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This is the published version of a paper presented at *Paper 61, ISAP Symposium and 53rd Asphalt Peterson Conference, Jackson Hole, WY. USA. July 18-21, 2016..*

Citation for the original published paper:

Khan, A. (2016)

Modelling coalescence process during breaking of bitumen emulsions

In: International Society for Asphalt Pavements (ISAP) (ed.), , Paper 61 (pp. 1-12).

N.B. When citing this work, cite the original published paper.

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Modelling coalescence process during breaking of bitumen emulsions

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ABSTRACT:

Cold mix bitumen emulsion technology is getting a lot of focus by the road industries since a few decades due to the diminished environmental impacts and reduced energy associated with it. The durability and mechanical performance of cold asphalt mixtures very much depend on the breaking, coalescence and phase separation processes in bitumen emulsions; however, the exact nature of the breaking mechanism of bitumen emulsion is not completely understood today. During coalescence or relaxation process, two bitumen droplets are completely fused into a unique spherical droplet and their kinetic is usually recorded in terms of time, denoted as relaxation time or $\tau_{\text{relaxation}}$. In this work, a two dimensional Phase Field model was used to simulate the coalescence process of two bitumen droplets in water phase. The numerical model is based on Finite Element Method and solves Navier-Stokes system of equations coupled with the Cahn-Hilliard equation. The model predictions are validated by direct comparison with the experimental measurements performed in our previous work. Moreover, the study was extended to the small size (order μm) bitumen droplets which are difficult to produce and handle via experimental methods.

Keywords: Cold Mix Asphalts; Bitumen Emulsions; Coalescence; Breaking Mechanism; Relaxation; Phase Field; Finite Element Method.

1. Introduction

The durability and mechanical performance of cold asphalt mixtures very much depend on the breaking and phase separation processes in bitumen emulsions. Bitumen emulsions start breaking or rupturing when coming into contact with aggregates. Thereafter, a phase separation by flocculation and coalescence should quickly occur that leads to partial loss of water in emulsion, Figure 1. A continuous film of bituminous binder is developed during coagulation that covers the aggregates either completely or partially, followed by a curing or setting of the binder. This process eventually determines the mechanical strength of the cold asphalt mixture [1-2]. Generally, due to the hydrophilic nature of the stone surface, the affinity for water at the aggregate surface is higher as compared to bitumen. The stone surface can be modified with emulsifiers and other additives which displace water and improve adhesion between the binder and the aggregates.

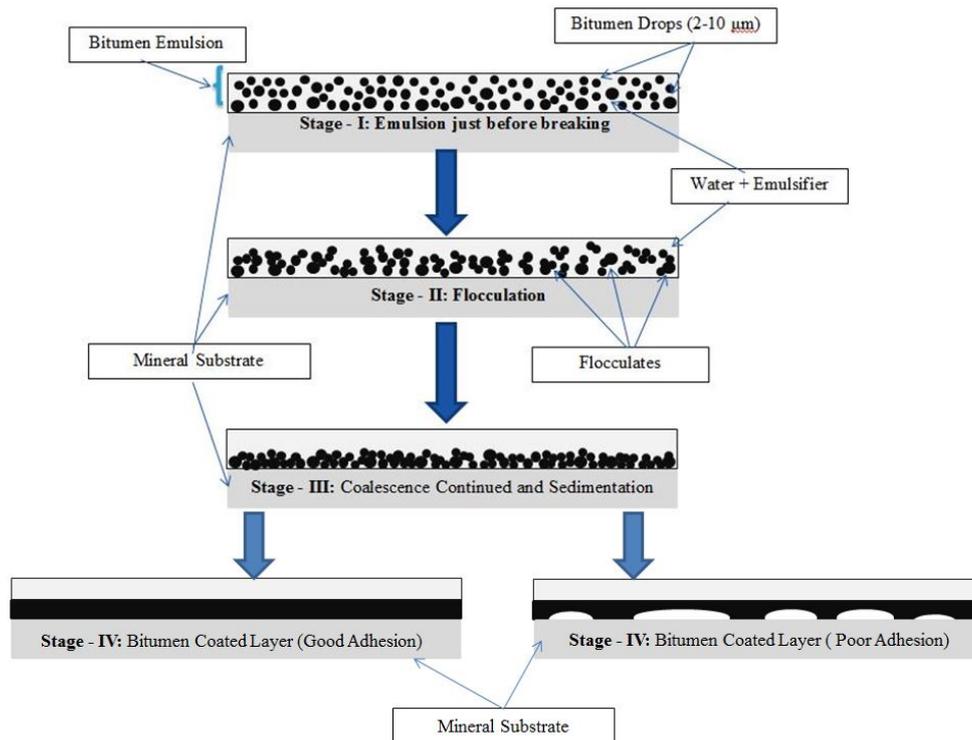


Figure 1. Breaking and Coalescence process in bitumen emulsion and bitumen interactions with aggregates.

A complete coating of aggregates by bitumen can be achieved if the breaking process in bitumen emulsion is controlled and any pre-mature coalescence is avoided which may result in a poor coating. This pre-mature coalescence can be due to high reactivity of stone surface, too quick breaking of the emulsion or too much fines in the stone material. Moreover, any physio-chemical change at the bitumen-water interface may also lead to destabilize the bitumen emulsions. For example, due to variation in pH, or ion exchange due to variation in salt concentrations or viscosity changes due to temperature. The current study suggests a modelling approach which could become useful for understanding the breaking and coalescence process.

Today, the breaking and coalescence mechanisms of bitumen emulsions are still not fully understood due to their complexities and lack of fundamental experimental methods and existing models. However, in the past years, efforts have been made in defining relationships for understanding the bitumen emulsions [3]. The kinetics of relaxation process were investigated by the authors via laboratory experiments for different grades of bitumen, temperature variations, bitumen droplet sizes and water phase additives e.g. emulsifiers and adhesion promoters. This parametric study was based on measuring relaxation times for two bitumen droplets in a water phase [4-5].

The first objective of the study described in the current paper is to develop a numerical model which is able to reproduce the coalescence experiments between two bitumen droplets in a water phase solution. The second objective of this research is to estimate the coalescence behavior of small size ($\sim \mu\text{m}$) bitumen droplets, which are not possible to study experimentally due to experimental limitations.

In this paper, the used methodology is described in the next section followed by a section containing the preliminary results along with a discussion. In the final section of this paper, the research work is summarized and conclusions are given.

2. Governing Equations and Numerical Methodology

Bitumen droplets coalesce in a water phase via viscous and surface tension driven flows, which are well described by continuum conservation laws for momentum and mass [6-7]. Inertial and gravitational forces are insignificant with respect to viscous forces due to high fluid viscosities and very small length scale. Therefore, these flows can be completely described by the Navier-Stokes equations for an incompressible fluid. Hence, in this work, the coupled Cahn-Hilliard/Navier-Stokes system of Phase Field [8-9] was used to model the coalescence process of two bitumen droplets in a water phase. The Comsol Multiphysics v5.1 tool was used to develop the Finite Element Method (FEM) base model. Later on the model was calibrated to reproduce the experimental results of bitumen droplet coalescence in terms of relaxation time [4]. Moreover, the study was extended to the small size (order μm) bitumen droplets which are difficult to produce and handle via experimental methods. A small description about the experimental measurements has been given in next paragraph that would be helpful for a better understanding.

The experimental setup for studying the coalescence process consists of a camera, light source, climate chamber with optically transparent walls, a transparent glass container with two L-shaped probes, temperature and humidity sensors and a Peltier heating panel connected to a temperature bath. Bitumen drops of 1-4 mm diameter are attached to the L-shaped probes and brought into contact with each other to initiate coalescence, after filling the glass container with relevant solvents (aqueous phase with and without selected additives). The process of droplets coalescing when brought together is recorded by a video camera. Then coalescence times are determined from the images and used to assess effects of variables such as binder grades, droplet sizes, test temperatures, types of emulsifiers, additives (salts at various concentrations, adhesion promoters etc.), pH and solvents on the process [4].

2.1. Theoretical Background

Phase field interfaces use the incompressible formulation of the Navier-Stokes equations:

$$\underbrace{\rho \frac{\partial \mathbf{u}}{\partial t}}_{(1)} + \underbrace{\rho(\mathbf{u} \cdot \nabla) \mathbf{u}}_{(2)} = \underbrace{\nabla \cdot [-p\mathbf{I}]}_{(3)} + \underbrace{\mu(\nabla \mathbf{u} + \nabla \mathbf{u}^T)}_{(3)} + \underbrace{F_{st} + F_{ext} + F}_{(4)} \quad \text{Equation 2.1}$$

$$\text{and } \nabla \cdot \mathbf{u} = 0 \quad \text{Equation 2.2}$$

Equation 2.2 represents continuity equation for incompressible fluids ($\frac{\partial \rho}{\partial t} = 0$), where, ρ is fluid density, \mathbf{u} is fluid velocity, ∇ is the gradient operator, p is fluid pressure and μ is fluid dynamic viscosity. The different terms correspond to the inertial forces (1), pressure forces (2), viscous forces (3), and the external forces applied to the fluid (4). The three forces on the right hand side of equation 2.1 are due to surface tension, a force due to an external contribution of the free energy and a user defined volume force. F_{st} is the surface tension force used for the phase field method which is implemented as a body force.

$$F_{st} = \left[G - \frac{\partial f}{\partial \Phi} \right] \nabla \Phi \quad \text{Equation 2.3}$$

Where G is the chemical potential (SI unit: J/m³) defined in the Equations 2.1 and 2.3 for the phase field method and $\frac{\partial f}{\partial \Phi}$ is a user defined source of free energy.

F_{ext} is external force or user defined volume force and it arises due to user defined source of free energy which is computed as Equation 2.4 given below;

$$F_{ext} = \left[\frac{\partial f}{\partial \Phi} \right] \nabla \Phi \quad \text{Equation 2.4}$$

Moreover, in the phase field method to track the interface, the convective Cahn-Hilliard equation can be written as:

$$\frac{\partial \Phi}{\partial t} + \mathbf{u} \cdot \nabla \Phi = \nabla \cdot \frac{\gamma \lambda}{\varepsilon^2} \nabla \Psi \quad \text{Equation 2.5}$$

Whereas free energy, Ψ is given by

$$\Psi = - \nabla \cdot \varepsilon^2 \nabla \Phi + (\Phi^2 - 1) \Phi + \left(\frac{\varepsilon^2}{\lambda} \right) \frac{\partial f_{ext}}{\partial \Phi} \quad \text{Equation 2.6}$$

Where the quantity λ (SI unit: N) is the mixing energy density and ε (SI unit: m) is a capillary width that scales with the thickness of the interface. These two parameters are related to the interfacial tension coefficient, σ (SI unit: N/m) as shown in the equation below;

$$\sigma = \frac{2\sqrt{2} \lambda}{\varepsilon} \quad \text{Equation 2.7}$$

and γ (eq.2.5) is related to ε through $\gamma = \chi \varepsilon^3$, where χ is the mobility tuning parameter. The volume fraction (V_f) of Fluid 2 is computed as

$$V_f = \min(\max([\Phi + 1]/2, 0), 1) \quad \text{Equation 2.8}$$

Where the min and max operators are used in a way that volume fraction has lower and upper limit of 0 and 1 respectively.

The density can also be defined as

$$\rho = \rho_1 + (\rho_2 - \rho_1) V_f \quad \text{Equation 2.9}$$

and the dynamic viscosity is

$$\mu = \mu_1 + (\mu_2 - \mu_1) V_f \quad \text{Equation 2.10}$$

where ρ_1 and ρ_2 are the densities and μ_1 and μ_2 are the dynamic viscosities of Fluid 1 and Fluid 2 respectively.

The mean curvature (SI unit: 1/m) of the interface can be computed by entering the following equation:

$$\kappa = 2(1 + \Phi)(1 - \Phi) \frac{G}{\sigma} \quad \text{Equation 2.11}$$

Where G is the chemical potential which is defined as:

$$G = \lambda \left(- \nabla^2 \Phi + \frac{\Phi(\Phi-1)}{\varepsilon^2} \right) + \frac{\partial f}{\partial \Phi} \quad \text{Equation 2.12}$$

2.2. Model Parameters

The Navier-Stokes equations are always solved together with the continuity equation while modeling fluid interface. The Navier-Stokes equations represent the conservation of momentum, while the continuity equation represents the conservation of mass. The initial surface shape completes the mathematical specification of the problem and mathematical form of governing equations provides an important insight to viscous driven relaxation process. These systems of equations are solved numerically by FEM.

The considered geometry consists of a solid rectangle and two circles of equal radius representing two droplets inside the rectangle box as shown below in Figure 2. For the following simulations, triangular elements are used for the Finite Element mesh.

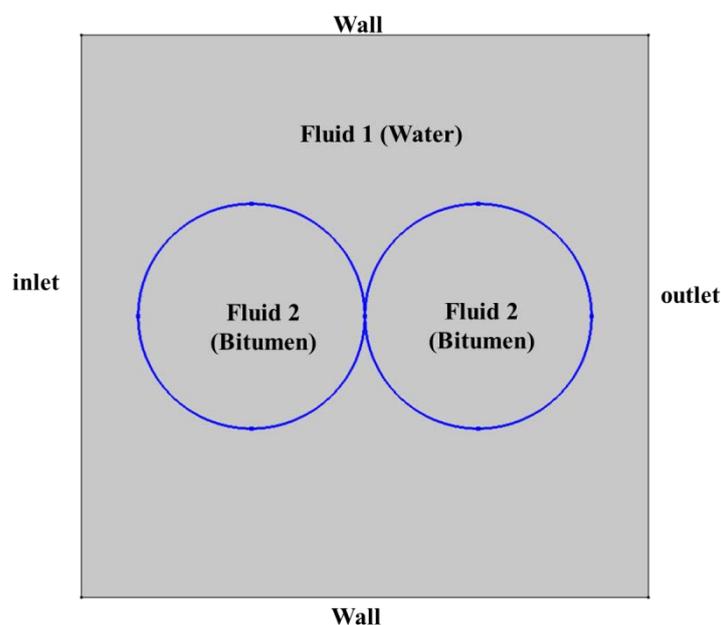


Figure 2. Model geometry and domain boundaries for bitumen droplet coalescence.

2.3. Materials Properties

In this study, an emulsion grade of unaged and unmodified straight run Nynas bitumen with penetration grade 70/100 was used. The basic physical properties and chemical constituents according to Iatroscan MK 6S chromatography analysis and the SARA (Saturates, Aromatics, Resins and Asphaltenes) fractions for Nynas bitumen 70/100 are shown in Tables 1 and 2 respectively.

Table 1. Properties of Nynas Bitumen 70/100.

Bitumen Grade	Penetration (mm/10)	Softening point (°C)	Dyn. Viscosity [Pa.s] @ 60°C	Kin. Viscosity [mm ² /s] @ 135°C	Fraass breaking point [°C]
70/100	87	46	90	239	-10

Table 2. Generic Fractions of Iatroscan Analysis (SARA Fractions).

Bitumen Grade	Saturates [% wt.]	Aromatics [% wt.]	Resins [% wt.]	Asphaltenes [% wt.]
70/100	6.4	48.6	26.2	18.8

Surface free energies of water [1] and Nynas bitumen 70/100 are presented in Table 3 below. The surface free energy of binder has been calculated based on sessile drop contact angle method at KTH-Royal Institute of Technology Sweden [10]. Furthermore, complex viscosity (Pa.s) data of this bitumen grade is plotted in Figure 3 and it was measured using DSR (Dynamic Shear Rheometer) at Nynas Bitumen AB.

Table 3. Surface free energy data of the water and bitumen phases.

Binder	γ^{LW} (mN/m)	γ^+ (mN/m)	γ^- (mN/m)	γ^{Total} (mN/m)
Water	21.8	25.5	25.5	72.8
Bitumen 70/100	42.4	0.15	0.15	42.7

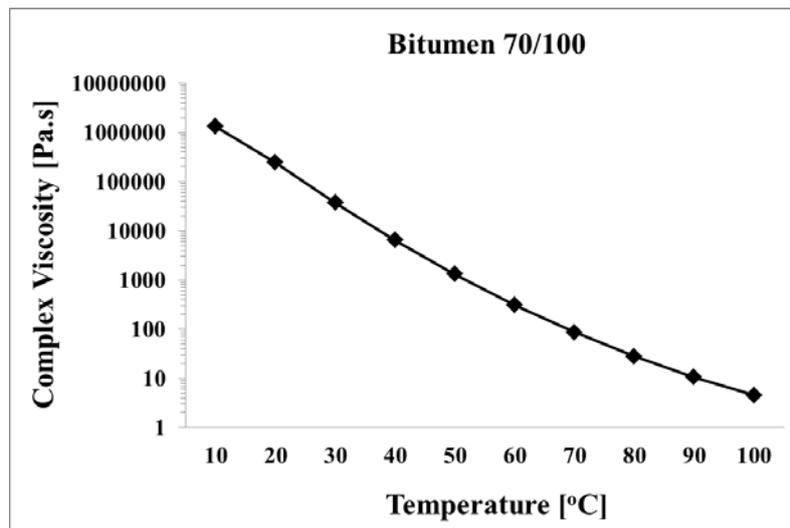


Figure 3. DSR (Dynamic Shear Rheometer) complex viscosity (Pa.s) of bitumen 70/100.

Viscosity of bitumen 70/100 is plotted on logarithmic scale as shown in Figure 3. The region of interest will be focusing on lower temperature range viscosities, as coalescence experiments were performed at 30 °C, 40 °C and 50 °C.

3. Results and Discussion

A two dimension (2D) Phase Field model with an incompressible laminar two phase flow has been developed to study hydro-dynamics of bitumen droplets coalescence. The relaxation time “ $\tau_{relaxation}$ ”, of bitumen droplets was evaluated numerically as a function of temperature of the water phase. Experimental results from previous studies [4-5,10] have been used to calibrate the numerical model. Experimental results of bigger size droplets were initially reproduced using Phase Field method. Later on phase field parameters were set for different

ranges of droplet sizes at different temperatures. At each temperatures, experimentally measured $\tau_{relaxation}$, has been compared with the numerically solved coalescence results. Moreover, with the calibrated model, extrapolated results for small droplet size have been extrapolated.

The time required for shape relaxation which causes two bitumen droplets to fuse into a unique one, called relaxation or coalescence time. According to literature studies about bitumen droplet coalescence [3,6,11], the characteristic time for shape relaxation is controlled by a competition between surface tension and viscous dissipation and can be represented as

$$\tau_{relaxation} \propto \frac{\eta R}{\gamma}, \quad \text{Equation 3.1}$$

where η is the droplet viscosity, R is the characteristic radius of the droplet and γ is the droplet surface tension.

In the early stages of coalescence process, droplets shrink little bit faster as compared to the final stages which can be observed both in experimental as well as numerical results as shown on Figures 4-6. Ratio between surface energy (Table 3) and viscosity (Figure 3) at different temperatures explains the kinetics of the coalescence process. Higher and lower values of this ratio mean soft and hard binder phase, respectively resulting in extremely fast and extremely slow coalescence process.

Results in Figures 4-6 belong to bitumen 70/100 droplets coalescence at three different temperatures (30, 40 and 50 °C respectively). For complete relaxation, two droplets must merge into a unique spherical droplet; however, in this case simulated results are plotted until the experimental relaxation cut. Experimentally, A complete fusion of two bitumen droplets has been observed for a H/L ratio equal to 0.8 (H represents the thickness or height of the contact bridge between two droplets and L is the horizontal length or width of two droplets which is equal to 4 times the radius of two equal size droplets at t_0 , i.e. before the start of relaxation). This ratio is thus used to determine the relaxation time obtained numerically. Consequently, the growth of contact-bridge between two droplets has shown almost similar shape profile in both experimental and numerical methods at different time steps, which revealed that numerical model parameters are well calibrated.

According to the results presented in Figures 4-6, the coalescence or relaxation times of bitumen 70/100 with an average droplet size of 1.5mm in a water phase at 30 °C, 40 °C and 50 °C are 4230 s, 800 s and 200 s, respectively. A rough estimate with H/L ratio equal to 0.8, fusion times for 30 °C, 40 °C and 50 °C are 4230 s, 800s and 200s, respectively. In other words, $\tau_{relaxation}$ of bitumen 70/100 has reduced from 30 °C to 50 °C, for the similar size droplets. Height, H(t) and Length, L(t) of merging droplet can be measured as a function of relaxation time at constant temperature.

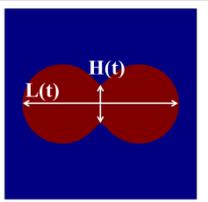
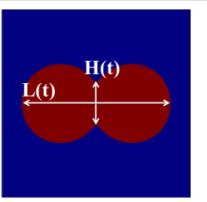
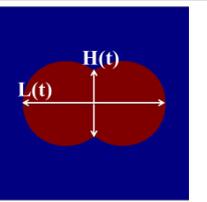
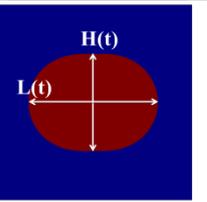
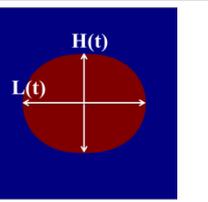
(a) Experimental					
(b) Numerical Method					
Relaxation Time, τ (s)	$\tau = 0$ s $H(t) = 0.705$ mm $L(t) = 2.854$ mm	$\tau = 158$ s $H(t) = 0.799$ mm $L(t) = 2.750$ mm	$\tau = 900$ s $H(t) = 1.209$ mm $L(t) = 2.548$ mm	$\tau = 3300$ s $H(t) = 1.728$ mm $L(t) = 2.283$ mm	$\tau = 4230$ s $H(t) = 1.800$ mm $L(t) = 2.231$ mm

Figure 4. Coalescence of bitumen 70/100 droplets (diameter~1.5 mm) in water phase at 30°C; via (a), Laboratory Experiment and (b), Numerical Method.

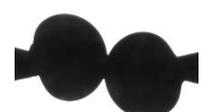
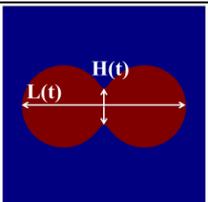
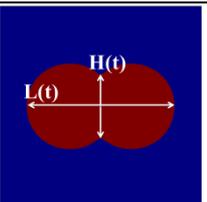
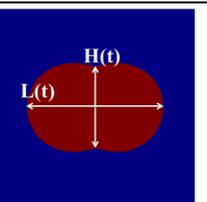
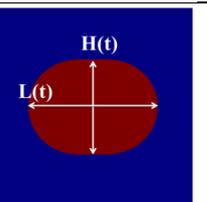
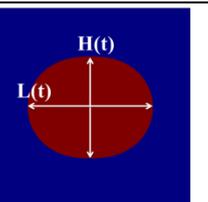
(a) Experimental					
(b) Numerical Method					
Relaxation Time, τ (s)	$\tau = 0$ s $H(t) = 0.648$ mm $L(t) = 2.906$ mm	$\tau = 168$ s $H(t) = 1.141$ mm $L(t) = 2.631$ mm	$\tau = 365$ s $H(t) = 1.494$ mm $L(t) = 2.460$ mm	$\tau = 592$ s $H(t) = 1.723$ mm $L(t) = 2.330$ mm	$\tau = 800$ s $H(t) = 1.816$ mm $L(t) = 2.257$ mm

Figure 5. Coalescence of bitumen 70/100 droplets (diameter~1.5 mm) in water phase at 40°C; via (a), Laboratory Experiment and (b), Numerical Method.

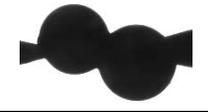
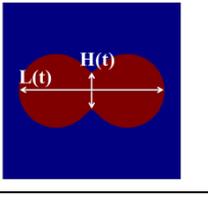
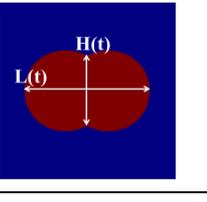
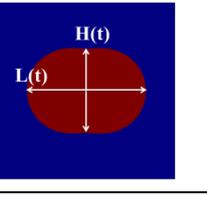
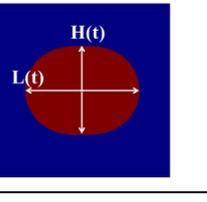
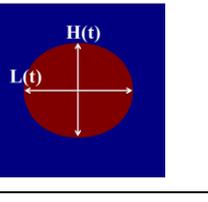
(a) Experimental					
(b) Numerical Method					
Relaxation Time, τ (s)	$\tau = 0$ s $H(t) = 0.721$ mm $L(t) = 2.755$ mm	$\tau = 60$ s $H(t) = 1.364$ mm $L(t) = 2.377$ mm	$\tau = 120$ s $H(t) = 1.598$ mm $L(t) = 2.247$ mm	$\tau = 180$ s $H(t) = 1.723$ mm $L(t) = 2.184$ mm	$\tau = 200$ s $H(t) = 1.614$ mm $L(t) = 1.993$ mm

Figure 6. Coalescence of bitumen 70/100 droplets (diameter~1.5 mm) in water phase at 50°C; via (a), Laboratory Experiment and (b), Numerical Method.

A Phase Field model was constructed in such a way that in the limit of an infinitesimal interface thickness (the so-called sharp interface limit) the correct interfacial dynamics are reproduced. The kinetic parameter or so-called mobility parameter' $\chi = \frac{\gamma}{\varepsilon^2}$ in the convective Cahn-Hilliard (Equation 2.5) is related to the atomic mobilities of the constituting elements which ensure the conservation of mass and as well as diffusion flux balance at the interface.

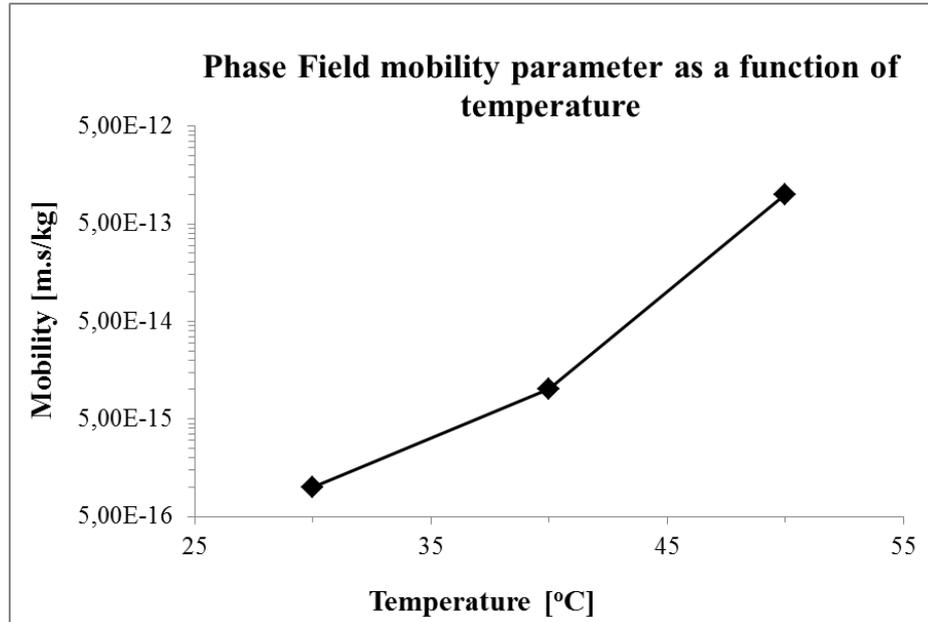


Figure 7. Phase Field mobility parameter as a function of temperature.

For bitumen 70/100, the mobility parameter was modified at each temperature to achieve the similar coalescence results as found experimentally. For example, at 30 °C, 1.5 mm diameter droplets with interface thickness ($\varepsilon = 0.5\text{mm}$), the fitting mobility was found to be 1E^{-15} m.s/kg. The calibrated mobility parameters as a function of temperature are plotted in Figure 7. As seems plausible, the fitted mobility parameters increased with increasing temperature at constant interface thickness. The plotted data values in Figures 7-9, are achieved through numerical simulations using Phase Field method and all calculations are based on experimentally measured viscosity, interfacial energy and relaxation times of same bitumen droplets. However, for very small sizes, data was extrapolated.

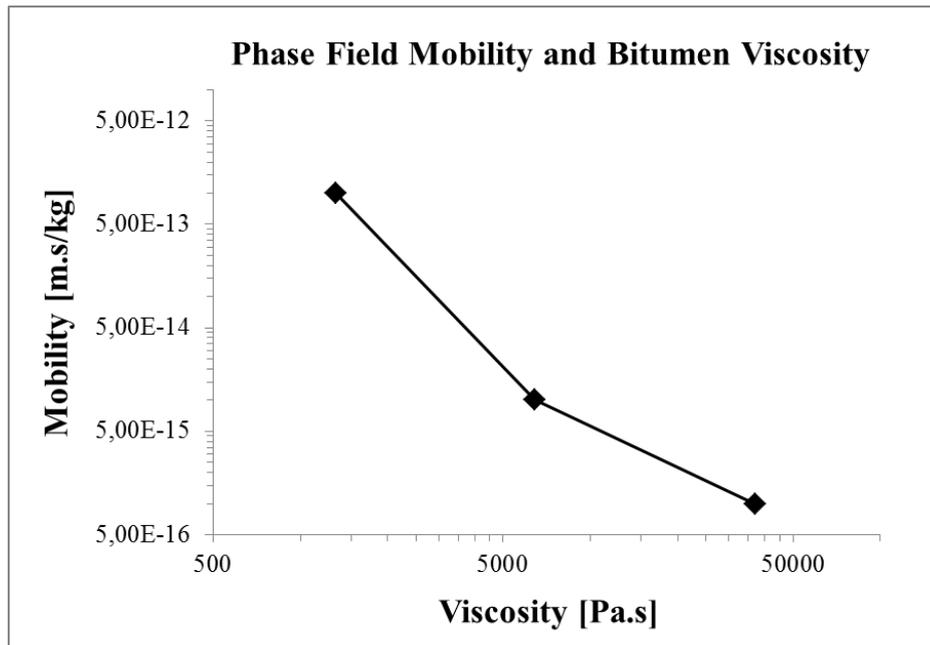


Figure 8. Phase Field mobility parameter variation with bitumen viscosity

Correspondingly, the mobility parameter can also be linked to the bitumen viscosity by keeping the interface thickness constant, as presented in Figure 8, which shows that at lower viscosity, mobility is higher as compared to higher viscosities. Furthermore, different interface thickness parameters were used for the different ranges of droplet size. The reason for this is that in the FEM model, the interface thickness is controlled by the mesh size. In case of smaller droplet size, a finer mesh size is used along with smaller thickness of the interface. In this work, mobility parameters were determined according to experiments by fixing the interface thickness with respect to the mesh size.

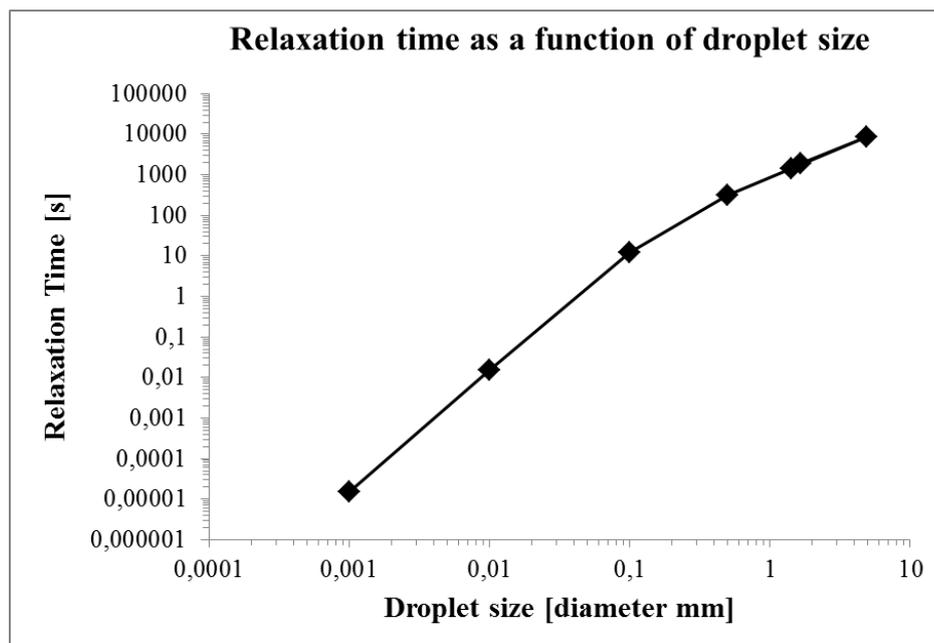


Figure 9. Relaxation time as a function of bitumen droplet size.

The model was validated at 30 °C to calculate the relaxation time for bitumen 70/100 droplets of different sizes ranges from μm to mm as plotted in Figure 9. Relaxation times as a function of droplet sizes verify Equation 3.1 as well as very much close to what we have found in our experimental work; however, these values are not representing the real emulsion systems which contain emulsifiers, pH levels and other additives in the water phase. Additionally, the small droplet sizes that have been extrapolated without experimental validation, as the main reason behind developing the model was to complement the (larger) scale size which is currently possible within the experimental set-up. Model is able to reproduce coalescence experiments of large scale bitumen droplets only in pure water without any surfactants and additives present. The extrapolation of small scale droplets for coalescence study using this model is still very limited and far away from the real emulsion systems. For instance bitumen from other crude sources, effects of additives and other modifications such as polymer could be included and validated with the experimental study in future to accurately mimic the emulsion systems.

4. Summary and Conclusions

A two dimensional computational strategy for the simulation of laminar two phase flow is presented to study bitumen droplet coalescence in water phase. In this work, the coupled Cahn-Hilliard/Navier-Stokes system of Phase Field was used to model the coalescence process. From this, the following conclusions can be drawn:

- The growth of contact-bridge between two droplets has shown almost similar shape profile in both experimental and numerical methods at different time steps, which revealed that the numerical model parameters are well calibrated.
- The model showed that mobility parameters increase with increasing temperature at constant interface thickness, which seems sensible.
- Additionally, the mobility proved to be inversely proportional to the bitumen viscosity at constant interface thickness, which also seems logical.
- The model can relate the relaxation times directly proportional to the droplet sizes.

Results obtained with the presented numerical method for systematic investigation of the coalescence of pairs of bitumen droplets exposed to water phase with different temperatures were consistent with both theoretical expectations and previous findings. To overcome the laboratory limitations and assist in the determination of appropriate scaling functions, a computational method based on Navier Stokes simulations, incorporating surface free energy parameters could be a possible approach. Further research could thus focus on the development of a three-dimensional multiphase model to study coalescence processes in more detail, incorporating bitumen from different crude origins as well as other emulsion like environments including effects of surfactants, pH and other additives such as mineral fillers and salts.

Acknowledgement

This research work was performed at KTH-Royal Institute of Technology Stockholm, Sweden and sponsored by Nynas Bitumen AB.

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