



# Moving boundary models for the growth of crystalline deposits from undetected leakages of industrial process liquors

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## ABSTRACT

In this study, a computational model which simulates the growth of crystalline deposits from dripping salt solution is developed and validated. This problem is of interest to the nuclear industry where the morphology of deposited material impacts on its associated criticality risk. An existing model for simulating geological-stalagmite formations is adapted to the case of dripping salt-solutions which form thin films of fluid that precipitate out over time, forming accumulations. The implementation of a CFD Volume-of-Fluid multiphase model is developed such that the fluid-flow is coupled to the crystallisation kinetics and a moving-boundary model is used for describing the size and shape of growing crystalline deposits. The fluid-flow and forming accumulation are fully coupled, with the model able to account for solute diffusion and solvent evaporation. Results are in good agreement with experimental data for surrogate salt-solutions. Numerical results are presented to assess the sensitivity to process and environmental parameters.

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## 1. Introduction

Modelling the growth of crystalline deposits plays an important role in a wide range of chemical process driven industries. Applications of computational mass deposition models are diverse; being applied for production and optimisation purposes, to inform process safety and maintenance, and the simulation of formation growth occurring in nature. Computational mass deposition models can relate to the simulation of a variety of physical processes, however here we are specifically interested in the transfer of mass which is contained within a fluid which then deposits on a solid surface. This could be due to either crystallisation or another mass transfer mechanism. Whilst there are some key physical differences between crystal deposition and other mass transfer mechanisms,

the techniques that are used to model them are often similar. Mass deposition models are inherently difficult to solve as it is important that multiple effects are coupled in an accurate and robust manner. The fluid dynamics of the solution flow, crystallisation kinetics and fluid interaction with depositing solids typically need to be considered. For example, in very thin films topological changes from deposited mass can often have a large impact on the flow as the fluid is required to move over or around the body.

A large area of research when modelling crystallisation and mass transfer relates to maintenance, optimisation and future design of equipment (Heath and Livk, 2006; Al-Rashed et al., 2013; Weber et al., 2013). A significant portion of this research relates to capturing fouling phenomena within industrial equipment (Radu et al., 2014; Jun and Puri, 2005). In these situations mass transfer occurs and deposits are formed in pipes or on walls of the equipment. These deposits can affect both the local fluid properties, such as the pressure and velocity, and the heat transfer to and from the equipment walls, as shown by Mayer et al. (2013) when investigating deposition in pipe flows.

The previous works demonstrate the coupling of mass transfer and fluid flow models, they do not specifically concern crystallisation, and do not incorporate crystal growth kinetics within the

*Abbreviations:* NNL, National Nuclear Laboratory; CFD, computational fluid dynamics; PUREX, plutonium uranium redox extraction; VOF, volume of fluid; UDF, user defined function.

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### Nomenclature

$\alpha$	volume fraction
$\Delta l$	length of linear element (m)
$\mu$	dynamic viscosity ( $\text{Pa s}^{-1}$ )
$\nu$	kinematic viscosity ( $\text{m}^2 \text{s}^{-1}$ )
$\rho$	density of solution ( $\text{kg m}^{-3}$ )
$\theta$	angle of inclination from the horizontal
$A_a$	surface area of gaseous inlet
$A_l$	surface area of liquid inlet
$B$	width of film (m)
$c$	concentration ( $\text{mol m}^{-3}$ )
$D$	diffusivity ( $\text{m}^2 \text{s}^{-1}$ )
$E$	evaporative flux ( $\text{kg m}^{-2} \text{s}^{-1}$ )
$F$	rate of deposition ( $\text{mol m}^{-2} \text{s}^{-1}$ )
$G$	growth rate ( $\text{m s}^{-1}$ )
$h$	film height (m)
$K$	overall growth coefficient ( $\text{m s}^{-1}$ )
$k_d$	constant of mass transfer ( $\text{m s}^{-1}$ )
$k_r$	constant of surface integration ( $\text{m s}^{-1}$ )
$M_s$	molar mass of substance in solid form ( $\text{kg mol}^{-1}$ )
$p$	pressure (Pa)
$p_{atm}$	atmospheric pressure (Pa)
$Q$	mass flow rate of liquid ( $\text{kg s}^{-1}$ )
$Q_a$	mass flow rate of air ( $\text{kg s}^{-1}$ )
$T$	solution temperature in the domain
$T_{in}$	solution temperature prior to entry into domain
$\mathbf{g}$	acceleration due to gravity ( $\text{m s}^{-2}$ )
$\mathbf{n}$	unit normal vector
$\mathbf{u}$	fluid velocity ( $\text{m s}^{-1}$ )

### Subscript

$aq$	substance in aqueous form
$i$	index of discrete element $i$
$l$	substance is in liquid form
$q$	phase $q$
$s$	substance is in solid form

models. Li et al. (2010) posed the growth of a single crystal as a Stefan-type problem solving a scalar diffusion equation for the solution concentration. Crystal kinetics were applied on the crystal surface, such that the growth was proportional to the solution's supersaturation at the surface. This work allowed the modelling of dendritic formations suspended within a stagnant fluid through a front tracking technique. Chen et al. (2009) posed a similar approach for suspended dendritic growth which solved for advection terms in the fluid equations, enabling to capture the motion of a moving solution. These models assume the crystal remains suspended at a fixed, stationary location within the solution. Studies that include crystal kinetic models, such that the crystals are free to move within solution include Heath and Livk (2006), Falola and Borissova (2012) and Cheng et al. (2012). These authors solve population balance models along with a single phase fluid model. The population balance allows the size distribution of the crystals to be captured throughout an industrial batch reactor, this in turn couples to crystal kinetic models to describe the growth. Whilst allowing the mapping of a nonhomogeneous mixture, the crystals in these materials have no impact on the fluid flow and as such, for thin fluid flow, these models do not account for the topological changes on the flow caused by the deposited crystalline mass.

The work in this study relates to modelling crystallisation deposits found in the nuclear industry. In nuclear reprocessing, a popular method of retrieving reusable fuels from spent solid fuel

is the PUREX cycle. This method involves the dissolution of solid plutonium and uranium fuels in nitric acids. This solution is then transported across the plant where it undergoes varying chemical processes. Further details on the PUREX cycle can be found in Phillips (1999). It has been found that in the event of an undetected equipment malfunction, leaking droplets of process liquor can coalesce to form thin liquid films. Solvent loss through evaporation and cooling of the solution through changes in ambient conditions can cause the liquid films to enter a supersaturated state where by crystalline deposits form. Incidents have occurred where the crystalline deposits covering an area of  $1 \text{ m}^2$  and  $30 \text{ cm}$  high, have been observed, details of which can be found in Burrows et al. (2006) and a report by the Health and Safety Executive (2007). These details are of great importance as it is known that for a deposit of heavy metal, the criticality risk of the solid formation depends on both its size and shape.

The purpose of the work here is to model the crystallisation of thin films of a surrogate salt solution. This aim is to offer insight into the size and shape of crystalline deposits which may occur from sustained droplets of industrial process liquor, arising from a long term undetected equipment or pipe leakage, for varying process and environmental conditions. A surrogate salt solution of sodium nitrate is considered as experiments can be conducted conveniently to validate the model. To the best of the authors knowledge, no previous studies have been carried out in order to model this problem.

The NNL have carried out a series of experiments such that a simulant solution of sodium nitrate was dripped onto inclined steel plates (Fig. 1(a)). Over time the drops of solution crystallise and a crystalline formation builds up on the surface of the plate (Fig. 1(b)). Various process parameters were considered in order to develop an understanding of their impact on the resultant crystalline formation.

The experimental work identified a diverse range of formations for changes in input parameters. Impinging droplets of a saturated simulant solution of sodium nitrate (8 M) formed tower-type formations (Fig. 2(a)). For lower concentration solutions (5 M), ring-like configurations were formed (Fig. 2(b)). The criticality implications of these formations change quite significantly for small changes in experimental parameters.

This study will investigate an appropriate model to describe the growth of the formations, and how varying parameters affect the growth and resultant size and shape. Examining the experimental formations it is clear that the crystal mass deposited is large in comparison to the fluid film depth and has a large impact on the flow field. As such, a single phase approach would not be appropriate. It was noted that when observing the deposition of highly concentration salt solutions, similarities could be drawn between these and underground geological stalagmites, albeit on a much smaller timescale. There are a number of published models for predicting the formation of stalagmites overtime (Romanov et al., 2008; Baker and Bradley, 2010; Short et al., 2005a, 2005b; Kaufmann, 2003; Kaufmann and Dreybrodt, 2004). In this study, we adapt a previous geological stalagmite model, Romanov et al. (2008), for the purpose of modelling tower formations formed from supersaturated salt solution. The model uses moving boundary techniques for capturing the crystal growth with a simple analytical model describing the thin laminar film flow. Results from this model offer insight into the crystal deposition behaviour of highly concentrated salt solution. The model neglects certain important physics. As such, a further model is developed using a coupled moving boundary and CFD technique to account for the fluid flow and crystallisation kinetics. This new framework allows the addition of several previously absent physical effects such as diffusion and evaporation. As evaporation is now included, solutions which are not initially in a supersaturated state can also be considered.

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