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(Environmental) Impact of Inhibitors Applied in the Geothermal Sector in the Netherlands

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ABSTRACT

At this moment there are 18 geothermal sites in operation in the Netherlands. Nearly all sites dose corrosion inhibitor at the bottom of the production well to protect the carbon steel based production and injection wells from corrosion. Though these corrosion inhibitors are considered effective as they significantly reduce the corrosion rates, the environmental impact can also be significant as these chemicals have a biocide capacity. Three leakage routes of inhibitors into the environment were found that had an average risk or high level after a Hazard Operability (HAZOP) analysis. These three leakage routes were all found in the injection well, as the injection well has overpressure and is not monitored continuously.

The first leakage route is towards the production aquifer after re-injection in the injection well. It is unknown how much residual inhibitor will reach the production aquifer, as monitoring of the injection well (including inhibitor concentration) is not performed continuously. The second and third risks are also situated in the injection well. For these risks also no continuous online monitoring is performed in the injection well. As this well is operating under pressurized conditions (2 - 50 bar), there is the risk of unnoticed leakages for a period of up to three years to fresh and brackish aquifers. Therefore, both ecosystems and other extractions (e.g. drinking water extraction) in these aquifers might be influenced. In the case of leakages, not only the inhibitor itself is considered toxic but also the production fluid, so leakages to the surroundings should be prevented.

We, therefore, recommend that more frequent monitoring of the injection well should be installed and that the concentration of inhibitor throughout the whole installation should also be monitored. Next, we recommend investigating the use of other corrosion-resistant well materials to, possibly, reduce the inhibitor dosage in geothermal systems. Fortunately, the sector is already acting upon these findings, as processes for better monitoring are in the pipeline and alternative well materials are recently implemented in some geothermal installations in The Netherlands.

1. INTRODUCTION

The use of geothermal energy for heat supply purposes has developed tremendously in the recent ten years in the Netherlands. The first geothermal site started operation in 2007, and at this moment there are 18 geothermal sites active. Even more, the Netherlands has developed a masterplan geothermal energy. (1) This masterplan describes the strong ambition to increase the number of sites to ~175 in 2030 and ~700 in 2050: an exponential increase to 200 PJ/y in 2050. As the sector is relatively young, it is important to learn the lessons now and mitigate any upcoming risks such that a sustainable future can be built.

One of the risks in the geothermal sector is the integrity decrease of the geothermal installation. As posed by the National Mines Inspectorate (SodM), State Supervision of Mines (SSM), Dutch geothermal projects are coping with corrosion problems. This research focusses on the corrosion problems within the Netherlands. The insights provided can also be transferred to other countries as there are more countries which suffer from comparable corrosivity problems.

To secure economic and sustainable geothermal heat production, integrity degrading corrosion processes have to be controlled and limited. For this purpose, the geothermal sector can use MOC-method. This method states that mitigation actions should be taken in the order of Mechanical (e.g. choice of well and above ground corrosion-resistant material), Operational (e.g. nitrogen blanket to prevent oxygen intake and corrosion monitoring) and Chemical (e.g. dosing of inhibitors). In the Netherlands, corrosion is observed in current operating geothermal wells for which mechanical and operational measures are already in place (well material, nitrogen blanket). Therefore, our research focusses on chemical corrosion protection: the use of inhibitors (corrosion, scaling, biological growth) in the geothermal sector in the Netherlands. Specifically, the technical and the environmental impact of corrosion inhibitors in the geothermal sector in the Netherlands are investigated, as currently, these are the only ones used. It seems that the sector is, however, moving towards more mechanical and operational ways for new geothermal systems to prevent corrosion of the system. Therefore, we also share some insights on mechanical and operational mitigation measures against corrosion, which are currently investigated within the Dutch geothermal sector.

2. PROCESS CONFIGURATION

In order to understand how corrosion in the Dutch geothermal systems takes place, knowledge on the geothermal installation and components present in the geothermal fluid has to be obtained. Currently, all wells in the Netherlands are constructed of carbon steel: a combination of carbon (C) and iron (Fe). (6), (7) **Figure 2-1** shows a schematic representation of a geothermal installation. On the left side, the production well is drawn and on the right side the injection well; the reservoir depth is between 1500 - 3000 m. As the static water level is 50 - 240 m below ground level, an electric submersible pump (ESP) is used to pump the fluid to the surface. This pumping results in under pressure from the point of the ESP on at the production site. After the hot fluid (60 - 100 °C) pumps to the earth surface, it is filtered through the first set of filters and passed through a heat exchanger. The exchanger releases heat to the environment via a separate fluid stream. Downstream of the heat exchanger the cooled geothermal fluid filters through the second set of filters. Pumping this cooled fluid (20 - 40 °C) into the injection well leads to an overpressure (2 - 50 bar) at the injection site (**Table 1**). (2)

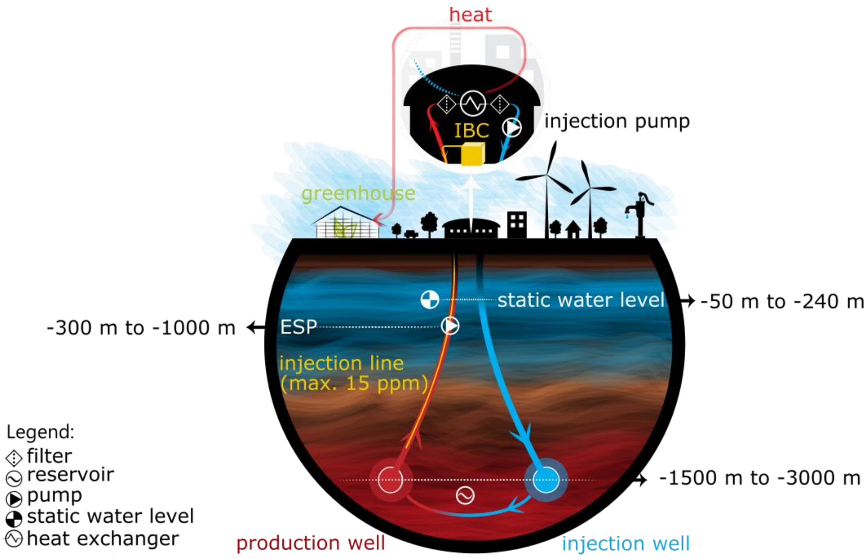

Figure 2-1: process configuration of geothermal installation in the Netherlands.

Table 1: Properties of geothermal fluid in the Netherlands. (2)

Property	Value	unit
Temperature $_{producer}$	60 - 100	°C
Temperature $_{injector}$	20 - 40	°C
Pressure $_{producer}$	3.5 - 25	bar
Pressure $_{injector}$	2 - 50	bar
pH	5.3 - 6.7	-
TDS (total dissolved solid)	81 - 240	g/L
Flow	100 - 390	m^3/h
carbon dioxide	0 – 57	Mol%gas
methane	5.6 – 93	Mol%gas
oxygen	0 – 0.065	Mol%gas

Right before the first set of filters, the geothermal fluid is continuously monitored on pH, temperature and oxidation-reduction potential (ORP). Additionally, also coupons, Linear Polarization Resistance probes (LPR) and water analysis measurements are used to monitor the corrosion rate of the geothermal fluid at this specific point. (4), (5) At almost all geothermal locations in the Netherlands, an intermediate bulk container (IBC) of 1 m^3 is present above ground, containing concentrated corrosion inhibitor. This corrosion inhibitor is continuously dosed to the production well, just above the reservoir level. (3) Baker Hughes, one of the inhibitor suppliers, also detects the concentration of inhibitor above ground. Downstream of the second filters no monitoring of (components in) the system is performed. The only insight in case corrosion is taking place when the ESP is pulled for maintenance purposes. At this moment, visual inspections to both the production and injection sides are performed. This so-called "log-campaign" is performed every 3-5 years. Thus, it may take three years before corrosion on the injection side is detected. The Dutch geothermal sector does not have a structural sector-wide corrosion management plan, which especially on the injection side, seems a miss. We, therefore, recommend drafting such a corrosion management plan and evaluate outcomes via an external audit.

3. CORROSION

The fluid of Dutch geothermal systems is slightly acidic (pH: 5.3 - 6.7) and contains high amounts of salt (Table 1). The total dissolved solids (TDS) are in fact 2 - 7 times (81 - 240 g/L) the concentration of seawater. Both carbon dioxide (0 - 57 mol%gas) and methane (5.6 - 93 mol%gas) are present at almost all of the locations, which at some locations results in degassing to harvest methane. In principle, there is no oxygen present in the whole system, and the addition of a nitrogen blanket mitigates possible oxygen intake. (2)

Table 2: Components in the geothermal fluid in the Netherlands. (2)

Component	Value	unit
Cadmium	<0.001 – 0.05	mg/L
Calcium	2.7 – 16	g/L
Chromium	<0.005 – 2.6	mg/L
Chloride	37 - 160	g/L
Fluoride	0.7 - 500	mg/L
Inhibitor	5 - 15	mg/L
Iron	5.6 – 190	mg/L
Lead	<0.05 – 13	mg/L
Magnesium	400 - 2000	mg/L
Phosphate	0.6 - 500	mg/L
Potassium	100 - 2200	mg/L
Sodium	17 – 32	g/L
Sulphate	56 - 2900	mg/L
Nickel	<0.01 – 2.4	mg/L

Due to the presence of carbon dioxide (CO_2) in the geothermal fluid, the pH of the fluid decreases to 3.5 - 6 (8): carbon dioxide reacts with water forming protons (acid; H^+) and carbonate (HCO^{3-}) (1). (4) (8) It is this acid that oxidizes the iron of the carbon steel well material, after which dihydrogen gas (H_2) forms and the iron dissolves in the aqueous fluid. (2) This oxidation reaction is catalyzed (accelerated) by high temperatures and chloride content present in the geothermal water: creating a highly corrosive fluid.

$$CO_2 + H_2O \rightleftharpoons H^+ (aq) + HCO_3^- (aq) \qquad (1)$$

$$Fe^0 (s) + 2\,H^+ \longrightarrow Fe^{2+} (aq) + H_2 \qquad (2)$$

Corrosion can cause, e.g. decrease of the thickness of the well material and leakages to the surrounding layers, decrease of heat capacity and precipitation of radioactive lead. (2) For Dutch geothermal systems, general corrosion and under certain circumstances crevice corrosion around crevices present between adjacent parts of casing and coupling is considered to be the most likely forms of corrosion. MIC (Microbiologically Influenced Corrosion) was not thought to take place as no traces of hydrogen sulphide were found (**Table 2**). However, recent studies (published after finalizing our investigations) for six Dutch geothermal systems showed that various types of bacteria are actually present in the production water. (16) As the most common species were the Sulphur metabolizing bacteria, it was concluded that these systems contain risks for the occurrence of MIC. MIC is corrosion induced by biological activity. One of the best-known groups of microorganisms which are involved in corrosion is Sulphur metabolizing bacteria. These organisms produce hydrogen sulphide and/or sulphuric acid, which is the cause of corrosion to the well material. The article further describes that these bacteria are also capable of decomposing corrosion inhibitors. This decomposing is in line with our information that the currently used corrosion inhibitors have biocidal activity. (4), (5)

4. CORROSION MITIGATION

4.1 Introduction

Currently, the geothermal sector in the Netherlands, only doses corrosion inhibitors which also have biocidal activity. Scaling inhibitors are not used anymore, as scaling is mitigated partially operationally by reinjection of carbon dioxide and partially as a result of corrosion inhibitor dosing. From the moment of adding corrosion inhibitors to geothermal wells in the Netherlands, both the rate of corrosion of the well material and the precipitation of radioactive lead have decreased substantially. (9)

Corrosion inhibitors are continuously dosed at the bottom of the production well with a concentration between 4 - 15 parts per million (ppm). However, the average concentration used is 10 ppm. Either Nalco Water or Baker Hughes supply the corrosion inhibitors. (4), (5) In lab tests performed by the inhibitor suppliers an 88,5% - 90% corrosion protection level was found at dosing levels of around 10 ppm for simulated lab scale systems. The right concentration of dosage is currently selected via an empirical approach, where both the manager of the geothermal site as well as the inhibitor supplier together define the right concentration. The geothermal sector, however, is moving towards site-specific dosage concentration to have the best protection against corrosion, while the amount of unnatural substance inside the system minimalizes.

Corrosion inhibitors are a cocktail of multiple compounds, but all are based on an "active component", which is less than <5 wt.% present within the cocktail. This main active component often has a nitrogen-based polar head on one side, and an apolar carbon-based hydrophobic tail on the other side (**Figure 4-1**). Upon addition of inhibitors, these "filming amines" compete with the acidic water, by adsorbing at the metal surface via their polar heads. Once, more inhibitors bind to the surface, a strong layer (film) is formed, as a result of strengthening (ionic and van der Waals) interactions between the filming amines (**Figure 4-2**). Depending on the dosed concentration and properties of the inhibitor, either a monolayer or a double layer forms. These layers cannot be penetrated easily by the acidic water anymore, resulting in a reduced corrosion rate. In some cases it is also suggested that these amines form pseudo-complexes with the (oxidized) iron atoms, also forming a protective film. (6), (7), (9), (11), (12), (17)

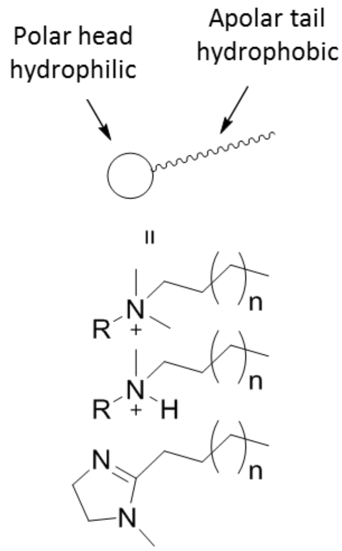

Figure 4-1: Active component in corrosion inhibitor cocktail. Such a filming amine often has an amine-based polar head and a carbon-based apolar tail. (7), (9)

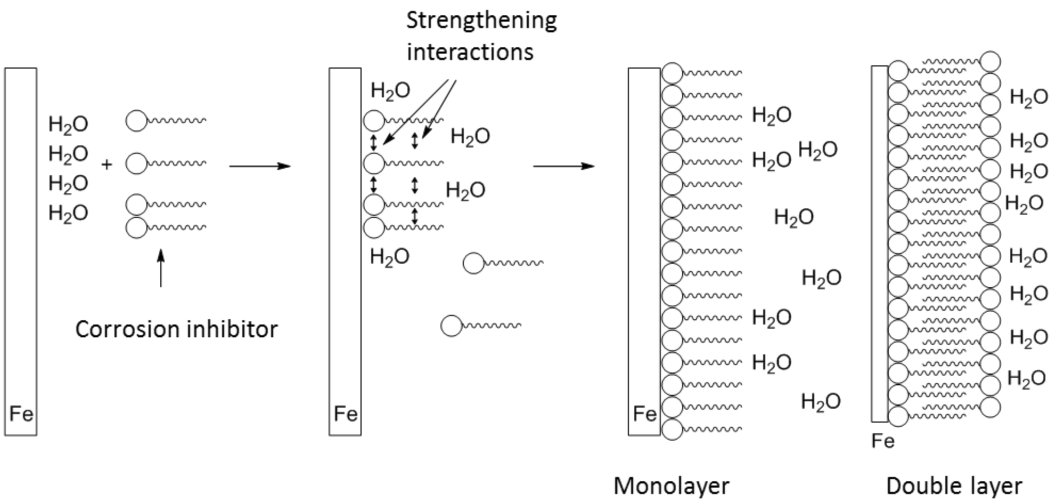

Figure 4-2: Principle of protection against corrosion using a corrosion inhibitor. (6), (11), (12)

Desorption of inhibitors from the carbon steel surface can occur in four ways:

- due to bad properties (adsorption/sticking/interaction) of the inhibitor components;
- through competition with other components present in the water stream;
- through shear stress, which is dependent on temperature and flow;
- due to low coverage of the surface as a result of too low concentration or insufficient mixing.

All geothermal systems have different properties which can all contribute to desorption. It is important to work with the earlier mentioned site-specific dosage of inhibitors to have better protection against corrosion, while the amount of unnatural substance inside the system minimalizes.

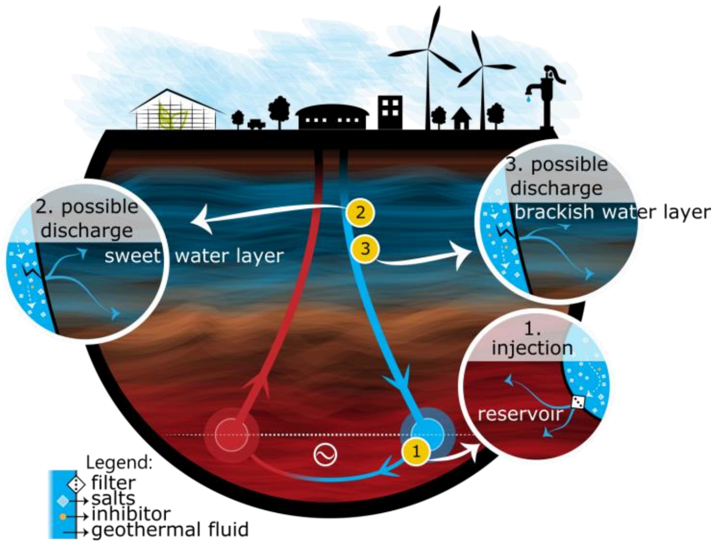

Figure 4-3: Three identified risks of leakage of inhibitor into the environment.

4.2 Hazard Operability Analysis (HAZOP)

We conducted a Hazard Operability Analysis (HAZOP) to determine possible pathways through which leakage of inhibitor towards the environment could occur for three scenarios. The scenarios were production site, above-ground installation and injection site. During this risk analysis session, focusing on operational risks, it became clear that there is a small chance of leakage on the production side. This site has under pressure, induced by the ESP. The installation above ground is in almost all cases completely built on a liquid-proof floor. Spills are automatically transferred to a waste basin, which is deposited to a chemical waste company. Next, it was discovered that it is unknown what the concentration of inhibitor is throughout the whole installation. Thus, it is unknown if there is still an inhibitor present and protecting against corrosion at the injection site. Moreover the possible presence of bacteria also leads to inactivation of the inhibitors, which lowers the inhibitor concentration even more. As a result, the main risks of leakage of inhibitor into the environment that were found during the HAZOP-session have been identified at the injection site (Figure 4-3):

1. injection into the geothermal reservoir (very high-risk level);
2. discharge along with trajectory injection well into drinking water layer (average risk level);
3. discharge, along with trajectory injection well into brackish water layer (average risk level).

For the three risks identified above, the impact on the environment of identified discharge/injection risks has been evaluated in different aquifers. Both consulted inhibitor suppliers state that the inhibitors are relatively stable under the geothermal conditions in the Netherlands. Information is not available regarding the stability of inhibitors under the geothermal conditions in time, neither is information on the toxicity of (degradation products of) inhibitors under anaerobic conditions available. (4) Even more, commonly used classifications for chemicals as cefas and Wassergefährdungsklasse are contradictory (**Table 3**). For example, Cortron CK990-G has the gold (least toxic) classification in cefas. It scores a 2 in WGK, where CRW93133 has a silver classification in cefas where it is marked 1 (least toxic) in WGK. (4), (5)

Table 3: Cefas and Wassergefährdungsklasse (WGK) classifications of corrosion inhibitors. (4), (5)

Product name	**Cefas**	**WGK**
Cortron CK990-G	gold (least toxic)	2
CRW83133	silver	1 (least toxic)

Thus, at this stage, it is unknown what the degradation products and environmental impacts are of the used inhibitors in the Dutch geothermal systems. We therefore strongly recommend the drafting of European guidelines for inhibitors under anaerobic geothermal conditions. As no unambiguous information is available regarding the toxicity of the inhibitors, we decided to use the signaling value of unknown substances in the Dutch drinking water guideline (Drinkwaterbesluit) as reference. (14) The signaling value for unknown substances is 0.001 ppm. Plotting the signaling value of the inhibitor (both total cocktail dosed and the 5% active compound, which is the most toxic within the mixture) and of components in the geothermal fluid versus the maximum concentration of all of these components (**Table 2**) results in the graph displayed in **Figure 4-4**. It can be seen that the relative concentration (compared to drinking water signaling value) of the total inhibitor is 20000 times above the allowed concentration and 20 times higher than the next compound (Pb) in the production fluid. The 5% active compound is between 100 and 1000 times higher than the allowed concentration, which is in between the concentration of chloride and sodium. From **Figure 4-4**, it becomes evident that all components within the geothermal fluid are high above the allowed concentrations in the Drinkwaterbesluit. For this reason, it is best to prevent leakage of the geothermal fluid into the environment.

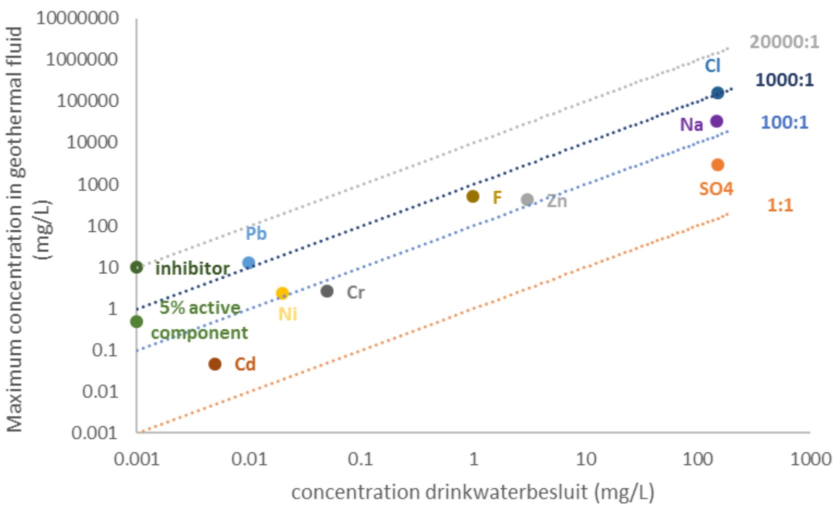

Figure 4-4: Relative concentrations of components present in the geothermal fluid (x-axes: concentration drinking water guideline (Drinkwaterbesluit); y-axes: maximum concentration of the component in geothermal fluid). (8), (14)

4.3 Transport modelling

Next, we modelled the transport of inhibitors for the three identified risks on leakage. For this modelling, we used Modflow and MT3DMS and the following worst-case assumption:

- no retardation of inhibitor;
- no biodegradation;
- 25 m^3/d leakage flow;
- 3 years before the next log-campaign;
- active use of groundwater layers.

Risk 1: leakage of inhibitors in the geothermal reservoir

If the formation of the geothermal reservoir is considered as a receptor, then injection of the inhibitors through the injection well can have a direct effect on the geothermal reservoir. In this reservoir, a concentration of 0.001 ppm (dosed inhibitor concentration of 10 ppm) spreads radially from the injection well into the geothermal reservoir over a distance of circa 740 meters. It must be stated that the concentration of 0.001 ppm does not reach the extraction well. There is neither sufficient knowledge of the biology and hydrochemistry of the geothermal reservoir nor of the behaviour of the inhibitors to say something on the environmental impact of the inhibitors on the reservoir.

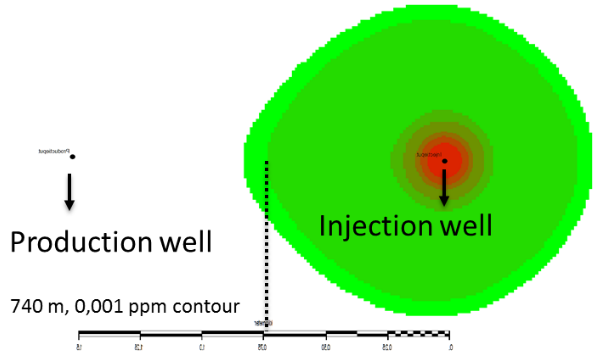

Figure 4-5: Modelling result of risk 1 where the transport of inhibitor from the injection well towards the production well is shown. The production well is not reached with a concentration of ≥ 0.001 ppm within 30 years.

Risk 2 and 3: discharge of inhibitors in fresh drinking water and brackish water aquifers

We first calculated the inhibitor spread without other injection or extraction wells inside a fresh/brackish aquifer. We did it with a natural gradient in hydraulic heads, a dosed inhibitor concentration of 10 ppm and a contour line set at 0.001 ppm. Our worst-case scenario assumes that it may take three years before corrosion on the injection site is detected via the log-campaign. Within these three years, our modelling indicates that the leakage of the inhibitor spreads with a maximum of 175 meters and a concentration of 0.001 ppm. It must be noted that the most toxic component is only for < 5% present in the inhibitor cocktail, which corresponds to a concentration of 0.05 parts per billion (ppb).

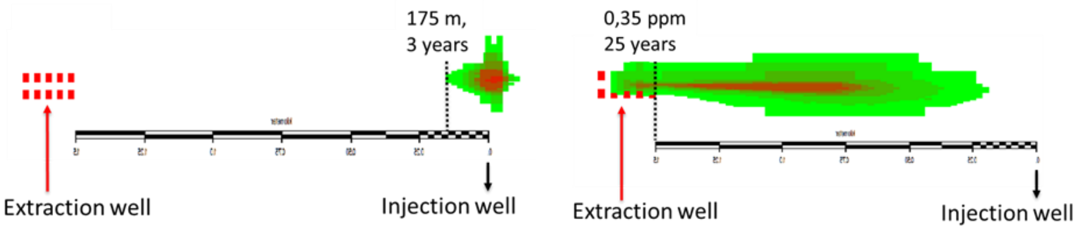

Figure 4-6: Modelling result of risk 2 (and 3) where the transport of inhibitor from the injection well towards the extraction well is shown. The inhibitors can travel up to 175 meters with a concentration of 0.001 ppm towards the extraction well within three years (left). Worst case simulations show that the inhibitor reaches the extraction well with a concentration of 0.35 ppm in 25 years (right).

The spread of inhibitors through leakage in a freshwater aquifer close to a freshwater extraction well will not directly reach the extraction well. Wells cannot be placed within the 25 years zone. The 25 years zone is the contour surrounding a freshwater extraction, where the travel path of groundwater to the well is 25 years. Modelling shows that after 25 years, the inhibitor reaches the extraction well and in the calculated worst-case scenario, the concentration is 0.35 ppm (starting concentration of 10 ppm). It must be noted that the most toxic component is only for < 5 % present in the inhibitor cocktail, which corresponds to a concentration of 0.0175 ppm. The same can be said for the release of liquid with inhibitors in a brackish water layer. Because extractions are more abundant in freshwater than brackish water, the possible impact of leakage of inhibitors in the freshwater extraction wells is larger than for brackish water.

There are plausible effects of inhibitors on ecology in both fresh and brackish groundwater. For example, it is known that inhibitors have a negative effect on biological growth. Also, the salinity and heavy metal concentrations affect the biological growth. The order and magnitude of these effects are unfortunately relatively unknown as already stated before, which is why more research on this topic is necessary.

4.4 Future insights

Up to date, carbon steel based materials combined with the use of inhibitor has mostly been the default choice for well construction. However, when considering the full life-cycle and the MOC-method, the mechanical well design should be the first priority for maintaining geothermal well integrity. Also, the material choice can be of great importance when it comes to corrosion prevention. Carbon steel based materials are the lowest capital cost option but are more susceptible to general corrosion than high-grade materials such as 13% chrome steel (13Cr) or super duplex 2507. To illustrate this, we performed a simulation experiment to evaluate general corrosion rates of various materials with OLI stream analyzer (Figure 4-7). (18) The simulated corrosion rate for standard carbon steel is 1.5 mm/year, 13Cr steel has a corrosion rate of 0.014 mm/year, and super duplex 2507 has a corrosion rate of 0.001 mm/year. As the maximal accepted corrosion rate is 0.075 mm/year, both chromium and duplex materials may be good alternatives to the currently used carbon steel. (4), (5) Unfortunately, the 13Cr material is thought to be more susceptible to pitting corrosion, and solid super duplex pipes are very expensive. For future geothermal projects in the Netherlands, the sector, however, must look for alternative materials such as super duplex 2507, (duplex) coated carbon steel or glass-reinforced epoxy (GRE). (3), (19)

In addition, also new investigations for early warning systems to protect the groundwater around geothermal wells are in place.. KWR proposed a monitoring system based on glass fiber and EM measurements, which can detect leakage of the production/injection site in an early stage. (20) Currently, KWR and Witteveen+Bos together investigate if this proposal is a feasible option in terms of design, applicability, costs and benefits.

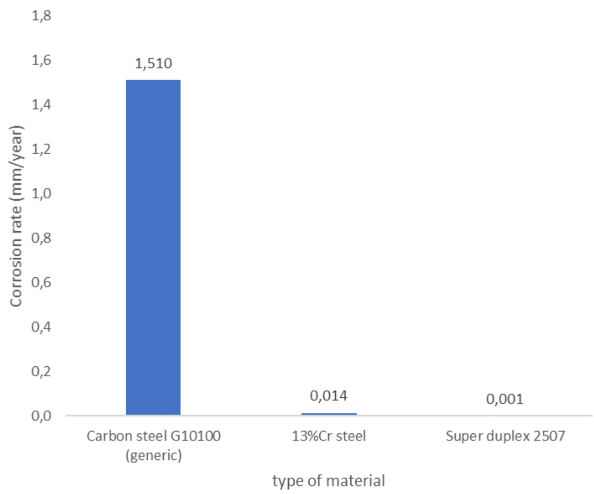

Figure 4-7: Corrosion rate simulation with OLI stream analyzer (18) with three materials under the same conditions. The carbon steel has a high corrosion rate (1.5 mm/year), where the chromium enriched steel (0.014 mm/year) and the super duplex 2507 (0.001 mm/year) show a much lower corrosion rate.

[a] Conditions: Flow: 250 m^3/h, Temperature: 90 °C, Pressure: 20 bar, Diameter: 35 cm, water-gas ratio: 1, ions (mg/L): Sodium (45000), Potassium (650), Calcium (6000), Magnesium (1000), Strontium (350), Barium (5), Iron (80), Lithium (10), Chloride (80000), Sulfate (330), Bicarbonate (10); gasses (mol%): Carbon dioxide (5,0), Nitrogen (1), Methane (90), Ethane (1.5).

5. RECOMMENDATIONS

The following recommendations are made from the analysis of mitigating measures:

1. Drafting of a structural used corrosion management plan for the whole geothermal sector and processing in a risk-based inspection plan.

2. Set up a quality control of the corrosion management plan (audit).

3. Set up monitoring to determine the integrity of injection wells.

4. Drafting of European guidelines for inhibitors under anaerobic conditions.

5. To conduct research on the following inhibitor-related topics: the optimal concentration of inhibitor throughout the installation, the amount of inhibitor that reaches the reservoir, the degradation of inhibitors (incl. toxicity of degradation products), the diffusion rate of inhibitors and the effect of (degradation products of) inhibitors on the environment.

6. In addition, research into the use of more corrosion-resistant materials is recommended. as a result of which the dosage of inhibitors is expected to reduce.

6. CONCLUSIONS

Inhibitors, as applied in the existing Dutch geothermal facilities, have a positive effect on the environment considering the currently used carbon steel based materials. Their protective effect against corrosion result in a reduced risk of leakage. In addition, precipitation of radioactive lead in the above-ground installations is also reduced.

However, there are three risks identified of possible leakage of inhibitors on the injection site. We performed simulations with the worst-case approach, which shows that the simulations are overrated. The simulations for the first risk, leakage of inhibitor in the reservoir, shows that in a worst case situation the inhibitor travels 740 m with a concentration of 0.001 ppm and does not reach the production well over a period of 30 years. For the other two worst case scenarios', leakage to sweet or brackish water layer, inhibitors can be found with a maximum concentration of 0.35 ppm of inhibitor (0.0175 ppm toxic component) at a drinking water well after 25 years. This value is higher than the signaling value of the Dutch Drinkwaterbesluit, and therefore the operator of a geothermal facility should monitor and / mitigate the risk. Based on the generic geohydrological modelling, it can be stated that there is no effect of injection of inhibitors in the geothermal reservoir on other geothermal extraction wells. Other extractions from fresh or brackish aquifers can be influenced by leakage of inhibitors in the reservoirs. It should be noted that the degradability of (breakdown products of) inhibitors under geothermal conditions in reservoirs or aquifers is unknown and that this should be better investigated. However, corrosion inhibitors have a biocidal activity and therefore impact on the subsurface biology is expected.

We are happy to see that the sector is already acting upon the findings of our investigations, as processes for better monitoring are in the pipeline and alternative well materials are recently implemented in some installations in The Netherlands.

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