EXPIRED

IAPWS Certified Research Need - ICRN 1

Evaluation of Binary Nucleation Models

The IAPWS Working Group - Power Cycle Chemistry has examined the published work in the area of binary nucleation models for condensing steam.

IAPWS recognizes that there is a requirement for work to be pursued in this field and has prepared this document to assist potential investigators obtain sponsorship. Specifically both theoretical and experimental studies are needed. Theoretical studies are required to extend the binary model to relevant power plant systems. Experimental work is required for the following systems: NH_3-H_2O, NaOH-H_2O, H_2SO_4-H_2O, HCl-H_2O, NaCl-H_2O, Na_2SO_4-H_2O, NH_4Cl-H_2O, $(NH_4)_2$ SO_4-H_2O, morpholine-H_2O and cyclohexylamine-H_2O.

Although encouraging this work, IAPWS is not able under its statutes to provide financial support. The IAPWS contact can provide any further development information and will act as a liaison between research groups.

Issued by the

International Association for the Properties of Water and Steam

President: **J.R. Cooper**
Queen Mary and Westfield College
London E1 4NS England

Executive Secretary: **Dr. R.B. Dooley**
Electric Power Research Institute
Palo Alto, California 94304 USA

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Evaluation of Binary Nucleation Models

In the recent years, the condensation process occurring in low pressure stages of power plants has received renewed interest. In particular, the research efforts have been drawn toward the study of the role of contaminants (mainly low volatility species) in the nucleation mechanism. Several experimental observations show a preferential condensation of chloride ions (Cl^-) in the first condensate (percentage of condensation $\leq$3%) (Lindsay and Lee, 1981; Kantola, 1982; Allmon, 1983; Jonas et al., 1984; Auerswald et al., 1985; Pinacci et al., 1992).

Particular attention has been devoted to the characterization of deposits of sodium chloride and sodium hydroxide. The first is known to promote the pitting corrosion and corrosion fatigue of turbine blades while the latter substance is one of the main causes for stress corrosion cracking.

It is worth to mention the work by Lindsay and Lee (1981) in which the possible mechanisms for the condensation of low volatility impurities in steam turbines are discussed. The authors pointed out that the factors which play a major role are those pertaining to the thermodynamics of the condensed phase, to the vapor phase solubilities of low volatility species and to the homogeneous and heterogeneous condensation processes. They emphasize the lack of conclusive experimental data on the vapor phase solubility of low volatility species. This brings about another very important issue which is related to the knowledge of the chemical form/state of the low volatility species in the vapor phase (fully or partially ionized, or in the molecular form) (Turner, 1990). Work has still to be done toward this characterization. The study by Allmon et al. (1983) is along the direction to gain a thorough understanding on the behavior of low volatility species in the vapor phase. The author studied the equilibrium repartition coefficient of sodium hydroxide, sodium chloride and ammonium chloride in the vapor phase. Kantola (1982) presented some preliminary experimental observations on the effect of impurities on the nucleation process during the steam condensation.

The effect of contaminants on the nucleation process is of concern also to people in the aerosol business that deal with the kinetics of formation of particles (for example, Wyslouzil et al. 1991a and 1991b).

From the preliminary analysis of the literature it appears that the understanding of the effect of low volatility contaminants is not completely understood and that to date there is only an understanding of a small part of the complete physical picture.

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Two main approaches have been used, as reported in the literature, to describe the nucleation phenomenon in the presence of contaminants. The first approach consists in extending the classical nucleation theory to account for the presence of low volatility species, which are assumed to be completely dissociated giving rise to some sort of solvated ions (Thomson model) (for example, refer to Singh et al., 1982). The solvated ions play the role of the foreign prenucleation centers which largely enhance the nucleation process. In the second approach, probably more realistic, the nucleation is pictured as a clustering process around the heteromolecular species (for example, refer to Wislouzil et al., 1991a). The two approaches differ in the construction of the Gibbs function of the critical (growing) nucleus.

Extended Homogeneous Nucleation Model (Thomson)

The Gibbs function of formation of the nucleus/droplet is expressed as follows:

$$\Delta G = \Delta G_C + \Delta G_S + \Delta G_e \qquad (1)$$

G_C is the chemical potential contribution:

$$\Delta G_C = -(4/3)\,\pi\, r^3\, \rho\, k T \ln S \qquad (2)$$

where S represents the supersaturation ratio, k is the Boltzmann constant and ρ is molecular density. ΔG_S is the surface energy contribution that, taking into account the dependence of the surface tension on curvature (Tolman model - Vogelsberger, 1980) takes the following form:

$$\Delta G_S = 4\pi r^2 \sigma_\infty (1+2(\delta/r))^{-1} \qquad (3)$$

where σ_∞ is the surface tension for a flat surface, and δ is the Tolman's coefficient which is a measure of the thickness of the interface layer. ΔG_e is the electrostatic contribution (Briant and Burton, 1976; Chan, 1980; Rabeony and Mirabel, 1986) which should account more specifically for the presence of the ionic species. This is the term debated in the literature; the typical form for it is:

$$\Delta G_e = (Q^2/2)\,(1-1/\varepsilon)\,(1/r - 1/r_1) \qquad (4)$$

where Q is the ionic charge (in E.S. units), ε is the relative dielectric constant, r and r_1 are the cluster and the ionic radii, respectively. It must be emphasized that the different formulations available from the literature do not have good agreement with the experimental data.

Binary Model (Flageollet-Daniel et al., 1983; Ray et al., 1986; Wyslouzil et al., 1991a)

Typically the heteromolecular species are pictured as hydrates which are the solvated form of the undissociated molecular species. The Gibbs function of the microcluster is then calculated as follows:

$$\Delta \underline{G} = \underline{N_1} (\mu_1 - \Phi_1) + \underline{N_2} (\mu_2 - \Phi_2) + \sigma \underline{A} \qquad (5)$$

where $\Delta \underline{G}$ is the Gibbs function of formation of one "molecule" of a microcluster, μ and Φ are the chemical potentials in the liquid and vapor phase; $\underline{N}_1$ and $\underline{N}_2$ are the number of molecules of water and heteromolecular present in the cluster. The last term represents the contribution of the surface energy.

Jaecker-Voirol et al. (1987) considered that in the steam the more likely species would be water, contaminants and hydrates, as indicated in Fig. 1 (h represents the number of water molecules of in the hydrates).

Then they proposed a different equation for the Gibbs function equation to take into account for the hydrates, further assuming that the hydrates are in equilibrium with the "free" chemical species. The Gibbs function of formation of the droplet, $\Delta G''$, is therefore computed as the droplet could be formed by $\underline{n}_1$ molecules of water, $\underline{n_2}$ molecules of the heteromolecular substance and $\underline{n}_h$ hydrates:

$$\Delta \underline{G''} = \underline{n}_2 (\mu_1 - \Phi_1) + \underline{n}_2 (\underline{\mu_2} - \Phi_2) + \Sigma \underline{n}_h (\mu_h - \Phi_h) + \sigma \underline{A} \qquad (6)$$

where μ and Φ are the chemical potential in the liquid and vapor phase; n_1, n_2 and n_h are the number of the "free" molecules of water, contaminant and hydrate$_h$ contributing to the formation of the droplet.

Assuming that the system is at thermodynamical equilibrium:

$$\mu_h = \underline{h}\, \mu_1 + \mu_2 \qquad (7a)$$

$$\Phi_h = \underline{h}\, \Phi_1 + \Phi_2 \qquad (7b)$$

Jaecker-Voirol et al. (1987) obtained the following relation for ΔG^* and ΔG:

$$\exp (- \Delta \underline{G''} / \underline{kT}) = \underline{C} \exp (- \Delta \underline{G} / \underline{kT}) \qquad (8)$$

where C is expressed by:

$$\underline{C} = \frac{1+\underline{K}_1\underline{P}w+....+\underline{K}_1\underline{K}_2...\underline{K}_h\,(\underline{P}w)^{\underline{h}}}{1+\underline{K}_1\,\underline{P}_1+....+\underline{K}_1\underline{K}_2...\underline{K}_h\,(\underline{P}_1)^{\underline{h}}}\quad \underline{n}_2 \qquad (9)$$

where the $\underline{K}_j$ represents the equilibrium constant for the hydrate formation process by the addition of one more water molecule to the hydrate$_{j-1}$. $\underline{P}_W$ is the equilibrium vapor pressure of the water over a solution with the same composition as the droplet and $\underline{P}_1$ is the actual partial pressure of the molecules of water in the vapor phase. All the pressures are in atmospheres. The Gibbs function variation due to the addition of one water molecule to the hydrate$_{h-1}$, $\Delta\underline{G}^\circ_h$, is expressed by the following equation:

$$\Delta\underline{G}^\circ_h = kT\ln P_W + 4\pi\sigma\,(3\underline{v}_1/4\pi)^{2/3}\underline{n}_1^{\,-1/3} = -k\underline{T}\,\mathrm{l\,n}\underline{K}_h \qquad (10)$$

where v_1 is the molecular volume of the water in the liquid phase.

This model has been successfully used to compute the nucleation rate for the system H_2SO_4-H_2O, overestimated by the classical (Thomson) model.

As recorded by our literature search, the only systems investigated by this approach have been the H_2SO_4-H_2O, HNO_3-H_2O and CH_3HSO_4-H_2O. Further they have been considered only at normal temperatures and pressures, around 25°C and 0.1 MPa.

Needed Research Activities

Theoretical

Based on the literature already available, a critical evaluation of the model is required with reference to the steam turbine environment. A further search is required to verify that all the information available in the literature, including other fields, on the binary models studies has been traced.

Theoretical studies need to be made to extend this theory to the relevant systems mentioned above without an inert gas carrier and for pressure and temperature ranges significant for the power industry.

Experimental (thermodynamics)

Based on a critical evaluation of the relations available in the literature for the dependence of temperature-concentration-radius on the thermodynamic parameters of aqueous solutions, a literature search and/or laboratory measurements are needed to complete the data base. It must be emphasized that the liquid "bulk" properties have to be carefully considered for their application to the

models" the liquid phase of interest is normally of molecular dimensions and therefore it should be characterized by properties that differ significantly from that of the "bulk".

The following parameters should then be considered, as well as their temperature and curvature dependence:

i) surface tension, σ
ii) density,
iii) molar volume, v
iv) fugacity, f

When phase transitions or chemical reactions are of interest for a specific contaminant-water system, the Gibbs functions will also need to be considered.

The systems that need investigation will be: NH_3-H_2O, NaOH-H_2O, H_2SO_4-H_2O, HCl-H_2O, NaCl-H_2O, Na_2SO_4-H_2O, NH_4 Cl-H_2O, $(NH_4)_2SO_4$-H_2O, morpholine-H_2O and cyclohexylamine-H_2O. Further systems of interest (not normally investigated) are SiO_2-H_2O and oxides (Fe,Ni,Cu,Cr)-H_2O. Priority should be given to the NaCl-H_2O and NaOH-H_2O.

Experimental (kinetics)

Laboratory measurements on the nucleation and growth rate are required for the systems mentioned above.

The first step of this activity will be the review of the experimental facilities available all around the world, evaluating the characteristics of each facility and its performances. Some specific experimentation has to be done on the systems abovementioned and this data used for the parameters estimation and to validate the models.

IAPWS Contact: Dr. F. Sigon
ENEL
Thermal and Nuclear Research Center
Via Rubattino 54
Milano 20134. Italy
Telephone: 39-2-7224 3044
Fax: 39-2-7224 3915

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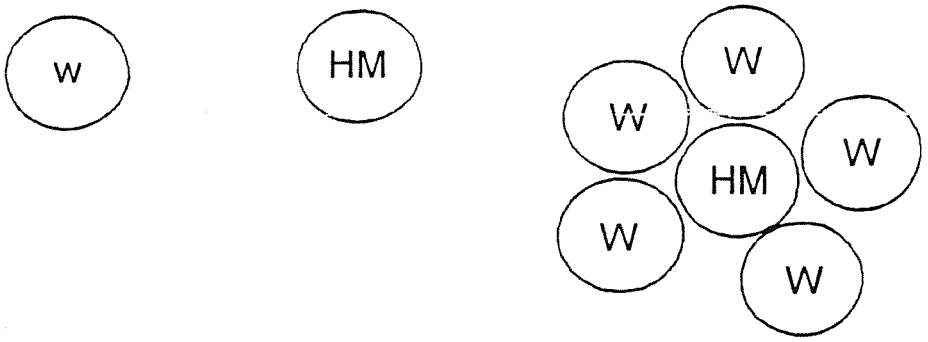
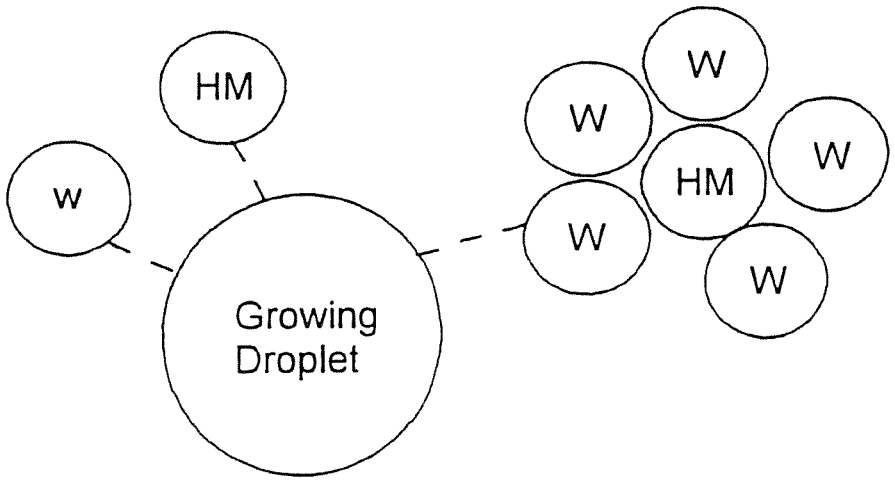


W: H_2O

HM:HCl, H_2SO_4, NH_4Cl, NaCl, ...

(a)



(b)

Fig.1 Schematization of the nucleation process starting out from free molecular species only (a) and with hydrates too (b).

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