LIQUID-LIQUID PHASE SEPARATION IN GRAVITY SETTLERS: MEASUREMENTS, POPULATION BALANCE MODELING AND CFD SIMULATIONS

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Certificate

This is to certify that the thesis entitled **"Liquid-Liquid Phase Separation in Gravity Settlers: Measurements, Population Balance Modeling and CFD Simulations"** being submitted by **Mr. Abhijeet H. Thaker** to the **Indian Institute of Technology Delhi, New Delhi** for the award of the degree of **Doctor of Philosophy** is a bonafide record of original research work carried out by him under my supervision in conformity with rules and regulations of the Institute. The results contained in this thesis have not been submitted, in part or in full, to any other University or Institute for the award of any Degree or Diploma.

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Abstract

Separation of liquid-liquid dispersion is an integral part of a variety of processes; for example, crude oil processing (crude desalting), hydrometallurgy (solvent extraction), etc. Mixer–settlers are widely used in the solvent extraction because of their advantages over the other equipment. In a mixer–settler, two immiscible liquid phases enter the mixer, and the organic phase is dispersed into the aqueous phase in the mixer (used for extraction) and then the dispersion flows into a large-sized gravity settler. The dispersion undergoes binary and interfacial coalescence that occur within the dispersion band inside the settler and results in phase separation. While there exist many reports available on mixer operations, a limited information reported on the separation phenomena in gravity settlers in the literature. The separation performance inside the settler depends on operating parameters (e.g., dispersion flow rate, dispersed phase volume fraction and drop size), geometrical parameters (e.g., settling area and dispersion inlet position) and rates of interfacial and binary coalescence. Therefore, an understanding the role of aforementioned parameters in phase disengagement is very important to improve the separation performance of settlers, which in turn can help in the optimization and design of settlers. Further, for the design and scale-up of gravity settlers, it is important to develop the CFD models that account for the binary and interfacial coalescence processes.

In the present work, experimental investigations of liquid-liquid disengagement are performed in a laboratory-scale continuous gravity settler. The effects of different operating and geometrical parameters on the phase disengagement are investigated. Based on the measurements and the data reported in the literature, an empirical correlation is proposed to predict the dispersion–band height. Further, to develop an understanding of the drop coalescence process, the micro-scale experiments are performed to measure the frequency of interfacial and binary coalescence in the presence of single isolated drops and multiple drops.

These measurements provide crucial database required to develop kernels for the interfacial and binary coalescence rates. Such kernels are important to predict the drop size distribution using the population balance (PBM) or integrated CFD+PBM models.

Further, the Eulerian multi-phase model is developed in the open-source CFD code OpenFOAM to simulate the gravity separation process. In addition, a stand-alone PBM is developed and integrated with the multi-phase CFD model to predict the drop size distribution and dispersed phase volume fraction within the dispersion band. Moreover, since the interfacial coalescence governs the separation process in the settler, the interfacial coalescence module is incorporated in the CFD model. Initially, the developed CFD+PBM model is used to simulate the batch settler to build confidence on the predictive capabilities in the relatively simple system (due to the absence of convective flow in comparison to the continuous gravity settler). To validate the developed model, the batch settling experiments are performed in a laboratory-scale batch settler. Later, the CFD+PBM solver is used to perform the simulations of liquid-liquid disengagement in the continuous-gravity settler to predict the dispersion band height, drop size and dispersed phase volume fraction distribution within the dispersion band and predictions are validated with measurements. The computational model developed in this work will be useful to predict the separation process in gravity settlers and to optimize their separation performance.

सार

तरल-तरल फैलाव का विभाजन विभिन्न प्रक्रियाओं का एक अभिन्न अंग है; उदाहरण के लिए, कच्चे तेल के प्रसंस्करण (क्रूड डीसाल्टिंग), हांइड्रोमेटलरजी (सॉल्वेंट एक्सट्रैक्शन), इत्यादि। मिक्सर-सेटलर्स का व्यापक रूप से सॉल्वेंट एक्सट्रैक्शन में उपयोग किया जाता है क्योंकि अन्य उपकरणों से उनके ज्यादा फायदे हैं। मिक्सर-सेटलर में, दो अमिश्रणीय तरल चरण मिक्सर में प्रवेश करते हैं, और कार्बनिक चरण मिक्सर में जलीय चरण में फैलाया जाता है (एक्सट्रैक्शन के लिए इस्तेमाल किया जाता है) और फिर फैलाव एक बड़े आकार के गुरुत्वाकर्षण सेटलर में बह जाता है। सेटलर में, फैलाव बाइनरी और इंटरफेसियल संघीकरण प्रक्रिया से गुजरता है जो प्रक्रिया फैलाव बैंड के भीतर होता है और चरण अलगाव में परिणाम होता है। जैसेकि, मिक्सर संचालन पर कई रिपोर्टें मौजूद हैं, साहित्य में, गुरुत्वाकर्षण के आधारित अलगाव की घटनाओं पर एक सीमित जानकारी की सूचना दी गई है। सेटलर के अंदर विभाजन का कार्य परिचालन मापदंडों (जैसे, फैलाव प्रवाह दर, फैलाव चरण मात्रा और ड्रॉप आकार), ज्यामितीय मापदंडों (जैसे, क्षेत्र और फैलाव इनलेट की परिस्थिति) और बाइनरी और इंटरफेसियल संघीकरण की दर पर निर्भर करता है। इसलिए, चरण विभाजन में पूर्वोक्त मापदंडों की भूमिका को समझना, विभाजन का कार्य को बेहतर बनाने के लिए बहुत महत्वपूर्ण है, जो सेटलर की डिजाइन में मदद कर सकता है। इसके अलावा, गुरुत्वाकर्षण सेटलर्स के डिजाइन और स्केल-अप के लिए, सीएफडी मॉडल विकसित करना महत्वपूर्ण है जिसमे बाइनरी और इंटरफेसियल संघीकरण प्रक्रियाओं को लागु किया गया हे।

वर्तमान काम में, तरल-तरल विभाजन की प्रयोगात्मक जांच प्रयोगशाला-पैमाने पर निरंतर गुरुत्वाकर्षण सेटलर में की गयी है। चरण विभाजन पर विभिन्न ऑपरेटिंग और ज्यामितीय मापदंडों के प्रभावों की जांच की जाती है। माप और साहित्य में रिपोर्ट किए गए आंकड़ों के आधार पर, फैलाव-बैंड की ऊंचाई की भविष्यवाणी करने के लिए एक अनुभवजन्य सहसंबंध प्रस्तावित किया गया हे। इसके

अलावा, ड्रॉप संघीकरण प्रक्रिया की समझ विकसित करने के लिए, एकल-स्केल ड्रॉप्स और मल्टीपल ड्रॉप्स की उपस्थिति में बाइनरी और इंटरफेसियल संघीकरण की आवृत्ति को मापने के लिए माइक्रो-स्केल प्रयोग किए गए हैं। ये माप आवश्यक डेटाबेस प्रदान करते हैं जो कि बाइनरी और इंटरफेसियल संघीकरण दर के समीकरण विकसित करने के लिए आवश्यक हैं। इस तरह की समीकरण जनसंख्या के संतुलन (पीबीएम) या एकीकृत सीएफडी + पीबीएम मॉडल का उपयोग करके ड्रॉप आकार के वितरण की भविष्यवाणी करने के लिए महत्वपूर्ण हे।

इसके अलावा, औइलेरियन बहु-चरण मॉडल को खुले-स्रोत सीऍफ़डी कोड OpenFOAM में गुरुत्वाकर्षण विभाजन प्रक्रिया का अनुकरण करने के लिए विकसित किया गया है। इसके अलावा, एक स्टैंड-अलोन पीबीएम ड्रॉप आकार के वितरण और फैलाव बैंड के भीतर चरण मात्रा अंश का अनुमान लगाने के लिए बहु-चरण सीएफडी मॉडल के साथ विकसित और एकीकृत किया गया है। इसके अलावा, चूंकि इंटरफेसियल संघीकरण सेटलर में विभाजन प्रक्रिया को नियंत्रित करता है, इसलिए सीएफडी मॉडल में इंटरफेसियल संघीकरण मॉड्यूल को शामिल किया गया है। प्रारंभ में, विकसित सीएफडी + पीबीएम मॉडल का उपयोग बैच सेटलर को अपेक्षाकृत सरल प्रणाली (निरंतर गुरुत्व बसने की तुलना में संवहन प्रवाह की अनुपस्थिति के कारण) में अनुमानित क्षमताओं पर विश्वास बनाने के लिए किया गया है। विकसित मॉडल को मान्य करने के लिए, प्रयोगशाला बैच सेटलर के के प्रयोग किये गए हे। बाद में, सीऍफ़डी + पीबीएम सॉल्वर का उपयोग फैलाव बैंड की ऊंचाई, ड्रॉप आकार और फैलाव बैंड के भीतर बिखरे हुए चरण मात्रा अंश वितरण की भविष्यवाणी करने के लिए निरंतर-गुरुत्व सेटलर में तरल-तरल विभाजन के सिमुलेशन को करने के लिए किया गया है और मापों के साथ पूर्वानुमान लगाया गया है। इस काम में विकसित कम्प्यूटेशनल मॉडल गुरुत्वाकर्षण सेटलर्स में विभाजन प्रक्रिया की भविष्यवाणी करने और उनके विभाजन प्रदर्शन का अनुकूल करने के लिए उपयोगी होगा।

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Nomenclature

A_s	settling area, m^2
A_{cs}	settler cross-sectional area, m^2
A_d	surface area of drop, m^2
A_H	Hamaker constant[1], 2.15×10^{-21} kg m^2/s^2
B_b	birth due to breakage, -
B_c	birth due to coalescence, -
C_D	drag coefficient, -
d_{32}	Sauter mean diameter, m
d_d	drop diameter, m
d_h	hydraulic diameter of settler cross–section, m
D_b	loss due to breakage, -
D_c	loss due to coalescence, -
Eö	Eötvös number (as defined in Table 2.3), –
F_b	buoyancy force, kg m/s^2
F_σ	interfacial tension force, kg m/s^2
g	gravitational acceleration, m/s^2
H	height of interface from the bottom, m
H_{db}	height of dispersion band, m
H_b	height of dispersion inlet opening baffle, m
H_t	height of end plate, m
h	height of settler, m
h_o	height of dispersion inlet baffle opening, m
l_s	length of settler, m
m_d	mass of a drop, kg
M	momentum exchange between the phases,
N	number of drops, –
N_D	dispersion number (as defined in Table 2.3), –
n	drop number density, -
p_{in}	pressure inside a drop, N/m
p_{out}	pressure outside a drop, N/m
P	pressure, N/m^2
r_f	radius of draining film, m
r_{bc}	rate of binary coalescence, m^3/s
r_{ic}	rate of interfacial coalescence, m^3/s
Re	Reynolds number, -
Q_d	dispersed phase flow rate, L/h (m^3/s in correlation (Eq.2.11)
Q_t	total flow rate, L/h
Re	Reynolds number, –
r	drop radius, m
t	time, s
t_{ic}	time of interfacial coalescence, s
t_{RM}	phase residence time in mixer, s
t_{RS}	phase residence time in settler, s
t_{coal}	coalescence time, s
t_d	continuous–phase film drainage time, s
u_r	relative velocity, m/s
$\bar{U}$	average velocity, m/s

V	volume of settler, m^3
V_d	volume of a droplet, m^3
u_r	rise velocity, m/s (used in Eq. 2.7)
V_m	volume of mixer, m^3
v_{in}	dispersed phase velocity at inlet, m/s
v_r	terminal rise velocity, m/s
w_o	width of dispersion inlet opening, m
x^*	dimensionless settler length (l/l_s), –
y^*	dimensionless settler height (h/H_t), –

Greek Letters

α_O	dispersed phase volume fraction, –
α_A	continuous phase volume fraction, –
β	mass source of binary coalescence
Υ	mass source of interfacial coalescence
ρ_d	dispersed phase density, kg/m^3
ρ_c	continuous phase density, kg/m^3
$\Delta\rho$	density difference, kg/m^3
τ_0	time of drop coalescence, s
σ	interfacial tension, N/m
ε_c	dispersed phase volume fraction at coalescing interface, –
μ_c	continuous phase viscosity (kg/ms)
μ_d	dispersed-phase viscosity, kg/m s
Ψ	rate of interfacial coalescence per unit area, $m^3/s–m^2$
Γ	rate of binary coalescence, m^3/s
δ	separation distance between the two drops, m
δ_r	critical film thickness at rupture, m
σ	interfacial tension, N/m
σ^*	standard deviation
ω_I	frequency of interfacial coalescence, 1/s
ω_B	frequency of binary coalescence, 1/s
Φ_c	rate of drop collision, m^3/s
φ_e	coalescence efficiency, -
ε	rate of dissipation, m^2/s^3
$\Delta\rho$	density differences between the liquid phases, kg/m^3

Subscript/Superscript

in	inlet
bd	buoyancy–driven collision
sd	shear–driven collision

Acronyms

AI	Active Interface
CFD	Computational Fluid Dynamics
DSD	Drop Size Distribution
DW	Demineralized Water
DLVO	Derjaguin B., Landau L., Verwey E., Overbeek J.

EDL	Electrostatic Double Layer
PBM	Population Balance Modeling
PMMA	Poly Methyl Methacrylate
PI	Passive Interface
PISO	Pressure Implicit with Split Operator
PIMPLE	Pressure Implicit Method for Pressure Linked Equations
RPM	Revolution Per Minute
RDC	Rotating Disk Contactor
RMSD	Root Mean Square Deviation
SDS	Sodium Dodecyl Sulfate
SIMPLE	Semi Implicit Method for Pressure Linked Equations
TBP	Tri–Butyl Phosphate