A new approach to investigate hydrate deposition in gas-dominated flowlines

Esam Jassim, M. Abedinzadegan Abdi, Y. Muzychka

A new model describing the mechanism of the hydrate deposition based on the most recent particle dynamics theories is developed. The model splits the motion of the particle into two main regions: the turbulent and the sublayer regions.

The proposed model employs the following three main components to simulate thehydrate deposition: (a) computational fluid dynamics (CFD) technique is used to configure the flow field; (b) nucleation and growth models are incorporated in the simulation to predict the incipient hydrate particles size and growth rate; and finally (c) a novel approach of particle migration and deposition is used to determine how particles deposit and adhere to the walls of flow conduit.

The results predicted by the model show that the distance of the deposition decreases as the particle size increases. However, after certain size of particle, there is no impact on the deposition distance. This critical size particle is called “deposition critical size”. The experimental tests are shown to be in good agreement with the model predictions in terms of the following criteria: 1) Formation of hydrate particles are observed to be poly-dispersed since different sizes of particles are formed; 2) Studying the influence of the Reynolds number and pipe diameter, the deposition distance is found to be linearly corresponded to both.

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1. Introduction

Particles deposition is a process that plays a key role in many fields ranging from atmospheric applications to material sciences. In oil and gas pipelines field, accumulation of hydrate is one of the most challenging aspects in the flow assurance modeling.

Hydrate can pose a major risk in all high pressure natural gas transport lines including the connecting lines and manifold systems in all offshore production facilities. Marine transportation of compressed natural gas is one example where prediction of hydrate formation is a requirement for the safe transport of gas to and from ocean going ships. Production facility components such as chokes, velocity-controlled subsurface safety valves, and conventional valves and fittings can all act as restrictions to the flowing fluids resulting changes in flow conditions which could lead to the formation of hydrate particles accumulated eventually in the pipeline.

Hydrate plugs have disturbed the normal flow of natural gas and other reservoir fluids in the production and transportation lines and have claimed lives of personnel and resulted loss of property in oil and gas industries (Sloan, 1998). Hydrate propagation tends to gradually form a plug that separates the pipe into two pressure sections: a high pressure section between the well or high pressure gas source and the plug and a second section at low pressure between the plug and the gas recovery division. In the upstream section, a pipe blast can occur due to pressure rise. The plug can also behave as a projectile that destroys the pipe when the pressure difference between the upstream and downstream sections increases. Both events can endanger personnel safety and damage production equipment (Sloan, 2000).

Considering such significant economic risks in the gas and oil industry, a great deal of research has been conducted by petroleum industry to hinder this undesirable phenomenon. Annually an operating expense greater than $500 million is allocated to hydrate prevention (Lederhos et al., 1996), almost half of which spent for...
hydrate inhibition (Sloan, 2003). In addition, offshore operations expend approximately $1,000,000 per mile for insulation of subsea pipelines to prevent hydrates. Further, Lysne (1995) listed three incidents in which hydrate projectiles erupted from pipelines at offshore platforms and caused loss of three lives and over $7 million (US) in capital costs. Consequently, concerns have been expressed about the effect of hydrates on foundations of platforms and pipelines, as well as offshore drilling.

The basic behaviours of any flowing particles such as deposition, transport, lifetime, and optical influence, depend strongly on their size, their chemical composition, and on the nature of the carrier fluid. Thus the interaction of these parameters is the dominating size, their chemical composition, and on the nature of the carrier transport, lifetime, and optical in offshore drilling.

The basic behaviours of any flowing particles such as deposition, transport, lifetime, and optical influence, depend strongly on their size, their chemical composition, and on the nature of the carrier gas. Thus the interaction of these parameters is the dominating process that controls the nucleation, the size distribution and the gas. Thus the interaction of these parameters is the dominating process that controls the nucleation, the size distribution and the deposition of the flowing particles.

Because of simultaneous influences from fluid flow convection, particle-boundary hydrodynamic interaction, colloidal interaction, molecular diffusion, and external applied body forces, particle deposition onto surface is an extremely complicated process. Chein and Liao (2005) found that the particle could be suspended above or deposited on the wall, depending on the Hamaker constant, the surface potentials of the particle and wall, and the thickness of the electrical double layer (EDL).

Yang et al. (1998) divided the process of particle deposition into four steps according to the distance between the particle and the surface:

- The particle transport is controlled by the fluid convection and external body force when the distance is large.
- As the distance becomes comparable to the particle size, additional force acting on the particle is produced due to the presence of the wall. This force, known as particle-wall hydrodynamic interaction, reduces the particle mobility.
- When the distance becomes within the range of 1–100 nm, the van der Waals (VDW) forces and the electrical double-layer (EDL) force due to the interaction between the surface potentials of particle and wall start to affect the particle motion. These two interactions form the basis of the well-known Derjaguin–Landau–Verwey–Overbeek (DLVO) theory of colloidal stability (Derjaguin and Landau, 1941; Verwey and Overbeek, 1948).

As the distance becomes smaller, within 1 nm, traditional continuum mechanisms may not be applied to tackle the particle behaviour for such molecular dimension. Hence the particle is usually considered to adhere to the wall when the VDW force is attractive.

The adhesive distance cannot be calculated from theoretical considerations and is usually approximated as a constant (Chein and Liao, 2005). Krupp (1967) suggested a value of 0.4 nm while Israelachvili (1992) proposed the adhesion distance as 0.2 nm. As the above mentioned sub-models requires first to solve numerically the fluid flow governing equations, commercial software based on Computational Fluid Dynamics (CFD) technique is used for this purpose.

The fundamental concepts related to the nucleation phenomena, growth theory, and forces applied to the particle are...
well established and ready to be integrated. The comprehensive
models which are able to predict hydrate formation and accumu-
lation have also been successfully implemented. However, based on
our extensive literature survey and best knowledge, models
specifically developed to predict the most probable location for
hydrate deposition under conditions where natural gas with little
or no free water flows through restrictions in pipeline systems are
still unavailable and need to be addressed.

A comprehensive model has been built to predict the deposition
spots of hydrate. The model could be used to modulate the
collection phenomena, analyze the forces applied to a particle, and
describe the process of deposition in a new approach. The latter is
the main issue concerned in the present work.

In this paper the theory of the particle migration in gas flow and
the mechanism of the particle deposition are brie

4. The proposed model

The model consists of the following parts:

i) Computational fluid dynamic (CFD) technique to configure
the flow field;

ii) The new correlation of hydrate growth and distribution based
on the satisfaction of the Law of Mass action, developed by
Jassim (2008), is considered to predict the incipient hydrate
particles size and growth rate.

iii) The principle of particle deposition velocity is used to track
the particle motion in the turbulent regime;

iv) A novel approach to describe the particle behaviour near wall
region is implemented.

4.1. Sublayer region

The behaviour of the particle in the boundary layer region is influenced
by the fluid turbulence and properties, particle-wall interactions,
particle inertia, gravity in the case of horizontal flows, and local wall

The particle path could be explained from the resultant force
acting on the particles based on the force balance method. As
mentioned earlier, lift force is the main force affecting the particle
trajectories and the lift force resists the lifting force and jointly in
fluence the particle path.

4.2. Bouncing model

When a solid particle contacts a surface at low velocity, the
particle loses its kinetic energy by deforming itself and the surface.

For fully developed turbulent flow in a circular pipe of length \(L\),
the deposition velocity \(V_d\) may be considered as constant along the
tube, and the collection fraction can be express as (Crowe, 2006):

\[
\eta = 1 - \exp \left( - \frac{4V_d L}{U_g D} \right)
\]

Where, \(V_d\) is the mean velocity; \(U_g\) is the carrier gas
mean velocity; \(D\) is the pipe diameter, and \(k^+\) is the surface
roughness (which is zero for smooth surface).

\[
V_{dr} = \begin{cases}
0.0845c^{-2/3} + \frac{1}{2} \left[ \frac{(0.64k^+ - 0.5d_p)^2}{1 + \frac{d_p}{S}} \right]
\times \left[ 1 + 8e^{-(10/32)} \frac{0.072}{1 + \frac{d_p}{S} + \tau^+ g^+} \right]
& \text{if } V_{dr} < 0.14 \\
0.14 & \text{otherwise}
\end{cases}
\]

where, \(V_{dr}\) is the deposition velocity; \(d_p\), \(L_s^+ = \frac{3.08S}{d_p}\), \(S = \frac{d_p}{R}, \quad d_p^+ = \frac{d_p U_g}{k^+}, \quad U_g\) is the carrier gas
mean velocity; \(D\) is the pipe diameter. and \(k^+\) is the surface
roughness (which is zero for smooth surface).
At high velocities, part of kinetic energy is dissipated in the deformation process (plastic deformation), and part is converted elastically to kinetic energy of rebound (Hinds, 1999). If the rebound energy exceeds the adhesion energy, the particle will bounce away from the surface.

There are two approaches used in defining the conditions at which the particle could bounce. The first approach is based on the limiting adhesion or kinetic energy whereas the second approach defines a critical velocity $V_c$ for which bounce will occur if that velocity is exceeded.

Friedlander (1977) described the process of bouncing as a balance of energy before and after impact and proposed a relation between the velocities of the particle before and after impact. The expression is given as:

$$v_2 = \sqrt{v_1^2 - \frac{E(1 - e^2)}{m_p}}$$  \hspace{1cm} (7)

Where, $m_p$ is the mass of the particle and $e$ is the restitution coefficient. The critical approach velocity corresponding to $v_2 = 0$ is given by:

$$v_{1c} = \sqrt{\frac{2E}{m_p}\left(1 - \frac{e^2}{2}\right)}$$  \hspace{1cm} (8)

Where, $E$ is the surface potential energy given by (Dahneke, 1971):

$$E = \frac{d_pA}{12y_0}$$  \hspace{1cm} (9)

$A$ is the Hamaker constant; $y_0$ is the equilibrium separation distance of the particle from the surface, which depends on the scale of surface roughness. For smooth surfaces, the distance is usually assumed to be 0.4 nm (Hinds, 1999).

The restitution coefficient, "$e$" proposed by Legendre et al. (2005), was used in the analysis:

$$e = e_{max}\exp\left[\frac{\beta}{St}\right]$$  \hspace{1cm} (10)

$\beta$ is a parameter taking the value 35.

$e_{max}$ is 0.91, the maximum coefficient of restitution that can be reached by the particle for collisions in air for both solid particles and drop.

$St$ is defined as the Stokes number of the particle away from the wall by an order of boundary layer thickness.

$$St = \frac{\left(\rho_p + 0.5\rho_f\right)U_f d_p}{9\mu_g}$$  \hspace{1cm} (11)

4.3. Calculating the distance of the deposition

According to the proposed model, the procedure for calculating the distance traveled by a single particle before depositing on the wall could be summarized as follows:

- The deposition velocity is first determined using Eq. (6).
- The time required for the particle to reach the sublayer region can be found from:

$$t = \frac{y_i}{V_d}$$  \hspace{1cm} (12)

Where, $y_i$ is measured from the tube wall.

- Within this time, the hydrate particle grows from the original size $r_{p0}$ to a new size $r_p$. The correlation of particle radius as a function of time becomes (Jassim et al., 2008):

$$r_p^2 = \sqrt{e^{2\beta t}\frac{r_{p0}^2}{\rho_p} + \frac{\beta t}{\rho_p} (e^{2\beta t} - 1)}$$  \hspace{1cm} (13)

- Now, launching from rest, the particle velocity can be determined using the following formula, which analytically derived from the Equation of Motion (Jassim et al., 2008):

$$u_p = \left(\frac{u_g}{g\tau_v}\right) \times \left(1 - \exp(-t/\tau_v)\right)$$  \hspace{1cm} (14a)

where,

$$\frac{U_u - u(y_i)}{u} = 2.5\ln\frac{R}{y_i}$$  \hspace{1cm} (14b)

and the residence time,

$$\tau_v = \frac{\rho_p d_p^2 C_c}{18\mu_g}$$  \hspace{1cm} (14c)

- The traveling distance in the turbulent region becomes:

$$X_1 = u_p(y_i) t$$  \hspace{1cm} (15)

- In the sublayer region, the particles smaller than the thickness of the boundary layer could migrate further as a result of external forces. Hence, the total distance from the initial position would be:

$$X_t = X_1 + X_3$$  \hspace{1cm} (16)

- When particles larger than the sublayer thickness, the bouncing distance ($X_b$), the distance taken by the particle to settle as a result of rebound, is added to the distance traveled in the turbulent region.

$$X_t = X_1 + X_8$$  \hspace{1cm} (17)

4.4. Summary of the deposition model

The flowchart presented in Fig. 2 summarises the procedure of the particle migration and process of deposition in turbulent flow. As particles travel in the fully turbulent region, deposition velocity is evaluated (depending on their sizes) and used to determine collection factor, the elapsed time required to reach the boundary layer, and the new size of such particle before merging in the sublayer regime.

In the sublayer region, again the size of the particle is used to direct the phenomena of the deposition to the proper model of deposition process, which is either the balance of the forces experienced by the particle or the probability of bouncing. Hence, the extended distance acquired by the particle before settling occurs would be evaluated.

5. Experimental apparatus and procedure

5.1. Preliminary experiment setup

Before constructing the actual apparatus for natural gas/hydrocarbon tests, we decided to perform preliminary tests using saturated air as the working fluid. The purpose of these tests was to obtain a better understanding of the test system by monitoring ice particles formed and deposited in the transparent tubes. The following section outlines the system setup.
5.1.1. Apparatus and procedures

As illustrated in Fig. 3, the compressed air is saturated by bubbling it through the water container, cooled down through cooling coil to drop the temperature to just above 0 °C, and then passed through separator to separate the air stream from any liquid droplets that might be formed during the cooling process. After that, the air flows through the orifice, which is connected to the tube where the accumulation is monitored, and finally the stream passes through a flowmeter before exiting to the ambient. The tests were performed at the Faculty of Engineering and Applied Science of Memorial University of Newfoundland using the Environmental Chamber located in the Fluids Lab. The entire system but the air source and the saturated vessel were placed inside the environmental chamber.

The compressed air pressure was measured downstream of the compressor. The orifice inlet pressure and temperature were directly measured using the attached pressure gauges and temperature sensors. Information about pressure loss through water container, cooling coil, separator, and all connections were recorded. No temperature measurement sensor was placed at the

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**Fig. 2.** Flowchart describing process of particle migration and deposition in turbulent flow.

**Fig. 2.** (continued).

**Fig. 3.** Schematic of deposition apparatus using saturated air.

**Fig. 4.** Numerical prediction of temperature variation through an orifice.
orifice outlet as any measuring sensor in this position could perturb the air flow and affect the process of condensation and/or crystallization. The pressure and temperature after the orifice were calculated using the procedure detailed in Appendix A1.

The original plan was to observe ice particles formed on the tube wall. However, only water droplets (as opposed to ice particles) could be observed. Numerical analysis using computational fluid dynamics software (the Fluent software was used here) was conducted to predict the temperature after the orifice. Fig. 4 shows the variation of air temperature along the orifice region.

The plot proves that although the air temperature drops sharply within the orifice, it increases rapidly right after the orifice and reaches almost orifice's inlet temperature. Therefore, there is no sufficient time for water vapour to nucleate and crystallize. Raising the flow rate (by increasing the orifice inlet pressure) decreases the orifice exit temperature. However, the time that the fluid needs at this low temperature is still inadequate due to exposure to relatively high temperature of the chamber, i.e. the air stream is affected by a significant driving force. The temperature of the chamber was intentionally kept above zero to avoid plugging in the cooling coil. The observed water droplets however were an indication of where the ice particles were likely to form.

In order to see actual ice particles, it was concluded that the chamber temperature should be kept low enough to make the nuclei formed in the orifice exposed to a sufficient driving force in the tube suitable for stabilization and growth. The conclusions reached at this stage helped to plan and design the actual system built at the Centre for Marine CNG Inc. (see Section 5.2). The improved environmental chamber, shown in Fig. 5, was constructed such that the temperature of the cooling coil and the tube could be controlled independently in two separate compartments. In the two separate chambers the actual natural gas or a hydrocarbon gas with similar behaviour (e.g., propane) can be used as the working fluid in the tests for hydrate formation and deposition studies. The first chamber is set around equilibrium temperature and contains the cooling coil and separator while the second chamber contains the rest of the system where temperature could be set at sufficiently below the equilibrium temperature, so the

![Fig. 5. Schematic of improved experimental system using propane as carrier gas.](image)

![Fig. 6. Components of the gas hydrate apparatus: a) parts outside the chambers b) parts located in the small chamber (coil, separator, saturator); c) Parts located in the large chamber (orifice, pipe).](image)
formed hydrate could be under driving force as it moves through the tube. Thus, the nucleation process could be enhanced.

5.2. Improved experimental apparatus

The main objective of the experimental work was to establish, by physical evidence, the role played by the various forces discussed in previous chapters on particles deposition. The method adopted to examine this effect was to form hydrate particles as the result of a contraction to flow passage and record the distance required by the particle to deposit on the wall.

5.2.1. Flow circuit

The experimental apparatus is composed of transparent tubes approximately 2 m in length and diameters ranging from 6.35 to 19.05 mm (1/4" to 3/4"). The orifice had a diameter of 2.54 mm (0.1") and other equipment pieces are listed in Fig. 5. Two main test zones were suggested, namely: the starting or gas saturation loop, which is designated for cooling the gas and saturating it with water at the desired temperature, and the main hydrate formation loop, where the gas passes through the orifice and the tube, and the place where hydrate particles are deposited.

In the starting loop, the charged gas is mixed with the cool gas in a separator container (9) before being routed to the compressor. In order to bring the gas temperature down, it is then passed through the cooling coil (5) located in the large environmental chamber. This process continues until the compressor's exit temperature reaches a stable value. The gas, at this time, will be directed to the main loop by opening Valve (2).

The gas then moves through the Container (3) filled half with water to saturate the gas, the cooling coil to bring the gas temperature to the equilibrium level, and the Separator (6) to trap any droplet that could form during the cooling process in the coil. At this point, the gas is at equilibrium temperature and ready to flow through the system. It is worth to note here that the temperature of the small environmental chamber is adjusted around the gas hydrate formation temperature.

The pressure and temperature of the gas are monitored once again upstream of the orifice and the tube to confirm that its conditions are suitable for hydrate formation. The orifice and the tube are located in a separate chamber with an independent temperature control system. As the gas flows through the orifice its temperature falls rapidly. Although the gas temperature could increase after leaving the orifice, as concluded from the numerical simulation mentioned earlier, the low temperature of the second chamber will keep the gas below the equilibrium hydrate formation temperature. Fig. 6 shows some photos of the actual system.

1. Compressor
2. Valve
3. Gas saturator
4. Gas charging valve
5. Cooling coil
6. Separator
7. Valve
8. Flow meter
9. Liquid trap/filter

6. Water-air system

The geometry used in this study (Fig. 7) is a simplified version of the actual orifice-pipe system, with the axisymmetric assumption. The dimensions of the orifice and the pipe are shown in Fig. 7.

The boundary conditions for the saturated air case are listed in Table 1, which represents the data measured under the experiments' conditions.

Fig. 8 shows the temperature contours inside of the orifice. Due to the variable temperature within the orifice, ice particles with diverse critical sizes may form as a result of different driving forces. This can also be interpreted as the flow leaving the orifice may contain a wide range of particle sizes. This conclusion will be further analyzed in the subsequent discussions.

6.1. Nucleation and growth of ice particles

The data obtained from the FLUENT software was used to describe and estimate the process of nucleation and growth of the ice particles. Using mass transfer as the predominant process of nucleation and growth, parameters used for describing the processes are listed in Table 2.

The variation of the particle size and its growth rate with time for the conditions listed in Table 2 are now presented. Fig. 9 shows the growth rate of ice particles in micrometer scale as a function of time. The growth starts with a very fast rate after the ice particle becomes nucleated and slows down as the time elapses. That means the ice particle growth rate decreases with particles size. Hence, the time required for ice particles to grow to several microns is relatively long; see Fig. 10.
Particles formed in the orifice have different values of critical sizes. This could also be due to the fact that the temperature distribution inside the orifice is not uniform. The temperature of the flow inside the orifice ranges from 235 to 272 K. Accordingly, the critical sizes would be 11–13 nm.

Since the measurement of the actual nucleus (critical particle) size was not possible in this research, the analytical correlation of the Particle Distribution Function listed below was used (Jassim, 2008).

\[ n^*(t, r) = 1.65r^* \exp \left( \frac{F_r}{C_0} r^* \right) \times \frac{1}{r^*} \] (18)

where,

**Particle Fourier number** : \( F_r = \frac{\beta_1 t}{r_c^2} \) (19a)

**Reduced radius** : \( r^* = \frac{r}{r_c} \) (19b)

Fig. 11 illustrates the number of particles in dimensionless form (using Eq. (18)) as a function of dimensionless particle radius at time equal zero (nucleation time). The graph proves that the distribution of the particles contains different sizes; in particular particles with radii larger than the critical radius. It means that, analytically, the size of the particles just after the orifice are neither identical nor all at critical size, rather large sizes (several microns) could also exist. Besides, due to large number of particles formed, the probability of particle–particle attachment is possible and even larger sizes (up to hundreds of microns) could form.

Hence the assumption made here is based on the conclusion that the particles leaving the orifice have different sizes and the approach used to describe the deposition model in this study is based on this fundamental hypothesis.

### 6.2. Calculating deposition distance

Based on the procedure elaborated in Section 4.3, the traveling distance in the turbulent region becomes:

\[ X_t = u_p(y_1) \times t \] (20)

The total distance includes, as discussed in Section 4.3, the distance traveled by the particle, either due to sublayer forces or as a result of bouncing.
6.2.1. Trajectory of a single particle ($d_p < \delta$)

The following data, obtained from the actual saturated air test, is used as the initial conditions in the simulations to track and calculate the distance from the initial position of a single ice particle ($x_0, y_i$) to the point of resting on the wall ($X_t, 0$):

- Smooth pipe: $D_{pipe} = 20$ mm; $f = 0.023$; $Re \approx 32800$ and $\delta \approx 56.7 \mu$m

Fig. 12 illustrates the deposition of a 10-µm particle initially located at the centre of the pipe where $u(y_i)=U_c$. Using the conditions mentioned earlier, the results show that the particle travels $\sim 7$ m before entering the boundary layer (BL) region. In the BL, the particle moves $1$ mm then deposits on the wall. Although the particle migration in the BL region is minor for this particular size, very small particles tend to travel a significant distance in this region. For instance, particles with 0.1-µm diameter travel 5995 m in the turbulent flow and 1.5 m in the BL region.

6.2.2. Trajectory of single particle ($d_p > \delta$)

Particles having sizes greater than the sublayer thickness experience bouncing process as a result of collisions in the wall region. The total distance that the particle travels is now equivalent to the migration distance in the turbulent regime plus the distance traveled due to bouncing effect. Fig. 13 shows the deposition distance as a function of the particle size for particles diameters greater than the sublayer thickness. The three curves in this figure indicate in the order: the distance caused by bouncing, the distance traveled in turbulent region, and the total distance.
traveled in the turbulent region, and the total distance traveled by the particle. The figure also shows that as the particle becomes larger, the inertia of the particle increases and hence the bouncing distance will shrink.

A very important conclusion is shown in Fig. 14, which represents a combination of results shown in Figs. 12 and 13. The reduction in the total deposition distance will asymptotically approach a certain value. Hence the conclusion reached from Fig. 12, which stated that the deposition distance decreased dramatically following an increase in the particle size, should be corrected as follows: For particles larger than a certain diameter, called hereafter deposition critical size, there is no significant influence of the particle size on the deposition distance. Consequently, particles having sizes greater than the deposition critical size will deposit at the same position. In this example, (ice formation studies), particles larger than 200 μm will deposit almost at the same location (0.6 m). Therefore the deposition critical size for this example is about 200 μm. Therefore deposition will start at about 0.6 m and all smaller particles will travel further until they become large enough to start settling on the wall.

6.3. Experimental results and preliminary model validation

Several tests were conducted using four tubes and at different Reynolds numbers. The transparent tubes used for the tests were 1.83 m (6 ft) in length and their diameters were varied from ¼ to ¾ inches. The tests for each tube were conducted using three different flow rates controlled by changing orifice inlet pressure through controlling the air pressure at the compressor discharge. Position of the first droplets deposit on the wall was observed and the distance from the orifice to that position was measured.

Based on the procedure of the experiments described in Section 5, the temperature of the air at the exit of the orifice drops slightly below water freezing point. The temperature right after the orifice starts to increase rapidly and reaches almost the orifice inlet value within a short distance (≤ 2 cm). Theory and numerical analysis matched the observations. Both confirmed that the ice particles, if formed in the orifice, thawed out due to either the rapid increase in the pressure and temperature after the orifice or as a result of the heat transfer with the surrounding. The latter means that the ice particles maintained in the solid form during migration in the flow then melted after deposited on the tube wall. In the present analysis both situations are studied by including the bouncing effect for solid particles and ignoring such effect for water droplets.

During the tests the following parameters were recorded: exit air standard volume flow rate Qex, inlet P1 and T1; and system exit conditions Pex and T. The given data were reported in standard condition (101 325 Pa; 15 °C).

The measured data were compared with the model predictions to confirm the level of agreement and to validate the accuracy of the simulations. The model presumed that various sizes of particles, ranging from the critical size (r ≤ 11.5 nm) to several hundred microns (200 μm), are leaving the orifice. It is concluded from Fig. 14 that particles larger than this range have no influence on the deposition distance. Based on the assumption, the model is designed to predict the deposition distance of these sizes using the approach discussed in Section 4.3.

Fig. 15 illustrates the distance from the orifice to the location of deposition as a function of pipe size when Reynolds number is 32.8 × 10^3. The diagram shows a fair agreement between the experiment and the model prediction. As shown in Table 3 the distance of the deposition increases with the pipe size.

Finally, comparison of the ice particles with water droplets is presented by studying the influence of the bouncing process. Fig. 16 illustrates the deposition distance as a function of the pipe size with and without bouncing effect, i.e. assumption of particles remaining in the solid or liquid states, respectively. It could be concluded from the diagram that the discrepancy of the deposition distance predicted by the simulation for the liquid droplets and solid particles with that observed in the tests for all pipes is within a reasonable range. The percent errors in model prediction of deposition distance for the liquid droplets and ice solid particles with respect to the experimental results are listed in Table 4. The results show good agreement between the simulations prediction and the tests observation.

### Table 3
Percent error of deposition distance observed in experimental tests and predicted by simulation for Re = 32 800 (Fig. 15).

<table>
<thead>
<tr>
<th>Pipe Diameter (cm)</th>
<th>Model prediction (cm)</th>
<th>Test result (cm)</th>
<th>% discrepancy</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.36</td>
<td>19.1</td>
<td>15</td>
<td>21.5</td>
</tr>
<tr>
<td>9.53</td>
<td>28.7</td>
<td>24</td>
<td>16.4</td>
</tr>
<tr>
<td>12.70</td>
<td>38.2</td>
<td>40</td>
<td>4.7</td>
</tr>
<tr>
<td>19.05</td>
<td>57.3</td>
<td>55</td>
<td>4.0</td>
</tr>
</tbody>
</table>

### Table 4
Comparison of the deposition distance for ice and water particles observed in the tests and predicted by the simulation for Re = 32 800.

<table>
<thead>
<tr>
<th>Dpipe (mm)</th>
<th>Test result (cm)</th>
<th>Liquid droplet</th>
<th>Ice particle</th>
<th>% error of droplet</th>
<th>% error of ice particle</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.35</td>
<td>25</td>
<td>19.12</td>
<td>27.86</td>
<td>23.5</td>
<td>10.64</td>
</tr>
<tr>
<td>9.525</td>
<td>30</td>
<td>28.7</td>
<td>33.95</td>
<td>4.33</td>
<td>13.17</td>
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<td>12.7</td>
<td>40</td>
<td>38.2</td>
<td>41.93</td>
<td>4.5</td>
<td>4.8</td>
</tr>
<tr>
<td>19.05</td>
<td>55</td>
<td>57.3</td>
<td>59.6</td>
<td>4.2</td>
<td>8.4</td>
</tr>
</tbody>
</table>

### Table 5
Percent of uncertainty error for saturated air tests.

<table>
<thead>
<tr>
<th>Re</th>
<th>% error</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Size (3/4&quot;)</td>
</tr>
<tr>
<td>32 800</td>
<td>3.3</td>
</tr>
<tr>
<td>35 300</td>
<td>3.4</td>
</tr>
<tr>
<td>37 300</td>
<td>3.1</td>
</tr>
</tbody>
</table>

Fig. 16. Comparison of deposition distance predicted by simulation for different types of particles (assumption of particles remaining in ice or liquid forms when reaching wall) with experimental data as a function of the pipe size.
6.4. Analysis of uncertainty for saturated air experiments

There is a fact that uncertainties exist in virtually all computational, analytical, or experimental engineering analyses. The purpose of this section is to manifest the uncertainty analyses, which was used in the planning of experimental facilities. Details on the approach used in the study can be found in References (21,22).

During the tests, the independent variables or the measured parameters were: flow temperature and pressure \(T, P\), standard flow rate \(Q_0\), pipe diameter \(D_{pipe}\), and the standard temperature and pressure \(T_0, P_0\).

Therefore, the uncertainty equations of the deposition distance \(S_d\) become:

\[
S_d = S_d(Q_0, D_{pipe}, T, T_0, P, P_0)
\]

\[
U_{S_d} = \left[ \left( \frac{\partial S_d}{\partial Q_0} \right)^2 + \left( \frac{\partial S_d}{\partial D_{pipe}} \right)^2 + \left( \frac{\partial S_d}{\partial T} \right)^2 + \left( \frac{\partial S_d}{\partial T_0} \right)^2 + \left( \frac{\partial S_d}{\partial P} \right)^2 + \left( \frac{\partial S_d}{\partial P_0} \right)^2 \right]^{1/2}
\]

7. Propane-hydrate tests

Propane was used as the working fluid since the propane hydrate can form above water freezing temperature under relatively low pressures.

Table 6
Data recorded from a typical propane test.

<table>
<thead>
<tr>
<th>Suction</th>
<th>Discharge</th>
<th>Point 2</th>
<th>Point 3</th>
<th>Point 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>3</td>
<td>63</td>
<td>60</td>
<td>8.0</td>
</tr>
<tr>
<td>Pressure (kPa)</td>
<td>82.74</td>
<td>455</td>
<td>413.7</td>
<td>193</td>
</tr>
<tr>
<td>Standard flow rate ((m^3/s))</td>
<td>13 \times 10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Corrected flow rate ((m^3/s))</td>
<td>4.28 \times 10^{-3}</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 7
Boundary conditions for typical saturated propane simulations.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Values</th>
<th>Units</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inlet Pressure</td>
<td>1.95 \times 10^7</td>
<td>Pa</td>
<td>gauge</td>
</tr>
<tr>
<td>Total temperature</td>
<td>279</td>
<td>K</td>
<td></td>
</tr>
<tr>
<td>Exit Pressure</td>
<td>1.93 \times 10^{-5}</td>
<td>Pa</td>
<td>gauge</td>
</tr>
<tr>
<td>Total temperature</td>
<td>278</td>
<td>K</td>
<td></td>
</tr>
<tr>
<td>Lateral Wall Temperature</td>
<td>272</td>
<td>K</td>
<td>Isothermal</td>
</tr>
</tbody>
</table>

7.1. Test procedure

A compressor was used to circulate the gas within the system. The gas loops were charged with propane and the compressor was started. The maximum pressure recorded in the transparent pipe was around 293 kPa ab (~42.5 psi). The hydrate equilibrium temperature \(t_{eq}\) relative to this pressure is 3 °C (Carroll, 2003). The small chamber was adjusted to a temperature slightly above \(t_{eq}\) (6 °C) to prevent any hydrate formation in saturation zone. The temperature in the large chamber (hydrate formation zone) was set to a temperature below \(t_{eq}\) (~2 °C) to induce a driving force large enough to stabilize the nucleation and particle growth stable.

It should be noted here that the saturator was originally located before the cooling coil outside of the small chamber. However, the temperature of the gas heading to the saturator was observed to be excessively high making the gas oversaturated leading to excessive condensation in the saturation zone. The condensed water overloaded the filter separator and ended up as free liquid in the transparent tube in the hydrate formation zone. Since the purpose of this study was to investigate the hydrate formation in the absence of free water, it was decided to saturate the gas, under low temperature of saturation zone, by moving the saturator to downstream of the coil inside the small chamber.

The data shown in Table 6 were recorded during the tests and the flow rates were corrected using Eq. (A1.1) in Appendix A1. Table 6 shows the recording data and the corrected flow rate for a typical test using the \(\frac{3}{4}\)“ pipe. From the table, the following parameters were calculated:

\[
\text{Fig. 17. Hydrate particles deposit on the pipe wall (magnification factor 2.73).}
\]

\[
\text{Fig. 18. Propane hydrate growth rate as a function of particle size.}
\]
Gas superficial velocity: \( U_g = 15.02 \text{m/s} \);

Gas density: \( \rho = 5.6 \text{ kg/m}^3 \);

Reynolds number: \( \text{Re} = 14560 \)

These parameters, along with the environmental temperature inside the large chamber, were used as the boundary conditions in the simulation model to predict the deposition of the hydrate particles.

7.2. Observations

As shown in Fig. 17, propane hydrate with different particle sizes was formed. It is presumed that due to the particle–particle attachment (coalescence within the gas and on the tube wall), formation of various critical sizes, and different growth times, a range of particle sizes were formed.

After the completion of the test, the distance from the orifice to the location of the first observable particle was measured. Then the pipe was dismantled and abandoned for a while to assure there were no water droplets remaining in the pipe before doing the next test. This procedure was repeated three times for different flow-rates to investigate the influence of the Reynolds number on the deposition distance.

7.3. CFD and simulation results

The model of nucleation needs the distribution of fluid properties, e.g., temperature, velocity, pressure, along the pipe, the governing transport equations of air-vapour mixture. These properties were numerically determined using commercial software (the FLUENT software was used in this research).

The boundary conditions are listed in Table 7, which represents the data measured for typical test conditions.

7.4. Nucleation and growth of hydrate particles

The data obtained from FLUENT were used to describe the process of nucleation and growth of the hydrate particles. Mass transfer and hydrate formation reaction kinetics are the dominant processes in the nucleation and growth of the hydrate particles.

Fig. 18 shows the growth rate of hydrate particles in the micrometer scale as a function of size (diameter). Since mass transfer is the dominating factor of growth after the hydrate was just nucleated, the rate of growth will decrease as the size of hydrate grows. However, the reaction kinetics will start to be the controlling factor in the growth process as the particle size grows further. Therefore there is a dip (valley) in the growth rate diagram. That means that the elapsed time required for hydrate particles to grow to several hundreds of microns is relatively long but the trend dramatically change as the particle size increase beyond 100 microns and a sharp change of particle diameter is observed; see Fig. 19.

7.5. Deposition distance

The data of the measured deposition distance from the orifice exit to the first deposition spot were recorded with a measurement

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Change in the parameter</th>
<th>( \Delta \text{Re} )</th>
<th>( \Delta S_d )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flowrate ( (Q_0) )</td>
<td>±2%</td>
<td>306</td>
<td>3</td>
</tr>
<tr>
<td>Diameter ( (D_{pipe}) )</td>
<td>±1%</td>
<td>151</td>
<td>2</td>
</tr>
<tr>
<td>System Temperature ( (T) )</td>
<td>±1K</td>
<td>55</td>
<td>1.3</td>
</tr>
<tr>
<td>System Pressure ( (P) )</td>
<td>±13.8 kPa</td>
<td>53</td>
<td>1.3</td>
</tr>
<tr>
<td>Ambient Temperature ( (T_0) )</td>
<td>±1K</td>
<td>720</td>
<td>6.6</td>
</tr>
<tr>
<td>Ambient Pressure ( (P_0) )</td>
<td>±13.8 kPa</td>
<td>2109</td>
<td>19</td>
</tr>
<tr>
<td>( U_g )</td>
<td></td>
<td>20.5</td>
<td></td>
</tr>
</tbody>
</table>
tape with millimetre precisions. The deposition measurement data were compared with the results obtained from the simulation. Table 8 illustrates such comparison of deposition distances with the experiment data. The deposition measurement data were compared with the results obtained from the simulation. The deposition measurement data were compared with the results obtained from the simulation.

7.6. Uncertainty analysis

Following the procedure elaborated and using Eq. (22), the uncertainty in the deposition distance and the precision of each parameter are calculated. Tables 9–11 demonstrate the discrepancy in the deposition distance for each measuring parameter and the total uncertainty error for each test.

8. Grid independency

Since the boundary layer (δ) in the orifice is of interest in the present study, for all CFD analyses, a non-uniform mesh was used in the y direction, and the smallest elements were placed close to the orifice wall. The parameters used were: first row a = 0.16; aspect ratio, the ratio of the two consecutive mesh thicknesses, (t1/δ/ t1 = 1.2) and the depth of the boundary layer meshing region g; δ. Starting from the wall, the height of the first grid row should be less than the boundary layer thickness to describe the flow in that region more precisely.

A structured grid with 575 340 cells was used for the calculations. Resolution was found to be independent of cell dimensions as seen in Table 12. The table shows the number of grids that led to convergence for each case with total mass error in inlet/outlet mass flow. It is clearly shown that meshing with more than 575 340 grids did not significantly affect the mass residual for convergence, hence it can be concluded that the solutions were grid independent.

9. Conclusion

The particle deposition theory was proposed to predict the deposition of the hydrate particles formed in a gas/vapour dominated flow. Computational fluid dynamics was used to configure the carrier fluid through solving the Navier–Stokes equation for turbulent flow. A new approach was developed to estimate the distance of particle migration from the spot of formation to the point of deposition.

The concept of the particle deposition velocity is introduced to help to predict the trajectory of the particle motion in the turbulent region. The model presented in this research proposes a new approach to track the particle motion merged in the sublayer region using the forces acting on the particle. For particles with sizes larger than the sublayer thickness, the model introduces the influence of the bouncing concept to explain the near wall effects. This study also presented the apparatus employed to validate model results using water saturated air and propane.

The approach and observations of the tests concluded that particles within the boundary layer travel further in the flow direction as a result of external forces (lift, drag, gravity, and adhesion). The bouncing action is the dominant process causing large particles to further travel before being attached to the pipe wall.

The main conclusions of the research can be summarized as follows:

1. In the turbulent region, small particles are influenced by the main fluid velocity but the effect diminishes for relatively large particles as a result of the high particle inertia.
2. The distance of deposition decreases as the particle size increases. However, the analysis has introduced a certain size of particle in which further particle growth has no effect on the distance of deposition. Such size was called “deposition critical size”.
3. The experimental tests were in good agreement with the model predictions and that within the test conditions, the deposition distance is linearly proportional to the Reynolds number and pipe size.
4. Formation of hydrate particles are observed to be poly-dispersed since different sizes of particles are formed. This observation is consistent with the analytical correlation derived based on the law of Mass Action (Jassim, 2008).

Acknowledgments

The authors wish to express their gratitude to the: Propel Inc., Canada, Atlantic Innovation Fund (AIF) through Atlantic Canada Opportunities Agency (ACOA), Natural Racecourses Canada (NRCan) for financing this project.

Appendix A1

The target of the following analysis is the temperature at orifice exit (T2), Fig. A1.

- Determining actual mass flow:

  Using the ideal gas relation:
Appendix A2. Calculation of parameters in Table 2

\[ n_p = \frac{P}{RT} = \frac{150}{8.314 \times 272} = 0.067 \text{ kg mole/m}^3 \]

**Diffusion coefficient (Bird et al., 2002)**

Critical temperature and pressure for:
- Air: 133K; 39 atm.
- Water: 647.1K; 217.75 atm.

\[ D_{AB} = \frac{3.64 \times 10^{-4}}{1.5} \times \left( \frac{272}{\sqrt{133 \times 647.1}} \right)^{2.334} \times (39 \times 217.75)^{1/3} \times (133 \times 647.1)^{5/12} \times \left( \frac{1}{29} + \frac{1}{18} \right)^{1/2} \]

\[ D_{AB} = 0.16366 \text{ cm}^2/\text{s} \]

Sherwood number: assumed to be \( Sh = 2.0 \).

**Equilibrium water content \( y_{eq} \):**

Using the diagram (A2), the water content at 0 °C and 1.5 bar is:
\( \sim 0.002 \text{ kg/m}^3 \text{ air. The water content in molar unit can be calculated as follows:} \)

1 gr – mole of a gas = 22.4 L at atmospheric pressure and 0°C temperature

\[ y_{eq} = \frac{0.002 \times 10^3 \times 22.4 \times 10^{-3}}{18} = 2.49 \times 10^{-3} \text{ gr – mole of water/gr – air} \]

**Saturation ratio (S) and actual molar fraction \( y \):**

From Eqs. (4.8) and (4.9), (Jassim, 2008):

\[ S = 1 + \frac{\Delta H_f(T_{eq} - T)}{RT^2} \]

\[ S = \frac{y_{eq}}{y_{eqk}} \]

\[ 0 < S < 1 \]

Fig. A2. Water content of saturated air.
Critical radius ($r_c$):

The range of flow temperature inside the orifice is 235–272K. Based on that the range of the critical radius would be:

$$r_c = \frac{2 \times 0.033}{1000 \times \frac{8314}{273} \times T \times \ln(1.076)} = (11.5 - 13.37) \text{ nm}$$

**Appendix A3. Uncertainty analysis of measuring data**

The sensitivity of the flowmeter device used in the experiment is ±2%, the tolerance error in the size of the pipe is ±1%, the accuracy of temperature measurement is within ±1 K, and that of the pressure measurement is ±13.79 KPa (±2 PSI).

The measurement's errors influence the flow Reynolds number according to the following formula:

$$Re = \frac{U_g D_{pipe}}{v_g} = \frac{4Q}{\pi D_{pipe} v_g}$$ (A3.1)

where,

$$Q = Q_0 \times \frac{P_0}{P} \times \frac{T}{T_0}$$ (A3.2)

The actual flowrate is sensitive to the error of measurements in the standard flowrate, the system temperature and pressure, and the standard temperature and pressure. Besides, the viscosity of the fluid changes strongly with the temperature (White, 2003). All these parameters will disturb the magnitude of Reynolds number followed by the value of the deposition distance ($S_d$). The following equation, which represents the variation of kinematics viscosity with temperature for air, was used in the analysis:

$$v_g = -1.1555 \times 10^{-14}T^3 + 9.5728 \times 10^{-11}T^2 + 3.7604 \times 10^{-8}T - 3.4484 \times 10^{-6}$$ (A3.3)

Where, $T$ is temperature in K, and $v_g$ kinematics viscosity in m²/s. Table A3.1 shows such response in the values of Reynolds number and deposition distance.

Using Eq. (22), the total uncertainty in the deposition distance would be:

Table A3.2

<table>
<thead>
<tr>
<th>$Re$</th>
<th>$S_d$ (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3200</td>
<td>18.41</td>
</tr>
<tr>
<td>3500</td>
<td>18.7</td>
</tr>
<tr>
<td>3700</td>
<td>18.61</td>
</tr>
</tbody>
</table>

$U_d = 18.41 \text{ mm} = 1.841 \text{ cm}$

The measured deposition distance of this typical sample test was found to be 55 cm. Therefore, the percent error due to uncertainty is approximately ±3.3%.

Similar procedure is established for all saturated air tests and the results of the total uncertainty in deposition distance for each tests is calculated using Eq. (22). Table A3.2 summarizes the uncertainty value for different pipe size and at various Reynolds number while.

**References**


