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Effect of water flow characteristics on gypsum dissolution

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Abstract. Gypsum is one of the karstic rocks with many positive and negative characteristics. The most important defect of these rocks is solubility against water flow. The dissolution phenomenon in gypsum is accompanied by the release of the sulfate ion in water increasing its concentration, gradually. In this research, the effect of water flow temperature (T), Reynolds number (Re) and water head (H) on gypsum dissolution rate was evaluated, experimentally. In all of the experiments, gypsum samples were prepared approximately in the same dimensions and a circular hole was created inside the sample. Results showed that by increasing the temperature above $T/T_a = 1.30$ (T_a = temperature of the ambient air), the dissolution rate of gypsum was significantly increased. At the end of the experiment, the hole diameter (d) at the temperatures $T/T_a = 1.09$, 1.30, 1.52 and 1.74 has been increased about 90%, 137%, 141% and 148% from the initial hole diameter (d_i), respectively. The dissolution rate at $T/T_a = 1.09$ is about 50% less than the dissolution rate at temperatures $T/T_a = 1.52$ to 1.74. Moreover, the increasing rate of the outlet discharge for $H/d = 333$ is 1.89 times the increasing rate of the outlet discharge for $H/d = 167$.

1 Introduction

About 15% of the Earth's surface consists of the karstic rocks [1]. Due to complexity of Karst areas, these areas have been considered problematic sites for the construction of hydraulic structures like dams. Karstification is the process of the evolution of the voids within rocks due to dissolution. This dissolution leads to the evolution of a pipe system within the subsurface [2]. Details of the dissolution kinetics of limestone and gypsum were studied by Bock [3], Liu and Nancollas [4], Nonveiller [5], Martinez *et al.* [6], Dreybrodt and Eisenlohr [7], Romanov *et al.* [8] and Jeschke *et al.* [9]. The engineering conceptions of the karstic fractures have been investigated by James [10], Breznik [11] and Milanovic' [12]. The first models of one-dimensional (1D) fractures have been considered by James [10]. Also, some studies about 2D models have been performed by Dreybrodt and Siemers [13]. Among the soluble rocks, gypsum ($CaSO_4 \cdot 2H_2O$) dissolves rapidly in water flow and the cave systems responsible for the subsidence are constantly enlarging, causing a continuing subsidence problem. The equation for gypsum dissolution is as follows:

$$CaSO_4 \cdot 2H_2O \rightarrow Ca^{++} + SO_4^{--} + 2H_2O. \tag{1}$$
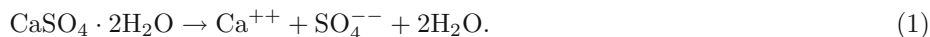
The calcium ion is usually combined with the impurities after the formation of the combination in water or soil, but the sulfate ion will remain suspended in water. The concentration of the sulfate ion will increase, gradually. The reason for this rise is not to be combined of the sulfate ion with the impurities inside the water. Because of the importance of dissolution, one of the reasons of dissolution of gypsum will be considered which the change in sulfate ion is. The rate of the dissolution of many substances is determined by the rate of transport of the soluble components across the boundary layer attached to the dissolving solid. The mathematical equation of the mass transport in the dissolution phenomenon is [14]

$$\frac{dM}{dt} = KA(c_s - c), \tag{2}$$

where, M is the mass dissolved in time t, A is the area exposed to the dissolution, c_s is the solubility of the material, c is the concentration of material in solution at time t and K is the dissolution rate constant. Equation (2) represents the dissolution kinetics of gypsum (calcium sulphate dihydrate), calcium carbonate and sodium chloride. However, anhydrite, in common with a few other soluble chemical compounds, is different.

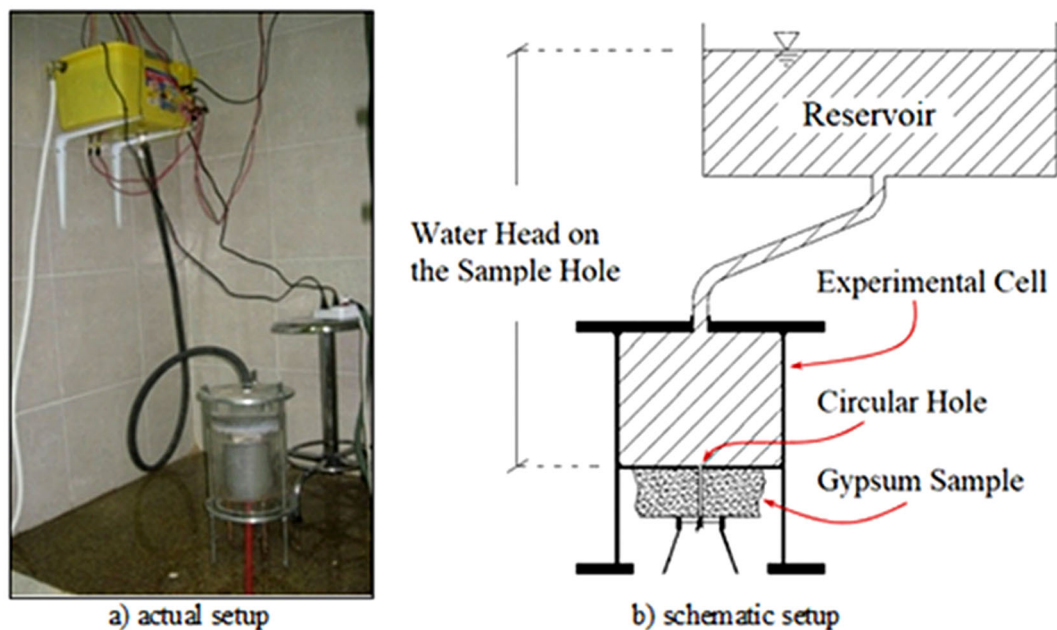
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Fig. 1. Experimental setup.

Numerous researchers, including Calcano and Alzura [15], Shaffer [16], James and Lupton [17], Gobran and Miyamoto [18] Gale [19], Palmer [20], Nafie [21], Klimchouk [22], Al-Mufty [23], Marion and Farren [24], Raines and Dewers [25], Dreybrodt *et al.* [26], Klimchouk and Andrejchuk [27], Andre and Rajaram [28], Hoxha *et al.* [29], Sadrekarimi *et al.* [30], Pfingsten *et al.* [31], Detwiler [32], Colombani [33], Burgos-Cara *et al.* [34], Kaufmann *et al.* [35], Reitman *et al.* [36], have studied the dissolution of the gypsum rocks. Bolan *et al.* [37] studied the dissolution of limestone and gypsum by using calcium and sulphate ions. They reported the differences in the rate of dissolution between the gypsum sources. Cooper and Calow [38] and Cooper [39] studied the dissolution rate of gypsum with a physical model. They used balls of gypsum and also, gypsum sample with a hole for evaluating of dissolution rate. Porter *et al.* [40] evaluated the gypsum dissolution technique in a steady-flow, a fluctuating-flow, and a mixed-flow environment by comparing the dissolution rate to direct flow measurements with an Acoustic Doppler Velocimeter (ADV). Colombani and Bert [41] used a physical model, characterized the microscopic transfer rate at the solid-liquid interface and showed that it was not an averaged value deduced from quantities measured far from the surface as in macroscopic-dissolution experiments. They found that the microscopic transfer rate is two times lower than the value obtained from macroscopic experiments. Al-Rawi *et al.* [42] investigated the influence of some parameters on the amount and rate of dissolution of gypseous rock samples. Their results showed that the rate of gypsum dissolution increased with increasing the water temperature, and increasing the flow velocity also increases the rate of gypsum dissolution. Also, they showed that the dissolution rate of gypsum depends to the different parameters including water temperature, flow velocity, hole diameter in the gypsum rock and water head. Behnamtalab [43–45] studied the dissolution rate of gypsum. Pachon-Rodriguez and Colombani [46] measured the dissolution of gypsum of various quarries in water and in aqueous solutions of salts inhibiting dissolution, and of the dissolution of anhydrite in water experimentally.

In contrast to, the previous studies, where the effects of different parameters on the dissolution of gypsum rocks were evaluated, in the present research, the effect of water flowing temperature, head and Reynolds number (Re) through gypsum on its dissolution rate are evaluated experimentally.

2 Experimental setup

Experiments were carried out in an experimental cell where the water flow enters the cell from a reservoir (fig. 1). For studying the dissolution rate of gypsum, the gypsum sample was placed in the experimental cell and water from the reservoir entered the experimental cell and passed through the hole created inside the gypsum sample.

The experimental cell was built from a Plexiglas cylinder and its top surface has been sealed with two metallic and rubber plates so that the pressured water enters the upper part of the cell without leaking from the sides. The cell internal diameter is 168 millimeters. The water from a specified level in the reservoir enters the experimental cell by a flexible tube. Also, the water surface level was constant during each experiment. The reservoir was used to produce
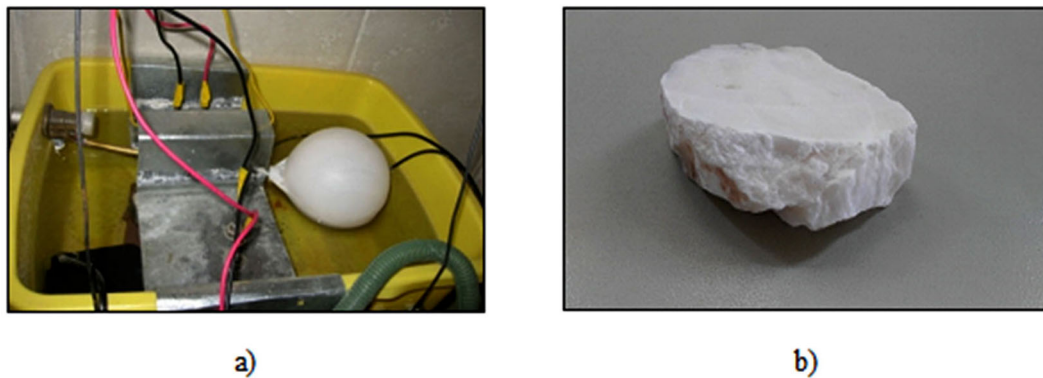
Fig. 2. Reservoir and gypsum sample.

water flow with the specified temperature and the specified water head. To do so, the four thermal elements were installed inside the reservoir by a thermostat which controls the reservoir water temperature. Because the elements warm only the environment around themselves, it was necessary to cycle the water inside the reservoir to distribute uniformly the water temperature. Therefore, two mixer devices are installed at two different levels and different directions inside the reservoir to turn the water inside the reservoir (fig. 2(a)). In this research, the gypsum samples were extracted from Hashemabad mine in Sabzevar city, in the east of Iran. The gypsum samples have 30 or 50 mm thickness and either a 2 mm hole or a 6 mm hole ($d = 2$ or 6 mm) created in the center. All samples were separated from a huge mass of gypsum rock. The dimensions of the samples, except for their thickness, had no particular shape, but the samples were cut so that they could be embedded inside of the experimental cell (fig. 2(b)).

The study of the literature shows that the different parameters including the velocity of the water, diameter of the fractures or holes in the karstic rocks, head, temperature, water density, viscosity and gravity acceleration are effective on the dissolution rate of the karstic rocks. By dimensional analysis, it was determined that non-dimensional parameters the Re (Reynolds number), H/d (head to hole diameter), T/T_a (water temperature to ambient air temperature) are more effective factors to the dissolution rate. Therefore, in this research, the effect of T/T_a, Re and H/d on the gypsum dissolution rate was studied. To do so, four different T/T_a, equal to 1.09, 1.30, 1.52 and 1.74, five different Re, equal to 500, 4200, 4800 and 6000, and two different H/d, equal to 167 and 333, were used.

By dissolution of the gypsum sample during the experiments, the hole diameter inside the gypsum sample will be increased and as a result, Re will be increased. In the experiments in which the effect of Re was studied, in order to keep this parameter constant during each experiment, based on the Reynolds equation ($Re = Vd/v$), because of increasing the d parameter, V should be decreased. Decreasing the velocity of the water flow inside the gypsum sample was performed by decreasing the outlet discharge from the gypsum sample. Therefore, after some minutes during the experiment, the outlet discharge from the gypsum sample was decreased by the control valve installed in the outlet flow to achieve desired Re. Moreover, the duration time of each experiment or total time (t_t) was considered to be 120 minutes from the entrance time of water into the gypsum sample.

It should be noted that in each experiment, the volume flow rate (Q) that exits from the hole of the gypsum sample was recorded in different times (t) during the experiment. Also, the gypsum sample weight and the hole diameter of the gypsum sample in the outlet surface were measured during the experiments. Also, the concentration of the sulfate ion in the water that passes through the sample was measured in the water temperature effect experiments. In order to determine the concentration of the sulfate ion in any experiment, three samples of water are extracted including the water before the experiment and the outlet water from the cell after 60 and 120 minutes. The concentration of the sulfate ion in the sample was measured by a spectrophotometer. The accuracy of the spectrophotometer that was used in this research was the maximum of 5% sulphate ion and 2 mg/l.

3 Results and discussions

3.1 Effect of water temperature

In the present work, the outlet velocity of the flow from the hole inside the gypsum sample on the bottom surface was calculated by the Bernoulli equation. Based on fig. 1(b), two points are selected including the water surface in the reservoir and the bottom surface of the sample to apply in the Bernoulli equation. The head loss (h_f) of the entering flow to the sample was also considered as $0.5V^2/2g$ [47], where V is the velocity in the hole inside the gypsum sample. The pressure and velocity at the water surface in the reservoir and the pressure at the bottom surface of the sample were assumed to be zero. By applying these values in the Bernoulli equation, the velocity in the hole inside the gypsum sample will be calculated.
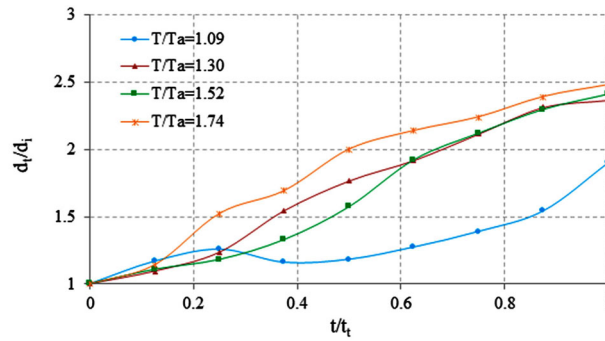
Fig. 3. Calculated average diameter of the hole at the different times and temperatures (d_t = theoretical diameter, d_i = initial diameter).

Table 1. Outlet discharge and the theoretical average diameter during the experiment. (Q_i = initial discharge).

t/t_t	$T/T_a = 1.09$		$T/T_a = 1.30$		$T/T_a = 1.52$		$T/T_a = 1.74$	
	Q/Q_i	d_t/d_i	Q/Q_i	d_t/d_i	Q/Q_i	d_t/d_i	Q/Q_i	d_t/d_i
0.000	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
0.125	1.36	1.17	1.20	1.09	1.23	1.11	1.30	1.14
0.250	1.58	1.26	1.53	1.23	1.39	1.18	2.32	1.52
0.375	1.34	1.16	2.38	1.54	1.77	1.33	2.87	1.69
0.500	1.40	1.18	3.12	1.77	2.47	1.57	4.00	2.00
0.625	1.62	1.27	3.68	1.92	3.69	1.92	4.57	2.14
0.750	1.93	1.39	4.47	2.11	4.49	2.12	5.02	2.24
0.875	2.38	1.54	5.35	2.31	5.27	2.30	5.72	2.39
1.000	3.60	1.90	5.60	2.37	5.80	2.41	6.15	2.48

The hole diameter increases by dissolution and as a result, the outlet discharge from the sample increases. In the experiments, the outlet discharge from the sample was recorded every 15 minutes. By using the outlet discharge, the theoretical average diameter (d_t) of the hole after the dissolution can be calculated. In table 1, the outlet discharge and the theoretical average diameter are presented every 15 minutes during the experiment. In addition, the variation of the average diameter of the hole at the different T/T_a and t/t_t during the experiments have been shown in fig. 3. Based on the figure, the hole diameter at $T/T_a = 1.09$ increases with lower rate than those at $T/T_a = 1.30$ to $T/T_a = 1.74$. At the end of the experiment, the hole diameter at $T/T_a = 1.09$, 1.30, 1.52 and 1.74 increased by 90%, 137%, 141% and 148% with respect to the initial hole diameter, respectively.

Based on table 1, the increasing of the outlet discharge after about $t/t_t = 0.250$ occurred. The increasing rate of the outlet discharge from $T/T_a = 1.30$ to $T/T_a = 1.74$ was higher than $T/T_a = 1.09$. For calculating the dissolution rate of gypsum, the dissolved volume of the gypsum sample was calculated based on the average diameter of the hole before and after the experiment in different times during the experiment (eq. (3)). Then, the average surface of the gypsum sample which subjected to the dissolution phenomenon was computed by eq. (4). Finally, the dissolution rate can be obtained by eq. (5):

$$\text{Dissolved volume} = \frac{\pi(d^2 - d_i{}^2)}{4} \times h, \tag{3}$$

$$\text{Average surface in exposure of dissolution} = \frac{(\pi d - \pi d_i) \times h}{2}, \tag{4}$$

$$\text{Dissolution rate} = \frac{\left(\frac{\text{Dissolved volume}}{\text{Surface in exposure of dissolution}}\right)}{\text{Time}}, \tag{5}$$

where d_i and d are the diameter of the hole in the gypsum sample before and after the experiment, respectively, and h is the thickness of the gypsum sample. Figure 4 illustrates the dissolution rate of the gypsum samples in different temperatures and times during the experiment.

As is shown in fig. 4, the dissolution rate for temperatures between $T/T_a = 1.30$ and $T/T_a = 1.74$ is closed to the one number while the dissolution rate is different at $T/T_a = 1.09$. The dissolution rate at $T/T_a = 1.09$ is about
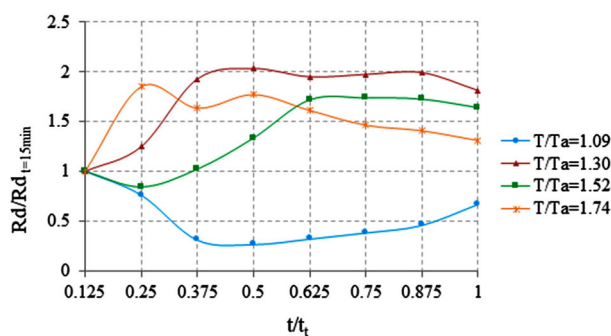
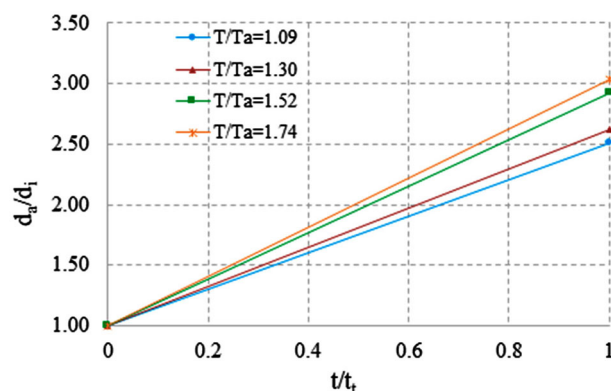**Fig. 4.** Dissolution rate at different times and temperatures.

Fig. 5. Actual diameter of hole at different times and temperatures (d_a = actual diameter).

50% of the dissolution rate for temperatures between $T/T_a = 1.30$ and $T/T_a = 1.74$. It shown that by increasing the water temperature from $T/T_a = 1.09$ to $T/T_a = 1.30$ or $T/T_a = 1.74$, the dissolution rate will be increased by two times. Also, according to fig. 4, the dissolution rates are very much different at the beginning of the experiment, but gradually, the rates are closed to a constant range and eventually closed to a constant value. These values of dissolution rates show that gypsum is very weak against water flow especially at high temperatures. The type of flow in all experiments is turbulent but Re was different along the experiments.

Results also showed that the higher water temperature caused that increasing of the hole diameter progress with a higher rate but gradually, these rates are closed to a constant value. The variation of the average of the actual hole diameter has been presented in fig. 5.

Figure 5 shows that the variation of the diameter at a temperature between $T/T_a = 1.30$ and $T/T_a = 1.74$ is more than $T/T_a = 1.09$. Also, at a temperature more than $T/T_a = 1.52$, the variation of the diameter of the hole can be negligible. The increment of d_a at $T/T_a = 1.09, 1.30, 1.52$ and 1.74 relative to the initial d (d_i) are about 151%, 162%, 192% and 203%, respectively.

In the present experimental research, the concentration of the sulfate ion of the water was measured at three times during of each experiment, in the beginning, middle and end. Figure 6 presents the variation of the concentration of the sulfate ion at different times and temperatures.

Based on fig. 6, the sulphate ion concentration for temperatures between $T/T_a = 1.30$ and $T/T_a = 1.74$ is close to the constant value. Also, according to the outlet discharge variation, the variation of the outlet discharge is almost the same at temperatures higher than $T/T_a = 1.30$. Another remarkable point in the graph is the rate of concentration change for sulfate ion in the temperatures $T/T_a = 1.30$ and $T/T_a = 1.52$ during the experiment. In fact, the slope of both curves is very much higher than the slope of the concentration curves at $T/T_a = 1.09$ and $T/T_a = 1.74$. It can be concluded that the variation rate of the sulfate ion concentration is very high from $T/T_a = 1.30$ to $T/T_a = 1.74$ but, the water temperature does not function as a very powerful catalyzer at other higher or lower temperatures.

3.2 Effect of Reynolds number and water head

In order to study the effect of the Reynolds number on the dissolution rate of gypsum, different Re were considered. The Reynolds numbers in the experiments are in the range from 500 to 6000 that cover laminar and turbulent flows.

As mentioned before, to keep Re constant, the outlet discharge from the gypsum sample changes. In fig. 7, the variation of the outlet discharge from the gypsum sample has been presented.
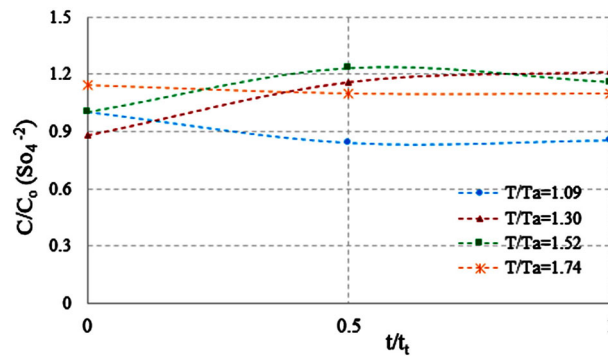
Fig. 6. Sulphate ion concentration at different times and temperatures.

a) H/d=167 (water head =1 m)

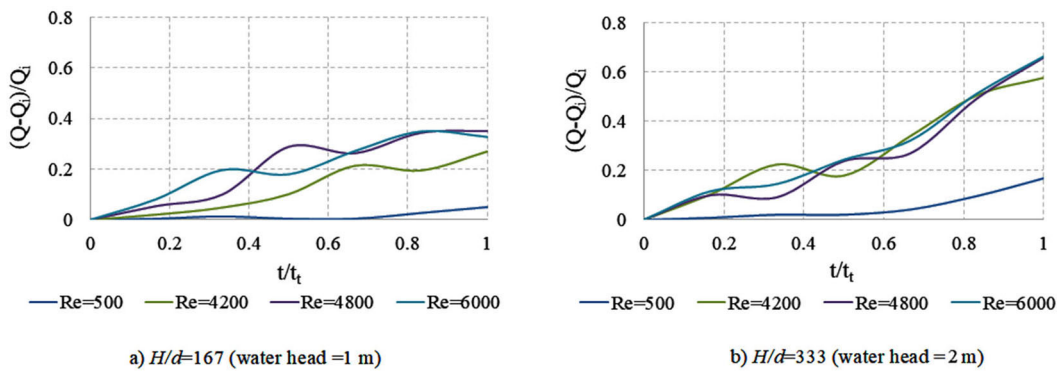b) H/d=333 (water head =2 m)

Fig. 7. Rate of discharge increasing for different Re and water head.

Based on fig. 7, the variation rate of the outlet discharge for laminar flow ($Re = 500$) is negligible while for $Re = 4200$, 4800 and 6000, the variation rate is considerable. The increase of the water head on the gypsum sample at the same Reynolds numbers causes the variation rate of the outlet discharge to increase. Figure 7 shows that the outlet discharge for constant Re is decreasing. Each of the curves in the above figure were fitted by a straight line. The slope of the lines for $H/d = 167$ and $Re = 500$ and 6000 were 0.0405 and 0.3398, respectively, and for $H/d = 333$ were 0.1497 and 0.6340, respectively. The comparison of the slope of the lines shows that by increasing Re, the variation rate of the outlet discharge will be increased. For $H/d = 167$ and $Re = 500$ (laminar flow), the increasing rate of the discharge relative to the initial discharge is 5% and for $H/d = 167$ and $Re = 6000$ (turbulent flow) it is 23%. Also, for $H/d = 333$ and $Re = 500$ (laminar flow), the increasing rate of the discharge relative to the initial discharge is 16%, and for $H/d = 333$ and $Re = 6000$ (turbulent flow)it is 49%. This shows that the regime flow change has a significant role on the increasing rate of the outlet discharge. The average slopes of the fitted straight lines slope at the different Re, for $H/d = 167$ and 333 are 0.262 and 0.495, respectively, which shows that the increasing rate of the outlet discharge for $H/d = 333$ is 1.89 times the increasing rate of the outlet discharge for $H/d = 167$. Also, this figure shows that the flow regime has a considerable effect on the outlet discharge from the gypsum sample. In fig. 7, along some curves, there are oscillations that are due to manual regulation of the outlet discharge by controlling the outlet valve.

The assessment of Re and the water head effect on the dissolution of gypsum can be performed by calculating the theoretical diameter (d_t) of the hole inside the gypsum sample. In the experiments, since Re is constant, the outlet velocity of the flow from the hole inside the gypsum sample will be calculated by Re. Then, by the discharge equation ($Q = VA$), the theoretical diameter of the hole will be computed. In fig. 8, the variation of the theoretical diameter of the hole has been presented for different Re and H/d.

According to fig. 8, for all Re, the theoretical diameter of the hole is increasing for all the duration of the experiment. Also, the water head on the gypsum sample has a significant effect on the variation rate of the theoretical diameter of the hole. The averages of the theoretical hole diameter for different Re at $H/d = 167$ and 333 are 0.122 and 0.219, respectively, which shows that the variation rate of the theoretical hole diameter for $H/d = 333$ is 1.80 times the variation rate of the theoretical hole diameter for $H/d = 167$.

To calculate the dissolution rate of gypsum, the dry weight of the gypsum sample before and after each experiment was measured. By using the density of the gypsum sample, the dissolved volume of the gypsum sample can be computed. The dissolution rate of the gypsum sample is calculated by dividing the dissolved volume by the dissolved area and the time duration of the experiment. The dissolved area is the area that is exposed to the dissolution phenomenon.
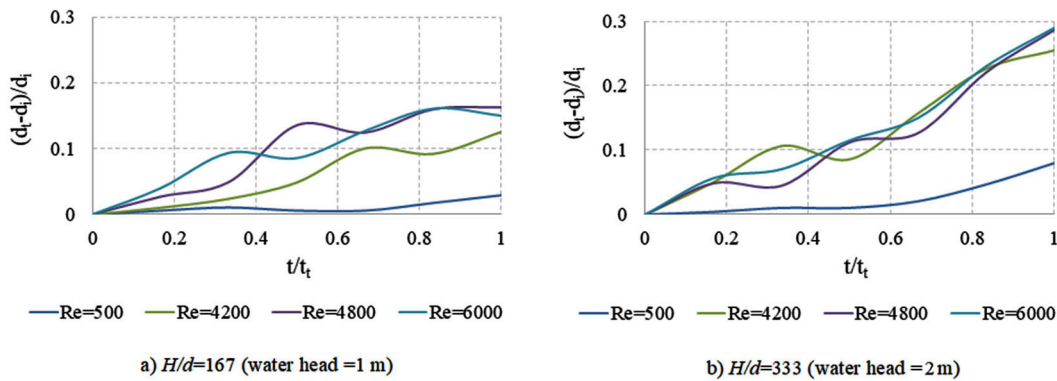
a) $H/d=167$ (water head $=1$ m)   b) $H/d=333$ (water head $=2$ m)

Fig. 8. Rate of theoretical hole diameter increasing for different Re and H/d.

Table 2. Characteristics of the gypsum sample before and after the experiment.

| H/d	Re	Sample length (mm)	Before the experiment		After the experiment		Dissolved volume (mm^3)	Dissolved Area (mm^2)
			d_i (mm)	W_i (gr)	d (mm)	W (gr)		
167	500	33.42	6.4	629.4	6.583	629.2	62	682
	4200	32.5	6.66	659	7.499	658.4	303	723
	4800	28.54	6.545	863.6	7.607	862.8	337	634
	6000	39.62	6.547	718.1	7.531	716.5	431	876
333	500	20.9	6.74	619.8	7.282	618.1	125	460
	4200	20.8	7.21	357.9	9.052	355.2	489	531
	4800	23.94	6.93	348.6	8.923	344.9	594	596
	6000	26.60	6.97	355.2	8.988	350.7	673	667
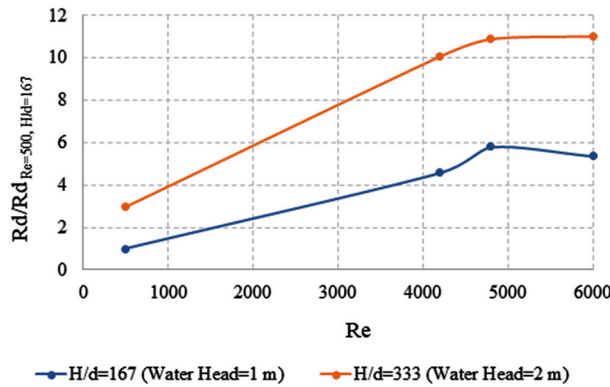
Fig. 9. Dissolution rate for different Re and H/d.

The dissolved area is computed by the average hole diameter before and after the experiment and thickness of the gypsum sample. In table 2, the characteristics of the gypsum samples in each experiment are presented. The density of the gypsum sample was $2.274\,\mathrm{gr/cm^3}$.

Figure 9 presents the dissolution rate of the gypsum sample for different Re and water head. Based on this figure, Re of the flow through the sample and the water head on the sample are considerable effect on the dissolution rate. For $H/d = 167$, by increasing Re, the dissolution rate increases mildly while for $H/d = 333$, by increasing the Reynolds number, the dissolution rate increases heavily. By increasing Re from 500 to 6000, for $H/d = 167$ and 333, the dissolution rate increases 4.81 and 8.86 mm/day, respectively, which shows that the head is very influential.

By changing the regime of the flow from laminar to transitional, the dissolution rate variation is negligible but by changing the regime of the flow from transitional to turbulent, the dissolution rate variation is considerable. One of the major changes of the flow characteristics after the regime changes is the viscosity of the fluid that depends on the Reynolds number. In the laminar flow, the viscosity is nearly constant but in the turbulent flow, a term of the turbulent viscosity is added to the total viscosity that changes with different parameters.

Also, fig. 9 shows that by increasing the water head on the gypsum sample, the dissolution rate increases for all Re.

4 Conclusions

The dissolution rate of the gypsum rock sample was evaluated experimentally for different temperatures, Reynolds numbers and water heads. The experiments were carried out using an experimental cell and a reservoir. The water flow passes through the hole created inside the gypsum samples that is embedded inside the experimental cell. The water temperatures in the experiments were $T/T_a = 1.09$, 1.30, 1.52 and 1.74, the Reynolds numbers were 500, 4200, 4800 and 6000, the normalized water head were $H/d = 167$ and 333. The results show that:

– At the end of the experiment, the hole diameter at the temperature $T/T_a = 1.09$ increases by 90% with respect to the initial hole diameter while at the temperatures $T/T_a = 1.30$, 1.52 and 1.74, the hole diameter increases by 137%, 141% and 148% with respect to the initial hole diameter, respectively.
– The dissolution rate at $T/T_a = 1.09$ is about 50% of the dissolution rate for the temperatures $T/T_a = 1.30$ to $T/T_a = 1.74$.
– The variation rate of the sulfate ion concentration is very high from $T/T_a = 1.30$ to $T/T_a = 1.74$, but the water temperature does not function as a very powerful catalyzer at other higher or lower temperatures.
– The Reynolds number of the flow through the sample and the water head on the sample have a considerable effect on the dissolution rate.
– For $H/d = 167$ and $Re = 500$ (laminar flow), the increasing rate of discharge relative to the initial discharge is 5% and for $H/d = 167$ and $Re = 6000$ (turbulent flow) is 23%. Also, for $H/d = 333$ and $Re = 500$ (laminar flow), the increasing rate of discharge relative to the initial discharge is 16% and for $H/d = 333$ and $Re = 6000$ (turbulent flow) it is 49%.
– The increasing rate of the outlet discharge for $H/d = 333$ is 1.89 times the increasing rate of the outlet discharge for $H/d = 167$.
– The variation rate of the theoretical hole diameter for $H/d = 333$ is 1.80 times the variation rate of the theoretical hole diameter for $H/d = 167$.
– By increasing Re from 500 to 6000, for $H/d = 167$ and 333, the dissolution rate increases 4.81 and 8.86 mm/day, respectively, which shows that the head is very influential.
– By changing the regime of the flow from laminar to transitional, the dissolution rate variation is negligible but by changing the regime of the flow from transitional to the turbulent, the dissolution rate variation is considerable. One of the major changes of the flow characteristics after the regime changes is the viscosity of the fluid that dependes on the Reynolds number. In the laminar flow, the viscosity is nearly constant but in the turbulent flow, a term of the turbulent viscosity is added to the total viscosity that changes with different parameters.

Notation list

A	Flow cross section	Q_i	Initial discharge
d	Hole diameter	Rd	Dissolution rate
d_i	Initial hole diameter	Re	Reynolds number
d_t	Theoretical hole diameter	t	Time
d_a	Actual hole diameter	T	Water temperature
g	Gravity acceleration	T_a	Ambient air temperature
H	Water head	t_t	Total time
h_f	Head loss	V	Velocity
Q	Discharge	υ	Kinematic viscosity

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