Reply:

In this letter, we respond to the points raised by Huang in his letter commenting on our article "Population balance model for nucleation, growth, aggregation and breakage of hydrate particles in turbulent flow."[1] We appreciate the detailed attention that Huang has paid to our work and his comments on our expressions for agglomeration and breakage rates. Below are our response to his comments in the order the author makes them.

First comment

van de Ven and Mason[2] define a collision efficiency α, which is the fraction of particle collisions leading to agglomeration, in the form:

$$\alpha = k\left(\frac{H}{36\pi\mu\gamma\bar{r}^3}\right)^b \qquad (1)$$

where k is the collision-efficiency prefactor, dependent on the London wavelength; H is the Hamaker constant; μ is the fluid viscosity; γ is the shear rate, and b is an exponent, which will be discussed later on in our response. The efficiency, α, is dimensionless.

In our work, due to the lack of experimental data on the desired type of hydrate, k and H were not available, as also pointed out by Huang. For this reason, we decided to fit the two together, effectively rearranging Eq. 1 to:

$$\alpha = kH^b\left(\frac{1}{36\pi\mu\gamma\bar{r}^3}\right)^b = k_3\left(\frac{1}{36\pi\mu\gamma\bar{r}^3}\right)^b. \qquad (2)$$

We thus named the product kH^b : k_3. This should only be seen as a convenient notation (k_3 was quantified in our article by fitting the PBM model results) and does not affect the basic principles; α remains dimensionless. In general, we agree with Huang that constants and parameters with dimensions should be avoided, especially ones for which the dimensions depend on fits to experimental results.

We note that the correct expression for k_3 from Eq. 2 is: $k_3 = kH^b = \left(36\pi\mu\gamma\bar{r}^3\right)^b\alpha$, rather than the expression given in Huang in the phrase: "It is clear that: $k_3 = 36\pi\mu\gamma\bar{r}^3\alpha$."

In the same comment Huang also mentions that "viscosity is not listed," this is true, but we did state that water was used as the carrier phase, the viscosity of which is well known. Other experimental conditions were the same as in Wang et al.[3] cited in our article.

Second comment

The second comment of Huang relates to the power b, which was found by van de Ven and Mason[2] by plotting the collision efficiency against the so-called flow number (the ratio between the attractive force, e.g., van der Waals forces, and the hydrodynamic force, acting on the particle).

We would like to comment that b 0.18 was not found from experiments in Ref. 2, as Huang indicates, but from solutions of the fundamental equations for particle movement in the fluid flow at the stage of doublet formation. It is true that the particle radius was lower than 4 μm, as Huang states. However, it has not been shown that the power of 0.18 cannot be used for larger particles. On the contrary, we can refer to at least two papers, namely Soos et al.[4] and Wang et al.,[5] who used the same power in their expressions for collision efficiency for the maximum agglomerate size of 35 and 40 μm. Both the models were in excellent agreement with the experimental data. Thus, Eq. 1 is likely to be valid also for larger particles.

Further Huang writes "While in Balakin et al.'s paper, the size of particle is much larger, that r covers the range from 0 to 400 μm." This is not so. In the main part of the work we only deal with sizes below 200 μm. Only our sensitivity studies involved particles of sizes above 250 μm, and this was done just to verify how the model behaves over a very wide range and was not related to the actual modeling of the physical process.

Huang refers to the articles by Midi[6] and and Zhu et al.[7] from which he concludes that particles with sizes above 100–250 μm cannot form agglomerates due to van der Waals attraction, because the inertial effects dominate. We agree that there is a certain agglomerate size above which this statement is true. However, we have carefully studied both of these references, and in the first one,[6] we have found that this statement is true only for particles above 250 μm (not 100–250 μm, as Huang states). In the second reference,[7] there is no quantification of the limiting agglomerate size. As mentioned above, the particle size in our model does not exceed 200 μm, thus, according to Ref. 6, our particle size is within the size range where the van der Waals interaction is important.

We can only agree with the final statement in this comment that the statistics for the experimental data we use[3] is rather low. There were no better data for the recirculating systems pump-pipeline with low-pressure hydrate particles available.

Third comment

Huang states: "Thus the coefficient k_4 are non-dimensionalized, that $g = k_4\left(\frac{G}{k_b}\right)\bar{r}$ in which k_b 1/s is used to match the dimension of the shear rate," referring to the work of Barthelmes et al.[8] for this. k_4 will not be nondimensionalized by this procedure, it will still have the SI units of m^{-3} s^{-1}, but dividing the shear rate by an arbitrary scale in this way will get rid of the variable b in the dimension of k_4. Moreover, this can be done without further changing the values in our article, as also suggested by Huang. Although others[5,9,10] have done the same as we have done, we agree that Huang's suggestion is a good one.

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