

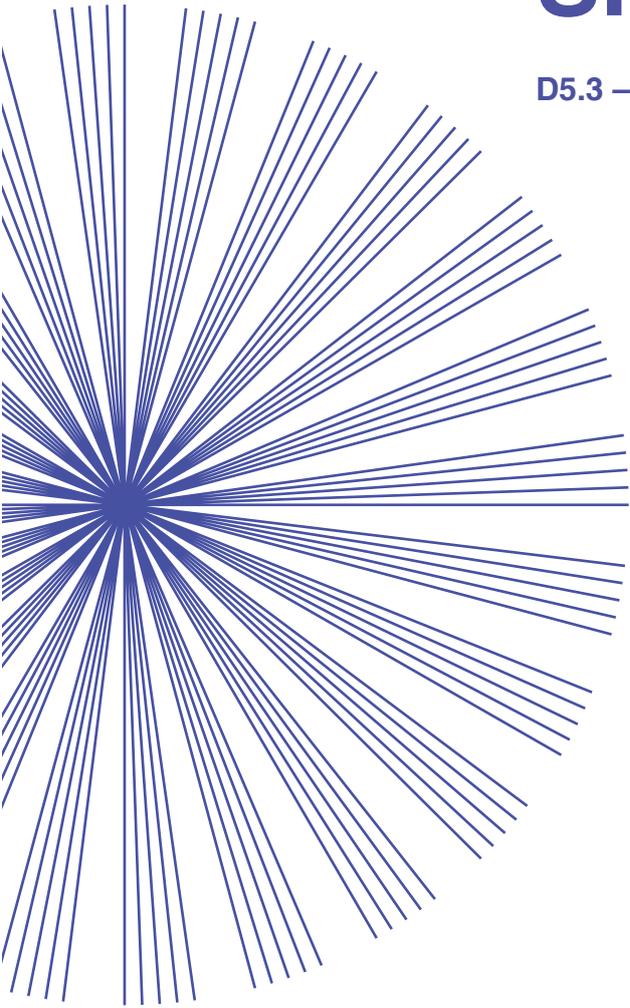
# Geothermal Communities

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GeoCom – FP7 CONCERTO – 239515

## REINJECTION TO SANDSTONE RESERVOIRS TECHNOLOGY SHOWCASE

D5.3 – Draft version



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GeoCom

WP5.3

Reinjection monitoring and modelling

Draft version

# **TOPICS OF REINJECTION MONITORING AND MODELLING**

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## 1. Introduction

Reinjection is a complex multi-parameter procedure used during the utilization of geothermal energy. Reinjection means the replacing of used geothermal fluids into geological formations.

The reinjection of water into Upper Pannonian sandstone layers at a pressure above 100 bars has a long history in the Hungarian oil industry. Therefore, this technology is uneconomical and unsustainable on the long term for thermal waters! The operation of the first economically sustainable thermal water reinjection well in Hungary began in 1998 in the geothermal system of Hódmezővásárhely. Since then, more than 2.8 million m<sup>3</sup> of water has been reinjected at a pressure of 2-5 bars.

Currently, there are a high number of geothermal fields with operating reinjection systems all over the world. However, due to its complexity, it cannot be considered a routine procedure since it requires serious expertise and continuous research. At the same time, a number of international and Hungarian examples show that reinjection can be realized by adequate operation and construction.

The objective of the current sub-programme is the collection of experiences related to the operation of thermal water production and reinjection systems, and the elaboration of dimensioning, construction and filtration methods of pairs and triplets of wells.

The purpose and favourable features of reinjection from the aspect of reservoir operation:

- The maintenance of reservoir energy, prevention of pressure draw-down
- Disposal of used geothermal fluids, prevention of surface heat and chemical pollution
- Ensuring the economical long-term use of the geothermal reservoir.

The production had to be reduced significantly in geothermal fields with no reinjection. Reinjection facilitates the maintenance of reservoir pressures and the extension of the lifetime of geothermal systems. Therefore, the reinjection of the recovered thermal water is a fundamental task.

The increase of the production potential was observed after the beginning of reinjection in case of several high-enthalpy vapour-dominated systems (Geysers, Larderello, Matsukawa etc.) (Stefansson, 1997)

In the geothermal fields of Tianjin and Beijing in the P.R. of China, the use of geothermal energy and the production of thermal water increased significantly since the 1980s. As a result, the water levels of reservoirs decreased drastically, and therefore, a significant part of recovered thermal waters has been reinjected in the past 10 years. Due to the reinjections, the water levels have become stable, and in some areas, they began to rise slowly (Kun et al, 2008).

Without reinjection, only a small part of the thermal energy in place in geothermal reservoirs can be recovered. Accordingly, the current objective is to use as much reinjected fluid in production as possible. The geological features of the reservoir, the location of and the distance between the wells play a major role in this. We can state that around 85% of the reinjected fluid can be recovered by a favourable geometric configuration. In the case of peripheral reinjection, we can calculate with a return rate of 30%. During reinjection, we increase the natural refilling of the system, thus we contribute to the better recovery of the reservoir's energy. (Stefansson, 1997)

### 3. Theory of thermal water reinjection, water treatment procedures

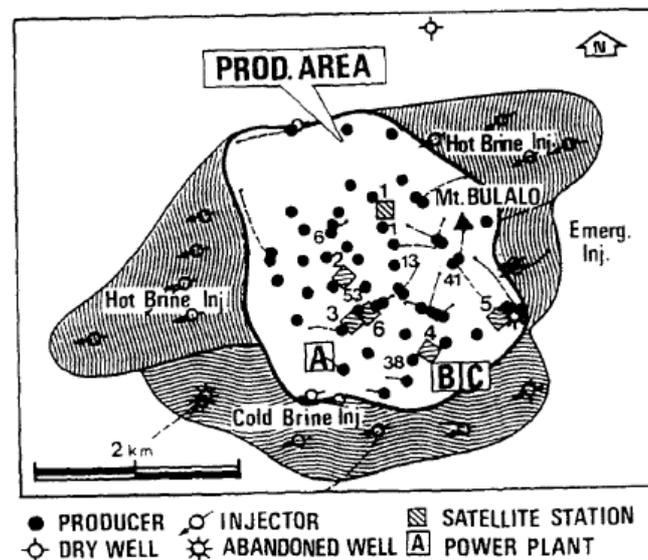
#### 3.1. International practices of reinjection

The detailed knowledge of the reservoir characteristics is necessary for the long-term maintenance of reservoirs and the practical implementation of the reinjection procedure. Therefore, the construction of pairs/triplets of production-reinjection wells must be preceded by accurate tests and modelling. The tests mean the investigation of the hydrodynamic characteristics of geothermal reservoirs. We must estimate the possible flowrate, drawdown, temperature, pressure, chemism and gas content of the water. (Seibt and Kellner, 2003)

The definition of the spatial relation of and the distance between the production and reinjection wells is one of the most critical tasks during the planning of reinjection. Currently, there is no universally accepted rule for the location of reinjection wells.

James (1979) defined the first rule of reinjection, according to which “production and reinjection wells are interchangeable”. Accordingly, there are no production and reinjection wells, only wells. This model assumes the uniform distribution of wells in the geothermal field. The development of this model would modify the production and reinjection wells in a way that they produce in the given period in certain parts of the reservoir and reinject in others, and the reinjection wells would be relocated after a certain amount of time.

Based on the relation of wells, we separate two basic types in case of a big number of wells. The first arrangement type includes production wells in the middle of the geothermal field, and reinjection wells on the periphery, e.g. Geysers (California) or the Bulalo (The Philippines) geothermal fields (fig. 1).



**Fig. 1:** The arrangement of production and reinjection wells in Bulalo. (Benavidez et al., 1988.)

The other type of arrangement consists of production wells surrounding several reinjection wells located in the middle of the geothermal field, e.g. the geothermal field of Larderello in Italy (Stefansson, 1997).

For the definition of the distance between production and reinjection pairs of wells, the following factors must be considered (Kun et al., 2008):

- The type of the geothermal reservoir
- Geological features of the geothermal field
- Reservoir permeability and thickness
- Fluid flow direction
- Temperature difference of the reservoir and the reinjected fluid
- Flowrate of reinjection.

According to Kun and his colleagues (2008), in case of low-enthalpy geothermal systems we do not have to worry about the cooling of the reservoir, if the distance between the production and reinjection wells does not exceed 100 m and the reinjection remains within a certain area. On the contrary, Antics's (2002) numerical modelling of low-enthalpy porous permeable sand/sandstone layers showed that the distance between the production and reinjection wells should be at least 500 m in order to avoid a thermal breakthrough.

We must note that the bigger the distance between the production and the reinjection well the higher will be the construction costs of the surface pipeline and the lower the rate of fluid reinjected into the production well. The prevention of a thermal breakthrough is the priority objective during the definition of distance.

Beside the definition of the distance between wells, the method of reinjection is worth defining as well, meaning either a direct reinjection into the production layer, or into a shallower layer in hydraulic connection with the production layer.

The task following the detailed planning of reinjection is the incorporation of geological information gathered during the drilling, and of geophysical, production and tracing test results into the theoretical/computer model created earlier.

Tracing tests are the most common means for the analysis of the communication between wells. These tests can be excellent indicators of a thermal breakthrough, since they provide information about the streamline and speed of the geothermal fluid between the production and reinjection wells. In case of fractured reservoirs, these tests define the volume of fractures. The results of tracing tests can be also used for the estimation of the reservoir cooling due to reinjection. (Kun et al., 2008):

Investigations into clastic sediments show that the potential injectivity index (the thermal water injection flowrate that is possible at a certain pressure) can be concluded from the results of the geological investigations and the production tests. (Seibt and Kellner, 2003)

The permanent monitoring is another important element of the practical implementation of reinjection during the operation of geothermal reservoirs. This must include the monitoring of the reservoir pressure, temperature, quantity of production, and the chemical features of the reinjected water. The objective of monitoring is to prevent an irreversible pollution of the well area and a thermal breakthrough, and to discover on time the changes in the geothermal system. The production and reinjection strategy can be modified according to the acquired data.

### Common difficulties and problems during reinjection

The fear of a thermal breakthrough was the most common argument against reinjection. The thermal breakthrough time is the time from initial injection until a significant cooling of the water observed in a producing well as a result of the activity of the reinjection well. (Mádlné, 2006) The length of the thermal breakthrough time is calculated with tracing tests and thermodynamic models. In case of tracing tests, it is important that a quick chemical breakthrough does not necessarily mean a quick thermal breakthrough as well. In general, there is a connection between chemistry and the thermal breakthrough in a form that the time of the thermal breakthrough is longer than the one of the chemical breakthrough. (Sigurdsson et al., 1995; Stefansson, 1997)

A real thermal breakthrough due to reinjected cold water has been documented only for a few geothermal fields. The enthalpy decline of the produced fluid is frequently connected to the thermal breakthrough. In fact, the enthalpy change is the result of the pressure change of the two-phase reservoir. The cooling of geothermal reservoirs cannot be considered a result of reinjection in either of the fields. Experience shows that a thermal breakthrough can be avoided with a flexible reinjection strategy, and it will apply only to certain pairs of wells, while the temperature drop will remain relatively small within the whole reservoir. (Stefansson, 1997)

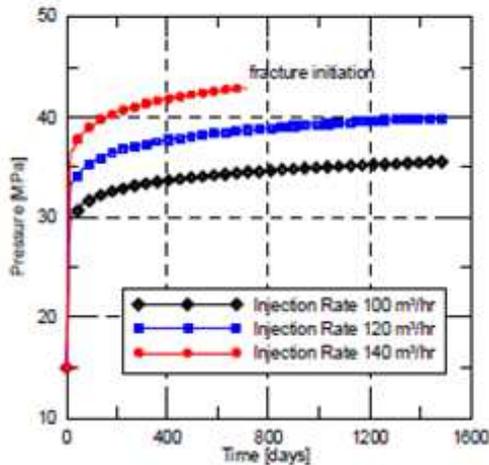
During the operation of the reinjection well, it is important to spare the well and to avoid sudden starts and stops. Otherwise, the formations near the filtered areas of the reinjection well can be ruined due to the sudden temperature and pressure drops. In the case of low solid, unconsolidated sandstones, the zone around the well is ductile, and it is characterized by the ruining of formations, the crash of the hole wall and the migration of solids (sanding). In solid sandstone reservoirs, fractures can develop due to the increased pore pressure and penetrate deep into the formation with the expansion of the cooled zone around the well. The development of fractures can accelerate the occurrence of thermal breakthroughs.

The stress conditions of the reservoir formation can change as a result of the increase of the pore pressure caused by reinjection. The part of the total stress carried by the solid rock matrix is called effective stress, and it represents normal stress minus the pore fluid pressure. Due to the increase of pressure, the centre of Mohr's circle shifts towards the origin. Furthermore, the envelope curve of the actual formation shifts towards the  $\sigma_n$  axis (figure 8) due to drop of the temperature.

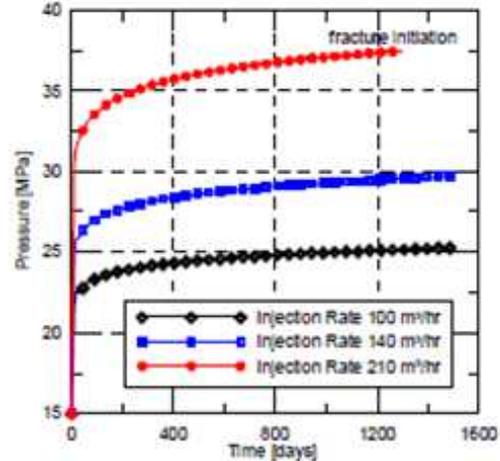
For the definition of the impact of reinjection on the mechanical behaviour of a porous sandstone body, Wojnarowski and Rewis (2003) have created a numerical model the mathematical formula of which contains the flow equations describing the mechanical behaviour of the fluid flow, thermal flow and the formation. They presented the different reinjection flowrates in relation to the time expired and the pressure according to the results of the model. The model also provided information about the fractures initiated in relation to days expired and the quantity of reinjected water in formations with different permeabilities. (Figures 9/a, b and c)

The results show that the lower the permeability of the porous formation the higher pressure is needed for the reinjection of the same quantity of water, and that fracturing is quicker by smaller flowrate as well.

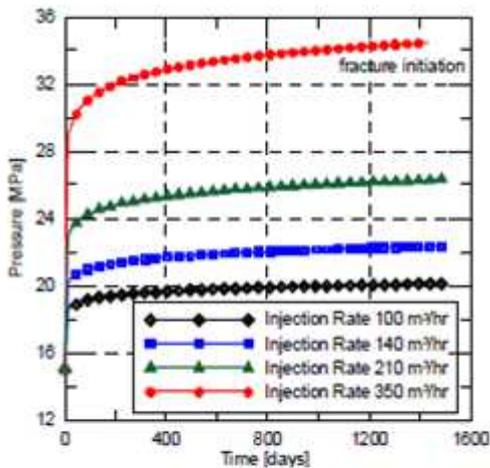
Fig. 9a. Simulated well pressure for three different injection rates and a rock permeability of 250 mD



9/a ábra: a szimulált kútnyomások 3 különböző visszasajtolási hozam mellett és 250mD permeabilitással (Wojnarowski és Rewis, 2003)



9/b ábra: a szimulált kútnyomások 3 különböző visszasajtolási hozam mellett és 500mD permeabilitással (Wojnarowski és Davis, 2003)



9/c ábra: a szimulált kútnyomások 3 különböző visszasajtolási hozam mellett és 1000mD permeabilitással (Wojnarowski és Davis, 2003)

Fig. 9c. Simulated well pressure for three different injection rates and a rock permeability of 1000 mD

We must special attention to the reinjection, as economic operation is guaranteed only when the thermal water can be reinjected over longer periods of time at little expenditure of energy, i.e. no blockings occur in the injection horizon.

In the case of low-enthalpy sandstone reservoirs, plugging can be caused by physical, chemical and biological factors. The reductions of the permeability (and injectivity) are

mainly due to solids in the water (Seibt and Kellner 2003; Ungemach 2003) that may originate from:

- Mobilisations of particles due to too high injection and production flowrates. The mobilisation of particles can be regarded as a mechanical process induced by the hydrodynamic force of the fluid. It has been demonstrated that there is a critical velocity above which the process is initiated, and this velocity depends on the salt content of the fluid. The higher the salt concentration of the injected fluid, the higher the critical velocity. The hydrodynamic force can reduce the permeability by 50%, but it is still much more insignificant than the chemical effect. (Ochi and Vernoux, 1998)
- The cooled-down injected fluid is chemically incompatible with the formation matrix and fluid. It is common that the reinjected fluid has a different chemical composition than the in-situ reservoir fluids. This chemical difference is rarely enough to influence the reinjection in a considerable manner in case the fluid is reinjected into the source reservoir. However, the thermodynamic changes (cooling, high pressures, degassing and related pH increase) of the heat-depleted brine may cause adverse thermochemical situations such as silica and carbonate scale (Vetter and Kandarpa, 1982a; Ungemach and Roque, 1988). In case the salt content of the reinjected fluid does not reach a critical salt concentration (CSC), clay particles may migrate at core constrictions causing permeability reduction. The critical salt concentration strongly depends on the nature and temperature of solute cations. (Khilar and Fogler, 1983). However, this theory applies to clean or almost clean waters, while reinjected thermal waters are mainly saline or brine.
- Oxidation and corrosion products from the transmission pipelines. Zn and Fe sulphides and oxides, silicate and quartz have been found near the reinjection wells of the Tianjin geothermal field, assumed to be parts of the pipeline. (Kun et al., 2008):
- Microbiological effects: Microbiological effects can be feared whenever the growth of sulphate reducing bacteria may be favoured by sulphate-rich formation waters and cold temperatures, a topic investigated on North Sea injector wells. Here, the damage occurs under the form of organic slime, material blocking pore entries. (Rosnes, 1990)
- Technical incompatibility.

The permeability reduction of well areas can begin during the drilling phase of reinjection wells due to the physical and chemical interactions between the components of the slush and the reservoir body. The suspended clay particles in the drilling fluid can significantly reduce the initial injectivity of the well due to the ingress of silt and clay particles stuck on the wall into the porous formation. The different chemical substances added to the drilling fluid can be thermodynamically instable and cause permeability reduction when reacting with each other or the reservoir body. (Vetter and Kandarpa, 1982b)

The permeability reduction of wells can be caused by suspended matters plugging the perforations and pore entries, their surface filtration can play a major role in the lifetime of reinjection wells.

A common argument against reinjection is that it significantly increases the investment and operation costs of the geothermal field. This is based on the obvious fact that the drilling of reinjection wells and the construction of the surface thermal network for wastewater delivery require a significant capital investment. This is true for short-term projects. However, this statement is not true for long-term projects. With a production of 10 to 20 years from a geothermal reservoir, it can be often seen that the quantity of the available water limits the quantity of extractable energy. Under these circumstances, reinjection will increase the lifetime of the reservoir and enable a more efficient energy extraction. (Stefansson, 1997)

In Hungary, the costs for the surface disposal of used thermal waters amount to approx. HUF 50/m<sup>3</sup> due to wastewater fines and contributions. In 2008, the reinjection costs of the geothermal system in Hódmezővásárhely amounted to HUF 31/m<sup>3</sup> (electric energy, surface filtering and maintenance costs, well maintenance). The reinjection in Hódmezővásárhely costed HUF 245 million, HUF 200 million of which being the price of the 2000-meter deep reinjection well. (Kurunczi, 2008)

It is important to mention that the economical use of a reinjection well on a long term requires the conservation of the well's direct geological environment. This means that the amount of the reinjected fluid above a certain limit can only be increased through the increase of the input energy and at the sacrifice of the formation's damaging, especially in the case of porous sandstone reservoirs. The maximum flowrate of reinjection of the geothermal system in Hódmezővásárhely is 40 m<sup>3</sup>/h, thus the quantity of recoverable water is also limited.

One production and two reinjection wells are required for an optimal thermal energy system, allowing the resting of reinjection wells, and the quantity of produced water would be less limited by reinjection in case of a simultaneous operation of the two reinjection wells, meaning that the profitability and return period of the system would improve. Accordingly, a proper operation would result in the return of the reinjection costs on a long term.

Currently, the only measurement results we have are the ones of the reinjection system in Hódmezővásárhely.

### Environmental aspects of reinjection

Geothermal energy is considered as an environmentally friendly energy. It has favourable indicators compared to fossil energy sources, e.g. in regard to the emission of greenhouse gasses. (Mádlné et al., 2008)

The surface disposal of used thermal waters can result in several environmental problems e.g. salt and heat load, disturbances in the water balance (decrease of the supply and reservoir pressure) etc. The used water is mainly discharged in public utility channels, on-surface open channels or rivers and living waters. (Kurunczi, 2008)

There are several examples for countries with coastlines to dispose the used thermal waters into the sea, e.g. in Ahuachapán (El Salvador) and Tiwi (The Philippines); however, these solutions were terminated on most locations in favour of long-term environmental objectives, and reinjection has been started. We must mention that there are cases when the surface disposal of used thermal waters had a positive impact on the environment. An

environmental impact assessment was composed before the operation of the geothermal field in Nesjavellir, Iceland, which showed that the surface disposal of thermal water near the Lake Thingvallavatn would improve the living conditions of the lake's fishes. During the operation of the geothermal field in Svartsengi, Iceland, a lake with considerable dimensions was created with the surface disposal of thermal waters, which has become a popular medicinal and recreational location. (Stefansson, 1997)

Reinjection is an excellent solution to avoid above-ground pollution and to maintain the reservoir energy. It is important for the reinjection to be carried out in a closed system and for the reinjected fluid to have a proper quality in order to avoid subsurface pollution.

The damage originating from suspended solid particles relates to particle and pore parameters (size, shape, concentration, tortuosity) and the various forces (hydrodynamic, retention) involved in the deposition, capture and release mechanisms.

The solid particles are retained on the surface of a matrix grain, are wedged between the convex surfaces of two grains, accumulated at the pore entries or they in pore "bellies" formed by several grains. The most important retention forces are the hydrodynamic forces of the fluid, the friction forces in the case of particles wedging between two grains, and the surface forces either repulsive or attractive depending on the physico-chemical nature of the suspension. Chemical forces and subsequent chemical bonding can play a role in case of colloidal particles. (Ungemach, 2003)

Different mechanisms facilitate the accumulation of migrating particles from the reinjected fluid or the formation, and influence the reduction of permeability as well. In porous formations, the basic mechanisms are interception, sedimentation, filtering according to size, and diffusion. The interception takes place, when the particles cannot follow the tortuous, at a microscopic scale, flow path and collide with the grains. During sedimentation, due to the solid-liquid density contrast, gravity becomes dominant and the particle, which moves at a velocity lower than fluid velocity, sediments. This process is regulated by the Stokes sedimentation law. The filtering mechanism occurs when the radius of the particle is bigger than the one of the pore entry. Diffusion can basically occur with particles smaller than 1 micron. (Ochi and Vernoux, 1998)

Deplugging of plugged pore spaces can occur as well. It can occur either spontaneously due to the natural flow conditions or artificially as a consequence of, operator generated, hydrodynamic changes such as flow-pressure pulses or flow reversal (backwashing).

Table 1 presents the classification of deep filtration types of suspended solid particles (after Herzig, 1970).

<b>Filtration type</b>	Mechanical	Physico-chemical	Colloidal
<b>Particle size</b>	7-30 $\mu\text{m}$	1-3 $\mu\text{m}$	<0.1 $\mu\text{m}$
<b>Retention sites</b>	constrictions, crevices, caverns	surface	surface
<b>Retention forces</b>	frictions, fluid pressure	Van der Waals, electrokinetic	Van der Waals, electrokinetic, chemical bounding
<b>Mechanism</b>	sedimentation, direct interception	direct interception	direct interception, diffusion
<b>Spontaneous deplugging</b>	unlikely	possible	possible

Provoked deplugging

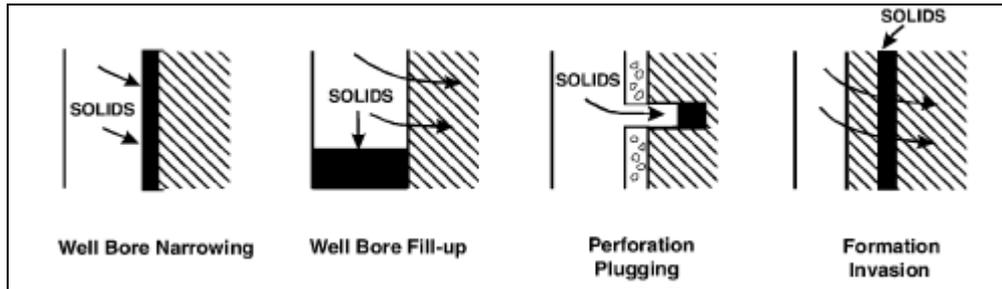
flow reversal

increase in flowrate

increase in flowrate

**Table 1:** Filtration types of suspended solid particles (Herzig, 1970)

There are four types of well and formation impairment caused by solid particles (fig. 3). (Barkman and Davidson, 1972)



**Fig. 3:** Types of well and formation impairment mechanisms (Barkman and Davidson, 1972)

- Wellbore narrowing. Particles are fixed against the wellface. This damage can be removed by reversing flow from injection to production (backwashing), or by acidizing.
- Wellbore fill-up. Particles are sedimented downhole by gravity reducing the length of the filtered section.
- Perforation plugging. It can be removed with backwashing or acid treatment.
- Reduction of formation permeability. Solid particles start bridging at a distance from the wellbore, where velocities cannot sustain entrainment any longer.

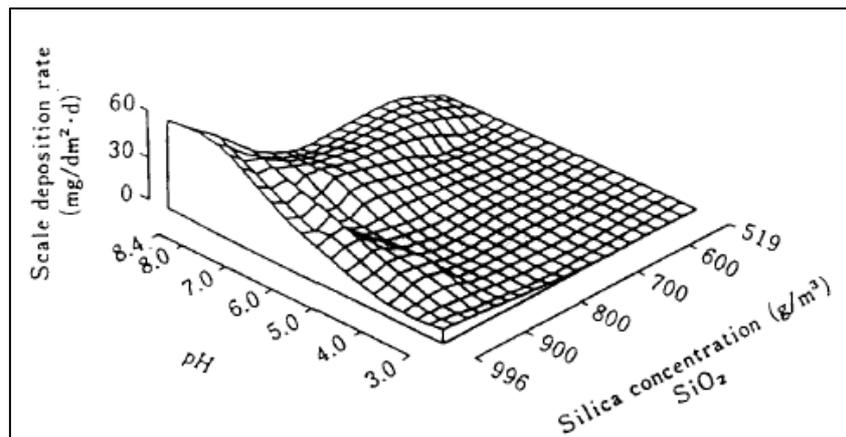
The majority of the low- and medium-enthalpy geothermal reservoirs of Hungary can be found in Tertiary Upper Pannonian sandstone formations. For a significant increase of geothermal energy utilization (for which the natural conditions are available), the research of reinjection into sandstone is a priority issue.

### 3.2. Physics, chemistry of reinjection

Despite the fact that we do not know the answers to many questions regarding reinjection, some Hungarian and international examples show that reinjection into these formations can be successful and technically solvable, if reinjection is preceded by accurate geological and hydrogeological research and economic feasibility prognoses.

In the case of high-enthalpy geothermal reservoirs (mainly fractured igneous rocks), silica scaling is a major problem of reinjection. Silica scaling is a complex process and depends on the following factors: temperature, pH, SiO<sub>2</sub> concentration and the concentration of coexisting dissolved matter.

Hirowatari and Yamauchi (1990) carried out an analyses in the geothermal field in Hatchobaru, where they used the non-condensable gases from the geothermal power station for the pH decrease of the separated brine. The pH decreased from 7.0 to 5.2 (at 59° C), and the deposition rate to 1/30 compared to the original brine. (Fig. 4) This is a very promising result; however, we must mention that the non-condensing gases of the power station were enough for the treatment of only 10% of the separated waters.



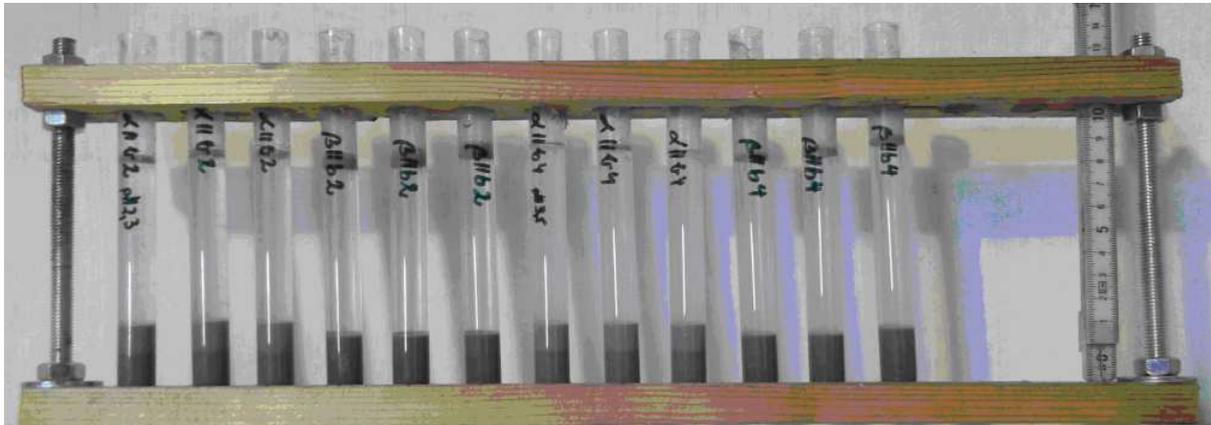
**Fig. 4:** Connection between the deposition, pH and concentration of silica.  
(Hirowatari and Yamauchi, 1990)

Based on experiments in Japan, The Philippines, New Zealand and Iceland, it can be stated the reinjection of fluids with a high silica content cannot be realized on a long term, since the injectivity of reinjection wells decreases quickly due to silica scaling. (Stefansson, 1997)

#### Sediment volume analyses

The measurement of the sediment volume is an excellent and simple method to analyze the bonding of grains in different solutions and the eventual suspensions. This consists of mixing the same amount of rocks into identical quantities of water in identical vessels, shaking them up and leaving them to deposit, and finally measuring the height of the sediment (figure 5). The increase of the sediment volume means the increase of the binding

force of grains, while the decrease of the sediment volume means the opposite. The particles not bonding completely are depositing independently, and the Brown movement can place them into all the free places, resulting in a compact sediment. In case the particles have the habit to bond, they bond during deposition into different grains that cannot fill every place, resulting in unconsolidated sediment and a greater sediment volume.



**Fig. 5.** Sediment volumes with different solutions.

The rock samples were taken from drillings near the reinjection location and from the reinjection depth. The core samples were ground, and the powder analyzed by means of XRF and XRD spectroscopy. The two ground materials were used for the sediment analyses; their symbols:  $\alpha$  and  $\beta$  rock samples.

For the XRF analyses, we placed the original powder samples into the sample holder and measured them. For the XRD analyses, we measured 0.2-0.2 grams from the macroscopically homogeneous samples and pulverized them in a mortar to reach a grain size of approx. 10 $\mu$ m. The resulting powder specimens were placed on a standard glass specimen holder with a standard background.

We used two water samples: Sample I was taken from the production well, sample II from the buffer reservoir of the reinjection well. We carried out two water treatments with both water samples.

**A.** The reduction treatment. The vessel with the original water sample was completely full when taken into the lab, and it remained so until its application. The content of the opened vessels were used and the residues emptied. Every measurement was carried out from freshly opened vessels as quickly as possible. The objective was to avoid the contact of the water sample and the oxygen from the air, i.e. to keep the water in a reductive state.

**B.** The oxidation treatment. For two hours, we bubbled air through the original water sample. The experiments were carried out from freshly bubbled samples reducing by that the disturbing effects of long-term slow reactions. The objective was to model the geothermal utilization and to allow the contact of the water sample with the oxygen from the air, i.e. to keep the water in an oxidative state.

For the pH analyses, water samples adjusted to pH values of **2, 4, 6, 8 and 10** were used for our sedimentation analyses. The accuracy of adjustment was  $\pm 0.2$  pH measured with a glass electrode.

The tests were carried out in 20-ml test tubes, with 2 g of rock sample and 10 ml of water in each tube. After an intensive shake-up and two hours of deposition, the tubes were placed in the frame in a way that the values read from the pictures taken of the tubes with the camera to be comparable. We took pictures of the marked test tubes by permanent artificial daylight. Marking for example *αa2*.

The relative height of the sediment in comparison to the frame was measured with the IrfanView software according to the stored picture.

### The effect of pH on sedimentation

For rock sample  $\alpha$ , we can see two well-separated sedimentation phases. The reason for this is that the sample is the mixture of two rocks with significantly different surface characteristics or grain sizes. We analyzed the phases separately and together as well. For rock sample  $\beta$ , we see on sedimentation phase.

### Reduction tests

Reductive circumstances mean that the vessel with the original water sample was completely full when taken into the lab, and it remained so until its application. The content of the opened vessels were used and the residues emptied. Every measurement was carried out from freshly opened vessels as quickly as possible. The objective was to avoid the contact of the water sample and the oxygen from the air, i.e. to keep the water in a reductive state.

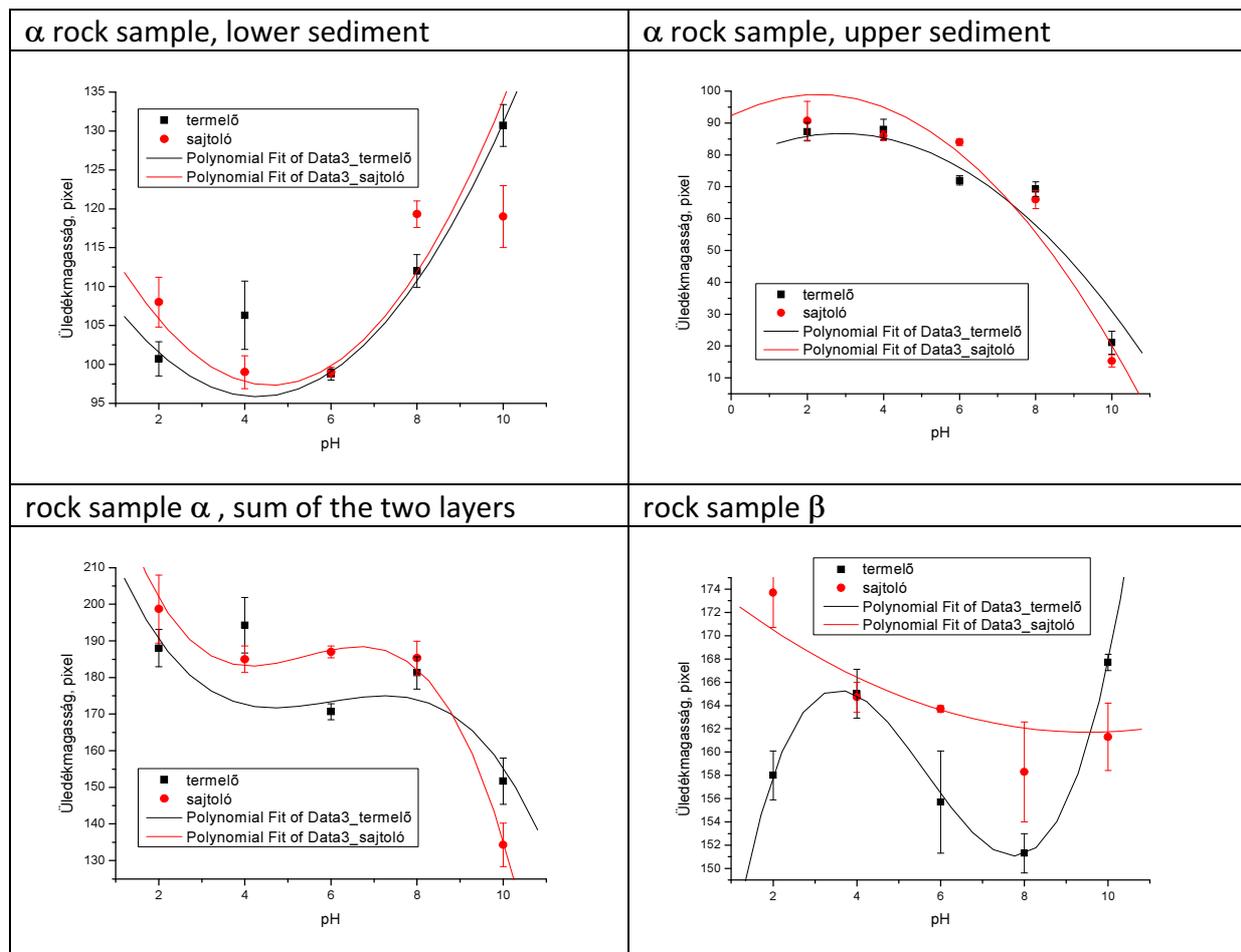
For rock sample  $\beta$ , the sediment volume of the water sample taken from the reinjection well showed a not categorical minimum at approx. 9 pH. This means that the binding force between the particles is the smallest and they can be moved in the easiest way at this pH value. The sediment volume of the water sample taken from the production well shows a categorical minimum at approx. 8 pH. This means that the binding force between the particles is the smallest at this pH value, but the sediment volume shows a maximum meaning that the grains bind strongly with the water of the production well. When decreasing the pH value to be smaller than 4, the attraction force between the particles decreases.

For rock sample  $\alpha$ , the sediment volume of the lower sediment of both water samples shows a minimum at approx. 4.5 pH, meaning that the binding force between the particles is the smallest and they can be moved in the easiest way at this pH value. When decreasing and increasing the pH values, the binding forces between the grains grow, the isoelectric point of these particles is at this point.

For rock sample  $\alpha$ , the volume of the upper sediment of both water samples monotonically decreases at an almost identical velocity with the increase of the pH value. These grains have obviously a negative charge (most likely clay mineral grains) and they bind easily after loosing their negative charge due to hydrogen ions; while the repulsive force between the grains significantly grows with alkalization. In this case, the small grains can easily move during the alkalization of the water.

For rock sample  $\alpha$ , the sediment volume changes identically at the sum of the two sediments for both water samples, it decreases with the increase of the pH value. Obviously, we see the resultant of the two separate curves, meaning that the sediment volume shows a minimum at a pH value around 4-5, thus the adhesion is relatively poor. The maximum is at a pH value of approx. 7, meaning a good adhesion of grains. By further alkalizing the system, we see a drastic drop indicating a weakening adhesion of grains.

Figure 6 shows the result of the reduction analyses of the sediment volume.



**Fig. 6.** Sediment volume curves with reductive water samples

### Oxidation tests

Oxidative conditions meant that we bubbled air through the original water sample for two hours. The experiments were carried out from freshly bubbled samples reducing by that the disturbing effects of long-term slow reactions. The objective was to model the geothermal utilization and to allow the contact of the water sample with the oxygen from the air, i.e. to keep the water in an oxidative state.

For rock sample  $\beta$ , the sediment volume of the water sample taken from the reinjection well showed a not categorical minimum at approx. 4 pH. This means that the binding force between the particles is the smallest and they can be moved in the easiest way at this pH value.

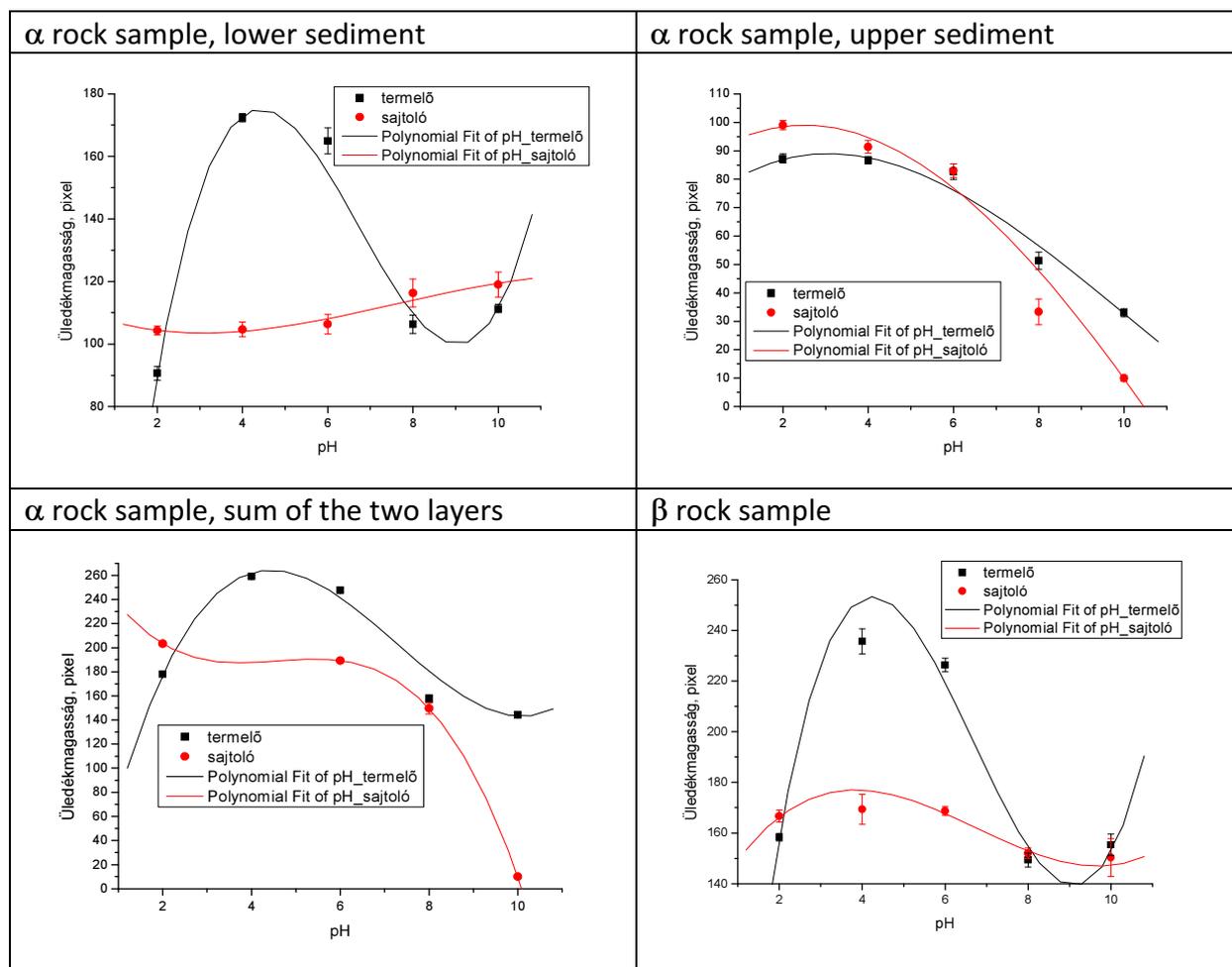
The sediment volume for both water samples shows a categorical minimum at approx. 9 pH.

This means that the binding force between the particles is the smallest at this pH value. The sediment volume of the production well shows a maximum at 4 pH, meaning that the grains bind strongly with the water of the production well, and when decreasing the pH value, the attraction force between the particles decreases again.

For rock sample  $\alpha$ , the water samples of the production and of the reinjection well are very different concerning the lower sediment. In the case of the production well, the sediment volume shows a maximum at approx. 4.5 pH, at an almost identical pH value. This means that the binding force between the particles is the biggest and they are the hardest to move at this pH value. When decreasing and increasing the pH values, the binding forces between the grains grow. The analyses of the water from the reinjection well show that the binding force depends only to a small degree from the pH value, the sediment volume increases with its increase, and the binding force between the grains decreases.

For rock sample  $\alpha$ , the volume of the upper sediment of both water samples monotonically decreases at an almost identical velocity with the increase of the pH value. These clay mineral grains have a permanent negative charge, which increases with the increasing pH value. This means that they bind easily after loosing their negative charge due to hydrogen ions; while the repulsive force between the grains significantly grows with alkalization. In this case, the small grains can easily move during the alkalization of the water.

For rock sample  $\alpha$ , the sediment volume shows a maximum at around 5 pH in case of the sum of the two sediments of the production well. This means that the binding force of grains is very strong, while at an increasing or decreasing pH value, the sediment volume and the binding force of particles decrease as well. The particles have the smallest charge around 5 pH. In a more acidic medium they have a positive charge, and a negative charge when alkalized, thus they repulse each other. The curve for the reinjection well has a monotonic decreasing character, with a not very categorical plateau of 6 pH, indicating a good adhesion of the grains. By further alkalizing the system, we see a drastic drop indicating a weakening adhesion of grains. Figure 7 shows the result of the oxidation tests of the sediment volume.



**Fig. 7.** Sediment volume curves with oxidative water samples

### The effect of the rock samples on the pH-dependence of the sediment volume

#### *Production well, reductive conditions*

- In the case of rock sample  $\alpha$ , we experience a small increase of the sediment volume in relation to the pH for the lower sediment layer, meaning that the increase of the pH value results in the increase of the binding force in reductive conditions, in the case of the production well.
- In the case of rock sample  $\alpha$ , we experience a decrease of the sediment volume in relation to the pH for the upper sediment layer and therefore along the plateau between the weak pH values 4 and 8 on the cumulative curve. This means that in the case of the production well, in reductive conditions, the increase of the pH value results in the decrease of the binding force in the alkaline range in case of a very smooth fraction.
- In the case of rock sample  $\beta$ , we experience a small increase of the sediment volume in relation to the pH in an alkaline range by a peak of 4 pH. This means that the increase of the pH value results in the increase of the binding force in alkaline range

in the case of the production well, in reductive conditions, but it shows a minimum at neutral values.

#### *Production well, oxidative conditions*

- In the case of rock sample  $\alpha$ , for the lower sediment layer and the complete sediment, and in the case of rock sample  $\beta$ , we experience a categorical maximum in the sediment volume at 4-5 pH. This means that in the case of the production well, the binding force shows a maximum value in a slightly acidic range in oxidative conditions.
- In the case of rock sample  $\alpha$ , we experience a decrease of the sediment volume in relation to the pH for the upper sediment layer. This means that in the case of the production well, in oxidative conditions, the increase of the pH value results in the decrease of the binding force in the alkaline range in case of a very smooth fraction.

#### *Reinjection well, reductive conditions*

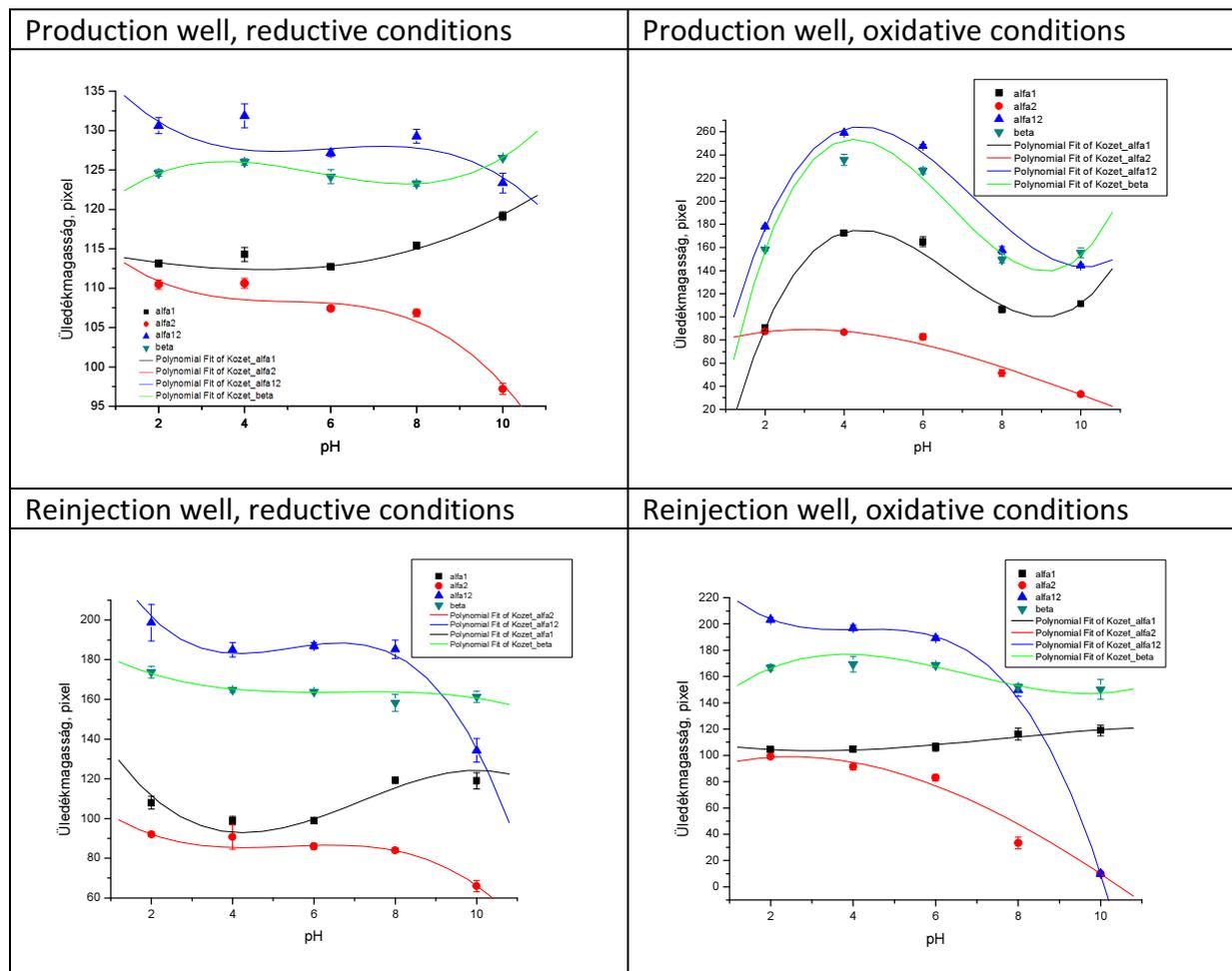
- In the case of rock sample  $\alpha$ , we experience a small increase of the sediment volume in relation to the pH for the lower sediment layer above the minimum of 4 pH, meaning that the increase of the pH value results in the increase of the binding force in reductive conditions in the case of the reinjection well.
- In the case of rock sample  $\beta$ , we experience a small increase of the sediment volume in relation to the pH, meaning that the increase of the pH value results in the increase of the binding force in the alkaline range in reductive conditions in the case of the reinjection well.
- In the case of rock sample  $\alpha$ , we experience a decrease of the sediment volume in relation to the pH for the upper sediment layer and therefore along the plateau between the weak pH values 4 and 8 on the cumulative curve. This means that in the case of the reinjection well, in reductive conditions, the increase of the pH value results in the decrease of the binding force in the alkaline range in case of a very smooth fraction.

#### *Reinjection well, oxidative conditions*

- In the case of rock sample  $\alpha$ , we experience a small increase of the sediment volume in relation to the pH for the lower sediment layer, meaning that the increase of the pH value results in the increase of the binding force in oxidative conditions, in the case of the reinjection well.
- In the case of rock sample  $\beta$ , we experience a small decrease of the sediment volume in relation to the pH, meaning that the increase of the pH value results in the decrease of the binding force in the alkaline range in oxidative conditions, in the case of the reinjection well.
- In the case of rock sample  $\alpha$ , we experience a decrease of the sediment volume in relation to the pH for the upper sediment layer and therefore along the plateau between the weak pH values 4 and 8 on the cumulative curve. This means that in the case of the production well, in reductive conditions, the increase of the pH value

results in the decrease of the binding force in the alkaline range in case of a very smooth fraction.

Figure 8 shows the results of the pH-dependence of the sediment volume for the different rock samples



**Figure 8.** Sediment volume curves for different rock samples

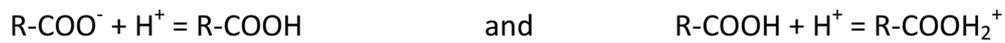
### Chemical processes during reinjection

The acid-base reaction can significantly modify the dissolution balance. Everyone knows karstic phenomena. This is when limestone dissolves in the acidic rainwater and leaves caves when washed out. However, everyone has seen stalactite caves too, where limestone precipitates from an alkaline solution (supersaturated solution) after losing a little bit of carbon dioxide. In this case, there are small differences of a few tenths of a percent between the concentrations, but in a state close to balance lasting for a long time, these small differences have a huge impact as well. The same processes can take place in the pores of the rocks, expanding them during dissolution and narrowing them during precipitation. Acidification usually helps dissolution and alkalization the precipitation, but this is not that obvious, as we will see below.

The oxidization of organic compounds has an important effect. The oxidized elements of the oxidation series, e.g. the aldehydes, ketones and organic acids can be present in the complex compounds as ligands. This means that they have a free electron pair that they can share with electron-deficient ions, such as the heavy metal ions. This outlines the effect that the

more oxidized organic molecules create a complex with the heavy metal ions and decrease the concentration of free metal ions in the solution. Due to the decreasing concentration of metal ions, the ions can change from a solid phase into solution.

Let us examine the process of complex formation. A complex is formed, when a ligand can easily share its electron pair. This requires a loosely bound pair of electrons. Let us take a look at an organic acid and its conditions in an watery medium. R is a group with an optional carbon content:



The first equation (higher probability) takes place from left to right, if we acidify the solution; and from right to left, if we alkalize the solution. The second equation is rare, it takes place in a very acidic medium, but the directions are similar to the previous equation. The carboxylate ion ( $\text{R-COO}^-$ ) is more complexing, since an oxygen with a negative charge is more likely to share its electron pair than a neutral, protonated form of uncharged carboxylic acid. What is the conclusion? The complexing tendency strongly depends on the pH of the formation. The complex formation decreases when acidified, and increases when alkalized. From here, we only have to expand these ideas to the reinjection system, and to combine the information about the pH and the complex formation. If the pH decreases, thus the system acidifies, the complex formation is confined, but the transition metal oxyhydroxides dissolve easier. If the solution is becoming alkaline, the complex formation comes to the fore, but the transition metal oxyhydroxides dissolve harder.

Which one is the strongest process? This depends on the quantity and quality of the complexing ligands in the solution. Many strong complexing agents in the solution can dissolve non-soluble compounds in an alkaline medium. With less and ligands in the solution, they can dissolve only in an acidic medium. Accordingly, we can state that the pH and the alteration of the oxidation state can seriously shift the dissolution balances in the system, but also from point to point due to the local modification of the pH.

There are several more processes in a complicated system like this. Here, we discussed the most simple and most general ones, and we can measure and present several other processes in the light of unique compositions.

### Summary of the results

During our analyses, we used two different ground materials (marked with  $\alpha$  and  $\beta$ ), and analyzed them with water samples from geothermal production and reinjection wells. We examined original (no contact with the oxygen from the air) and treated (two-hour bubbling with atmospheric air) water samples. For the pH analyses, water samples adjusted to pH values of 2, 4, 6, 8 and 10 were used for our sedimentation analyses.

### *Summary of our results*

For one of the rock samples, we observed a solid phase depositing at two different velocities, one with larger and one with a much smaller grain size. According to the results of

the XRD and XRF analyses carried out on the rock samples, we can state that the dominant components in both samples are quartz, 10 Å-phase (most probably a well-organized 2 M muscovite) and dolomite. As an accessory, plagioclase feldspar close to albite in composition, chlorite and calcite are safely identifiable, and traces of goethite cannot be excluded either. Sample  $\alpha$  contains hematite and rutile as well. For sample  $\alpha$ , the shape of double reflection at 7 Å and the abnormal intensity ratio of chlorite presume the presence of kaolinite. The result of these deviations is the two phases of sample  $\alpha$ .

In relation to the pH, significant differences were demonstrated for the untreated water samples, the aerated thermal waters and the rock samples as well. With the increasing pH, we obtained a decreasing sediment volume, meaning that alkalization decreases the binding force of grains. In several cases, we experienced extreme values in the analyzed range. This means that there are pH ranges that produce maximally and minimally adhesive particles. More analyses on more rock samples would be necessary in relation to the pH for the deduction of generalities. The details were evaluated separately when discussing the different treatments.

Both wells show that the pH value of the aerated sample is significantly higher than the one of the original water sample. The explanation for this is that the intensive bubble aeration eliminates a great amount of carbon dioxide from the system, resulting in the reduction of carbonic acid and hydrogen ion concentration. The original pH of the reinjection well is lower, explained by the fact that the sample oxidizes while travelling in the pipeline and touching the surfaces, and eventually contacting the oxygen of the air in the buffer tank, in a way that a much smaller amount of carbon dioxide leaves the system compared to the intensive bubble aeration method. The acidifying effect of oxidation can predominate beside the effect of carbonic acid generating a much bigger alteration of the pH.

#### *The results of the analysis of titration curves*

In the case of acid titration, we do not find any difference between the samples. The measured inflection points show that the small amount of matter found at the beginning corresponds to the carbonic acid balance.

In the case of the alkaline titration, we see that among the first inflection point inorganic compounds – mainly complex ones – there are several ones where we can talk about protonation in this range. These can be aluminium or iron complexes with low- or moderate-molecular-weight organic ligands. The second inflection point shows the transformation of carbonate ions into hydrocarbonate.

In the case of the alkaline titration, we find a significant difference between the waters of the production and the reinjection well.

### Theoretical summary of plugging during reinjection

According to the above-mentioned facts, one reason for plugging can be that the grains move from their places and the flowing fluid moves them until they get stuck in a tight cross section. The physical cause of plugging is quite easy to estimate and calculate. The grains can migrate easily in case of high-speed water flows, and they can get stuck later. Accordingly, the best solution is to stream the fluid as slow and as steady as possible during reinjection. It is more complicated to define the degree of grain migration and the strength of the binding force keeping them in place or trying to move them into the liquid phase. In the chapters above, we tried to provide several elementary correlations and guidance to this issue.

Another possible reason is the separation of some sort of solid precipitation from the fluid phase, continuously narrowing the pores. These chemical processes are very complicated, and we must take into account the actions of the water on the surface. These processes were described in detail, and the complexity of the problem is visible.

In short, we can state that we dispose of many information, basic knowledge and correlations about reinjection. However, due to the complexity and interaction of the processes, we can find well-specific results only through the analysis of data acquired from the actual object and with model experiments.

Based on these facts, we see that fluids with chemical characteristics identical to their reservoir rocks are worth being reinjected, reducing the above-ground processes influencing water quality to their possible minimum.

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