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A novel factorial design search to determine realizable constant sets for a multi-mechanism model of mixing sensitive precipitation



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ABSTRACT

A mechanistic model for the reactive precipitation of amine mono- and di-hydrochloride (AHC) salts was developed in the first stage of this work (Komrakova and Maluta, 2016). In a companion paper, an extensive experimental investigation of the reaction products is reported (Ershad, 2013). The experimental work shows a sudden shift from mono- to di- production as the blend strength of 4, 4'-methylene dianiline (MDA) decreases, and a strong dependence of di-concentration on excess HCl in the feed for the high blend strength experiments. Due to the complexity of the reaction-precipitation system, eleven constants related to physical properties and rates were identified in the model. An extensive search of the literature returned only realistic ranges for most of these constants. In this work, a novel method for reducing the uncertainty in the space defined by these eleven constants is presented. The trends in the experimental data were successfully reproduced and the model results provide a number of important insights into the two complex reaction outcomes. A factorial design search for similar loosely bounded parameter spaces is proposed.

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1. Introduction

This paper will follow two main threads of investigation: the first has the goal of increasing our understanding of the amine hydrochloride (AHC) system and solving the model specifically designed for that purpose; the second is to present a protocol which was successfully used to define realizable limits on the set of physical and reaction rate constants associated with the model. To begin, we provide some context for parameter estimation procedures in the larger field of reaction modeling.

According to Schwaab et al. (2008), "Parameter estimation procedures are very important in the chemical engineering field for development of mathematical models, since design, optimization and advanced control of chemical processes depends on model parameter values obtained from experimental data. Model nonlinearity makes the estimation of parameters and the statistical analysis of parameter estimates more difficult and more challenging." Before moving to a brief review of the solution approaches which have already been

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http://dx.doi.org/10.1016/j.compchemeng.2017.06.014 0098-1354/© 2017 Elsevier Ltd. All rights reserved. proposed for parameter estimation, it is useful to emphasize the difference between a constant which has inherent physical meaning and a single possible value (e.g. the solubility of A in B), and a fitting parameter, which is used to align a set of data with a proposed equation form. Depending on the equation form, fitting parameters will take on different values. In the commentary by Schwaab et al. (2008), the line between a constant and a parameter is blurred, possibly based on the argument that any model form is subject to the choices made by the modeler. Throughout this paper, we will attempt to maintain the distinction between an immutable physical property of the system, and a model parameter which is affected by the underlying assumptions and assumed model form. Secondly, we note that the equations for reactive precipitation are non-linear, highly coupled, and present a number of numerical challenges.

In the literature, there is an abundance of methods for the local and global optimization of parameters to fit an existing data set as closely as possible (Horst et al., 2000). The general optimization problem aims to minimize one or more objective functions (Boyd and Vandenberghe, 2004) in the feasible design space subject to a series of inequality and equality constraints (Marler and Arora, 2004). In our study, the feasible design space is unknown and the constraints are only loosely defined due to the scarcity of experimental data.

Nomenclature		
b ₀	Initial half width of the jet (m)	
Bnucl	Primary homogeneous nucleation rate $(m^{-3} s^{-1})$	
C _{C n}	Molar density of n^{th} particle type (kmol m ⁻³)	
Ci	Concentration of i th species in the bulk (kmol m ^{-3})	
Ci a	Molar concentration of i^{th} species in the side <i>a</i>	
- 1, <i>u</i>	$(kmol m^{-3})$	
C	Molar concentration of i^{th} species in the side h	
$C_{1,b}$	$(kmolm^{-3})$	
C	Concentration of i th species at the end of the jet	
C _{i,e}	$(kmol m^{-3})$	
C	(KIIIOIIII ⁻) Molec concentration of i th encoder entrained from	
$C_{i,E}$	woral concentration of 1^{m} species entranied from the heile (law element)	
C	(ine bulk (kinoi in ³)	
Cn	Concentration of n ^{cm} sait type in liquid phase (mono-	
D	or di-) (kmol m^{-3})	
D _{AB}	Diffusion coefficient $(m^2 s^{-1})$	
Gn	Growth rate of the particles (mono- or di-) (m s ^{-1})	
k _{1,f}	Forward reaction rate for reaction $1 (m^3 \text{ kmol}^{-1} \text{ s}^{-1})$	
$k_{1,r}$	Reverse reaction rate for reaction $1 (s^{-1})$	
k _{2,f}	Forward reaction rate for reaction $2(m^3 \text{ kmol}^{-1} \text{ s}^{-1})$	
k _{2,r}	Reverse reaction rate for reaction $2(s^{-1})$	
k _B	Boltzmann constant (JK ⁻¹)	
Ke	Kinematic momentum flux in the jet $(m^4 s^{-2})$	
K_{eq}^{-1}	Inverse of the equilibrium constant (kmol m ⁻³)	
Kn	Solubility of n th particle type (kmol m ⁻³)	
K _{σ,n}	Surface tension parameter for n th particle type (–)	
l	Characteristic diameter of the particles (mono- or	
	di-) (m)	
$\ell_{0,n}$	Critical nuclei size for the n th particle type (m)	
N _A	Avogadro's number (kmol ⁻¹)	
'n _{i.g}	Rate of molar generation of i th species per unit vol-	
,0	ume (kmol m ^{-3} s ^{-1})	
$n_{n,a}(\ell)$	Number concentration of n th particle type with	
.,()	characteristic diameter ℓ in the side q (m ⁻⁴)	
$n_{n,h}(\ell)$	Number concentration of n th particle type with	
11,0 (*)	characteristic diameter ℓ in the side $h(m^{-4})$	
$\mathbf{p}(\ell)$	Number concentration of p^{th} particle type with	
$\Pi_{n,e}(c)$	Number concentration of it particle type with	
(a)	characteristic diameter ℓ exiting the jet (m ⁻⁺)	
$n_{n,E}(\ell)$	Number concentration of n th particle type with	
	characteristic diameter ℓ entrained from the bulk	
(a)	(m^{-1})	
$n_n(\ell)$	Number concentration of n th particle type with	
	characteristic diameter ℓ in the bulk (m ⁻⁴)	
r _n	Hydrodynamic radius of the particle (m)	
Sh	Sherwood number (–)	
Sn	Supersaturation of the product that produces n th	
	particle type (–)	
Т	Temperature (K)	
V	Volumetric flow rate in the jet (m ³ s ⁻¹)	
V _{CIIR}	Volume of the reactor (m ³)	
v _E	Entrainment velocity (m s^{-1})	
Ve	Total volume flow rate exiting the jet (m ³ s ⁻¹)	
Ϋ́ _E	Entrainment volumetric flow rate from the bulk to	
-	the jet $(m^3 s^{-1})$	
V _{iet}	Volume of the radial jet (m^3)	
Vmn	Molar volume of n^{th} particle type (m^3)	
Vs	Exchange velocity (m s^{-1})	
Vs	Exchange volumetric flow rate $(m^3 s^{-1})$	
Vx	Downstream velocity profile in the jet (m s^{-1})	
V _x max	Maximum downstream fluid velocity (m s ^{-1})	

y-average of the jet downstream velocity (m s^{-1})

V_{x,max}

νx

х	Radial/downstream coordinate of the radial jet (m)
x ₀	Radius of the inlet round jet (m)
У	Width coordinate of the jet (m)
y*(x)	Half width of the jet at x (m)
Z	Coefficient for the exchange velocity between sides of the jet (-)
Δ	Limit on the x-coordinate to overcome singularities (m)
μ	Dynamic viscosity of the liquid (Pa s)
$v_{D,n}$	Dissociation number for n^{th} particle type (–)
σ_0^{-1}	Dimensionless initial spreading rate of the turbulent free jet (-)
σn	Surface tension of n^{th} particle type (N m ⁻¹)
Thull	Residence time in the bulk (s)
	Expansion angle of the jet (\circ)
Ψ	Arimuthal/angular accordinate of the redial ist (red)
ω	Azimuthai/angular coordinate of the radial jet (rad)

The highly coupled, stiff equations led to instabilities in the code, a number of unconverged solutions, and solutions converged to non-physical values. This made fixed boundaries of the parameter space difficult to define using optimization. Several local minima are known to exist, which further complicates the possibility of seeking a globally optimized solution point (Forgó, 1988; Hansen and Walster, 2003; Horst and Tuy, 1993; Horst and Pardalos, 1995; Parsopoulos and Vrahatis, 2002; Pintér, 1996; Rao, 2009; Schwefel, 1995; Törn and Zilinskas, 1989).

For these reasons, a multidimensional search based on factorial designs was proposed as another way to define the feasible design space (domain) and to obtain preliminary information on the model response to the change in constants and parameters. The full factorial search was selected to investigate the domain and the codomain of the model constants, to identify the most relevant parameters and constants and thus help the modeler to prioritize the experiments needed to determine those constants and parameters. The factorial search was chosen for two reasons: first, because of its ability to provide information on the effects of the controllable factors (both model constants and parameters) and their interactions on the response variables and second, because of the simplicity and robustness of the method.

2. Modeling the amine hydrochloride (AHC) system

The product composition and particle size distributions of amine-hydrochloride salts are the result of many complex physical processes and their interactions (Ershad, 2013), as shown in Fig. 1 which adapted and updated for this study, starting from a similar scheme by (Schwarzer and Peukert, 2004a). These processes give rise to a highly-coupled system of stiff differential equations, so the effect of the parameters on the outputs is difficult to predict a priori. A way to determine and understand the response of the model to changes in the input parameters and unknown physical constants was sought.

2.1. Modelling the reaction

The reactive precipitation of AHC salts was experimentally studied in a confined impinging jet reactor (CIJR). Results showed that in the reaction of 4, 4'-methylene dianiline (MDA) with hydrogen chloride (HCl) two different salts are produced: methylene dianiline monohydrochloride (MDA.HCl, also called mono-) and methylene dianiline dihydrochloride (MDA.2HCl, also called di-). The reaction and precipitation of AHC salts requires a high local concentration of MDA. The mean concentration of MDA (the blend Download English Version:

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