

## DISSOLUTION RATE OF SODIUM SALT SCALES IN FALLING FILM EVAPORATORS

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### ABSTRACT

In pulp mills, fouling can be a significant problem in the black liquor evaporation plant. Part of the black liquor evaporation plant works as a crystallizer where large amounts of sodium carbonate and sodium sulphate will saturate and precipitate.

In this work, the dissolution rate of sodium scales was experimentally investigated and modelled. Experiments were conducted in a pilot falling film evaporator where a sodium carbonate and sodium sulphate solution was evaporated on the outside of a vertical stainless steel tube to form the scales. The aim was to relate the dissolution rate to wetting degree (i.e. the mass flow rate per unit circumference) and temperature of the wash liquid.

An effective experimental method to measure the removal of sodium scales in falling film evaporators was developed and a 1<sup>st</sup> order model with respect to the difference in mass fraction between the scaling layer surface and the bulk of the wash liquid was shown to fit the experimental data well.

### INTRODUCTION

Fouling is an important issue for the pulp and paper industry. One of the most important problem areas is in the so-called black liquor evaporation plant. Black liquor, which is a residual stream from the pulp production process, needs to be recycled for both environmental and economic reasons. In the liquor recovery cycle, the black liquor is concentrated in a multistage evaporation plant prior to combustion in a recovery boiler where the chemicals needed for the pulping process are regenerated and, at the same time, steam is produced.

Black liquor is a complex viscous alkaline solution of organic and inorganic components. Fouling of the evaporators is mainly caused by the precipitation of inorganic water-soluble sodium salts present in the black liquor. The sodium scales are mostly in the form of double salts of sodium carbonate and sodium sulphate. The Na<sub>2</sub>CO<sub>3</sub>-Na<sub>2</sub>SO<sub>4</sub> system will form different salts depending on the sodium carbonate/sodium sulphate ratio and also depending on the temperature (Shi et al., 2003, DeMartini, 2007). The crystal species generally encountered in black liquor evaporators are either Burkeite or Sodium Sulphate Dicarboxylate and these crystals have different propensity to form scales on the heat transfer surface (Gourdon, 2009). Previous research has mainly focused on how to prevent

fouling by optimizing operating conditions rather than how to efficiently remove scales (Euhus et al., 2003, Haag and Stigson, 2003, Frederick Jr. et al., 2004, Gourdon, 2009).

Cleaning of the evaporator surface is, of course, also an important part of the pulp mill operation. In the black liquor evaporators with the most severe fouling, the cleaning is frequently performed during the continuous operation of the mill regardless of the degree of fouling. Today, however, very little fundamental knowledge exists of how to remove scaling and the cleaning process is based on heuristics (Schmidl and Frederick, 1998). The key factor to achieve an efficient cleaning cycle is to reduce the concentrations of the scaling species in the wash liquid below the solubility limit and to maintain those low concentrations for a sufficient period of time. No general data have been found regarding how long this period should be. Thus, there is a need for basic knowledge of the dissolution process of sodium salts in order to create general guidelines regarding efficient evaporator washing.

The removal mechanism during cleaning can, in general, include both dissolving the crystal deposits and mechanically removing the crystals either by breaking the adhesion bonds between the crystal and the surface or by breaking the cohesion bond between crystals. The thermodynamic driving force of the dissolution is the concentration difference between the concentration in the wash liquid and the solubility. The solubility, in turn, depends on the temperature and the composition of the fluid. The mechanical removal of deposits is dependent on physical properties such as temperature and flow rate (Plett, 1985).

A more efficient washing can reduce the time needed for cleaning and reduce the use of wash liquid as well. A good washing model can also provide a reliable way to determine when the heat surfaces are clean.

Our laboratory has been engaged in fundamental studies of the black liquor evaporation process, both concerning heat transfer and fouling mitigation for the past several years (e.g. (Johansson et al., 2009, Gourdon et al., 2010)). That work has been focused around a pilot falling-film evaporator facility, described below.

In this paper the dissolution of sodium carbonate and sodium sulphate salts was investigated. Experiments were performed in a falling-film facility and the dissolution rate of a scaling layer formed on the outside of a tube was studied under different operating conditions. In this study

the parameters of interest were the washing liquid temperature and wetting degree (i.e. flow rate per unit circumference). The goal was to produce a washing model that relates the rate of dissolution to the studied parameters.

### Dissolution of Solids

The dissolution of a solid into a liquid film is a common process and the rate of solid removal is generally described as a first order reaction with the difference in concentration between the solid-liquid interface and the bulk of the liquid as the driving force (Kramers and Kreyger, 1956):

$$\frac{dm}{dt} = kA(c_i - c_b) \quad (1)$$

The total mass of the solid species  $m$ , in this case the scaling layer, will decrease in time as mass is dissolved and transported from the surface. The driving force for the mass transport is the difference in concentration between the solid-liquid interface,  $c_i$ , and the bulk of the solvent,  $c_b$ .  $A$  is the surface area of the solid-liquid interface and  $k$  is a rate constant. Similar expressions where the transport coefficient  $k$  is expressed as the diffusion coefficient divided by the boundary layer thickness were developed over 100 years ago (Noyes and Whitney, 1897, Nernst and Brunner, 1904) and are still used. Common for these expressions is that the dissolution process is assumed to be limited by the diffusion rate of liquid solvent to the solid crystal which is broken up (Dokoumetzidis and Macheras, 2006). Equation 1 can also be interpreted to describe the counter-diffusion of dissolved ions from the crystal lattice out in the solvent (Murray, 1987, Grant et al., 1996, Grant et al., 1997).

### MODELLING APPROACH

In this work, two models for the removal of the sodium carbonate-sodium sulphate crystal scaling layer from the outer surface of a tube were experimentally evaluated for the dissolution process. The simplest approach was a 1<sup>st</sup> order model, where the hypothesis was that the dissolution process only depends on diffusion and was therefore modeled with a first order expression (Equation 1). In the second model suggestion, the diffusion-reaction model, a reaction term was added to account for the transformation of crystals into ions. Both models assumed a bulk flow that was perfectly mixed. The models did not include the sodium carbonate's equilibrium with sodium-hydrogen-carbonate and does not take into account any effects of the pH-value. Literature data from Shi et al. (2003) for the solubility limit of the double-salts,  $\omega^*$ , was used in this study.

#### 1<sup>st</sup> Order Model

In the 1<sup>st</sup> order model the rate determining dissolution mechanism was considered to be the diffusion of ions from the surface of the scaling layer through a boundary layer and into the bulk flow (Figure 1). When the dissolved scales have diffused through the boundary layer they were regarded as part of the bulk flow, thus resistance of mass transport in x-direction could be neglected in the bulk and the bulk concentration (or mass fraction) would be constant. The mass fraction at the solid-liquid interface was assumed

to be constant and equal to saturation at the current temperature.

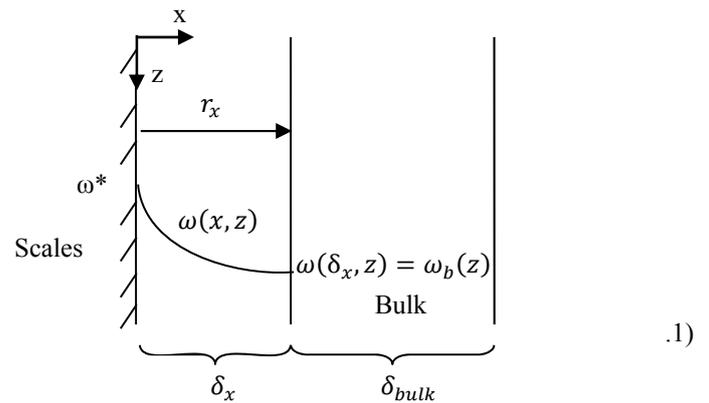


Fig. 1 Schematic representation of the mass fraction profile in the boundary layer between the solid scales and the wash liquid.

The dissolution of the scales into free ions in the wash liquid are dependent on the diffusion rate and if there are any scales left on the tube to be dissolved. The dissolution rate is modeled as follows:

$$r_x = K(\omega^* - \omega_b) \quad (2)$$

$K$  is a transport coefficient in  $\text{kg/m}^2\text{s}$  that takes into account the transportation of dissolved scales from the scaling layer to the bulk flow and thus include the boundary layer thickness  $\delta_x$ . The unit of  $K$  is a result of the driving force of the dissolution mechanism being expressed using mass fractions i.e. as mass salt per mass solution.  $K$  can, however, easily be expressed with the more commonly used unit meter per second if divided by the density of the solution.

### Diffusion-Reaction Model

A second mechanism in form of a reaction expression can be added to describe the dissolution process and, in particular, the breakage of the crystal structure into ions. The film was therefore divided into a reaction boundary layer in addition to the previous diffusion boundary layer and bulk flow (Figure 2). The two boundary layers were considered to have separate mass fraction profiles and different thicknesses.

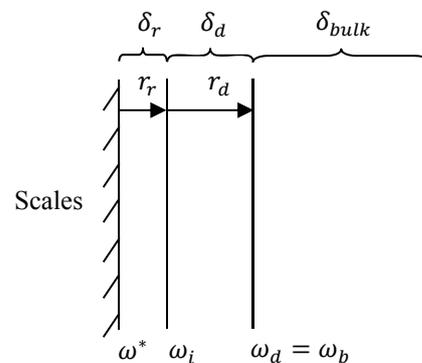


Fig. 2 Division of the falling film into two separate reaction and diffusion layers and a bulk flow.

In the reaction boundary layer, the rate limiting mechanism for the transportation of salt was assumed to be the transformation of crystals into ions and the process was described with a reaction expression:

$$r_r = K_r(\omega^* - \omega_i)^n \quad (3)$$

where  $K_r$  is a reaction coefficient. The liberated ions are then transported through the diffusion boundary layer where the transport mechanism is diffusion and modeled as follows:

$$r_d = K_d(\omega_i - \omega_d) \quad (4)$$

where  $K_d$  is a transport coefficient. The bulk was as before considered to be perfectly mixed, thus the mass fraction was constant throughout the entire bulk.

## EXPERIMENTAL EVALUATION

### Apparatus

The experimental part of this work was performed in a pilot evaporator and a schematic of the experimental set-up is seen in Figure 3. The evaporator was a falling film type, heavily instrumented and the heat transfer surface consisted of a vertical tube, 60 mm in diameter and 4.5 m long, with the falling liquid film on the outside. Further details about the research apparatus and measurement equipment are given in (Gourdon, 2009) and (Johansson et al., 2005).

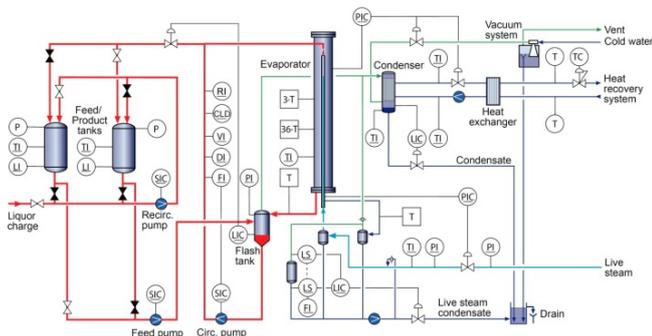


Fig. 3. Flow sheet of the research evaporator.

### Experimental Procedure

The scale build up was performed by evaporating a saline solution of sodium carbonate and sodium sulphate with a mole ratio of about 17:3. During precipitation this ratio increased, which resulted in a shift in the composition of the formed crystals. Because of this change in liquid composition, the composition of the formed crystals will also change although within the same type of crystal specie. The change in crystal composition should, however, not influence the subsequent washing if the process is only driven by dissolution as is the hypothesis in this paper.

To decrease the amount of salt needed, some of the wash liquid was reused in the following experiment. This, however, led to small differences in the molar ratio between the different experiments. The system was operated at atmospheric pressure on the solution side of the system, corresponding to 107 °C, and at a wetting degree of 1.01 kg/m s.

The density of the saline solution was raised during the course of evaporation and at a sufficient density level the first nucleation point was reached with the first precipitation of crystals. A formed scaling layer decreases the transferred heat and thus the thickness of the scaling layer could be related to the overall heat transfer coefficient. The evaporation process continued until the overall heat transfer coefficient had decreased from its initial value of about 2000 W/m<sup>2</sup> K down to approximately 500 W/m<sup>2</sup> K.

Before starting the washing sequence the tube was inspected visually and the thickness of the scaling layer was studied together with the distribution of the scales. Knowing the scale layer thickness is important and this was monitored at three different vertical positions, 0.15, 1.9 and 3.9 m from the inlet by visual observation through sight glasses (method is described in the subsequent section “Washing”).

**Washing.** For the washing, deionized water of different temperatures was used as wash liquid. A vessel of preheated wash liquid was placed above the top of the tube and the wetting degree was regulated with a control valve at the bottom of the heating barrel.

To monitor the removal of scales, video cameras were mounted at the three different vertical positions and the entire washing process was recorded. The video films supplied information of how the scales were removed, i.e. whether they were evenly dissolved or if there were parts of the tube that became clean sooner, if the flow of wash liquid covered the tube or not, when the tube was clean at the bottom and middle sight glasses. It also provided an alternative way to estimate the thickness of the scaling layer.

To further monitor the removal of scales, a device for measuring the scale thickness before and during washing was constructed and was installed at two different positions, 1.8 and 3.8 m from the inlet. The measurement equipment consists of a small flat disc that is fixed on the end of a metal rod which is pushed inwards until it reaches the surface of the scaling layer and contact is achieved. The scales are hard and thus the device should not affect the scales besides diverting the liquid flow for the short period of time of the measurement. The thickness is measured on the outside of the shell using Vernier calipers. The estimated accuracy of these measurement devices is ±0.2 mm.

To obtain the desired temperature, the hot tube was cooled with water on the inside prior to the actual washing. When the wash liquid had passed the tube it was collected in plastic containers of 1 dm<sup>3</sup>. The sampling time was recorded together with the temperature of the exiting wash liquid. When the tube was visually determined to be clean, the flow of wash liquid was stopped and the experiment ended. The temperature, density and weight of the outgoing wash liquid were collected. The salt mass fractions were measured by determining the dry solids mass fraction for a few of the 1 dm<sup>3</sup> samples. These measurements were then the basis of a correlation between mass fraction and density which was used to estimate the mass fraction in the outgoing wash liquid for the rest of the samples.

The temperature of the wash liquid was experimentally limited to 85 °C and, since the salts have poor solubility below 40 °C, the range from 45°C to 85 °C was investigated. This interval is also in the region of the warm and hot water systems at pulp mills. The wetting degree was limited to a maximum of 0.43 kg/m s. Insufficient wetting occurs below 0.12 kg/m s, thus wetting rates around 0.14 kg/m s, 0.29 kg/m s and 0.43 kg/m s were investigated.

### Calculation Method

MATLAB was used to model the washing behavior and dissolution rate. The tube was discretized into several cells in z-direction with rotational symmetry (Figure 4). The washing was simulated by taking short time steps. In each time step a mass element was introduced at the top and its path was integrated over the entire length instantaneously, i.e. a simplification was made which meant that the change in vertical scale thickness during the flow-time from top to bottom is ignored. The flow time is, however, in the order of a few seconds and thus this is assumed to be a reasonable simplification. In each of the cells in z-direction, all physical properties were assumed to be constant. Each cell had a mass of scales which decreased depending on the dissolution rate  $r$ . The presence of scales was modeled with a Boolean variable  $\theta$  that is equal to 1 for the cells with scales and 0 for the clean cells.

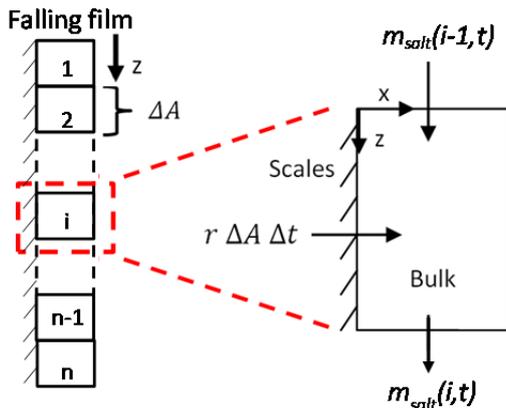


Fig. 4. The falling film was discretized in z-direction along the tube into  $n$  cells.

To simplify the calculations the interfacial area,  $A$ , between the scales and the wash liquid was assumed to be constant and equal to the heat transfer area of the tube. In reality it was varying depending on the thickness and roughness of the scaling layer. However, assuming smooth scales, the outer diameter including the experimentally measured scale thickness would only increase the area of by at most 5 % compared to the clean surface, making it a reasonable assumption.

The model used both internal and external mass balances. The mass balances within each cell ensured that the amount of salt dissolved from the tube is equal to the difference  $m_{salt}(i) - m_{salt}(i-1)$ . The external mass balance ensured that the assumed distribution of scales added up to the total amount of dissolved scales.

The salt mass fraction in the outgoing wash liquid was modeled and compared to the experimental results. Constant transport coefficients for each model and experiment were determined using a best fit approach to the experimental data. It was adjusted so it could simulate the outgoing wash liquid, i.e. match the salt mass fraction with time. For the diffusion-reaction model, two individual constants ( $K_d$  and  $K_r$ ) were adjusted without actually knowing the intermediate mass fraction,  $\omega_i$ . Starting from the diffusion coefficient being equal to the 1<sup>st</sup> order model transport coefficient,  $K$ , a set of plausible combinations of  $K_d$  and  $K_r$  were tested to find the best fit in the region of interest. This method was used for both 2<sup>nd</sup> and 3<sup>rd</sup> level reaction expressions (i.e.  $n=2, 3$ ).

### Scale Distribution

It is necessary to know the scale distribution on the tube if the washing is to be modeled since it will affect the contact area,  $A$ , between washing liquid and scales.

According to previous research (Gourdon et al., 2007), scales grow from the bottom and up. Since the scales acts as insulation material on the tube it is realistic to assume that the growth will slow down as the layer gets thicker and the heat transfer is hindered. This behavior was also confirmed during the experiments where the thicknesses of the scaling layer at the two lower vertical positions were about the same.

Even though the scale thickness was measured at two different vertical positions and visually observed at the top, the thickness at the rest of the tube was unknown. With a known mass of scales dissolved in the washing solution and measurements of the thickness at the three vertical positions, a reasonable scale distribution was estimated. The most difficult part to estimate was between the top and middle positions as these generally had vastly different initial thicknesses. A typical result of this procedure can be seen in Figure 5. The circles represent measured values and the line the estimated scales thickness.

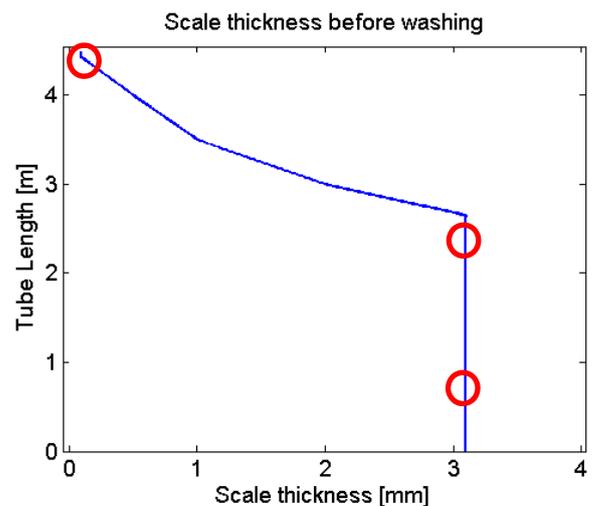


Fig. 5. A typical estimation of the scale distribution. Inlet is at 4.5 m. Top thickness (at 4.35 m) is estimated by visual observation, other two thicknesses are estimated by the measurement procedure described in the section “experimental procedure”.

## EXPERIMENTAL RESULTS AND DISCUSSION

The dissolution rate's dependence on temperature and wetting degree was experimentally evaluated. Eight experiments were conducted and are included in this study (see Table 1) of which a few will be presented more in detail below. The average temperature differences from inlet to outlet are also displayed in the table. In general it was observed that the mass fraction of dissolved salts in the outgoing liquid was increasing with temperature.

Table 1. Experimental conditions and basic results

#	$T_{avg,in}$ (°C)	$T_{avg,out}$ (°C)	$\Gamma$ (kg/m s)	Salt on tube (kg)	Water req. (dm <sup>3</sup> )
1	40	52	0.157	4.0	24
2	76	76	0.139	1.8	10
3	45	48	0.168	2.9	17
4	84	81	0.123	3.9	16
5	82	78	0.295	3.8	19
6	63	75	0.287	3.8	21
7	84	82	0.401	4.4	22
8	63	72	0.432	4.1	24

### Washing Behaviour

Figure 6 shows how the mass fraction of dissolved scales typically varied in the course of the cleaning procedure. Each column represents one sample. The x-axis shows the washing time in minutes and the sampling time vary somewhat due to variations in the volumetric flow rate. In particular, the first samples have longer sampling time as the flow velocity will be lower when the surface needs to be wetted, which is also one of the reasons not to include it in the region of interest. Part of this variation originated from a volume expansion caused by the dissolved scales. The y-axis shows the amount of dissolved scales per amount of solution. The dashed line represents the solubility limit which indicates the maximum possible amount of salt that can be dissolved in the wash liquid.

The mass fraction of salt with time usually had a shape of an inverted S (Figure 6). The mass of dissolved salt varied from a value close to the solubility limit down to a level of about 5 % of the maximum. At the end of the washing the mass fraction of salt levelled out above zero, i.e. even though the tube was clean the wash liquid still contained salt. Most likely, this was due to crystals in the bottom of the evaporator that was continuing to dissolve after the tube was clean.

The high mass fraction in the beginning of the washing does not have to be only due to scales dissolved from the tube, but could also be residual saline solution from other areas. For instance, even though the distributor on the top was drained from saturated solution it is likely that it still contained some residual solution when the washing began. In addition, the flow will not be fully developed in the beginning since it takes time to establish a fully developed film. Moreover, as the tube was cooled from the inside to achieve the desired temperature, vapour started to condense on the scaling layer. The scaling layer was therefore usually

covered with a thin, saturated layer of solution prior to washing.

### Dissolution of Scales

The scale thickness measurements revealed that for all experiments the scales at 1.8 m from the inlet started to dissolve rapidly already at the beginning of the washing (Figure 6). The rapid thickness reduction was more or less persistent throughout the whole washing. The scales at the lowest measurement position (3.8 m) started to dissolve with a much slower rate but later increased to the same rate as the one at 1.8 m.

The washing behaviour can be related to how the mass fraction of salts varied with time. The outgoing salt mass fraction changed only as a larger section of the tube became clean and not while the scaling layer got thinner (plateau period). This demonstrates that the thickness of the scaling layer probably is irrelevant for the dissolution rate and that it is only the surface of the scaling layer that is involved in the dissolution process. When parts of the tube become clean, the contact time between wash liquid and scales will be lower, resulting in less concentrated wash liquid (Figure 6).

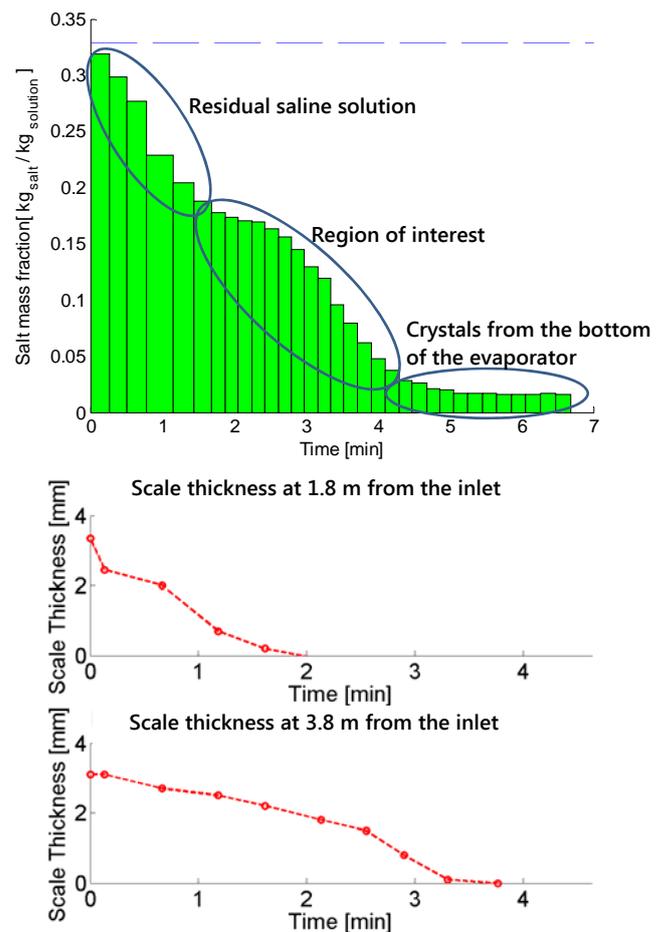


Fig. 6. Exp #8. Top: Measured salt mass fraction in outgoing washing liquid. Dashed line represents the solubility limit. Bottom: Thickness of the scaling layer with time at 1.8 and 3.8 m from the inlet. Dots represent measurements.

**Wetting Degree Dependence**

In Figure 7 the washing behavior for two different wetting rates are presented. For low wetting degrees, the wash liquid had high salt saturation degree during the plateau period whereas for the higher flow rates, the mass fraction on the plateau was lower. When the wetting degree was doubled from 0.123 kg/m s to 0.295 kg/m s, the washing time was cut in half but 19 % more wash liquid was required.

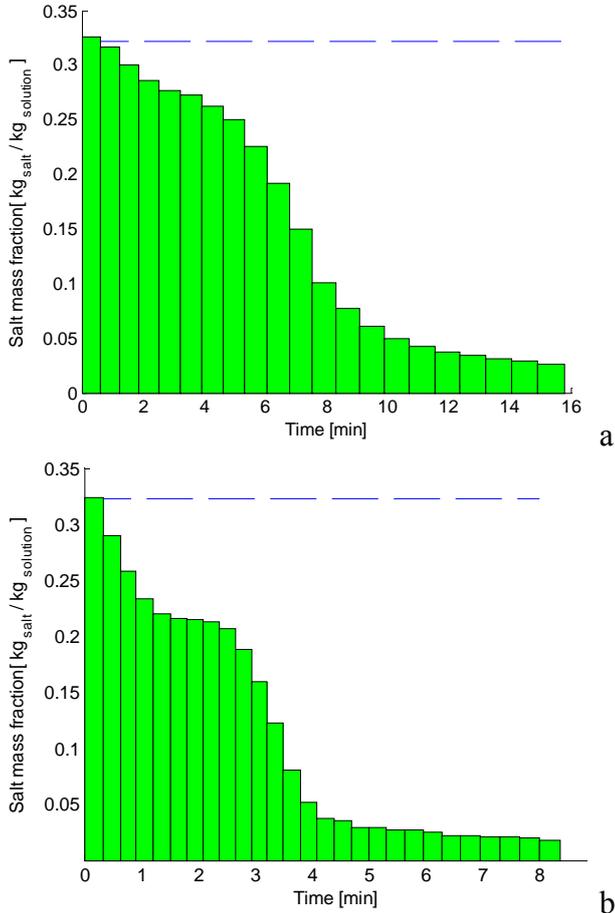


Fig. 7. Amount of dissolved salt with time for (a) Exp. #4 with  $\Gamma = 0.123 \text{ kg/m s}$  and (b) Exp. #5 with  $\Gamma = 0.295 \text{ kg/m s}$ . Dashed lines represent the solubility limit.

**MODELLING RESULTS AND DISCUSSION**

All proposed models simulated the washing in almost the same way for all experiments and therefore only two experiments will be discussed here. The models' predictions for experiment 5 can be seen in Figure 8. As displayed in Figure 6, it is the period in the middle that is of primary interest, excluding the initial period to remove residual saline solutions and the final period with dissolution of parts other than the heat transfer surface.

All proposed models were capable of simulating the characteristic washing behavior with about the same accuracy. The 1<sup>st</sup> order model somewhat under-predicted the results in the first part of the plateau period. The models showed larger differences when it came to predicting the dissolution rate at the two vertical measurement positions. As can be seen in Figure 8, the 1<sup>st</sup> order model provided the

best prediction at 1.8 m from the inlet. However, it is not obvious which is best at 3.8 m. For experiment 5 the 1<sup>st</sup> order model was the closest to predict when the tube would become clean, but the 3<sup>rd</sup> order model provided a better match during the washing and only deviated at the end. In general the 2<sup>nd</sup> and 3<sup>rd</sup> order models, however, did not significantly improve the prediction even though a second adjustable parameter was introduced. Based on this the further analysis was only done for the 1<sup>st</sup> order model.

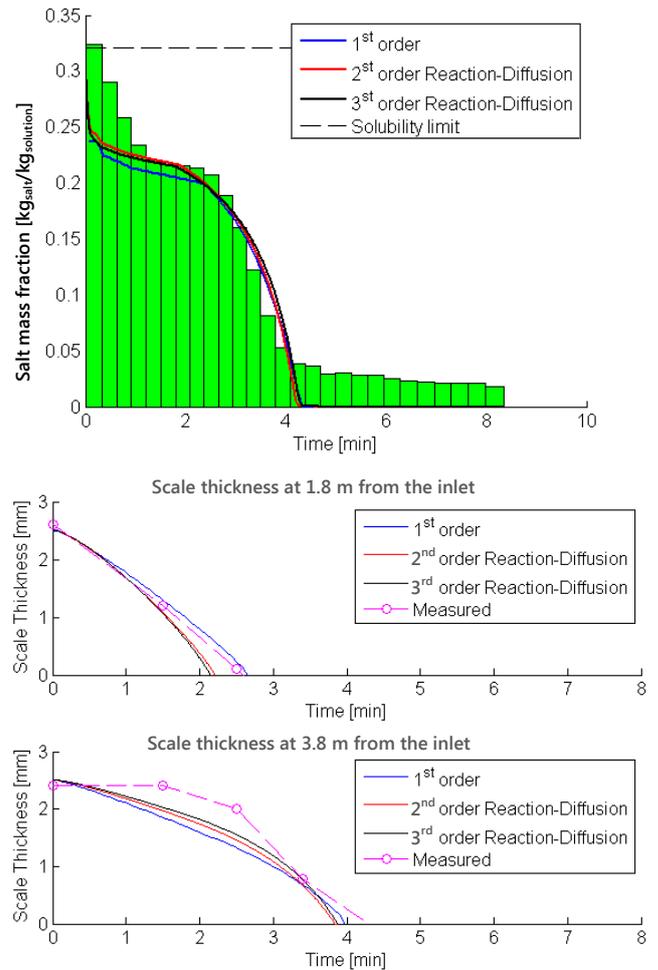


Fig. 8. Exp #5. Top: Measured salt mass fraction in outgoing washing liquid including model predictions. Dashed line represents the solubility limit. Bottom: Thickness of the scaling layer with time at 1.8 and 3.8 m from the inlet including model predictions. Dots represent measurements.

The washing behaviour for experiment 1 and 2 deviated from the others. The change in mass fraction followed an exponential curve with time rather than the previously mentioned inverse S-shape. In experiment 1 the wetting of the tube was uneven which led to an inhomogeneous tube cleaning. Due to malfunctioning equipment in experiment 2, the fouling operation had to be stopped prematurely which resulted in a significantly thinner scaling layer. The low amount of scales on the tube probably caused a different scale distribution which resulted in the exponential washing

shape. None of the proposed models were able to predict the characteristics of these first two experiments. The scale thickness was also not measured during the washing in these experiments, thus the models could only be compared to the time at which the tube was visually considered clean. Due to the lack of data and the deviating behavior it is difficult to tell if the adapted models can be used to simulate the washing in these two cases. If the scale distribution is estimated in another way than the procedure described above, it is possible to achieve a better fit, but since the real distribution is unknown it is difficult to tell if this approach is more accurate. These experiments have therefore been disregarded from further analysis.

### Parameter Optimisation

Since the 1<sup>st</sup> order model generally provided most consistency with the measurements in all the experiments further investigation of its transport coefficient was conducted. Table 2 shows the value of the fitted transport coefficient for the 1<sup>st</sup> order model. A perfect match was not necessary for the beginning and the end of the washing, as previously discussed. Therefore, the transport coefficients were adjusted to specifically match the salt content during the middle part of the washing.

Table 2. Transport coefficients for the 1<sup>st</sup> order model and experimental conditions for the modeled experiments.

#	$T_{avg,in}$ (°C)	$T_{avg,out}$ (°C)	$\Gamma$ (kg/m s)	$K$ $\left(\frac{kg_{solution}}{m^2 \cdot s}\right)$
1	40	52	0.157	-
2	76	76	0.139	-
3	45	48	0.168	0.070
4	84	81	0.123	0.155
5	82	78	0.295	0.150
6	63	75	0.287	0.120
7	84	84	0.401	0.160
8	63	72	0.432	0.130

Experiment 4, 5 and 7 had approximately the same temperature but different wetting degrees. By comparing the wetting degree and the transport coefficient, no correlation could be found. This indicates that the wetting degree seems to have no influence on the transport coefficient for the tested range from 0.123 to 0.401 kg/m s.

Experiment 3, 4, 5 and 7 were conducted at stable temperatures (i.e. inlet temperature approximately the same as the outlet temperature). If the transport coefficient in these experiments is plotted against the temperature it appears as the transport coefficient varies linearly with temperature (Figure 9). There is, however, limited data and more measurements are needed to conclude the true temperature dependence.

Experiment 6 and 8 had larger temperature differences between ingoing and outgoing wash liquid. The wash liquid also experienced large temperature variations with time during the wash. To account for these temperature fluctuations in time and thus improve the model, the constant transport coefficient was replaced by the linear,

temperature dependent expression for  $K$ . The temperature profile was unknown and therefore it was assumed that the ingoing wash liquid was heated directly to the outgoing temperature. This assumption is reasonable since the tube was the hottest on the top and the absence of scales in this part resulted in a high heat transfer. The modeling of the washing with this method leads to higher consistency with the measurements for both experiments 6 and 8. This can be regarded as a confirmation of the transport coefficient being temperature dependent and shows the importance of having the correct modeling temperature.

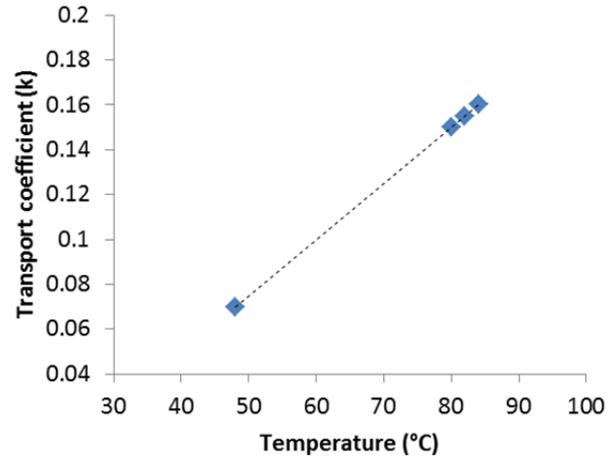


Fig. 9. Temperature dependence of the transport coefficient.

### FINAL REMARKS

Further work is needed before a general model and specific guidelines can be developed regarding washing in the industry. Industry often use diluted weak black liquor as wash liquid, which has significantly different properties compared to deionised water. How these properties affect the dissolution, especially the viscosity, needs to be investigated. Moreover the true temperature dependence is of importance for a more full-covering model as well as establishing in what flow region the wetting degree has limited effect on the transport coefficient.

Furthermore, to fully investigate and model the dissolution rate the type of scales covering the tube should be varied, i.e. whether burkeite, sodium sulphate dicarbonate or carbonate have crystallised.

### CONCLUSIONS

An effective method to study and measure dissolution rates of sodium salt scales in a falling film evaporator has been developed in this study.

A dissolution model was developed and successfully predicted the dissolution of sodium scales. The model was validated for the temperature interval 45-85 °C and wetting degrees of 0.13 kg/m s up to 0.44 kg/m s. The dissolution rate followed a first order expression with regards to the difference in mass fraction between the scaling layer surface and the bulk of the wash liquid. For each experiment one parameter, the transport coefficient was adjusted. By this, the model was able to predict the outgoing salt mass

fraction as well as the dissolution rate at two different positions on the tube, proving the validity of the model.

No dependence of wetting degree could be found for the transport coefficient in the studied flow region. As a consequence, the bulk concentration will decrease proportionally with increasing wetting rates. Thus, the dissolution rate will increase at higher wetting degrees, which means that the washing time will decrease with the flow rate, but more washing liquid is needed to dissolve the same amount of scales.

The transport coefficient was found to have strong positive temperature dependence. This implies that the washing will be much faster at a higher temperature and that less washing liquid will be needed. The accuracy of the model was also improved for experiments with fluctuating temperature with time when implementing a temperature dependent expression for the transport coefficient.

## NOMENCLATURE

$A$	Surface area of the solid-liquid interface, $m^2$
$c$	Concentration, $kg_{salt}/m^3$
$k$	Transport coefficient, $m/s$
$K$	Transport coefficient, $kg_{solution}/m^2s$
$m$	Mass of fouling layer, $kg$
$n$	Reaction order
$r$	Dissolution rate, $kg_{salt}/m^2s$
$t$	Time, $s$
$\Gamma$	Wetting degree, $kg/m s$
$\delta_d$	Thickness, $m$
$\theta$	Boolean variable
$\omega$	Mass fraction, $kg_{salt}/kg_{solution}$

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