EXPERIMENTAL AND THEORETICAL OBSERVATIONS OF SCALING RATE AND IMPLICATIONS FOR ANTI-SCALING PRACTISES

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Keywords: *geothermal power; above-ground plant; binary plant; silica scaling; stibnite scaling; colloid transport; brine chemistry; brine acidification; acid dosing; scale prevention; reinjectivity*

ABSTRACT

Mineral scaling in wells and power plant facilities is one of the main obstacles to increasing the effectiveness of utilization of the limited geothermal resources. Brine acidification has had considerable success in controlling silica polymerization and deposition in above-ground equipment. Organic inhibitors have also been trialed. Meanwhile, fouling by stibnite and loss of well injectivity can be persistent problems, particularly in binary or flash plant operation.

In this paper we present a summary of the experimental and theoretical studies of the fouling phenomena conducted at the University of Canterbury. An improved understanding of the interrelation between the chemical and hydrodynamic conditions of the scaling process and its dominant mechanisms was achieved. This contributes towards optimization of the anti-scaling and maintenance measures.

1. INTRODUCTION

1.1 Scaling and its control

The rate of mineral scaling in geothermal systems is significant when dissolved minerals separate from the fluid phase and polymerize into colloidal particles (Dunstall et al., 2000). This may take place when extraction of heat (or loss of CO_2 in the case of calcite) from geothermal fluid significantly decreases the minerals' solubility. Once the particles have formed their deposition, governed by their stability and transport to the walls, constitutes the major part of the scaling process.

Currently, scaling by one of the most abundant minerals in the geothermal fluid – silica – is controlled by acidifying the geothermal brine (Yoshitaka et al., 2013). An acid is added to the brine with the aim to lower its pH to 4-5. This prolongs the nucleation stage of the colloidal silica formation (Kokhanenko et al., 2012). As a result, effectively no colloidal particles form while the brine flows from the production to the re-injection wells.

Under the normal operation of the acid-dosing system no major scaling by deposition of the colloid silica occurs in the aboveground equipment (Addison et al., 2013). Meanwhile, deposition of silica is still likely to occur in reservoir formation. This can lead to a decrease in injectivity. In addition, there is a high risk of silica deposition in the re-injection wells and aboveground pathways (heat exchangers, injection pipe) during an abnormal operation of the acid-dosing system (e.g. start-up of the dosing plant) (Addison et al., 2013).

Moreover, the deposition of another dissolved mineral – stibnite – can contribute to the mineral scaling problem through a similar colloidal mechanism of deposition. In this case, the formation of the colloidal stibnite is accelerated by brine acidification (whether this acidification occurs by addition of an acidic concentrate or of a sulphuric acid). Even though this mineral is present in a lower concentration than silica still, over time, it can accumulate on the heat-exchanger surfaces and cause a sensible increase of their thermal resistances. Therefore, periodic cleaning of the heat-exchangers is required to maintain their efficiency. This naturally may affect the operational costs of the binary power plant.

In this paper we analyze general features of mineral scaling by colloidal deposition. The scaling rate, in this case, can be expressed as a product of the colloid stability and transport rate (Kokhanenko et al., 2013). The stability is the inverse of the probability that a particle bonds with another particle or with a stationary surface upon collision. It is determined by the chemical properties of the suspending fluid and particle size. The transport of the particles, depending on their size, can be dominated either by diffusion or by inertia. For both mechanisms the transport rate also depends on the flow and surface parameters. Below, we review the details of these relationships and draw practical conclusions.

1.2 Experimental studies of the colloidal silica deposition

A number of measurements of the geothermal scaling rate were conducted in different geothermal fields (Mroczek, 1994). The reported scaling rates, presented in Table 1, reflect variability in the corresponding hydrodynamic and chemical conditions.

Table 1: Field and laboratory scaling rates

Field/Experiment name	Silica scaling rate, $\times 10^{-7}$ kg/s/m^2
Wairakei, NZ	3.3
Ohaaki, NZ	0.42
Hveragerdi, Iceland	1.1
Verkhne-Mutnovskaya, Russia	25
Cylinder in a crossflow (max): 125nm particles	0.42
Pipe scaling experiments: 40 nm particles	0.17

While these first experiments provided insight into the effects of the flow rate and brine oversaturation on the scaling rate, they also lead to the contradictory conclusions. Therefore, laboratory experiments were designed and conducted both with the natural geothermal brine (Dunstall et al., 2000) and synthetic colloidal solutions (Kokhanenko et al., 2012, 2013, 2014) in order to improve the repeatability of the results and study the details of the scaling process.

Silica deposition onto carbon steel cylinders in a cross-flow of natural geothermal brine was studied by Dunstall et al. (2000). The deposition rate was found to increase with the particle size and flow rate (Fig. 1). In addition, the silica colloids formed ridges of scale parallel to the cylinder axis and perpendicular to the flow direction. The height of the silica ridges was found to vary significantly around the cylinder circumference. The maximum height of ~0.25 mm was measured at approximately 21° from the upstream stagnation point.

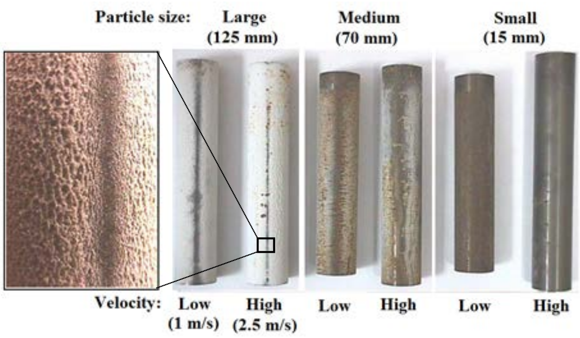

Figure 1: Geothermal silica scale on cylinders in a cross flow (Dunstall et al., 2000)

The rate of deposition of 125 nm particles from the flow with a bulk velocity of 2.5 m/s was calculated in (Kokhanenko et al., 2013) from the scale height distribution reported by Dunstall et al. (2000). The average deposition rate was evaluated to be $(1.6\pm0.6)\times10^{-9}$ kg/s per 1 m length of the ø25mm cylinder or $(4.2\pm1.6)\ \times10^{-8}$ kg/s/m^2. The dimensionless deposition velocity found by dividing this averaged value by the bulk concentration of the silica particles (c_p^0= 0.25 kg/m^3) and by the average shear velocity on the cylinder surface ($\upsilon_0 = 0.14\ \mathrm{m/s}$) was 1.2×10^{-6}.

Experimental studies of the silica scaling in mild steel pipes (Kokhanenko et al., 2012, 2013, and 2014) revealed similar particle size and flow rate effects. The higher deposition rate was associated with larger particles and higher flow rates. In particular, for 40 nm particles, the scaling rate was directly measured (by weighing the deposits) to be $1.7\ \cdot10^{-8}$ kg/s/m^2. When normalized with the corresponding shear velocity ($\upsilon_0 = 0.17\ \mathrm{m/s}$) and concentration of the silica particles (c_p^0 = 1.6 kg/m^3) this gave a dimensionless deposition rate of $6.3\ \cdot10^{-8}$.

Both the cylinder-in-cross flow and pipe scaling rates are presented in Table 1. They are generally smaller than the field scaling rates. Most likely, this is due to the much higher flow rates, and corresponding Reynolds (Re) numbers, observed in the power plant flow paths.

Similar to the cylinder in the cross-flow experiments, the deposition of silica in pipes was highly non-uniform. The growth of numerous relatively large (0.1-1mm) protrusions and a few yellowish deposits in the areas in-between them (Fig. 2) were observed. At high flow rates the deposition of larger silica particles formed spanwise consecutive ridges inclined towards the oncoming flow. These “tower” type protrusions most probably were formed by inertial deposition. The inertial mechanism of deposition is expected to cause faster growth of the leading edge of a protrusion. The diffusion, in turn, dominates the particle transport behind them, in the wake zones, where mixing and, therefore, mass transport are significantly accelerated.

In addition, this specific mechanism of particle transport by diffusion and inertia was confirmed by the scale distribution observed in the pipe scaling experiments (Kokhanenko et al., 2012, 2013, and 2014). Particularly, the rate of deposition increased with the distance from the pipe inlet – an unexpected outcome if particles are transported only by diffusion onto a smooth surface. In this later case, flow development along the pipe causes growth of the hydrodynamic and diffusion boundary layers. This must decrease the rate of particle transport by diffusion, and consequently decrease the scaling rate with the distance from the pipe inlet. The growth of the scale ridges on the pipe surface changes this by disturbing the boundary layer, decreasing its thickness and increasing particle transport downstream.

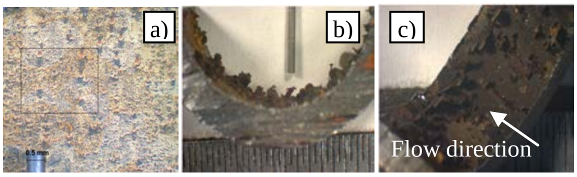

Figure 2: Silica scale on the internal surface of a carbon steel pipe: a) small particles/low flow rate and b, c) big particles/high flow rate

1.3 Stability of Colloidal Silica

The effects of the ionic strength (IS) and pH of the colloidal solutions on their long term behavior were investigated in our recent work (Kokhanenko et al., 2014). The particle growth for the conditions representative of the geothermal brine was observed with the dynamic light scattering (DLS) and electron microscopy. The systematic analysis of this data allowed us to determine that silica nanoparticles grow by reaction-limited cluster aggregation. The corresponding aggregative stability (W) was measured to be significantly higher $(10^8 < W < 10^9, \text{see Fig. 3})$ than $10^0 < W < 10^1$ predicted by the standard theory of colloid interactions established by Derjaguin and Landau (1941) and Verwey and Overbeek (1948). This DLVO theory describes the interactions between colloidal particles as a superposition of the repulsive electrostatic and attractive dispersive (molecular) forces by implementing the classical mean field approach. The detailed discussion of the DLVO theory can be found in Adamczyk et al. (1990) or more briefly in our previous publications (Kokhanenko et al., 2012, 2013, 2014).

The relationships between the aggregative stability and such parameters as the particle size, ionic strength and pH of the solution established experimentally in our previous paper (Kokhanenko et al., 2014) were compatible with the theory of the colloid stabilization by steric interactions (Kobayashi et al., 2005). The model of the “gel” layer, a centerpiece of this theory, was used to explain some of the discrepancies between the previous theory and observations of the colloid silica behavior.

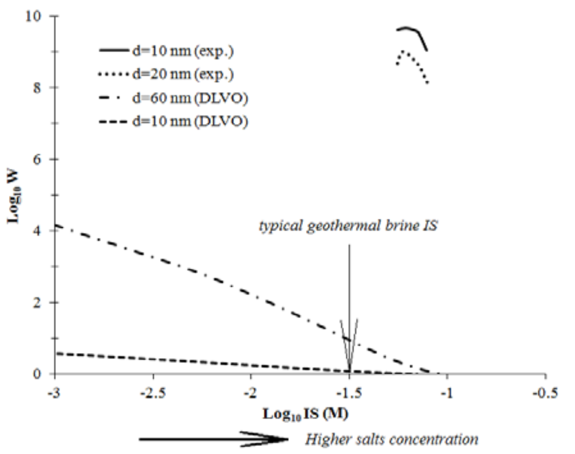

Figure 3: The experimental and theoretical stability (W) of the colloidal silica as a function of the solution ionic strength (IS) at pH=7.5

In contrast to the standard DLVO theory, the stability was determined to be higher for smaller particles. More complex dependence from the solution IS was revealed. On one hand, a decrease of the IS (as well as increase of pH) causes swelling of the "gel" layer, and consequently an increase in stability. On the other hand, it also accelerates the processes responsible for the rearrangement of the aggregates' surface curvature that effectively leads to a decrease of their stability.

The stability of silica colloids was found to be higher at higher solution pH. The increase of pH from 7.5 to 9.5 caused particle growth by aggregation to virtually cease in our experiments. The DLVO theory extended to include the steric mechanism of particle stabilisation predicted at least a twofold increase of colloid stability in the alkalized solution.

The pH dependence of the silica solubility in water was identified to cause specific effects on the colloid stability. Thus, the transient processes of the "gel" layer dissolution – re-growth caused by the change of the solution pH was identified to be responsible for the narrowing of the particle size distribution observed on this final stage of the sol study.

Finally, it can be shown that the aggregative stability of the silica colloids discussed above is approximately equal to the stability in the particle-wall collisions. Indeed, any surface with even a minor amount of accumulated scale cannot be considered as smooth. New incoming particles interact with the previously deposited ones. However, over time those particles that bonded to the wall become further cemented by precipitation of the monomeric silica (and/or deposition of smaller particles) in the gaps between them. This process tends to change local curvature of the scaled surface and thus the probability of the particle to attach to it. Coincidentally, the same process proceeds with the reaction limited aggregation of colloidal silica. Therefore, as a first approximation, the probability of particle adhesion to a wall can be determined as the inverse of the aggregative stability discussed above.

1.4 Particle transport mechanisms and the role of surface roughness

An extensive review of the current theory of particle transport onto a rough wall in turbulent flows was given by Guha (2008). The particles are transported by the two main mechanisms (Fig.4): diffusion and convection (the latter is sometimes called inertial transport). The non-dimensional particle deposition velocity ($V_{dep}^{+} = j/c_p^0 \upsilon_0$) is characterized by a different relationship to the particle size d_p (or particle relaxation time: $\tau_p^{+} = \frac{\rho_p d_p^2}{18\rho_l}\left(\frac{\upsilon_0}{\nu}\right)^2$) depending on which transport mechanism is dominant. The non-dimensional deposition velocity V_{dep}^{+} is the wall particle flux j normalized by the bulk concentration of particles c_p^0 and the fluid friction velocity $\upsilon_0 = \sqrt{\varepsilon/\rho_l}$ determined from flow conditions (here ε denotes wall shear stress). The particle relaxation time includes the particle ρ_p and water ρ_l densities (here $\rho_p = 1500\ kg/m^3$) and the water kinematic viscosity ν.

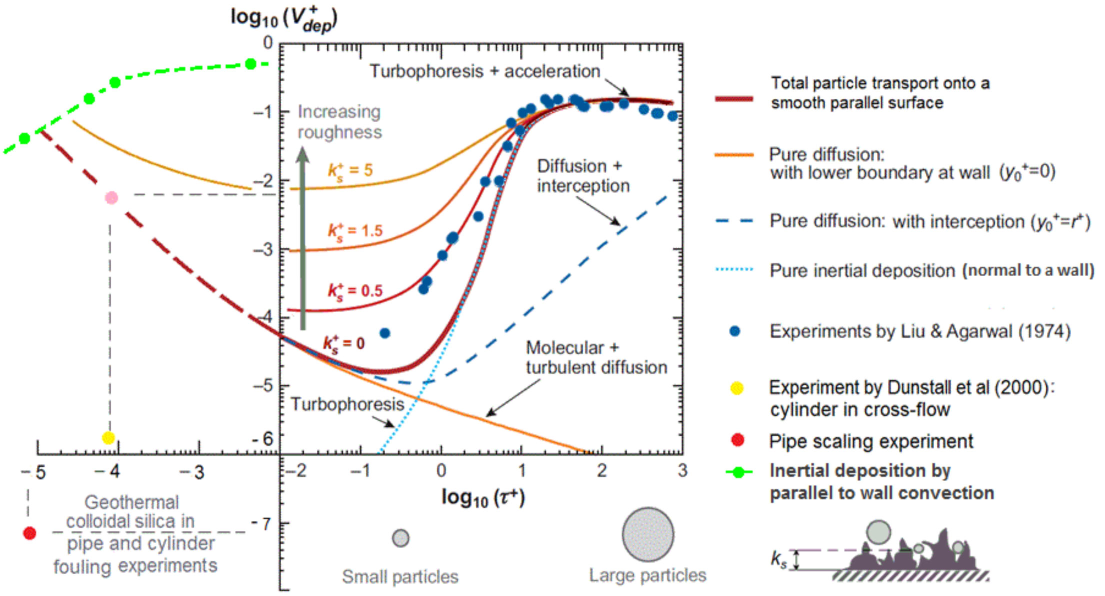

Figure 4: Classification of the particle transport mechanisms (Guha, 2008) and rate of the additional convection (Kokhanenko et al., 2014)

Smaller particles follow the fluid motion more closely than bigger particles, thus as they get closer to the wall they lose the y-component of their convective velocity much faster than bigger particles. For very small particles this eventually leads to the existence of a thin region close to the wall in which particle transport continues only by Brownian diffusion. This region is called the diffusion sublayer.

Guha (2008) has also shown that the particle deposition velocity is significantly affected by the roughness of the surface to which particles are transported (Fig.4). Real walls, having roughness elements protruding from their surfaces, experience higher mass transfer than ideal, perfectly smooth walls. The particles need to be transported through the diffusion sublayer over shorter distances to reach the roughness elements which have an effective height k_s. This increases the transport rate of smaller particles for which this diffusion sublayer exists (see Fig. 4 for $log_{10}\tau_p^+ \leq 0$).

The dimensionless scaling velocity measured in two experiments (Dunstall et al. 2000, Kokhanenko et al. 2012) was from 4 to 7 orders of magnitude smaller than V_{dep}^+ predicted by the particle transport theory for the corresponding values of τ_p^+ (Fig.4). This difference can be explained by the moderate stability of the colloidal silica discussed above. The theoretical scaling rate calculated as a product of the transport rate $V_{dep}^+$and attachment probability ($10^{-6} < [\ k = \frac{1}{W}] < 10^{-4}$) was found to be smaller than the corresponding experimental rates (Table 2).

Moreover, for the relevant particle size range, the theory of particle transport predicts a significant decrease of the deposition velocity with increasing particle size. The experimental scaling rates, however, revealed the opposite - faster scaling for larger particles. Scaling required both transport to the surface and attachment, and the experimental observations can be explained by the increase of the attachment probability with particle size. However, the standard DLVO theory predicts the opposite – higher stability for larger particles, whereas the theory of electrosteric stability may support this (see Fig.3), although it is not developed well enough to make a reliable conclusion regarding the stability dependence from the particle size.

Table 2: The experimental and theoretical scaling rates

Particle size	Experimental scaling rate, [1, 7]	k, [8]	k× [Transport rate in [9]]	k× [Transport rate in [9]+ additional convection]
125 nm	$1.2{\cdot}10^{-6}$	10^{-4}	$5{\cdot}10^{-7}$	$3.4{\cdot}10^{-6}$
40 nm	$6.3{\cdot}10^{-8}$	10^{-6}	$3{\cdot}10^{-9}$	$3.2{\cdot}10^{-9}$

A second possible explanation is that current theory of particle transport does not include all relevant transport mechanisms. We hypothesized in (Kokhanenko et al., 2013) that the inertia of particles in the diffusion dominated size range, although it has insignificant effect on convection normal to a wall, still can promote convection in the tangential (parallel to the wall) direction onto roughness elements protruding from the wall.

This hypothesis also explains the spatial distributions of the scale observed in the deposition experiments with the cylinders (Fig.1). The scaling rate can be higher at the locations with higher wall shear stress due to, first, smaller thickness of the diffusion boundary layer here, which makes the effect of existing surface roughness higher. Second, at the locations with higher wall shear stress particles have higher tangential velocity and thus more significant additional convection.

We have evaluated the tangential component of the transport (Kokhanenko et al., 2013) by first, finding particle flux onto an upstream side of a single scale protrusion. As expected it was found to increase with the particle size and flow rate (green data points in Fig.4). Next, it was multiplied by the experimentally derived surface density of the protrusions to give the estimated transport rate for this mechanism.

This simplified approach showed that additional convective particle transport is at least of the same order of magnitude as the total transport rate calculated in Guha (2008) (see Fig.4). The rate of this additional convection was shown to increase with the particle size. Its magnitude is significant enough to explain the discrepancy between the experimental and theoretical scaling rates for larger particles (Table 2). Therefore, this specific inertial mechanism of particle transport should be included into future numerical silica scaling simulation.

In addition, current theory of the particle transport outlined by Guha does not consider the effect of the roughness elements on the diffusion of the particles onto the surface patches in-between the protrusions. As discussed above the protrusions generate wake regions with more intense turbulent pulsations and mixing downstream from their location.

2. DEVELOPMENT OF THE ANTI-SCALING PRACTICES

Taken together, the experimental observations and theoretical calculations strongly suggest that such parameters as surface roughness and average colloid particle size have significant effects on the rate of the scaling process. The following discussion is general and applicable to any fouling mechanism with a predominantly colloidal mechanism of deposition.

Observations of particle transport in situations relevant to power plant (highly turbulent flow and rough wall surfaces) suggest that the roughness of the scale can be used as a maintenance criterion. According to our findings (Kokhanenko et al., 2013) the onset of the inertial deposition of the particles leads to a rapid growth of the roughness elements initially present on a wall surface. A random ensemble of these protrusions gets slightly more material deposited and due to the positive feedback – the greater the roughness the faster the deposition – they grow into more prominent protrusions.

These protrusions can arrange into sequential, equally spaced rows spanwise to the flow, at least under certain conditions, and we suspect an as-yet unidentified hydrodynamic cause for this. Once the protrusions grow beyond the viscous boundary layer and penetrate into the turbulent boundary layer they significantly increase rates of momentum, mass and energy transport in the near wall region. Along with the increase of the pressure drop this can result in a temporary increase of the heat transfer in heat exchangers. Over time, the accelerated fouling will surely lower it.

Therefore, if currently the decision to start heat exchanger maintenance is made based on the observed decrease of its efficiency, this new understanding may help develop more efficient maintenance criteria. Namely, instead of starting heat exchanger cleaning after a continuous film of contaminant accumulated on the surface and its efficiency dropped significantly to observe it through the temperature readings it may be preferable to clean it before the roughness protrusions grow big enough to accelerate the fouling. This may require a shorter time interval between cleaning cycles, but because much less deposit will need to be removed, an overall shorter idle time in heat exchanger operation can be achieved. Variations in the pressure drop across the heat exchanger, as well as its efficiency, may be proved useful in determining the optimal moment for the start of the heat exchanger maintenance.

When only very small particles are present in the brine, the rate of inertial transport is small and particle deposition is controlled by diffusion. Similar to the case of large particles discussed above, the positive feedback mechanism due to scale roughness growth exists - the colloidal particles need to be transported over a shorter distance, normal to the wall, to reach the protrusion. In this case the resultant scale ridges are seen to be smaller and more closely spaced. The transport to the protrusions and flat surfaces in-between is due to the same diffusion mechanism. These smaller protrusions still cause an increase of the diffusion transport downstream of their location, though not as significant as in the inertial deposition mode.

So, with inertial deposition the steady-state height of the scale protrusions is reached when inertial particle flux onto the protrusions is equal to the diffusion flux to the flat surfaces in between (which is accelerated by these protrusions). While for diffusion controlled transport, the steady-state scale roughness is established when diffusion flux onto protrusions is equal to the diffusion onto a flat surface (also enhanced by the presence of the roughness elements).

The morphology and hardness of the scale formed in our lab experiments were observed to depend on the particle size. Larger particles built up as a softer, more rough and non-uniform scale. This effect must be caused by the lower stability of the larger particles and reflects the similarity to the process of kinetic aggregation. Similarly, in the case of reaction limited aggregation, the higher stability of the particles means that they experience more collisions before they attach. This allows them to sample more locations on the surface of a wall (or an aggregate) before attaching and forming a denser scale (or an aggregate with a higher fractal dimension). Therefore, while smaller particles are less prone to deposition, due to their higher stability and less significant convective transport, they deposit as a denser, harder to clean and more uniform scale.

Particularly intense scaling could take place in pipe bends and valves (Mroczek, 1994). Higher shear stress and flow recirculation characteristic for these locations must cause this. The former determines higher transport rates and the latter implies that fluid resides much longer there, which allows the particles to grow larger and, as a result, to have higher likelihood to attach to a wall. For particularly critical cases, fouling at such locations can be mitigated by implementing active flow control. It may be feasible to stimulate ejection of the fluid vortices from the recirculation zones into the main flow. This can both decrease the residence (growth) time for the particles and reduce the average shear stresses on the equipment walls.

As mentioned earlier the size of the colloids effectively governs the rate of the above ground scaling processes. Moreover, it may be presumed to have a similar effect on the injectivity index decline. Larger colloidal particles, because of their lower mobility and stability, are expected to become trapped in the pores at a higher rate. Smaller particles suspended in brine can travel further away from the reinjection point into the formation. This increases the probability of their dissolution due to the brine reheating before they can affect the permeability of the formation. Therefore, it is expected that a brine with more numerous smaller particles will cause slower fouling and loss of injectivity than a brine with the same total oversaturation ratio but fewer, larger particles.

Up to this point we have considered the situations in which colloids formed in the heat exchangers, reinjection pipelines or wellbores. For certain conditions, formation of colloidal silica particles can also occur once the brine has reached the formation. If the pH of the acid-treated injected brine returned back to neutral (for instance by reaction with calcite) faster than its temperature then nucleation and consequent growth of the particles may start.

It is possible to control the brine parameters to affect the size distribution of the formed particles. For example if the brine is forced to reach its final oversaturation ratio more rapidly the corresponding nucleation process consumes more of the excess silica (or stibnite) and less of mineral is available for the later particle growth stage. This can be achieved by flashing the brine (Mroczek et al., 2013) or by running it through large plate heat exchangers.

Effectively, such brine would have many smaller particles and would cause less fouling in the aboveground and underground pathways. Whereas, in the situations where final oversaturation of the brine is reached gradually, as in the slow recovery of pH in the reinjected brine mentioned above, prolonged nucleation and slower growth may yield larger, more polydisperse particles.

3. CONCLUSIONS

A systematic analysis of the colloidal mechanism of scaling processes in geothermal systems was performed. The summary of the previous experimental and theoretical studies allowed us to establish the effects of the brine chemistry (pH and ionic strength), size of the colloidal particles present in the brine and flow parameters on the rate of scaling. By analyzing its two governing processes, transport and attachment of the colloidal particles to a surface, the rate of scaling was shown to be higher for: 1) larger particles, higher ionic strength and lower pH of the brine and 2) higher roughness of the surface.

A number of practical implications for the anti-scaling practices were drawn. First, the size of the scale roughness observed through the variations in the pressure drop and heat fluxes across the heat exchanger can be used as a key criterion for optimal maintenance cycle design.

Second, the deposition of smaller and more stable particles was found to form harder scale. Therefore, for some cases a compromise between the ease of cleaning and the rate of fouling could be made by controlling the particle size or brine pH and ionic strength.

Third, active or passive flow control can be used to mitigate scaling in the critical locations like pipe bends and valves.

Finally, the reduction of the formation permeability due to scaling brought about by the reinjected brine can be decreased by fine tuning the operating parameters during the colloid formation phase. For instance, if the brine is forced to reach its final mineral oversaturation faster it will form smaller particles that are less likely to cause formation pore occlusion.

ACKNOWLEDGEMENT

The authors are grateful to Mighty River Power for the generous financial support of this research.

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