

Analysis of Geothermal Reinjection Problems with Hydrogeochemical Modelling

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ABSTRACT

Reinjection of thermal waters is a key parameter for sustainability of geothermal systems. However, injection can face difficulties due to several factors, e.g. mineral precipitation, fines migration or biological processes, especially in case of clastic reservoirs. These processes can clog the well screens and the pores of the reservoir, which can result in the decrease of injectivity. Mineral precipitation, which is the focus of this paper, is controlled by hydrochemical and physical changes. In this study, we focus on injection problems at the geothermal site in Mezőberény, SE Hungary.

A geochemical model was set up to simulate the geothermal reinjection processes and to model the saturation of the minerals during injection, using changing parameters: fluid composition, rock composition, air contact, injection temperature and injection pressure. For hydrochemical modelling we use PHREEQC Version 3 with the phreeqc.dat database. Based on the results general and site-specific conclusions could be drawn: In general, high concentrations of Fe and Mn content play a role at the injection problems at the Mezőberény site through precipitation of goethite, hematite and Mn-oxides. Furthermore, calcite is also oversaturated, therefore able to form carbonate-scaling. The geochemical model was validated with XRD analysis performed on scale samples from the Mezőberény site: precipitation contains goethite, calcite and magnesioferrite, which confirms our model.

Our findings include that air contact radically increases the saturation of Mn minerals, slightly enhance saturation of Fe minerals and decreases the saturation indices of carbonates. Lower injection temperature enhances the saturation of Fe-Mn mineral, in contrast to carbonates. Injection pressure has a negligible effect on the saturation of minerals.

The reinjection of heat depleted thermal water into sandstones has a relatively short history in Hungary. With two decades of experience and several successful projects in SE Hungary (Orosháza, Hódmezővásárhely, Szeged), reinjection is successful. Compared with these geothermal systems, saturation indices and the amount of possible precipitation are higher at Mezőberény. By preventing contact with air, iron and manganese minerals would be less saturated and the possibility of precipitation would decrease.

1. INTRODUCTION

Reinjection into geothermal aquifers, especially siliciclastic-porous ones, can face several difficulties. One of them is the clogging of aquifer pores and well screens by suspended solids, like fines, corrosion products and scales (Ungemach, 2003). Scaling processes originate from mineral precipitation as a consequence of changes in hydrochemical and physical conditions.

This study is carried out to achieve two goals: 1. Identify the influence of the hydrogeologic-hydrodynamic parameters in the geothermal utilization for the mineral precipitation through analysing the saturation indices in the geothermal systems. 2. Identify the reasons of the reinjection problems at the geothermal site in Mezőberény (SE Hungary) through hydrogeochemical modelling.

2. STUDY AREA

The geothermal potential of the Pannonian Basin is well known and its utilization is wide spread. One of the main aquifers is the Pannonian sandstones formed in the Miocene-Pliocene, filling the basin. Geothermal reinjection into the sandstones has a relatively short tradition in Hungary, with only several operating reinjection sites (Szanyi, 2010). The geothermal site in Mezőberény was constructed in 2012, with the aim to utilize thermal water for district heating. One production well (B-115) with a depth of 2003 m and one reinjection well (K-116) with a depth of 2001 m form the geothermal doublet. During a three-week operation of the cascade system in 2012, injectivity radically dropped and since then the operation has been stopped. In 2017, mechanical and chemical cleaning was performed to remove the clogging material from the injection well, which had no substantial effect. A long-term solution to increase injectivity has still to be found (Brehme et al., 2019).

Two additional operating geothermal systems were examined in this study as successfully working reference systems. The site of Hódmezővásárhely and Orosháza are situated 60 and 40 km SW of Mezőberény (Figure 1).



Figure 1: Location of the investigated systems in Hungary (modified from <http://ontheworldmap.com/>)

Their systems target similar geological formations and have ongoing reinjection operations since 1998 (Hódmezővásárhely) and 2011 (Órosháza). As these systems consist of more than two wells, for the comparison only one doublet was chosen. Table 1 shows the basic parameters of the doublets.

Table 2: Basic parameters of the investigated geothermal systems

	Mezőberény		Hódmezővásárhely		Órosháza	
Well type	Production	Injection	Production	Injection	Production	Injection
Number	B-115	K-116	B-1092	B-1094	B-770	K-775
Year of drilling	2011	2012	1996	1997	2004	2011
Well end depth (m)	2003	2001	2013	1685,5	1560	1558-1565
Reservoir rock	Pannonian Sandstone					
Screened reservoir depth (m)	1826-1947	1643-1931	1832-1997	1473-1669	1415-1553	1475-1533
Length of opened sections (m)	27.5	75.5	58.38	79.12	66	43
Reservoir temperature (bottom hole temperature) (°C)	109.7	111.3	84.2	72	101.2	104.5
Outflow temperature (°C)	76.9	83,2	73.5	61	88.2	84.5

3. DATABASE

For the hydrogeochemical modelling, the following data of the three systems has been used, mostly originating from hydrogeological logbook of the wells: water composition, separated gas composition, bottom hole temperature, injection temperature and injection pressure. In some cases, the inputs are more than 20 years old, therefore a change in chemical composition (e.g. dilution) is possible.

Although the geological setting is similar, the hydrochemistry differs clearly. In case of Mezőberény, TDS is 1,5 times higher (8120 mg/l) than in Órosháza (production well: 4800 mg/l). It is mainly caused by an increased amount of HCO_3^- (4470 mg/l), Na^+ (1860 mg/l) and Cl^- (268 mg/l). Fe content is also higher in Mezőberény (production well: 7,5 mg/l). Phenol indices are equally high in all the cases (6000 and 7200 $\mu\text{g/l}$). Table 2 and Table 3 contain the results of the obligatory fluid and gas analysis performed after well completion.

Table 2: Composition of fluids (sampled and analysed after the well completion; NM: not measured, ND: not detectable) (VITUKI, 1996; VITUKI, 1997; VITUKI, 2004; VITUKI, 2011; VITUKI, 2011; VITUKI, 2012)

		MEZŐBERÉNY		HÓDMEZŐVÁSÁRHELY		OROSHÁZA	
Sample		B-115	K-116	B-1092	B-1094	B-770	K-775
Well type		Production	Injection	Production	Injection	Production	Injection
Location of sampling		surface	surface	surface	surface	surface	surface
Flow rate during sampling	l/min	310	285	487	1600	650	1000
Sample date		2011 September	2012 June	1996 March	1997 March	2004 December	2011 April
pH	-	7,5	7,6	7,4	8,1	7.55	8.04
Electrical Conductivity (20°C)	[μS/cm]	5600	5360	3089	NM	3550	3830
Total Dissolved Solids - TDS	[mg/l]	8120	7120	3470	1370	4667	4800
T	[°C]	76.9	75	73.5	65.8	88.2	84.5
HCO ₃	mg/l	4470	4570	2440	920	2917	3221
F		1.7	1.56	3.99	0.47	NM	2.16
Cl		268	148	25	31	30	24
Br		2.2	2.6	<0.05	0	NM	0.29
I		2.9	2.6	0.073	NM	NM	0.32
NO ₃		<1	<0.02	<1	0	<1	<1
SO ₄		20	19	17	ND	38	28
Na		1860	1770	900	290	1574	1294
K		30	28	13	3.2	18.6	16
Ca		13.7	17.2	9	4.9	5.9	10.3
Mg		3.7	5.9	5.3	1.5	<3	7.6
Fe		7.5	4.8	0.35	0.25	0.356	0.176
Mn		0.1	0.09	ND	ND	<0.01	0.01
Li		0.55	0.49	NM	ND	NM	NM
Ba		1.98	1.99	0.083	NM	0.023	0.283
SiO ₂		79	79	52.5	52	78.6	51.6
NH ₄		24.4	28	15.5	8.4	18.9	18.5
TOC		1360	1260	NM	3.6	NM	290.43
Carbonate hardness	(CaO mg/l)	28	38	25	10	14	32
COD	(O ₂ mg/l)	66.8	63.1	21	3.6	NM	NM
Phenol index	μg/l	6000	5540	NM	NM	7.2	5.3

Table 3: Separated gas composition (%) (VITUKI, 1996; VITUKI, 1997; VITUKI, 2004; VITUKI, 2011 VITUKI, 2011; VITUKI, 2012)

	Mezőberény		Hódmezővásárhely		Orosháza	
	B-115	K-116	B-770	K-775	B-1092	B-1094
O ₂	0.73	0.72	0.54	4.7	2.6	1.25
CO ₂	37.71	14.29	54.62	33.44	33.33	3.01
N ₂	2.63	3.3	6.29	22.07	27.41	40.28
CH ₄	58.93	81.66	38.56	39.72	36.99	55.46

4. METHODS AND MODEL PROPERTIES

In this study, chemical data from fluids and solids have been combined in different hydrogeochemical models covering three geothermal sites in Hungary. Hydrogeochemical modelling was performed using Notepad++ PHREEQC with phreeqc.dat database (Parkhurst and Appelo, 2013). Considering the saturation indices ($SI = \log_{10}(IAP/K_{\text{mineral}})$ where IAP =product of ion activities and K =solubility constant for the mineral) in the output files, assumptions about the possible precipitations can be made: If SI is positive, the mineral will probably precipitate over time. We used the “precipitate_only” command, to obtain information about the quantity of the forming scales (mol/l).

To test real scenarios, different parameters were varied: fluid composition, possible air contact, injection temperature, injection pressure, rock composition (Figure 2.):

1. Parameter: Fluid (water and gas) composition can naturally differ strongly at the different sites and different wells
2. Parameter: Free contact to air can happen due to the set-up of the system. If an open buffer tank is used, and the fluid can interact with the air after production and before injection
3. Parameter: Injection temperature depends on the type and quantity of the heat utilization
4. Parameter: Injection pressure depends on the amount and flow rate of reinjected water and the settings of the injection pumps
5. Parameter: Exact mineral composition of the reservoir layer can differ in the reservoir, which can have an effect on the fluid composition and the mineral saturation

In order to represent the geothermal cascade from production horizon to reinjection of cooled water, the following set-up of the model was chosen (Figure 2.):

- Model step 1: First, the fluid composition from production well is defined (composition, pH) at different temperatures (70°C, 50°C, 30°C).
- Model step 2: Air is added as a gas phase in some cases, to simulate the contact of fluid with air at the surface.
- Model step 3: Fluids of the injection well (water and gas composition) and the minerals in equilibrium with the fluid are defined separately from the production fluid. Temperature of the second fluid is the reservoir temperature measured downhole in the injection well.
- Model step 4: Mixing of the two fluids is modelled at different pressures (15 atm, 8 atm, 1 atm) in ten steps, with variable temperature caused by the mixing). Result from progressed mixing state (MIX5) is considered with approximately 85% production fluid and 15% injection well solution.

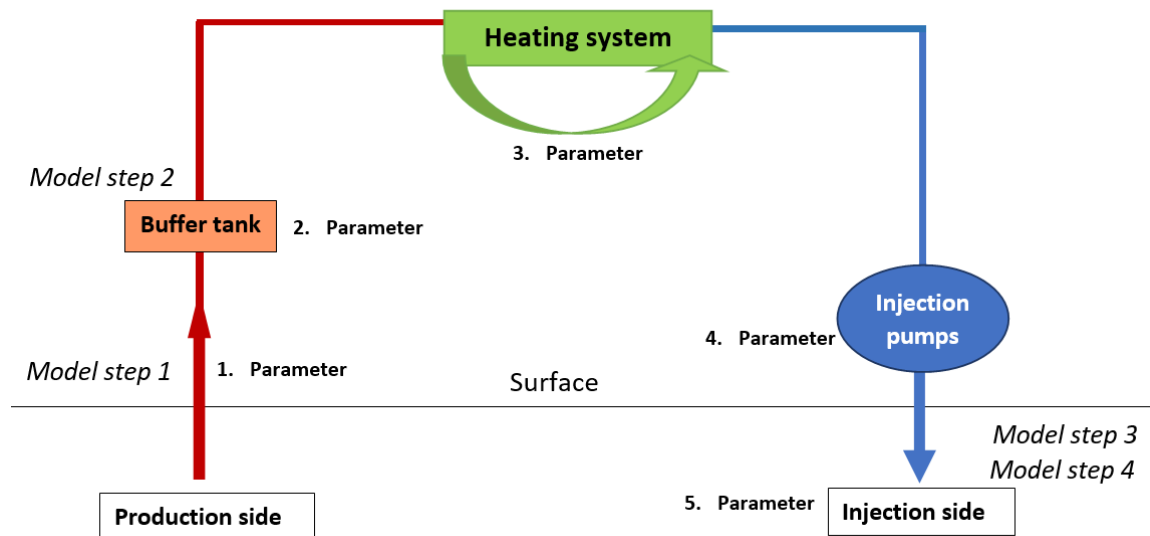


Figure 2: Schematic figure of the model layout

Reservoir rock minerals in this study are included as an ‘Equilibrium Phase’ because of the assumption that the reservoir fluid was able to solve all of the soluble minerals from the reservoir formation until equilibrium.

In case of Mezőberény no core samples were available. Thamóné Bozsó (2006) gives an average composition of the Újfalui and Zagyvai formation: quartz (SiO_2), K-feldspar (KAlSi_3O_8), chlorite (14A) ($\text{Mg}_5\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_8$), K-mica ($\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$), pyrite (FeS_2), goethite ($\text{FeO}(\text{OH})$). In GeoCom WP5.3 (2013) (project examining reinjection into sandstone) rock samples from the reservoir formation were examined on XRD and XRF analysis from similar depth. According to these measurements, the reservoir rock contains quartz, muscovite ($\text{KAl}_3\text{Si}_3\text{O}_{10}(\text{OH})_2$), dolomite $\text{CaMg}(\text{CO}_3)_2$, albite ($\text{NaAlSi}_3\text{O}_8$), chlorite, calcite and goethite. However, using the rock composition from literature (Thamóné Bozsó, 2006) the model did not reach acceptable calculation errors at reservoir circumstances. After several attempts, the composition from XRD analysis (mentioned above) with additional illite ($\text{K}_{0.6}\text{Mg}_{0.25}\text{Al}_{2.3}\text{Si}_{3.5}\text{O}_{10}(\text{OH})_2$) gave the fitting reservoir composition of quartz, K-mica, dolomite, albite, chlorite, calcite, goethite, and illite.

By modifying the basic model, the three geothermal systems were modelled assuming the scenario which is most close to the ‘real case’. Table 4 shows the basic input parameters.

Table 4: Parameters of the site specific modelling (*planned injection temperature)

	Mezőberény	Hódmezővásárhely	Orosháza
Injected fluid composition	B-115	B-1092	B-770
Reservoir fluid composition	K-116	B-1094	K-775
Injection temperature	50 °C*	20 °C	45 °C
Reservoir fluid temperature	111.7 °C	72 °C	104.5 °C
Air contact	yes	yes	no
Injection pressure	1 atm		
Reservoir mineral composition	quartz, K-mica, dolomite, albite, chlorite, calcite, goethite, illit		
Mixing state	MIX5		

5. RESULTS

The main results of the models are the saturation indices of the different minerals showing the possibility of precipitation and the quantity of the possible precipitated minerals. We examined the possibly precipitating minerals with a focus on their probability based on Corsi (1986) and Brehme (2018).

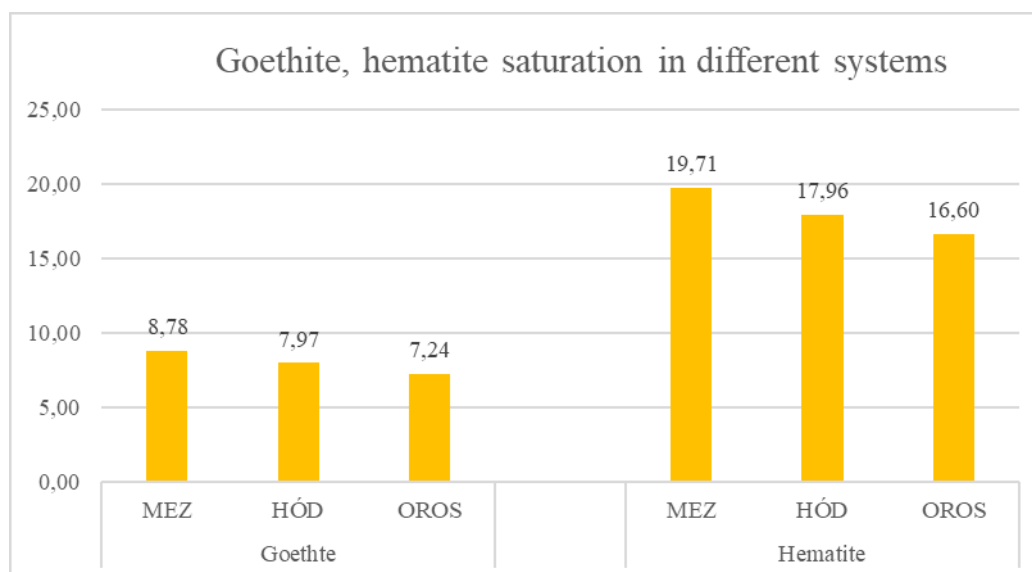
5.1. Influence of the injection parameters

Air contact enhance the saturation of Fe minerals on a small scale (increase with $\sim 0,3$), while Mn mineral saturation reacts radically into oversaturated state. For instance, using the Mezőberény fluid composition, saturation index (SI) of hausmannite (Mn_3O_4) changes from -9,90 up to 3,4. In contrast to that, carbonate saturation decreases with air contact. Saturation of amorphous silica (SiO_2) does not change with air contact, while the saturation index of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) is slightly higher. A reduce of injection temperature causes a decrease of the saturation of carbonates and hausmannite-pyrolusite (MnO_2), while the saturation of iron minerals and amorphous silica increases. The saturation of gypsum was not influenced by temperature decline.

According to the model, increasing injection pressure causes minimal differences in the saturation of the minerals. Changing injection pressure from 1 atm (default) to 15 atm leads to minimal changes with a magnitude of 0,01-0,03 SI. The influence of the reservoir mineral composition is more difficult to interpret. In presence of K-feldspar (which is a common component of sandstones) SI of goethite, hematite, amorphous silica and gypsum is decreased. With pyrolusite and manganite, manganese minerals have higher saturation.

5.2. Site specific results

Based on the modelling at the real case scenario (50 °C injection temperature, with air contact) in the Mezőberény geothermal system carbonate, iron and manganese minerals are in oversaturated state. In contrast to that, amorphous silica is slightly (SI = -0,43) and gypsum is highly undersaturated (SI= -4,49). Figure 3. shows SI of goethite and hematite; in both cases all the three systems are in an oversaturated state, but there is substantial difference with a highest saturation in Mezőberény. Saturation indices of calcite, aragonite and dolomite are plotted on Figure 4. In Hódmezővásárhely and Orosháza carbonates are in equilibrium or undersaturated state, while in Mezőberény carbonates are oversaturated with 0,42-1,07.

**Figure3: Saturation of iron minerals in different systems (MEZ=Mezőberény, HÓD=Hódmezővásárhely, OROS=Orosháza)**

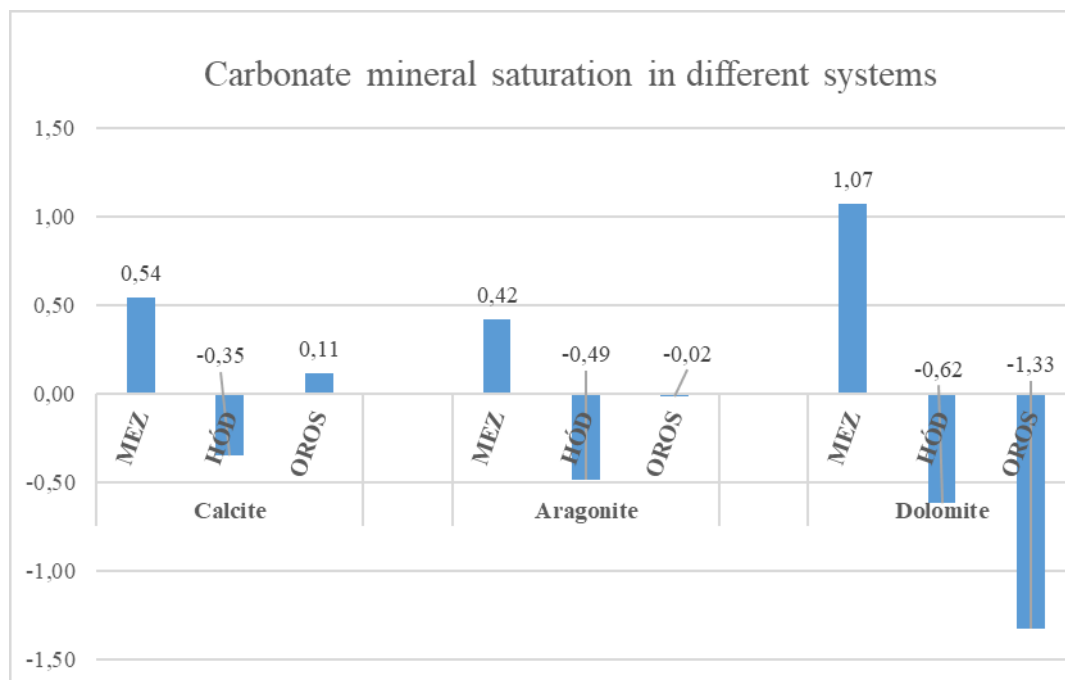


Figure 4: Saturation of carbonate minerals in different systems (MEZ=Mezőberény, HÓD=Hódmezővásárhely, OROS=Orosháza)

We revised the minerals with positive saturation indices, considering the quantity of the forming scale in mol/l (Figure 5). The results show similar trends as the qualitative approach but a more detailed analysis of on going processes: Generally, there are more precipitating minerals and the amounts of possibly forming scale are significantly higher in Mezőberény compared to Hódmezővásárhely and Orosháza. Calcite has the highest value of $1,165 \cdot 10^{-4}$ mol/l, followed by dolomite ($8,96 \cdot 10^{-5}$ mol/l), hematite ($5,738 \cdot 10^{-5}$ mol/l) and pyrolusite ($1,58 \cdot 10^{-5}$). In the other systems Hódmezővásárhely and Orosháza, only calcite shows a considerable amount of $2,838 \cdot 10^{-5}$ mol/l in Orosháza, hematite precipitates with $2,976 \cdot 10^{-6}$ in Hódmezővásárhely and with $2,708 \cdot 10^{-6}$ mol/l Orosháza.

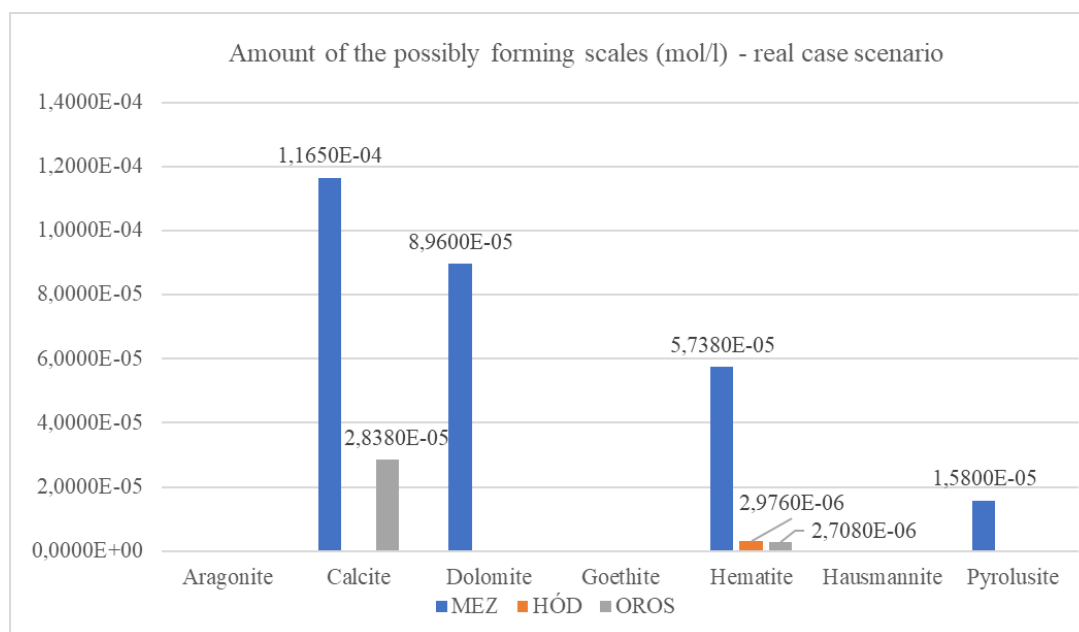


Figure 5: Amount of the possibly forming scale in mol/l (MEZ=Mezőberény, HÓD=Hódmezővásárhely, OROS=Orosháza)

5.3. Model validation

During other scientific studies of the project, five samples were taken from precipitation material from the short operation in 2012. According to the XRD analysis, samples contain goethite, calcite, magnesioferrite (MgFe_2O_4) (samples from normal operation) and siderite (FeCO_3) (samples from the well from the production test in 2017). Magnesioferrite can be a corrosion product, siderite can

be formed as a consequence of marl content in the reservoir near the screened sections. Presence of goethite and calcite does confirm our model calculations.

6. CONCLUSION

In this study, we examined geothermal reinjection problems due to mineral precipitation. We modelled the possible influence of changes in physical-hydrochemical conditions. In addition, the saturation state of minerals in Mezőberény was analysed and compared to two other successfully operating systems. Among the reasons, higher TDS and Fe, Mn, HCO_3 content could be identified. Considering these results, the following are recommended for the geothermal site in Mezőberény:

By preventing air contact, iron and manganese minerals would be less saturated and the possibility of precipitation would decrease. In addition, avoiding the degassing of CO_2 could prevent the decline of its partial pressure, which can help keeping the carbonates in solution. It could have an additional advantageous effect on microbial activity (not detailed in this study) as well, as oxygen enhances their activity too.

Increased injection temperature can reduce the saturation of the iron, and quartz as well, but based on the results, manganese minerals and carbonates would be more oversaturated.

For further advices, precipitated solids from recent operation stages should be analysed, and the operation could be adjusted to the results.

In addition to the engineering methods, application of scaling inhibitors is also a possibility to prevent the precipitation. Finding the proper inhibitor requires extended study to prevent the secondary effects.

The difficulties arising at the reinjection of geothermal fluids are of complex nature, inhibiting and monitoring scaling can contribute to the success of a long-term solution, in which hydrogeochemical modelling is a key method.

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