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EPIC – ENABLING PROCESS INNOVATION THROUGH COMPUTATION: A HIERARCHICAL MODELLING FRAMEWORK FOR INNOVATION

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ABSTRACT

Multiphase flows are ubiquitous in chemical and materials processing industries. In process design and performance analysis of such unit operations, the traditional approach has been to ignore fluid dynamical effects by invoking simplifying assumptions of homogeneity, but pay the price during scale-up of processes through expensive pilot scale experiments. The question addressed in this presentation is "Can Multiphase flow modelling Enable Process Innovation through Computation?", thus minimizing the need for expensive pilot scale development. An overview of the research in using CFD to develop experimentally validated multiphase flow process models, which span a range of scales from direct numerical simulation (DNS) to averaged two-fluid models (TFM) models that are in need of closure relations are presented with examples.

On the other hand, there is an immediate need to study flow of complex fluids of industrial importance, even with a larger uncertainty in models, as the alternative is pure guess work or expensive pilot testing. Such cases include the recent oil spill modelling, polymer blending processes involving melting, deformation and break-up, corrosion-erosion in pipelines and process vessels, mass transfer in packed beds with random and structured packings or in Sieve trays. In such studies the TFM framework forms the basis of flow models coupled with experimental validation of such predictions in an effort to develop *scale invariant closure models*.

INTRODUCTION

The goal of EPIC at LSU is to transform the next generation *engineered, manufacturing process systems* that can be deployed in energy production, environmental remediation, chemical and materials *manufacturing* operations to be energy efficient and environmentally friendly, while provinding advanced training to engineers to take that culture of sustained innovation to the workplace. EPIC is advancing the fundamental science of multiphase flows that governs many of these *engineered process system, through development of advanced, validated computational models for multiphase flows*. EPIC will *shift the design paradigm* of such processes from a largely empirical approach (as currently practiced) to one that makes full use of advances in computational, experimental and manufacturing technologies. In particular, the consortium will focus on investigation of multiphase transport in process systems in the presence of large scale heterogeneities and to manage such heterogeneities right from the conceptual design stage of the equipment itself. EPIC will be developing *experimentally validated high fidelity computational models* that enable exploration of a large design space in a cost effective manner. EPIC, focusing on industrially relevant test-beds for proof of concept, is converging technologies in High Performance Computing (HPC), high fidelity measurements (tomography, PIV), and high precision, complex manufacturing. Such convergence enables study of large complex design spaces on all scales, making possible new process designs. Davidson argues in a National Academies' report, as follows: "A case study of the economic benefit of the application of Computaional Fluid Dynamics (CFD) in one chemical and engineered-material company over a six-year period conservatively estimated that the application of CFD generated approximately a *six-fold return on the total investment*." Proctor & Gamble R & D director states, "We used to use HPC *modeling and simulation for autopsies*—to explain why things didn't work after they failed, but now we have the computing power *to get things done correctly up front* rather than wait for a catastrophic failure and then try and figure out what went wrong". These views encapsulate the general optimism that high fidelity computational models can help revolutionize the design of the next generation of green chemical processes. When constraints on emission and fuel standards were placed on the automotive industry newer designs emerged. Similarly as carbon tax and similar environmental regulations are put in place, the holistic cost functions will emerge making newer process designs imperative. Through several examples of ongoing work, this concept will be demonstrated.

An Opportunity for Innovations in Chemicals Manufacturing

Rapid globalization of businesses, volatility in energy prices, and emergence of low cost labor markets have changed the landscape of traditional chemical manufacturing and petroleum refining industries dramatically. In a recent address by Stephen Pryor, President, ExxonMobil Chemical Company on "Innovation and the Evolution of Chemical Feedstocks", he singled out the *boom in shale gas and liquids* in the north America as being the most dominant factor and a game changer that is revolutionizing the global chemical and petrochemical industries by evolving their advantaged feedstocks. American manufacturers use natural gas to fuel and power a wide variety of processes to produce a broad portfolio of manufactured goods – from a variety of performace monomers, specialty chemicals, pharmaceuticals, advanced materials agrochemicals and cosmetic consumer products. For some energy-intensive products, energy for both fuel and power needs and feedstocks account can represent 85% of total production costs.

The increased availability of cheap shale gas has increased the output in a range of eight manufacturing industries – paper, chemicals (excluding pharmaceuticals), plastic & rubber products, glass, iron & steel, aluminum, foundries, fabricated metal products. To guide the technology development that supports this tremendous boost to chemical manufacturing sector by the shale gas production revolution must be the top most priority for the US in the next few years.

A Case for Louisiana Industrial Commons

Louisiana has traditionally played an important role in the chemical and petrochemical industries as well as the upstream petroleum exploration and production. A report titled "The Economic Impact of the Chemical Industry on the Louisiana Economy", notes that chemicals are Louisiana's third largest export, shipping \$8.3 B (14.4% of total production in the state) in chemical products to other parts of the world in 2011. Petroleum and agricultural products are #1 and #2. Louisiana ranked #3 among the 50 states in terms of the total value of chemical shipments from LA (~\$58.2 B in 2010). With 19 operating refineries, Louisiana was second only to Texas in 2013 in both total and operating refinery capacity. The Louisiana Offshore Oil Port is the only port in the United States capable of offloading deep draft tankers. The U.S. Strategic Petroleum Reserve's two Louisiana facilities consist of 29 salt caverns capable of holding over 300 million barrels of crude oil. In 2011, Louisiana ranked second among the states in total energy consumption per capita, primarily because of the industrial sector (about 2/3th of consumption), which includes many refineries and petrochemical plants (www.eia.gov).

Despite the afore-mentioned positive economic impacts, US consumption patterns can be traced to the landscape of Mississippi river with its abundant water supply to sustain these industries, but also create significant environmental problems. Over one hundred refineries and chemical manufacturing facilities are intermixed with sugar refineries, metal processors, and coffee production facilities, revealing the demands of the nation. American consumers benefit from the myriad of products made possible by petro-chemistry, while pollution and waste often affect only the poorest communities. (Fig.1). Next-generation chemical plants and refineries must become cognizant of the long-term impacts on the environment and should address the issue efficiency, reduced footprints, reduced emissions in a global setting. In a report titled "The Future of the European Chemical Industry", the key to the "survival" of the chemical industry against the business globalization and increased competition from the developing nations was through "sustained innovation". Industrial commons are geographically rooted "collective R&D, engineering and manufacturing capabilities that sustain innovations. When located in a network, intellectual, financial and human capital flow between institutions at every phase of technology development, boosting the innovative capacity of all institutions involved.

Figure 1: Mississippi river chemical corridor shown by the locations of various chemical products at geographically distributed plants along the river. (Petrochemical America; Aperture 2012 © SCAPE)

The advantages of such industrial commons have been recognized in IT and Bioscience/Engineering industries. EPIC will demonstrate that such an industrial commons can also work with process industries with Baton Rouge as its focus.

To understand the role of universities in this landscape, an NRC study entitled, "International Benchmarking of U.S. Chemical Engineering Research Competitiveness" points out "*... Future U.S. leadership in chemical engineering is not guaranteed. Many factors could significantly affect the position of the U.S., and these include shifting funding priorities by federal agencies, reductions in industrial support of academic research in the United States, and decreases in talented foreign graduate students, among others*." This report points to an urgent need for rejuvenating research and training elements focused on *process innovation* to maintain US competitiveness in this sector. Future manufacturing processes for new products will likely involve *hybrid feedstocks* (a combination of fossil and bio-based feedstocks) to move toward sustainability and reduced carbon footprint. The goal of EPIC consortium is to *leverage a thematic University-Industry Collaboration* by using *advanced design tools* to addresses the implications of evolving feedstocks on process designs as well as related environmental concerns to achieve sustainability in this area.

Technology Roadmap

The accompanying technology road map (Fig 2a) envisions two metrics, one on the use of renewable energy sources and another on sustainable technology development. This divides the hydrocarbon-based economy into four quadrants and the current economy is on the 4th lower-left quadrant where reliance is on fossil fuel and the environmental sustainability takes a back seat. By incorporating hybrid feedstocks (i.e. increased dependence on renewable feedstocks), we move to the right (quadrant 3) and by adopting carbon sequestration (i.e. reduced emissions and improved environmental sustainability) we move to quadrant 2. A combination of these is needed to move to the ideal quadrant one. EPIC consortium will assist in the above-mentioned transformation by introducing process innovations that adopt hybrid feedstocks and account for reduced environmental impacts during the design process itself. The chemical value chain gets further enriched through the incorporation of new technologies that are tested at the pilot scales. Such technologies are the outcomes of a sustained innovation cycle that involves all potential stakeholders including both the technology developers and the end users during the research and testing stages. The proposed effort addresses the entire life cycle of various hydrocarbons-based feedstocks starting from subsurface production (technologies like hydraulic fracturing, in-situ conversion, carbon sequestration) to upgrading the chemical feedstock (methane/ethane to ethylene, gas-to-liquids). Validated simulation models can aid in both knowledge and technology diffusion, and pathways to adoption for the novel, more efficient, reduced-footprint, and greener technologies. Figure 2(b) shows the framework for process innovation that is built on multiphase, multiscale, multiphysics models combined with Advanced simulation, Advanced manufacturing and Advanced measurements as enabling technologies.

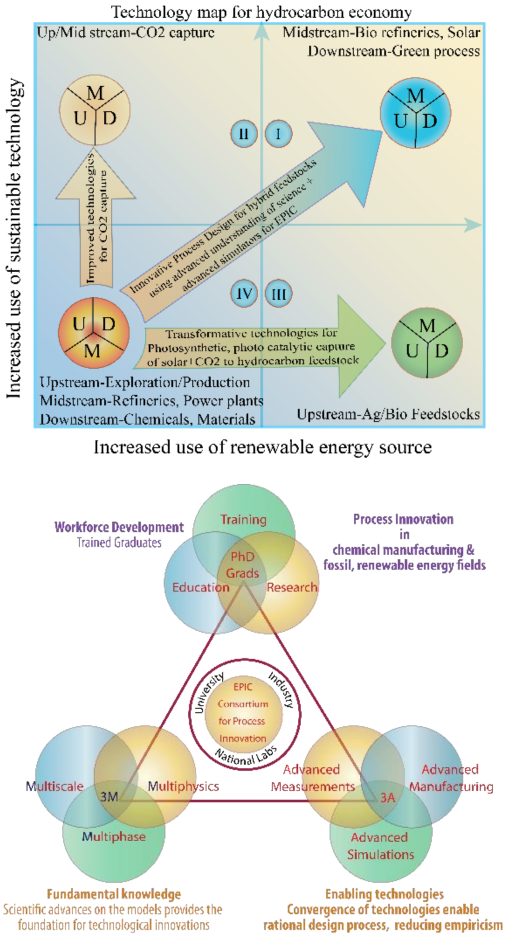

Figure 2: Top: Technology roadmap for innovation in chemical processing industries. Bottom: Schematic of a collaborative consortium involving universities, industries, and national laboratories in the area of process innovation in the chemical manufacturing and energy industries.

CONCEPTUAL MODELLING FRAMEWORK

Multiphase processes can be modelled on various scales from the molecular scale to equipment scale. Figure 3 shows the interrelations of models on various scales. The performance models on the equipment scale are the most widely used in plant operations and found in widely used plant scale simulators such as ASPEN/HYSYS.

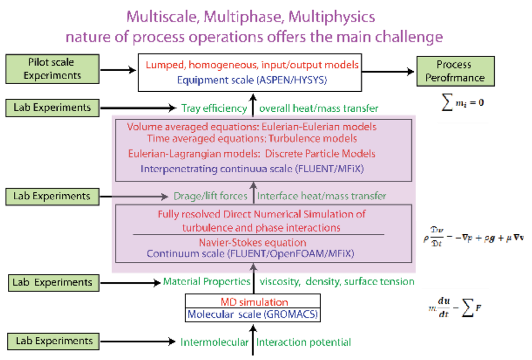

Figure 3: Multiscale nature of multiphase flows revealing the hierarchical nature of the model inter-relationships.

They often need drag coefficients, overall heat and mass transfer coefficients, effective reaction rate constants, tray efficiencies etc. as inputs, which are obtained either from more detailed models or from pilot scale experiments. The next level of resolution are models on the dispersed phase scales, sometimes called interpenetrating continua scale.

The Euler-Euler framework is a classic example of such models. The discrete element models (DEM) follow the Euler-Lagrange framework that are higher fidelity models for tracking the particle-particle interaction. Then the population balance models provide a formalism for tracking bubble/droplet breakup and coalescence. The Particle Resolved Direct Numerical Simulation (PR-DNS) is the next higher level of fidelity in the modelling heirarchy. At this level the modelling parameters needed are typically physical properties such as density and viscosity to support the constitutive equations that model modelcular phenomena on a continuum scale. They have the ability to predict the local variations in drag, heat and mass transfer coefficients on the particle scale. At an even deeper level than the continuum level models, we have Molecular Dynamics models and the Density Functional Theory models. Computations at the molecular level will enable innovation in materials synthesis and characterization, and provide information needed for the continuum models for transport processes. It is the DNS, DPM, DEM, Euler-Euler frameworks that will enable inovations in process equipment design for manufacturing of these chemicals and materials.

HEIRARCHICAL MODELLING FRAMEWORK

Equipment level lumped performance models – losing the impact of flow structure

A performance level model for a reactor, for example, will assume homogeneous flow conditions inside the process vessel, thus ignoring the impact of flow on. A simple mass balance for a continuously stirred tank reactor that is well mixed will have the form

$$V\frac{dC_i}{dt} = F(C_{in} - C_i) - r_i(C_i) \qquad (1)$$

Where r_i is the effective reaction rate expression, C_i is the homogeneous exit concentration from the reacor, C_{in} is the inlet concentration, F is the flow rate. Similar models can be developed for mass transfer in bubble columns that will require overl all mass transfer coefficients as inputs. As such vessel volumes V are scaled up, the flow pattern and residence time distributions are likely to change affecting such parameters as reaction rate constants and mass transfer coefficients.

Continuum models – losing the identity of molecules

In process equipments, for example, a more detailed model for flow and other transport and reaction processes can be developed through the following equations. For single phase, isothermal flow of a Newtonain fluid, such conservation law based models remain scale invariant and hence are more useful for process design innovations at various scales.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho\, \boldsymbol{u}) = 0 \qquad (2)$$

$$\frac{\partial \rho \boldsymbol{u}}{\partial t} + \nabla \cdot (\rho\, \boldsymbol{uu}) = -\nabla \mathrm{p} + \nabla \cdot \mu \nabla \mathbf{u} + \mathbf{F} \qquad (3)$$

$$\frac{\partial C_i}{\partial t} + \nabla \cdot (\rho\, \boldsymbol{u} C_i) = \nabla \cdot \mathrm{D_i} \nabla \mathbf{C_i} + r_i(C_i) \qquad (4)$$

Figure 4: Momentum and mass conservation equations for a single phase Newtonian fluid that is scale invariant.

These are complex, coupled partial differential equations that are difficult to solve at higher Reynolds numbers, necessitating the introduction of filtering procedures for turbulence and multiphase flow models, requiring closure relationships.

Direct Numerical Simulation frameworks

When a mobile second phase is introduced the coupling between the two phases can be handled in a rigorous manner using the DNS framework. The model equations are shown in Figures 5 & 6 for both fluid-particles and fluid-fluid systems. However, when the number of dispersed phase entities is very large such as in fluidization or bubble columns, tractability of the solution even using computers becomes a challenge. In such cases DNS probes with statistically meaningful number of particles or droplets can be used to discern the closure relations that are needed in the averaged model equations.

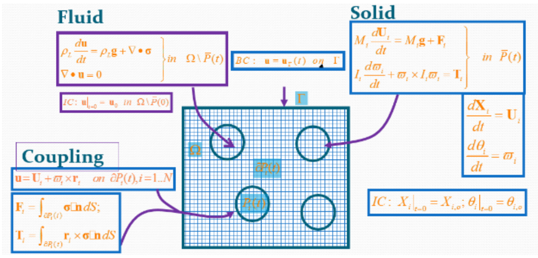

Figure 5: Fully resolved fluid-solid interaction models that track particle-fluid and particle-particle interactions, such as immersed boundary and fictitious domain methods.

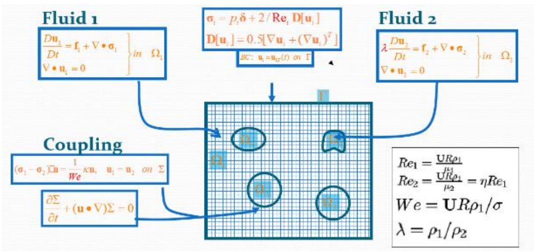

Figure 6: Fully resolved fluid-fluid interaction models that track the interface deformation, such as volume of fluid, level set methods.

Discrete Element Modelling framework

In the discrete element framework, shown in Figure 7, Euler-Lagrange formulation is used, where the fluid is treated in the averaged sense and particle identity are kept, but the interaction with the fluid is not fully resolved, but only through a drag closure relation.

(a)

Continuity equation

$$\frac{\partial}{\partial t}(\varepsilon\rho_g) + \nabla\cdot(\varepsilon\rho_g \mathbf{u}_g) = 0$$

Momentum equation:

$$\frac{\partial}{\partial t}(\varepsilon\rho_g\mathbf{u}_g) + \nabla\cdot(\varepsilon\rho_g\mathbf{u}_g\mathbf{u}_g) = -\varepsilon\nabla p + \nabla\cdot(\varepsilon\bar{\bar{\boldsymbol{\tau}}}_g) + \mathbf{S}_g + \varepsilon\rho_g\mathbf{g}$$

Momentum exchange source terms

$$\mathbf{S}_g = -\frac{1}{V}\int_V \sum_{k=1}^{NP} \frac{V_{p,k}\beta}{1-\varepsilon}(\mathbf{u}_g - \mathbf{u}_{p,k})\delta(\mathbf{x}-\mathbf{x}_{p,k})dV$$

(b)

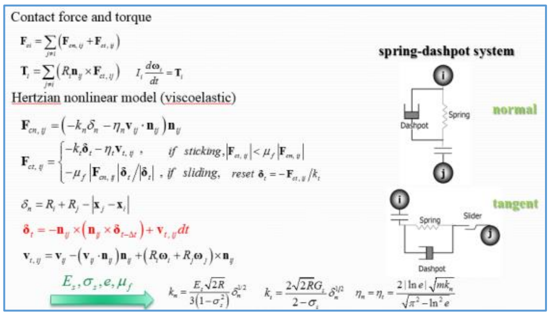

Figure 7: (a) Continuous phase is treated in the Eulerian framework with the feedback of momentum from the discrete phase. (b) Detailed modelling of particle-particle interaction model.

Euler-Euler interpenetrating continua – losing the discrete nature of dispersed phase

In this formulation, the identity of the discrete phase is lost as spatial or ensemble averaging of the continuum level equations are carried out to a scale larger than dispersed phase scale. Hence closure models are necessary. Typical model equations coded in commercial simulators have the following forms. The rigor in such ad-hoc extensions remains questionable.

$$\frac{\partial \alpha_i \rho_i}{\partial t} + \nabla\cdot(\alpha_i\rho_i\,\boldsymbol{u_i} - \Gamma_i \nabla\alpha_i) = 0, \quad i = \mathrm{L,G} \quad (5)$$

$$\frac{\partial \alpha_i\rho_i\boldsymbol{u_i}}{\partial t} + \nabla\cdot[\alpha_i(\rho_i\,\boldsymbol{u_i}\boldsymbol{u_i} - \mu_i(\nabla\boldsymbol{u_i} + \nabla\boldsymbol{u_i^T}))] = \alpha_i(\mathbf{B_i} - \nabla p) + \mathbf{M_i}, \quad i = L, G \quad (6)$$

$$\frac{\partial \alpha_i\rho_i Y_{ji}}{\partial t} + \nabla\cdot[\alpha_i(\rho_i\,\boldsymbol{u_i}\boldsymbol{Y_{ji}} - \Gamma_i\nabla Y_{ji})] = \sum_{k=1,k\neq i}^{N} m_{ik}^{j}, \quad i = L, G, \quad j = 1, \dots N \quad (7)$$

Figure 8: Momentum and mass conservation equations for Euler-Euler models. They need scale invariant closure models from dispersed to equipment scales.

ON-GOING CASE STUDIES AT EPIC

We have examined numerous physical problems involving multiphase flows, some of which include spontaneous structure formation and others include large industrial applications. Due to space limitations, details are not presented here. At the oral presentation detailed validation of model predictions against experimental data will be presented for the following specific case studies. (a) deep water oil-spill modelling involving oil-gas dispersion in sea water using Euler-Euler model with Population Balance models as well as the dynamics of single droplet in the presence of interface mass transfer of surfactant and hydrocarbons, and (b) dynamics of a slurry loop reactor. The design space exploration will be illustrated with (a) clarifier design and (b) a fractal distributor design.

First the DPM framework is used to study the dynamics of *suspension droplets* as they settle in a liquid. Experimental observations are documented in Machu et al. (2001). The questions are (a) when does a suspension containing particles in a liquid behave like a droplet with its own distinctive effective properties from the suspending fluid, (b) how does the progressively complicated dynamical structure predicted by the DPM framework compare with the experimentally documented phenomena and (c) what other new structures can be predicted by the DPM framework, not yet observed in any experiments. At a Reynolds number of 5, Fig 9 shows the breakup of a spherical suspension droplet into two droplets through the formation of a torus as an intermediate step. More complex phenomena will be presented in the oral session.

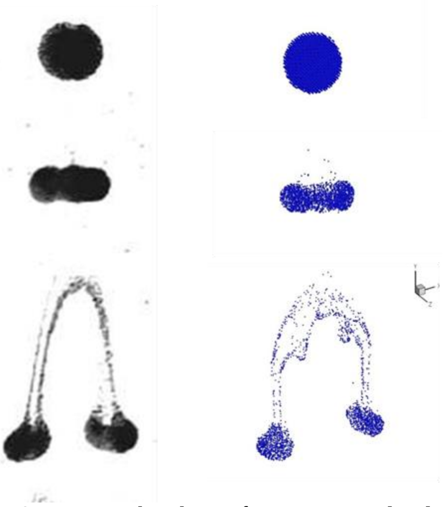

Figure 9: Dynamic break up of a suspension droplet using the DPM framework as compared to similar break up process from the experimental work of Machu, Meile et al. 2001

The next example is one where solids interaction is dominant and fluid dynamics effects are minimal. The physical setup is one where two different types of granular material are partially filled in a square container and the container is rotated about its axis at a constant speed. The particles are initially well mixed to achieve homogeneous dispersion. With time, the particles segregate spontaneously and the DEM framework is able to capture the phenomenon as seen in Fig. 10.

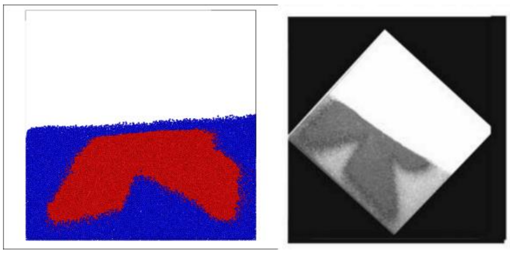

Figure 10: Spontaneous pattern formation in a rotating square drum containing two distinct spherical particles. Comparison of DEM predictions with experimentally observed pattern of Jain et al (2005).

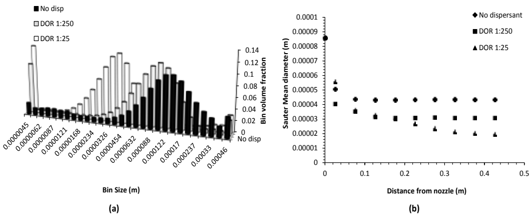

Figure 11: Droplet size distribution and mean diameter along the jet length of a turbulent oil jet in a submerged water in the presence of various Dispersant to Oil ratio, (DOR).

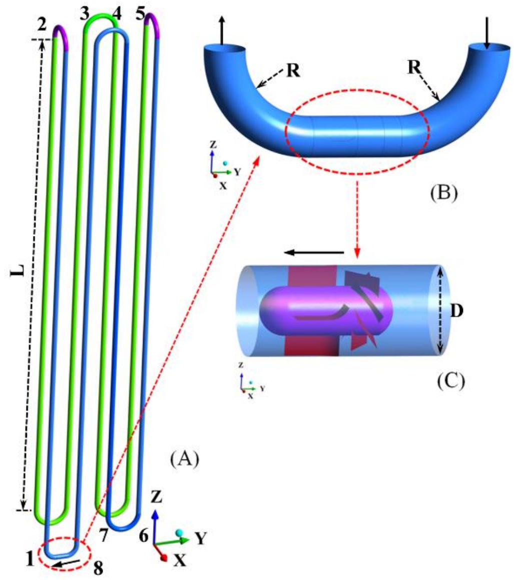

Figure 12: Polymerization loop reactor dynamics using Eulerian-Eulerian framework. This is an eight leg loop reactor with an internal pump, which under certain operating conditions gives rise to well-known oscillations in the operating regime of the reactor.

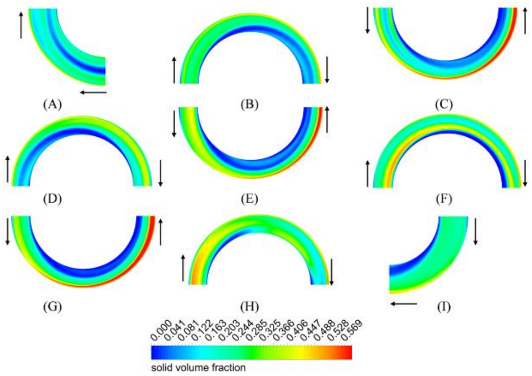

Figure 13: Contours of solid volume fraction on the middle plane of the bends. The letters (A) ~ (I) indicate the bend number indices from 1 to 9. The black arrows point the flow direction at the entrance and exit of the bends.

The next example demonstrates the TFM-PBM (Two-Fluid model with Population Balance Model) framework to model the droplet breakup from a turbulent jet that emanates from a nozzle. This phenomenon is akin to oil discharge in an undersea environment and quantitative data have become available recently from SINTEF. The Figure 11 below shows that the droplet size distribution shifts to lower size with increasing amounts of the dispersants used. Other results will be presented at the oral session.

Figures 12 and 13 above shows the 8-leg loop reactor using the Euler-Euler model to study the slug formation. Figure 13 shows the segregation of sloids around each bend. This is a strong function of solids concentration as well as particle size. Such a study reveals the mechanisms of slug formation which is detrimental to production capacity increases. The model can then be used to develop design alternatives to overcome such shortcomings.

CONCLUSION

A hierarchy of models of varying fidelity provide a framework for studying the dynamics of multiphase flows in process vessels. Some of the extensions found in current simulators for turbulent multiphase flows are ad-hoc extensions from our understanding of single phase turbulent flows. Theoretical advances are necessary to identify the proper averaging or filtering procedures and the structure of ensuing closure models. Validation of such models against carefully conducted experiments are also necessary. With advances in measurement techniques, such data for validation of multiphase models are within reach. Furthermore advances in manufacturing techniques enable design of process equipment that can manage the phase distributions inside vessels to any desired configurations. With sustained efforts, the reliability of models can be improved and innovative designs will begin to come out.

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