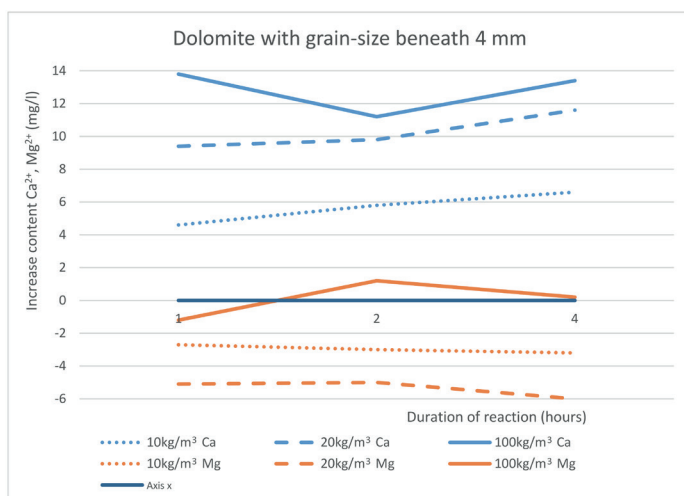


Fig. 10. The dependence of the increase of Ca and Mg from dolomite in the *water II* on its amount and contact time.

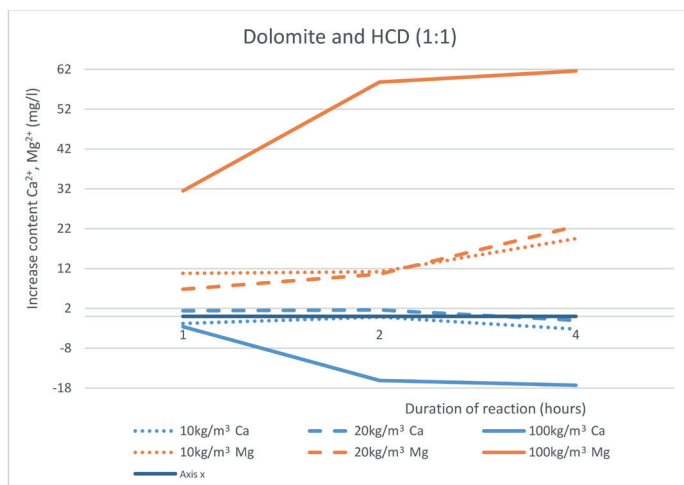


Fig. 11. The dependence of the increase of Ca and Mg from the mixture of dolomite and HCD in the *water II* on its amount and contact time.

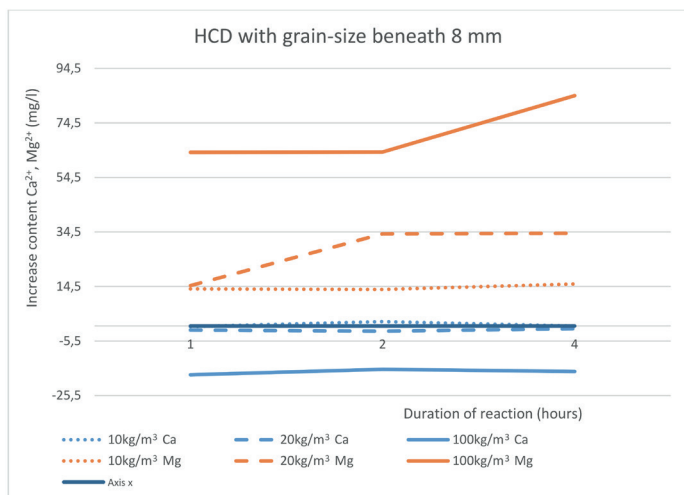


Fig. 12. Dependence of Ca and Mg increase in the *water sample II* from HCD, its amounts and duration of the mineralization process.

2 and 4 hours) and 20 kg/m³ of the water during 1 hour mineralization, the water hardness was within the range 1.90 to 1.98 mmol/l (medium hard water). At amounts of 20 kg/m³ (2 and 4 hours) and 100 kg/m³ of the water at 1 and 2 hours lasting mineralization, the water hardness was from 2.690 to 3.575 mmol/l (hard water). When applying HCD in quantity of 100 kg/m³ of the water and duration of static mineralization 4 hours the water hardness was 4.405 mmol/l (very hard water).

Table 17 presents that Ca and Mg contents - at the use of dolomite with the grain-size beneath 4 mm in amounts of 10, 20 and 100 kg/m³ of the water - were at comparison with the input *water II* in order: increase of Ca within the range of 4.6-6.6 mg/l and decrease of Mg within the range of 2.7-3.2 mg/l at amount of 10 kg/m³; increase of Ca from 9.4 to 11.6 mg/l and decrease of Mg from 5 to 6 mg/l at amount of 20 kg/m³, as well as increase of Ca from 11.2 to 13.8 mg/l and decrease of Mg by 1.2 mg/l (100 kg/m³, duration 1 hour), resp. increase of Mg from 0.2 to 1.2 mg/l in amount of 100 kg/m³ at duration of mineralization 2 and 4 hours - Fig. 10.

Contents of Ca and Mg at application of dolomite and HCD in a ratio of 1 : 1 in amounts of 10, 20 and 100 kg/m³ of the water were in comparison with the values in input *water II* in following order: Ca contents decreased at all dosings from 0.2 to 17.3 mg/l (except for the amount of mixture 20 kg/m³ and durations of 1 and 2 hours), while the Mg contents increased significantly with increasing amount of the mixture, namely: Mg from 10.8 to 19.5 mg/l at 10 kg/m³; Mg from 6.8 to 22.6 mg/l at 20 kg/m³ and Mg from 31.5 to 61.6 mg/l at 100 kg/m³ generally at all durations of static mineralization, but prevailing at duration of mineralization 2 and 4 hours - Fig. 11.

The Ca a Mg contents at application of the half-calcined dolomite with grain-size beneath 8 mm in amounts 10, 20 and 100 kg/m³ of the water were in comparison with values in the input *water II* in the order: Ca contents decreased at all amounts of HCD within the range of 0.2 to 17.9 mg/l (at amount of 10 kg/m³ they were from 0 to +1.6 mg/l at duration 2 and 4 hours), while the Mg contents radically increased with an increasing amount of HCD, namely: Mg in the range from 13.4 to 15.4 mg/l at 10 kg/m³; Mg from 14.8 to 34.0 mg/l at 20 kg/m³ and Mg from 63.7 to 84.5 mg/l at 100 kg/m³ generally at all durations of static mineralization - Fig. 12.

Table 18 reviews the differences in values of selected qualitative parameters of water samples

after 4 hours static mineralization in comparison to values in input water 11.

Table 18 documents that at application of dolomite with the grain-size beneath 4 mm with its increasing amount of 10, 20 and 100 kg/m³ after 4 hours of static mineralization of the *water 11* the Ca content and water hardness increased in comparison to input *water 11* in the order for Ca: +6.6; +11.6 and +13.4 mg/l and for the water hardness: +0.035; +0.045 and +0.343 mmol/l, while the Mg contents have decreased, resp. slightly increased in the order: -3.2; -6.0 and +0.2 mg/l.

Using the mixture of dolomite and HCD in a ratio of 1 : 1 with its increasing amount of 10, 20 and 100 kg/m³ after 4 hours static mineralization of the *water 11*, the Ca contents decreased in the order: -3.2; -1.0 and -17.3 mg/l. Simultaneously there significantly increased the Mg content and water hardness in comparison with the input *wa-*

ter 11 in the order for Mg: +19.5; +22.6 and +61.6 mg/l and water hardness: +0.720; +0.905 and +2.105 mmol/l.

Application of HCD with the grain-size 8 mm and its increasing amount 10, 20 and 100 kg/m³ after 4 hour lasting static mineralization of the *water 11* decreases the Ca content in the order: 0.0; -1.0 and -16.7 mg/l and distinctly increases the Mg content and the water hardness in comparison to input water sample 11 in the order for Mg: +15.4; +34.0 and +84.5 mg/l and for the water hardness: +0.635; +1.375 and +3.060 mmol/l.

Dynamic (flow) tests of mineralization of the water sample 11 in laboratory conditions

Table 19 presents the results of the dynamic mineralization tests of the *water 11* in the flow regime in laboratory conditions (the same as in the case of the *water sample 6*).

Tab. 18
The differences in the values of selected qualitative parameters of the water samples after 4 hours of lasting static mineralization of the *water 11*, compared to the input water values

Type of sample / sample quantity		pH	Hardness	Ca	Mg	Ca + Mg
(CO ₂ 0.03 % obj.)	kg/m ³	-	(mmol/l)	(mg/l)	(mg/l)	(mg/l)
Input water sample No. 11	-	6.32	1.345	26.1	16.9	43.0
Dolomite	10	+1.19	+0.035	+6.6	-3.2	+3.4
Dolomite + HCD – 1 : 1	10	+3.17	+0.720	-3.2	+19.5	+16.3
HCD	10	+2.68	+0.635	0.0	+15.4	+15.4
Input water sample 11	-	6.32	1.345	26.1	16.9	43.0
Dolomit	20	+1.31	+0.045	+11.6	-6.0	+5.6
Dolomite + HCD – 1 : 1	20	+3.32	+0.905	-1.0	+22.6	+21.6
HCD	20	+3.78	+1.375	-1.0	+34.0	+33.0
Input water sample 11	-	6.32	1.345	26.1	16.9	43.0
Dolomite	100	+1.66	+0.343	+13.4	+0.2	+13.6
Dolomite + HCD – 1 : 1	100	+3.49	+2.105	-17.3	+61.6	+44.3
HCD	100	+4.13	+3.060	-16.7	+84.5	+67.8

Tab. 19
Dynamic (flow) tests of mineralization of the *water 11* at 20-22°C in column

Sample type	pH	Ca and Mg (mmol/l)	Type of water	Ca (mg/l)	Mg (mg/l)
Water sample No. 11	6.32	1.345	Medium hard	26.1	16.9
CO ₂ 0,03 % obj.	Kolónové prietokové skúšky s vodou 11				
6.944 ml/min; D; 150 g	7.94	1.375	Medium hard	27.3	16.9
6.667 ml/min; D : HCD = 2 : 1; 150 g	8.62	2.270	Medium hard	21.6	42.1
6.849 ml/min; D : HCD = 1 : 1; 150 g	8.75	2.505	Hard	20.2	48.6
7.143 ml/min; D : HCD = 1 : 2; 150 g	8.80	2.955	Hard	18.8	60.4
6.757 ml/min; HCD; 150 g	9.09	3.575	Hard	17.8	76.1

Note: D – dolomite with the grain-size beneath 4 mm; HCD – half-calcined dolomite with the grain-size beneath 8 mm

Table 19 demonstrates that at the water flow of ca 7 ml/min. with application of dolomite, the water hardness increased to 1.375 mmol/l (medium hard water) with slightly increased Ca content (27.3 mg/l) and former value of Mg (16.9 mg/l) in comparison to input water 11. The increasing to the HCD share in the mixture with dolomite, there as a rule increases the water hardness, so at the dolomite to HCD ratio 2 : 1 the water is medium hard (2.270 mmol/l), at other ratios and at pure HCD the water is hard (the water hardness increases from 2.505 to 3.575 mmol/l). The contents of Ca with increasing share of HCD in the mixture lower to values from 21.6 to 17.8 mg/l, while the Mg contents significantly increase from 42.1 to 76.1 mg/l in relation to input water 11.

Obtained results of static and dynamic mineralization of the water samples with designation 6 and 11 in laboratory conditions demonstrate that kinematics of the water mineralization process depends of the water flow, resp. its duration, on water temperature, input contents of Ca and Mg in the water, grain-size and the amount of added dolomite and HCD mixture in kg/m³ of the water and their mutual relationship.

Conclusion

The laboratory tests of mineralization of the water in static and dynamic conditions at atmospheric pressure of CO₂ with various carbonatic rocks (dolomite, limestone, magnesite and HCD) have manifested that there is possible significantly increase the content of Ca and Mg in the water to values fulfilling or even overreaching the values stated by the Regulation of the Government of Slovak Republic No. 354/2010 Coll. as amended by the Regulation of the Government of the SR No. 496/2010 Coll. for drinking water. The highest enrichment was obtained using HCD and it related to magnesium increase. However, the HCD application caused the decrease of the Ca content. Therefore, the limestone or dolomite must be also used. For water mineralization in home wells we recommend using a mixture of limestone and HCD, while the HCD content in the mixture should be about 30 %. No significant differences were found from the point of view of the grain-size of the carbonate rock used. The Ca and Mg contents increase with longer duration of mineralization process. The recommended amount of added rock is from 20 to 100 kg.m⁻³ of the water.

The controlled addition of carbon dioxide to waters with deeply deficient calcium and magnesium contents when using dolomite (or half-calcined dolomite) at the amount of 20 kg/m³ causes a significant increase in the hardness and Ca and Mg concentrations in a relatively short time (up to 5 minutes) in mineralized water up to / above the recommended values according to the valid legislation.

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Zvýšenie kvality pitnej vody pridaním karbonátových hornín do nízko mineralizovanej podzemnej vody: návrh riešenia pre okres Krupina

V rámci riešenia projektov GEOHEALTH (www.geology.sk/geohealth) a LIFE FOR KRUPINA (www.geology.sk/lifeforkrupina) sa zistilo, že zdravotný stav obyvateľov Slovenskej republiky okresu Krupina je jeden z najnepriaznivejších v rámci celej Slovenskej republiky. Prejavuje sa to hlavne zvýšenou úmrtnosťou na kardiovaskulárne a onkologické ochorenia. Zvýšenú úmrtnosť spôsobuje hlavne nízko mineralizovaná podzemná voda s nízkym obsahom vápnika a horčíka, používaná na zásobovanie obyvateľov okresu Krupina pitnou vodou (Rapant et al., 2015, 2017). Obsah týchto dvoch dôležitých prvkov sa väčšinou pohybuje na úrovni odporúčanej hodnoty v NV č. 8/2016, NV č. 496/2010 a NV č. 354/2006 pre pitnú vodu, ale často je aj nižší ako odporúčané hodnoty. Hlavným cieľom realizovaného laboratórneho výskumu bolo navrhnuť optimálnu skladbu karbonátových hornín, ktoré by boli čo najvhodnejšie, ich optimálnu zrnitosť, potrebné množstvo a trvanie mineralizačného procesu s cieľom zvýšiť obsah Ca a Mg ich pridávaním do nízko mineralizovanej vody v individuálnych domových studniach v okrese Krupina.

V príspevku sú uvedené výsledky mineralizácie jednej vzorky *modelovej vody* a dvoch vzoriek reálnych vôd zo studní (*vzorky 6 a 11*) z okresu Krupina pri bežnom parciálnom tlaku vzduchu ($\text{CO}_2 \sim 0,03$ obj. %) a pri zvýšenej koncentrácii CO_2 vo vode v laboratórnych statických (neprietokových) a dynamických (prietokových) podmienkach s použitím karbonátových hornín (dolomit, magnezit, vápenec) a polovypáleného dolomitu (PVD) v množstve 10, 20 a 100 kg na 1 m³ vody pri trvaní procesu mineralizácie 1, 2 a 4 hodiny. Metodika mineralizácie vody bola v prvom štádiu overená na modelovej (destilovanej) vode aplikáciou **dolomitu**, **magnezitu** a **vápenca** pridávaných samostatne. Následne v druhom štádiu výskumu boli aplikované dolomit a PVD na dvoch reálnych vzorkách vody zo studní v okrese Krupina, a to samostatne aj v rôznych kombináciách, v statických (neprietokových) aj dynamických (prietokových) podmienkach.

Po mineralizácii *modelovej vody* (veľmi mäkká – destilovaná – voda s obsahom Ca 2,21 mg · l⁻¹ a Mg 1,34 mg · l⁻¹) v trvaní jednej hodiny **dolomitom**, **magnezitom** a **vápencom** samostatne v množstve 20 kg na 1 m³ vody pri pôsobení atmosférického CO_2 sa dosiahlo len malé zvýšenie kvalitatívnych ukazovateľov mineralizovanej vody. Dosiahnuté hodnoty tvrdosti vody boli v rozmedzí 0,315 až 0,405 mmol · l⁻¹ (veľmi mäkká voda) s obsahom Ca v rozmedzí 6,62 až 10,22 mg · l⁻¹, resp. Mg s obsahom v rozmedzí 1,94 až 5,84 mg · l⁻¹. Tým sa však nedosiahli odporúčané hodnoty (OH) podľa platných legislatívnych predpisov (nariadenia vlády č. 8/2016 Z. z., 496/2010 Z. z. a 354/2006 Z. z.). Veľmi mäkkú vodu – *modelovú vodu* – bolo možné efektívne mineralizovať v pomerne krátkom čase (do 5 minút) použitím dolomitu v množstve 20 kg na 1 m³ vody so súčasným riadeným dávkovaním CO_2 (zvýšenie koncentrácie CO_2 v upravovanej vode). Pri tom sa dosiahla tvrdosť vody 3,51 mmol · l⁻¹ (tvrdá voda) s obsahom Ca 35,3 mg · l⁻¹ a Mg 63,9 mg · l⁻¹.

Najpriaznivejšie výsledky po statickej mineralizácii reálnej vody vo vzťahu k vstupnej vzorke – *vzorka vody č. 6* zo studne na lokalite Kňazova dolina – s obsahom Mg nižším ako OH v laboratórnych podmienkach ($\text{CO}_2 \sim 0,03$ obj. %) boli dosiahnuté použitím dolomitu so zrnitosťou menšou ako 4 mm (10, 20 a 100 kg na 1 m³ vody) pri trvaní mineralizácie 1, 2 a 4 hodiny, pričom obsah Ca bol od 36,3 do 46,1 mg · l⁻¹ a Mg od 11,3 do 14,8 mg · l⁻¹. Výnimkou bola aplikácia dolomitu v množstve 10 kg na 1 m³ vody v trvaní jednej hodiny (obsah Mg vo vode bol nižší ako OH). Pri dynamických podmienkach mineralizácie *vzorky vody č. 6* aplikáciou dolomitu sa zvýšila tvrdosť mineralizovanej vody na 1,48 mmol · l⁻¹ (stredne tvrdá voda) so zvýšeným obsahom Ca 35,7 mg · l⁻¹ a Mg 14,3 mg · l⁻¹ oproti hodnotám vo vstupnej *vzorke vody č. 6*.

Zvýšený obsah vápnika (vyšší ako OH), resp. odporúčaný obsah Mg v skúmanej vode oproti vstupnej vzorke – *vzorka vody č. 11* zo studne na lokalite Hontianske Nemce – s obsahom Ca nižším ako OH bol po statickej mineralizácii v laboratórnych podmienkach ($\text{CO}_2 \sim 0,03$ obj. %) dosiahnutý pri všetkých množstvách dolomitu so zrnitosťou menšou ako 4 mm a pri všetkých časoch trvania mineralizácie. Obsah Ca bol od 30,7 do 39,9 mg · l⁻¹ a obsah Mg od 10,9 do 18,1 mg · l⁻¹, pričom obsah Ca s rastúcim množstvom dolomitu sa postupne zvyšoval. Pri dynamických podmienkach mineralizácie *vzorky vody č. 11* aplikáciou dolomitu sa vo vzťahu k vstupnej vzorke zvýšila tvrdosť mineralizovanej vody na 1,375 mmol · l⁻¹ (stredne tvrdá voda) s nepatrne zvýšeným obsahom Ca na 27,3 mg · l⁻¹ a pôvodnou hodnotou Mg 16,9 mg · l⁻¹.

Po zhrnutí výsledkov laboratórnych skúšok mineralizácie skúmaných vzoriek vody pri atmosférickom tlaku vzduchu (0,03 obj. % CO_2) sa ukazuje, že pokiaľ bude obsah Ca vo vode okolo 20 až 30 mg · l⁻¹ a Mg 5 až 10 mg · l⁻¹, potom na dosiahnutie odporúčaných hodnôt (Ca viac ako 30 mg · l⁻¹ a Mg od 10 do 30 mg · l⁻¹ v zmysle NV č. 8/2016 Z. z., 496/2010 Z. z. a 354/2006 Z. z.) po jej mineralizácii zmesou dolomitu a PVD je potrebné zastúpenie PVD v tejto zmesi minimálne 25 až 30 % pri celkovom množstve v rozmedzí od 20 do 100 kg na 1 m³ vody. Závisí to predovšetkým od jej prietoku (trvania mineralizácie) a teploty. Čím bude teplota vody vyššia a prietok vody nižší (trvanie mineralizácie dlhšie), tým budú výsledky jej mineralizácie priaznivejšie.

Riadená dotácia oxidu uhličitého do vody s hlboko podlimitným obsahom vápnika a horčíka pri použití dolomitu (prípadne PVD) v množstve 20 kg na 1 m³ vody spôsobí v pomerne krátkom čase (do 5 minút) podstatné zvýšenie jej tvrdosti a koncentrácie Ca a Mg v takto mineralizovanej vode až na odporúčané alebo vyššie hodnoty podľa platných predpisov a noriem.

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