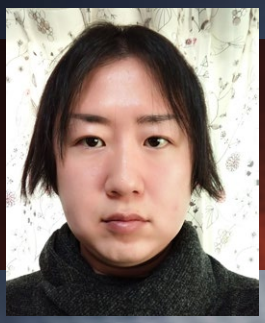

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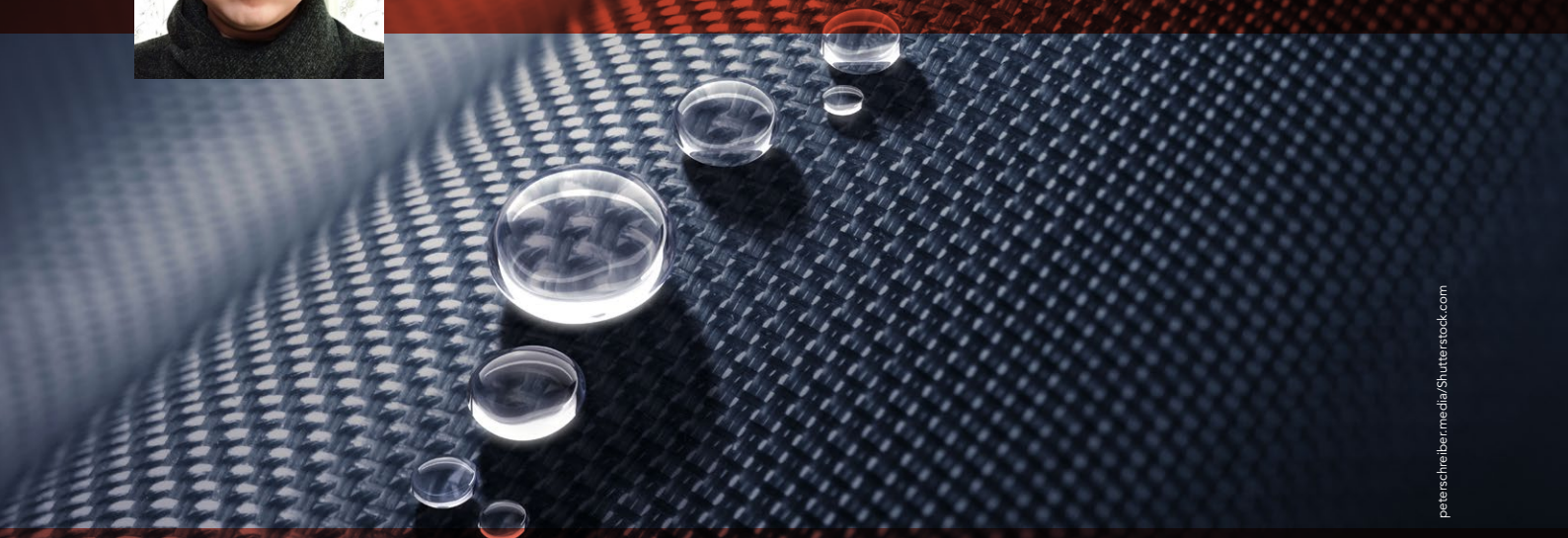

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Modelling Sea-Island Reversal in Viscoelastic Phase Separation

Research Objectives

Mr Kubo studies alternative ways to characterise so-called viscoelastic phase separations.

Detail

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Bio

Mr Yoshihide Kubo, MSc, studied at the Faculty of Science Department of Physics, Kagoshima University, and later received his master's degree at the Graduate School of Integrated Arts and Sciences, Hiroshima University. After graduating, he continued to conduct research at home whilst working as a programmer. Currently, Yoshihide Kubo is working for SHIKOKU ELECTRIC POWER CO., INC.

Collaborators

- Shinpei Tanaka
- Yoshihiro Yamazaki

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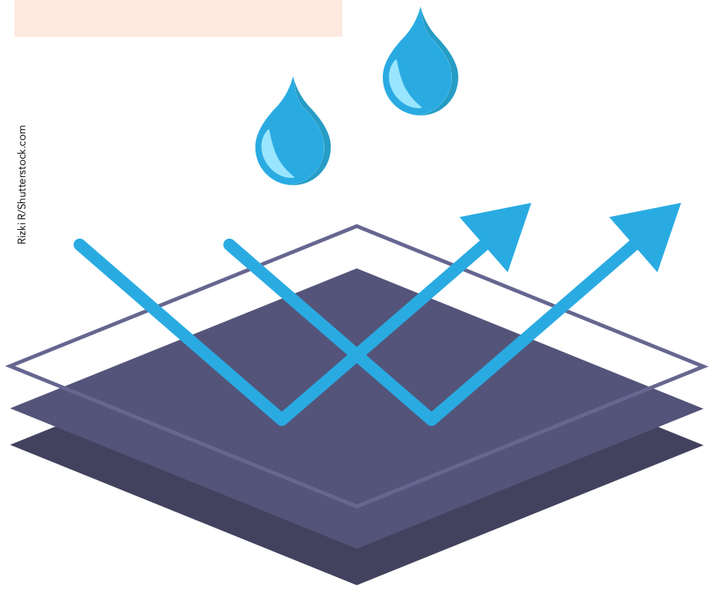
Personal Response

How could your approach be applied to improving manufacturing processes?

"If a material with gelation is used, a network structure can be spontaneously generated, and if it is solidified in that state, the structure can be made lightweight and sturdy, like the internal structure of bone. In addition, whether the reversal of the sea-island structure occurs depends on whether the mixed solution concentration is above or below the gel point. Also, even if the mixed solution concentration is too far from the gel point, it is difficult to observe a clear reversal of the island-sea structure, so it is crucial to pay attention to the mixed solution concentration during the manufacturing process."

Modelling Sea-Island Reversal in Viscoelastic Phase Separation

In the 1990s, researchers first discovered that within mixtures of viscoelastic materials, unique transitions known as phase separations can appear when the right conditions are applied. In his research, Mr Yoshihide Kubo, Hiroshima University alumnus, presents the first robust approach for modelling these systems, avoiding unnecessary complexities while producing results which can be easily reproduced in experiments. His work addresses a variety of long-standing hurdles which were faced by previous models and could help researchers to gain a better understanding of the diverse physical properties exhibited by soft materials.



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When two liquids with different physical properties are thoroughly mixed, the resulting fluid can appear to display its own unique properties, which remain uniform throughout its interior. However, when the right temperatures and concentrations are applied, the two fluids can suddenly separate again through a process named 'phase separation' – significantly altering the properties of the overall mixture. Previously, phase separation was known to occur in a wide variety of both solid and fluid systems, which could be well described by mathematical models. Yet in the 1990s, materials physicists discovered a new type of phase separation entirely.

Their findings relate to 'viscoelastic' materials. These materials are both resistant to flow – like honey and syrup – and move back to their original shapes when mechanical forces acting on them are removed – like rubber. This behaviour can be found during the phase separation of mixtures with 'asymmetric' dynamics, containing some slow-moving molecules and others travelling far more quickly. It is particularly common in many forms of 'soft matter': a family of deformable materials including polymers, liquids, and gels which behave in a similar way to solids until mechanical forces are applied.

In many cases, the asymmetric dynamics characteristic of viscoelastic phase separation originate from large size differences between the molecular components of fluid mixtures. Alternatively, they can arise in a variety of glass-forming materials where there are large temperature differences between hard, brittle glassy states, and softer rubbery states. Because of these unique properties, the models previously used to describe phase separation in solid and fluid systems could no longer be applied in this more complex case.

SEAS AND ISLANDS

Through early studies in the 1990s, researchers first observed that viscoelastic phase separation involves the rapid reversal of 'sea-island' structures, where smaller droplets of one fluid are suspended in larger fluids surrounding them. As time goes by, these seas and islands can flip around. As this happens, the larger fluid will condense into droplets or strings while the other fluid will expand to surround the former. In the process of transitioning the sea to an island, it becomes an island after the sea exhibits a network structure. The network structure and the reversal of the sea-island structure are observed during the transition period from the mixed state to the separated state in the viscoelastic phase separation.

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If applied to manufacturing processes, Mr Kubo's research could help develop methods for fabricating novel materials, such as hydrophobic surfaces which strongly repel water.

These pattern changes are strongly driven by a long-range connectivity between the molecules in each fluid. When environmental conditions pass certain thresholds, the nature of this connectivity can change suddenly, altering the physical properties of the liquid. Such unique properties are characteristic of 'sol-gel' processes, in which polymer-forming molecules suspended in a liquid transition into gels made up of integrated polymer networks. However, when viscoelastic phase separation was first discovered, its characteristics proved far more difficult to model than other types of phase separation. This problem has persisted until the present day. So far, these difficulties have prevented researchers and manufacturers from exploiting its unique properties for useful applications.

Viscoelastic phase separation involves the rapid reversal of 'sea-island' structures, where smaller droplets of one fluid are suspended in larger fluids surrounding them.

PROS AND CONS OF COMPLEX MODELS

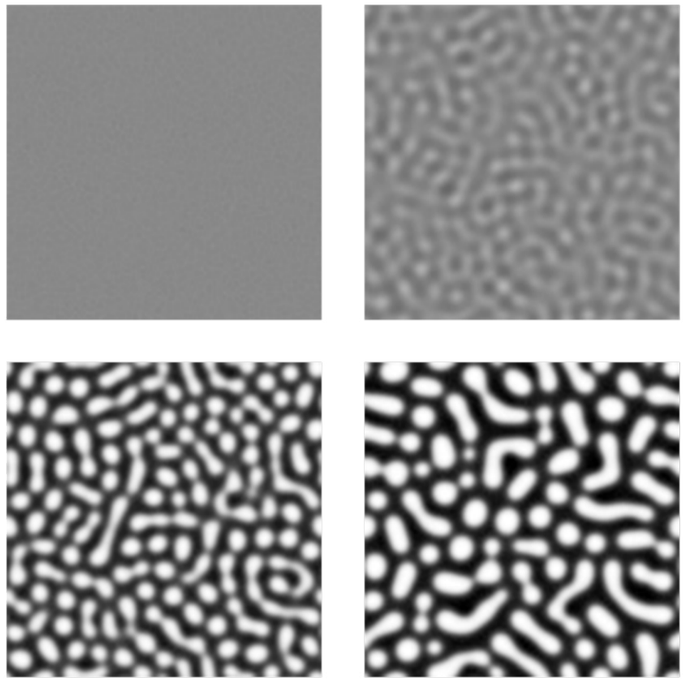
Despite these hardships, Mr Yoshihide Kubo's efforts to build a more reliable model have been founded on a strong mathematical basis. Among the first models created to study viscoelastic phase separation was the 'two-fluid' model – which considered how dynamical asymmetry arises within solvents made from large, complex polymer molecules, and smaller solvent molecules. This approach was strongly based around considerations of the connectivity between these particles. Therefore, the equations describing the motions of both materials expressed a link between the mechanical stresses applied to them, and the speeds at which they diffused.

Alternatively, other models considered the viscoelastic properties of mixtures during phase separation in a more direct way. While one model considered how mixtures relax after mechanical stresses are removed, another approach approximated their dynamics by treating interactions between particles as tiny springs – which can become disconnected during phase separation. Through further studies, researchers showed that this family of models can successfully predict the outcomes of real observations of viscoelastic phase separation. Yet despite this advantage, the complexity of these approaches made it difficult for researchers to integrate into their simulations – severely limiting their applicability.

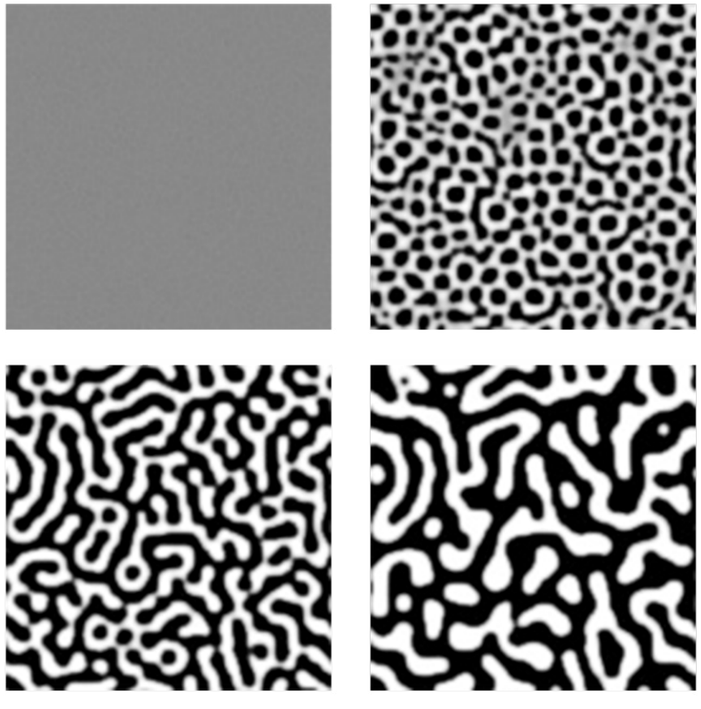
LIMITED SUCCESS WITH SIMPLER MODELS

At the same time as the development of these models, other studies were aiming to model viscoelastic phase separation in a more straightforward way. These efforts would be based on mathematical frameworks which directly approximate dynamical asymmetries, without accounting for the physical properties responsible for them. To do this, researchers in these studies introduced an equation named the 'mobility function', in which the motions of both fluids in the mixture depend on their concentrations instead of more complex underlying factors.

Using this approach, researchers were able to predict the evolution of



Numerical simulation result of normal phase separation.



Numerical simulation result of viscoelastic phase separation.

sea-island patterns characteristic of viscoelastic phase separation, without any need to consider the physics of viscoelasticity. Due to its simplicity, the most important outcomes of the process can be condensed down into a single equation, making it far easier for researchers to implement within fluid dynamics simulations. However, the simplicity afforded by the use of mobility functions came with its own disadvantages. With less of a basis in real-world phenomena, the outcomes of these models were far less likely to reflect real experimental results, making it more difficult for researchers to make reliable predictions from their simulations.

BUILDING A FREE ENERGY MODEL

Together, these two approaches to modelling the complexities of viscoelastic phase separation have enabled researchers to explore many of the phenomenon's unique characteristics since its initial discovery. Ultimately, however, their shortcomings have long called for a more sophisticated method, which is both accurate, and easy to implement into simulations. Through his latest research, Mr Kubo has discovered that these goals can be reached through a small alteration to a mathematical equation describing the 'free energy' of mixtures during viscoelastic phase separation.

Free energy is a term describing the maximum amount of energy which can be added to a thermodynamic system while keeping either its temperature and pressure constant, or temperature and volume constant. Through their calculations, Mr Kubo and his colleagues have proved that their proposed alteration to its description can keep the parameters representing the dynamics of a separating mixture constant, even as its physical properties evolve over time. Yet instead of modelling parameters like elastic relaxation or direct changes in particle dynamics, the team's free energy model considers a value named the 'chemical potential' – describing the amount of energy which is either absorbed or released as the quantities of particles undergoing a phase transition are varied.

ACCOUNTING FOR CHEMICAL POTENTIAL

Using their updated approach, Mr Kubo's team have now considered how the flows within a mixture can be enhanced during viscoelastic phase separation, causing a jump in its chemical potential. Importantly, if environmental conditions remain the same, such a jump can only occur within mixtures with mostly identical compositions. Therefore, by adjusting the composition of the mixture, the researchers were able to precisely control dynamic asymmetry in their simulations, without any need to consider more complex physical properties.

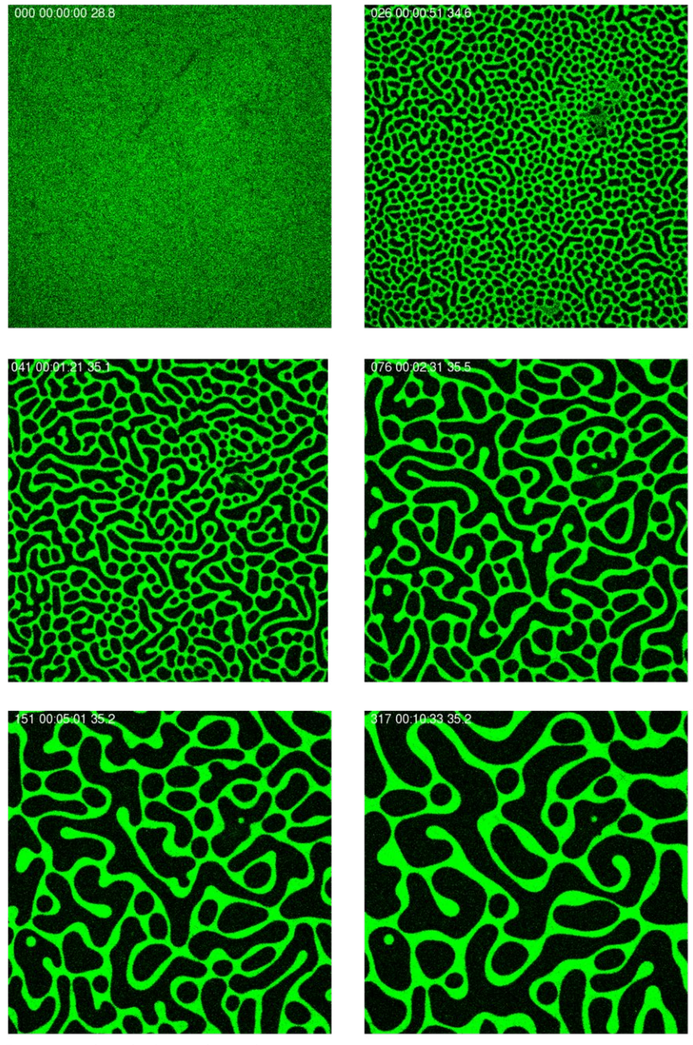
These advantages enabled the researchers to fine-tune their simulations to produce specific sea-island patterns, which, in turn, allowed them to engineer mixtures with specific physical properties. Their considerations of free energy and chemical potential even allowed them to reproduce the strange patterns found in solutions of dilute emulsifying fluids – which form rings around their island structures as phase transitions unfold. Overall, the team's results suggest the need for a rethink of the physics involved in viscoelastic phase separation. This would involve developing models centred around free energy – which is a static physical quantity, instead of other effects – such as dynamic asymmetry or the viscoelastic effect.

Kubo's model says that a large flow occurs at the transition concentration, suggesting that the phase transition due to the concentration change is the cause of the sea-island structure reversal. Furthermore, since it can be read that the free energy model of gel also has a jump in chemical potential, it may be necessary to link the reversal of the sea-island structure with gelation or a microscopic network similar to gel.

IMPROVING PROSPECTS FOR FUTURE RESEARCH

By accounting for Mr Kubo's discoveries in their models, researchers in future studies of viscoelastic phase separation could soon become far better equipped to understand how its unique characteristics can vary with material composition. They could also gain the ability to finely tune mixtures with dynamical asymmetries to exhibit specific properties during phase separation, making them suitable for a wide variety of applications.

If applied to manufacturing processes, such uses could include new methods for fabricating hydrophobic surfaces which strongly repel water, membranes which can filter liquid mixtures on microscopic scales, and pigment separation in chromatography.



Experimental result of viscoelastic phase separation.

Instead of modelling parameters like elastic relaxation or direct changes in particle dynamics, the team's free energy model considers a value named the 'chemical potential'.

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