

Research Proposal

To my view, the core interest of chemical engineering lies in creating a world that makes the best possible use of its limited resources. When crude oil was fit for nothing more than fuel, chemical engineers distilled it and processed it into everything from plastics to pharmaceuticals. Over time, chemical engineering has subsumed special interests in soft matter physics and biological engineering with the same over-arching goal.

In soft matter physics, there are unique opportunities to develop low-cost and high value-added materials by careful engineering at the microstructural level. However, there are always at least two major technical obstacles to viable commercial production (1) the material must be processed into the target microstructure and (2) processing must be sufficiently fast/efficient to keep the operation profitable. Rheology is the sub-discipline of soft matter physics that attends to these challenges, bridging the gap between knowledge of the microstructure and understanding of the flow behavior.

In my research as a rheologist, I am principally concerned with industrially-relevant complex fluids that are already being processed in enormous volumes - billions of tonnes per year – including commodity polymers [1, 2, 3, 4, 5], dense suspensions [6, 7], and wormlike micelles [8, 9, 10]. Improvements in the way such materials are processed can have enormous market impacts by reducing waste, increasing through-put, and/or enhancing the value of the final product. These topics will continue to be an important part of my research portfolio.

Like rheology, biological engineering is a core discipline in most chemical engineering departments today, but the broader study of biological systems extending to the level of whole ecosystems has recieved comparatively less attention. However, it is my view that the future of chemical engineering must eventually include biological systems engineering at this level. For example, the raw materials for a renewables revolution are primarily going to be sourced from an agricultural system under increasing strain from disease [11], pests [12], and pollinator decline [13]. Moreover, current trends in agricultrual expansion and invasive species are stressing the natural ecosystems we rely on for carbon capture, water purification, nutrient cycling, and more [14].

During the COVID-19 pandemic, I helped develop new tools for studying disease transmission in human populations [15, 16, 17]. As part of my future research, I would like to repurpose these tools to study ecosystems - both natural and agricultural - under stress from disease, invasive species, climate change, and more. Ultimately, ecosystems can be modelled as a special kind of reaction/diffusion problem, and as such are well within the core competency of the chemical engineer. But more importantly, maintaining healthy ecosystems is integral to the core interest of chemical engineering as a field: creating a world that makes the best possible use of its limited resources.

With these motivations in mind, I outline three prospective research topics that interest me.

First, as a continuation of my PhD and postdoctoral studies, I would like to study competing explanations of “die drool” phenomena in polymer extrusion. Second, there is an opportunity to fill a gap between theory and practice for high Reynolds number flows of many complex fluids. Third, I propose an example problem for modeling epizootic controls of invasive species.

1. Problems in Polymer Processing

In industry practice, polymer extrusion is often limited by flow instabilities and irregularities that mar the extrudate, including melt fracture and shark-skin [18], as well as die drool [19]. In each case, there are empirical strategies for mitigation, but theoretical understanding is still considered incomplete in many respects. Over the past few decades, there have been remarkable advances in both (1) CFD-friendly and microscopically accurate rheological models [3, 5] and (2) the power of computing resources needed to solve such models. From my experience working with companies like 3M and Unilever, I have seen that the trial-and-error stage of die design and product formulation is increasingly being shifted towards in-silico experiments. With the tools that I’ve developed during my PhD and postdoctoral studies, I believe that there is a unique opportunity to build a unified modeling framework to study “die drool” phenomena in polymer extrusion.

In polymer extrusion, “die drool” refers to the slow build-up of polymer melt at the die face, as shown in Figure 1, and it mars the extrudate when it drips down or chars onto the die face, for example. It is clear from the existing literature that the “drool” itself consists of chains that are shorter than those in the main extrudate, but it is not yet clear (1) how those short chains became sequestered at the outermost layer of the flow or (2) why the outermost layer of the flow (comprising mostly short chains) separates from the main extrudate onto the die face. The proposal that I outline here will focus on the first of these two questions, but in principal the modeling framework I propose is suitable to consider both questions.

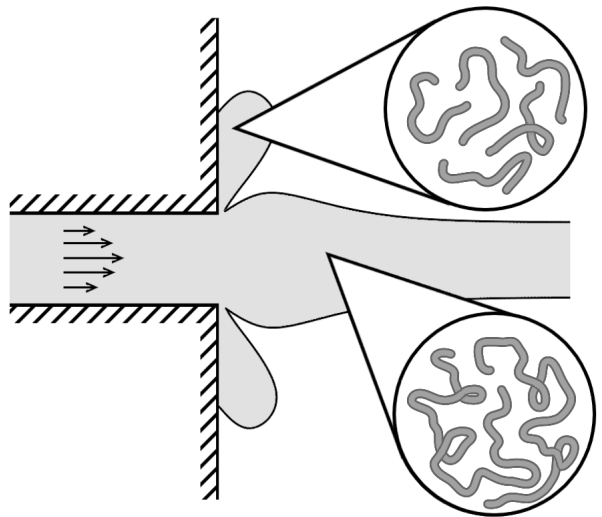


Figure 1: Cartoon of die drool accumulating on the face of an extruder

There are two prevailing hypotheses for how short chains become sequestered near the extruder walls: flow-induced fractionation and chain degradation [19]. In one instance, it is hypothesized that gradients in the stress drive long chains to migrate away from the wall, where shear rates are highest. In the second instance, it is hypothesized that polymer degradation (i.e. long chains breaking into short chains) can be significant near the extruder walls, due to a combination of high shear rates, high temperatures, and long residence times. It is difficult to distinguish between

these two hypotheses in an experimental setting, as each of these mechanisms could be dominant depending on the particulars of the melt composition, flow rate, and extruder geometry.

To gain a clearer understanding of die drool phenomena, I want to study a multi-fluid approximation of polydisperse blends with flow-induced scission in an annular flow. This study would combine my PhD work on multi-fluid models of polydisperse blends [4, 5] with my postdoctoral studies of breakable polymer rheology [8, 9]. The results of this study would yield both (1) experimentally testable predictions for the extrudate’s annular composition and (2) a broader picture for how these two mechanisms contribute (or even interact) to produce die drool under various flow conditions. This study will bring new insights to the mechanism behind die drool formation, and should also provide opportunities to improve existing mitigation strategies. The modeling framework including both degradation and fractionation may prove relevant to other polymer processing operations where temperatures and shear rates are high, including mixers, kneaders, and twin-screw extruders.

2. Reynolds Averaging for Complex Fluids

In recent decades, rheologists and soft matter physicists have made remarkable progress in building constitutive equations that connect the microscopic structure of a material to its bulk flow behavior. At one point, microscopically-derived constitutive models were only available for very simple non-Newtonian fluids, such as dilute suspensions of hard spheres, droplets, and polymers. Today, however, microscopically-derived constitutive models are available for all manner of complex fluids, including entangled linear polymers [20], wormlike micelles [8], dense suspensions [6], yield stress fluids [21], and even more exotic materials like entangled telechelic star polymers [22]. In principle, these models are useful for directly modeling industrially relevant fluids under industrially relevant flow conditions, but in practice this is not always true - many industrially relevant complex fluids (e.g. drilling muds, fracking fluids, emulsions, etc.) are processed at high Reynolds numbers (e.g. by use of large diameter pipes/vessels)

Typical bench-scale rheology experiments are not adequate for predicting a material’s rheology at high Reynolds numbers. For example, when processing a dense suspension, the particle loading is chosen to be as high as possible without jamming under the desired flow conditions. However, the critical shear rate for jamming under steady shear flow conditions (as might be measured in a laboratory setting) can not be used to predict the critical flow rate for jamming under turbulent flow conditions. In a turbulent flow, the chaotic “fluctuations” will lead to regions of flow becoming locally jammed, which can then percolate a jamming transition across the whole system.

In a sense, a turbulent flow imparts an additional level of “microstructure” to the fluid at length-scales and time-scales much smaller and much faster than those of the overall process. This separation of scales makes direct numerical simulation impractical - ideally, one would prefer to treat the turbulence itself as an additional level of microstructure to be averaged out.

The notion of turbulence as an additional level of microstructure (c.f. Figure 2) is used extensively for modeling Newtonian fluids at high Reynolds numbers [25, 23, 24], but at this point Reynolds-averaged constitutive models for complex fluids are mostly limited to simple non-Newtonian fluids, like the FENE-P model for dilute polymer solutions [26]. The process of constructing Reynolds averaged constitutive equations is difficult in practice but straightforward in principle; just as mesoscale constitutive equations rely on closure approximations informed by particle-based simulations, Reynolds-averaged constitutive equations rely on closure approximations informed by direct numerical simulation of the mesoscale constitutive equation in simple turbulent flows.

As a starting point on this project, I would like to develop Reynolds-averaged constitutive equations for dense suspensions, using recent models I helped develop during my postdoc [6, 7] to describe microstructure evolution at the mesoscale. These dense suspension models are, in my view, the first to contain sufficient microscopic detail to be useful for turbulent flow modeling: they have been successfully validated in reversing flows, shear-jamming flows, and flows with unsteady perturbations [6, 7]. Immediate applications for a Reynolds-averaged extension could include modeling of wellbore fluids, industrial concrete mixers, mudslides, and more.

3. Ecological modeling

The gypsy moth (Lepidoptera: Lymantriidae) is an invasive species that can cause enormous damage to ecosystems through defoliation of hardwood trees [27]. The moth was originally native to Europe, but was introduced to Massachusetts around 1870, whereafter it began to spread westwards [28]. In the early days, it was difficult to contain the spread of the gypsy moth population - there were few natural predators, and species-specific pathogens (e.g. *Lymantria dispar* multicapsid nuclear polyhedrosis virus or LdMNPV) are not a strong control method unless the moth population is already moderately high [29]. In this respect, viral controls were (and still are) helpful in the short term but not viable as a long-term control measure. Pathogens that are not species specific, like baculoviridae, can also be useful for short-term control of major outbreaks.

Today, the gypsy moth population continues to spread but it now poses a greatly reduced long-term risk to natural habitats, primarily due to a species-specific fungus from Japan (*Entomophaga maimaiga*) that was first established in New York in the late 1980s [30]. Unlike the viral pathogen LdMNPV, the fungal spores can persist in the environment for years, making it effective for long-term population control even when the moth population is low. At the leading edge of the gypsy moth invasion, however, this fungus is not yet established in the environment and there is still an

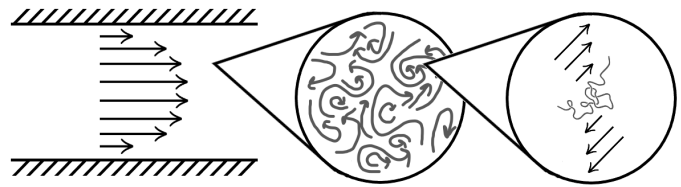


Figure 2: Turbulence as an additional layer of microstructure. Cartoon shows a polymer-based complex fluid as an example.

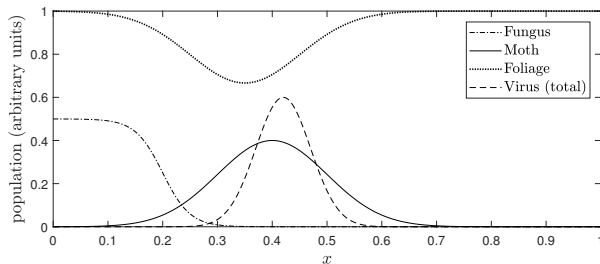


Figure 3: A cartoon figure for the spread of gypsy moths across a 1D landscape in the presence of fungal and viral control measures. Viruses are introduced (and reproduced) at the leading edge of the moth population to slow the spread, while a fungus trails the leading edge to keep the moth population in check long term. Defoliation (and the damage thereof) is primarily localized to the leading edge of the invasion.

abundance of foliage and an absence of natural predators. In the very long term, both the gypsy moth and its fungal pathogen will inevitably establish themselves across North America, and the fungal pathogen will keep population levels in check. But to reach this final state, what control measures best balance the total damage to our ecosystems against the total cost of containment? The question of optimal control is still an ongoing area of research.

To that end, as a first foray into ecosystem modeling, I would like to use sophisticated new epidemic modeling tools (like those in the pyRoss library I worked on for COVID-19 [15, 16, 17]) to study optimal control measures in systems involving invasive species and two complementary control pathogens: one with a long persistence time but a low reproduction rate (e.g. the fungus) and another with a high reproduction rate but a low persistence time (e.g. the virus). The gypsy moth and its pathogens are an appealing system for a first application, but I believe that the general modeling framework may prove interesting for other invasive species, where long-term biological control measures are still a subject of active research (e.g. cane toads, emerald ash borers, etc).

A cartoon describing an idealized version of this problem with one spatial dimension is shown in Figure 3. At position $x = 0$, the moth and its fungus are both endemic, and neither poses a long-term threat to the native habitat. At position $x = 1$, neither the moth nor its fungus are present. The leading edge of the moth invasion occurs around $x = 0.4$, where the virus is being introduced (and reproduced by the moths themselves) to slow the invasion. This is, in a sense, a kind of multi-component reaction/diffusion problem, except the reactions and diffusion are biological processes with distinct kinetics and time-delays particular to the system of interest.

The basic modeling framework required for this study can be easily extended to include additional species and additional control measures. The proposal outlined here is strategic for its ecological pertinence and relative simplicity, but the underlying tools for modeling and optimizing intervention protocols are easily extended to more complex systems with even broader impact.

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