

Application of fractal theory for characterisation of crystalline deposits

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Abstract

The objective of this investigation was to study the mechanisms of mixed salt crystallisation fouling on heat transfer surfaces and to characterise the structure of the deposits formed on the heat transfer surfaces. The effects of various operating parameters such as solution composition and hydrodynamics of the system on crystallisation fouling of mixtures of calcium sulphate and calcium carbonate, which are the most common constituents of scales formed on heat transfer surfaces, were studied experimentally. After clarification of the effects of operating parameters on the deposition process, deposits on the heat transfer surfaces were collected for analysis. The crystalline samples were analysed using Scanning Electron Microscopy, X-Ray Diffraction and Ion Chromatography techniques. It was found that crystallisation fouling on heat transfer surfaces exhibits fractal geometry. Therefore, the concept of fractal theory was used to quantify the structure of the deposits by introducing a new quantity called the fractal dimension.

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1. Introduction

Scale deposition is one of the most important and serious problems usually faced by heat transfer equipment during operation. This phenomenon is known as a major engineering problem in process industries, since deposits on heat transfer surfaces create a barrier to the transmission of heat, increase pressure drop and promote corrosion of tube material. These effects, therefore, reduce the performance of the heat transfer equipment significantly. Scales sometimes limit or block pumps, tubing, casing, flow lines, heaters, tanks and other heat transfer or production equipment and facilities. Fouling has hence for long been recognised as a serious constraint in the design and operation of heat transfer equipment.

Scale deposits are classified according to the physical and chemical processes that occur. Fouling occurring on the heat transfer surfaces of boilers and evaporators is usually a

crystalline deposition caused by precipitation from solution of mineral salts which have inverse solubility curves. Since the thermal conductivity of these crystalline deposits is very low, deposits of these salts will reduce the overall heat transfer coefficient significantly.

Mixed salt crystallisation fouling on heat transfer surfaces during convective heat transfer and sub-cooled flow boiling is a very complicated process. Its mechanism relates to many subjects such as momentum, heat and mass transfer, chemical kinetics, material science, etc. which makes it difficult to express in mathematical terms.

No information is available in the literature about characterisation of the deposits formed on the heat transfer surface by fractal analysis. The objective of this study was to investigate the crystallisation fouling phenomena for salt mixtures under convective heat transfer and sub-cooled flow boiling conditions, and to characterise the structure of the deposits formed on the heat transfer surface. In the present investigation, the effects of various operating parameters such as solution composition and hydrodynamics of the system on crystallisation fouling of mixtures of calcium sulphate and calcium carbonate were studied experimentally (Helalizadeh et al., 2005). After clarification of

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the effects of operating parameters on the deposition process, deposits formed on the heat transfer surfaces were collected for analysis. The crystalline samples were analysed using a high resolution Scanning Electron Microscopy (SEM), X-Ray Diffraction (XRD) and Ion Chromatography techniques. The concept of fractal theory was used to characterise the structure of the deposits, by introducing a new quantity called the fractal dimension.

2. Experimental equipment and procedure

The fouling experiments were performed in a flow circuit which included two parallel annular test heaters. Each test section consisted of an electrically heated cylindrical stainless steel rod. This test heater was manufactured by Ashland Chemicals according to specifications by Heat Transfer Research Incorporated (HTRI). The length of the heater was 400 mm with heated zone of 99.1 mm (216 mm from the beginning of the rod). It was located concentrically within a surrounding vertical pipe and the test liquid was flowing through the annulus in upward direction. A miniature stainless steel sheathed resistance wire was fitted into the centre of the heater rod. Four stainless steel sheathed E-type thermocouples were located closely below the surface of the heater rod. Three of these thermocouples were used to obtain information about the surface temperature. The remaining thermocouple was connected directly to a temperature controller. The test heater was connected to an adjustable power supply permitting variation of the heat flux between 0 and 450 kW/m². Experimental equipment and procedure are discussed in detail by Helalizadeh et al. (2000 and especially Helalizadeh, 2002).

In this investigation, aqueous solutions of mixtures of calcium sulphate and calcium carbonate, the most common constituents of crystals formed on heat transfer surfaces, were used as test solutions. The range of the pH values in the present investigation was between 6.5 and 7.5. Calcium nitrate and sodium carbonate were used to adjust the total alkalinity. The solubilities of the electrolytes in water were predicted using the SOLMINEQ88 software and the physical properties of the solutions were taken from the International Critical Tables (1926–1933). The criteria for selecting the salts were based on the solubility of the salts, the valence and size of the respective ions. The range of the experimental parameters in the present investigation is given in Table 1. For the present experimental conditions, heat fluxes and surface temperatures are high and, as a consequence, operating conditions involve boiling with different nucleate boiling fractions (Helalizadeh, 2002).

In the present investigation, calcium sulphate, calcium carbonate and mixed calcium sulphate/calcium carbonate solutions were used. After each run, deposits on the heat transfer surfaces

were collected for analysis, using SEM, XRD, Ion Chromatography and the concept of fractal theory.

3. Identification of crystalline solids

Solid deposits can be classified into crystalline or amorphous states. The crystalline state differs from the amorphous state by the regular arrangement of the constituting molecules, atoms or ions into some fixed locations known as lattice, which is the characteristic of the substance. Crystals are characterised by the repetition of their constituent building blocks in a three-dimensional array, quite unlike liquids in which the molecules are randomly placed. Therefore, a crystal structure is a symmetrical array of atoms, ions or molecules arranged in a repeating three-dimensional pattern.

Crystals can be classified into four types according to the kind of particles that make up the crystals and the forces that hold them together. Ionic crystals, molecular crystals, network crystals and metallic crystals are four types of crystalline solids.

Ionic crystals: An ionic crystal must accommodate ions of opposite charge and different size in the proper stoichiometric ratio and in such a way that ionic and electrostatic attractions outweigh electrostatic repulsion. In ionic crystals, positive and negative ions are held in the crystal arrangement by ionic and electrostatic attractions. Because these forces are strong, ionic substances are hard, brittle and have a high melting points.

Molecular crystals: Molecules occupy positions in crystals of covalent compounds. The intermolecular forces that hold the molecules in the crystal structure are not nearly as strong as the electrostatic forces that hold ionic crystals together.

Network crystals: In these crystals, atoms and molecules are joined by a network of covalent bonds. The entire crystal can be looked at as one giant molecule. In diamond, an example of this type of crystal, carbon atoms are bonded by covalent bonds into a three-dimensional structure.

Metallic crystals: The outer electrons of metal atoms are loosely held and move freely throughout a metallic crystal. The remainder of the metal atoms, positive ions occupy fixed positions in the crystal. The negative cloud of the freely moving electrons, sometimes called an electron gas or a sea of electrons, binds the crystal together. This binding force is called a metallic bond.

Identification of the above crystals by their structural configurations or physical properties can be accomplished by the following techniques:

1. Scanning Electron Microscopy (SEM)
2. Differential Thermal Analysis (DTA)
3. Polarised Light Microscopy (PLM)
4. X-Ray Diffraction (XRD)
5. Laser Light Scattering (LLS)

According to the solubility curves of calcium sulphate and calcium carbonate, deposits of these salts can occur in various ionic crystal structures (Helalizadeh et al., 2000). To characterise the deposits of this study, samples were analysed using a high resolution SEM, XRD and Ion Chromatography

Table 1
Range of investigated operating parameters

V (m/s)	T_b (°C)	\dot{q} (kW/m ²)	[CaSO ₄] (g/l)	[T.A.] (g/l)	P (kPa)
0.5–2	50–90	100–450	1–2.5	0.0–0.3	105–150

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