Institution: University of Leeds

Unit of Assessment: UOA 09

Title of case study: "Decreasing the environmental impact and delivering substantial manufacturing cost reductions for cellulosic film production in the UK and USA"

Period when the underpinning research was undertaken: 2010-2019

Details of staff conducting the underpinning research from the submitting unit:

Name(s):	Role(s) (e.g. job title):	Period(s) employed by submitting HEI:
Professor Mike Ries	Professor of Physics	2003-present

Period when the claimed impact occurred: 2010-2019

Is this case study continued from a case study submitted in 2014? No

1. Summary of the impact (indicative maximum 100 words)

Professor Mike Ries has collaborated with the global manufacturing company Futamura, the world's largest producer of renewable cellulosic films with a 65% share of the world's market. This collaboration has enabled Futamura to significantly decrease the quantity of chemicals used in the production of NatureFlex cellophane, saving 300 tonnes of carbon disulphide and 30 tonnes of caustic soda per annum. As a result, their UK and USA divisions' environmental impact has decreased, leading to a significant reduction in Futamura's environmental footprint.
In addition, a significant improvement (2%) in the cost efficiency of the process has been achieved, which equates to an annual cost saving of GBP1,300,000 while in parallel solving manufacturing problems such as the use of different wood pulp sources and improving film mechanical properties.

2. Underpinning research (indicative maximum 500 words)

Each year the amount of cellulose produced by nature outweighs, by factors of thousands, all the combined manufactured oil-based polymers. It is an almost inexhaustible source of raw material for environmentally friendly products and has the potential to lessen the need for conventional petroleum-based plastics significantly. Cellulose can be used to produce films, fibres, sponges, membranes and food thickeners.

Unfortunately, although the resources stem from nature and the final products are biodegradable, the whole process is not in reality "green" due to the solvents used. Current cellulose processing is one of the most polluting industrial chemical methods in use today. For example, the US Environmental Protection Agency (EPA) 'identified carbon disulphide (CS_2), carbonyl sulphide (COS), and toluene as the hazardous air pollutants (HAP) emitted in the largest quantities from viscose process operations' https://www.epa.gov/stationary-sources-air-pollution/cellulose-products-manufacturing-national-emission-standards.

For cellulosic films to compete with traditional plastic packaging, it is essential to improve both the environmental footprint and the cost efficiency of its production.

More sustainable solvents for cellulose processing [i, ii, iii]
An ionic liquid is a salt with a melting point below 100 °C. They are chemically and thermally stable, have negligible vapour pressure and are recyclable. A number of ionic liquids have been found to dissolve cellulose, with the result that they offer a route to an environmentally friendly process for cellulose production. With this aim, a collaboration was formed between the University of Leeds and

the Japanese cellulosic manufacturer Futamura, to determine what makes a good cellulose solvent. An investigation into carbohydrate ionic liquid solutions was carried out using nuclear magnetic resonance (NMR). This helped understand the mechanism of cellulose dissolution, which aided the future design of "green" solvents. This research resulted in two publications by **Ries**, one with Futamura **[1]** and one with a French research centre of excellence, Mines ParisTech **[2]**.

In cellulose processing, co-solvents and anti-solvents are used. Co-solvents are used to reduce the viscosity of cellulose solutions and lower production costs by diluting the more expensive solvent. Anti-solvents are liquids that bring the dissolved cellulose out of solution to form the cellulosic material, termed wet casting. A Leeds-led project using nuclear magnetic resonance spectroscopy was carried out to investigate ionic liquids combined with the co-solvent dimethyl sulfoxide (DMSO) and the anti-solvent water. This work mapped out important macroscopic properties such as viscosity and discovered how they depend on composition. The result of this joint work formed two publications, both with co-authors from Mines ParisTech **[3,4]**.

Futamura holds the intellectual property for a range of ionic liquids. A study carried out at the University of Leeds into these solvents revealed how their macroscopic properties depend on the chemical architecture. This generated an article co-authored with Futamura **[5],** which concluded that increasing the mass of the anion counterintuitively decreased the solution viscosity. This important finding is a central parameter for processing.

Fundamental understanding of the "Viscose" production steeping process [ii]
Futamura uses the "Viscose" process to manufacture cellophane. An NMR study was carried out at the University of Leeds on "Viscose" solutions from their UK plant, as part of a Royal Society Industry Fellowship. In this confidential study, NMR followed the reactions taking place in the "steeping" stage, allowing Futamura to optimise the balance of chemicals used in their process. As is typical of industrial work, this study is not in the public domain.

Understanding different behaviour of pulps from diverse sources [ii, iv, v, vi]
The wood pulp used to make cellophane consists of cellulose and a variety of other polysaccharides, depending on the source of wood used. A collaboration between the Leeds School of Physics and Astronomy and the Leeds Faculty of Biological Sciences, studying the interactions between cellulose and other biopolymers, found strong interactions between these biomolecules at certain stoichiometric ratios **[6]**. This work mapped out the mechanical properties of films made from these materials, which is essential information used in explaining the manufacturing problems at the Futamura US production site. Films produced from different wood pulp sources were breaking on the production plant. This was due to the different polysaccharide compositions, which could then be corrected for by the addition of "waste" hemicelluloses to the feedstock.

3. References to the research (indicative maximum of six references)

[1] **Ries, ME**; Radhi, A; Green, SM; Moffat, J; Budtova, T, Microscopic and Macroscopic Properties of Carbohydrate Solutions in the Ionic Liquid 1-Ethyl-3-methyl-imidazolium Acetate, Journal of Physical Chemistry B, 122, 8763, 2018.
Studies the dissolution mechanism of carbohydrates by an ionic liquid and reveals how the microscopic and macroscopic properties are affected by the solute. ***[ii, iii]***

[2] **Ries, ME**; Radhi, A; Keating, AS; Parker, O; Budtova, T, Diffusion of 1-Ethyl-3-methyl-imidazolium Acetate in Glucose, Cellobiose, and Cellulose Solutions, Biomacromolecules 15, 609, 2014.
Studies how the diffusion of ions is affected by the presence of a carbohydrate, this reveals how many ions are needed to dissolve an anhydroglucose unit. ***[ii]***

[3] Hall, CA; Le, KA; Rudaz, C; Radhi, A; Lovell, CS; Damion, RA; Budtova, T; **Ries, ME,** Macroscopic and Microscopic Study of 1-Ethyl-3-methyl-imidazolium Acetate-Water Mixtures Journal of Physical Chemistry B, 116, 12810, 2012.
Studies the effect of water, a cellulose anti-solvent, on the properties of an ionic liquid. ***[i]***

[4] Radhi, A; Le, KA; **Ries, ME;** Budtova, T, Macroscopic and Microscopic Study of 1-Ethyl-3-methyl-imidazolium Acetate-DMSO Mixtures Journal of Physical Chemistry B, 119, 1633, 2015.
Studies the effect of DMSO, a cellulose co-solvent, on the properties of an ionic liquid. ***[ii]***

[5] Green, SM**; Ries, ME**; Moffat, J; Budtova, T, NMR and Rheological Study of Anion Size Influence on the Properties of Two Imidazolium based Ionic Liquids Scientific Reports, 7, 8968, 2017.
Studies a variety of ionic liquids designed by Futamura for the dissolution of cellulose. ***[ii, iii]***

[6] Abou-Saleh, RH; Hernandez-Gomez, MC; Amsbury, S; Paniagua, C; Bourdon, M; Miyashima, S; Helariutta, Y; Fuller, M; Budtova, T; Connell, SD; **Ries, ME**; Benitez-Alfonso, Y, Interactions between callose and cellulose revealed through the analysis of biopolymer mixtures Nature Communications, vol. 9, 4538, 2018.
Studies the interactions between callose and cellulose and finds large differences in the mixture properties from the predictions of simple mixing rules. ***[ii, v, vi]***

Grants

[i] 2010 Engineering and Physical Science Research Council (EPSRC) Collaboration Fund EP/I500286/1 £10.5k, Dr Asanah Radhi 2 months.
[ii] 2013-2017 Royal Society Industry Fellowship (Grant number: IF120090) £113k + £40k from industry, Prof ME Ries.
[iii] 2013- 2017 EPSRC Case Award EP/I501495/1 (with Futamura) £67k + £23k from industry, Stephen Green.
[iv] 2014-2018 Internationally Funded PhD from Libya £75k, Wafa Ezzawam.
[v] 2015-2016 EPSRC Grant with Faculty of Biological Sciences EP/M027740/1 (FBS) £100k, Dr Candelas Paniagua.
[vi] 2016-2021 Leverhulme Trust with Faculty of Biological Sciences £353k, Dr Candelas Paniagua, Dr Radwa Abou-Saleh, Dr Sam Amsbury.

4. Details of the impact (indicative maximum 750 words)

The "Viscose" process and steeping

A long term and highly productive collaboration has been established between the University of Leeds and Futamura, the world's largest producer of cellulosic films with 65% of the market share and an annual turnover of USD783,000,000, through a wide variety of funding sources **[i - vi]**. Currently, 60% of Futamura's market share is NatureFlex™ film, a sustainable food packaging made using the Leeds process. Futamura aims to increase this to 90% of its production by 2025.

Futamura state in their supporting 2021 letter **[A]** that the Leeds support to develop a more environmentally friendly process for NatureFlex™ cellulose film (as a replacement for oil-based polymeric films) has impacted all their products. The letter describes how: *"The work we conducted'* with Leeds *'studying the effect of steeping caustic concentration on alkali cellulose reactivity has*

helped us to drive significantly improved process efficiency. The work has already had a significant impact on our industrial process."

Further, *the changes we have implemented to the process have contributed to significant and measurable increases in our process efficiency, by ~2%. The packaging film market is a highly competitive and price-sensitive market, where we find ourselves competing with significantly cheaper polyolefin-based plastic materials. Despite the "green" credentials of our Naturflex™ film range we often find the price can be prohibitive to people selecting our product over a plastic alternative, especially in cases where the plastic material matches the technical performance of our product. We made a decision some years ago to resist increasing the sales price of our product, and instead focus on driving internal efficiencies to help maintain or even grow our industrial margin, whilst maintaining sales volumes. The cost efficiency of the process achieved equates to an annual cost saving of £1.3M.* **[A]**

Customer testimonials

In their current brochure introducing customers to the NatureFlex™ film **[B]**, and on their website **[C]**, Futamura catalogue a number of recent customer testimonials on moving to NatureFlex™ cellulose film from oil-based polymer films, where a ***cost effective 'green'*** home compostable packing film has proved a significant customer selling point. Exemplar testimonials from end-users include:

- The newly launched **vegan Galaxy chocolate** in 2019 includes this statement on the back of the product: "*The outer card sleeve is widely recyclable. The clear NatureFlex™ film inside is made with wood fibre and is compostable within a few months and is suitable for garden and food waste collections*."
- **Ecoplaza** comment: "*We have found an environmentally alternative to plastic packaging, in the form of innovative compostable biomaterials*."
- **Teapigs**, teabag manufacturers, comment: "*Looks like plastic, but made from wood pulp, genius! Suitable for home compost*. **[B]**" "*Our tea sachets are home compostable and breakdown with 50 days"* **[B].** The company goes into more detail on their website **[D]**: "*until six months ago, we used a polypropylene plastic bag to hold our tea temples. However, knowing there must be a greener option, we did some research, and hey-presto, ALL our tea temples are now packed into* NatureFlex™ *- a fab material made from wood pulp that is fully compostable at home."*
- **Stoats**, oat bar manufacturers, comment: "*Green isn't just our favourite colour, it's our mind-set. (..) And we wrap our bars in biodegradable* NatureFlex™ *film, suitable for home composting."*

In a recent press release (27 February 2020) **[E]** Futamura announced that: "*Sustainable vegan chocolate producer, the **nu company**, has chosen Futamura's renewable and compostable* NatureFlex™ *packaging films for two of its latest vegan chocolate ranges of the brand 'nucao*'. Further, "*German Food Start-up the nu company, being eager to find a working alternative to conventional plastic packaging, opted for* NatureFlex™ *compostable cellulose films. These films are manufactured from responsibly sourced wood pulp and …. are certified for home composting. NatureFlex™ offers outstanding technical performance including high barrier to gases, moisture and oils, whilst offering renewability and an end-of-life option that diverts waste from landfill; it can go straight in the garden compost bin!"*

In a joint press release (April 2020) **[F]** Futamura and **Samova**, manufacturer of compostable tea sachet envelopes, in announced that: "*German Tea Company, Samova, has chosen Futamura's renewable and compostable* NatureFlex™ *films for its tea sachet envelopes.."* Esin Rager, Founder of Samova described this innovation **[F]**. "*After a long search, we are very pleased to have finally found a material,* NatureFlex™*, which meets our highest demands for environmentally responsible and a circular economy, while also offering even better product safety.*"

In 2019, **Selfridges [G,H]** announced that: "*This Christmas Selfridges is launching a sustainable alternative to plastic packaging across its own brand range of festive food using innovative home compostable cellulose film NatureFlex*™." Daniella Vega, Selfridges' Director of Sustainability, says: "*We know our customers share our concern for the environment and we're continually looking at ways in which we can address the sustainability of our products. This includes reducing our plastic use and introducing more sustainable products and packaging. NatureFlex*™ *is an incredible alternative; it looks just like plastic but can be easily popped into the food waste bin or home compost after use*."

In summary, Futamura's 2021 letter **[A]** states: "*A 2% improvement in process efficiency has a measurable and significant impact on our margin & profitability. The cost efficiency of the process achieved equates to an annual cost saving of £1.3M. This work has also enabled us to drive a more environmentally friendly process, by reducing the chemical burden of the process. In short a gain of 2% is quite rare, when comparing against other CI type work programmes, and this work has helped us to deliver a sustainable business performance".*

5. Sources to corroborate the impact (indicative maximum of 10 references)

[A] Letter from Futamura, New Technology Group Manager, Futamura Chemical UK Ltd (February 2021).

[B] Futamura's 'Introduction to NatureFlex' document including a number of customer testimonials.

[C] NatureFlex website with a number of customer testimonials for cellulose film https://www.natureflex.com/uk/testimonials-uk/

[D] Article on the Teapigs website https://www.teapigs.co.uk/blogs/news/how-teapigs-have-moved-away-from-excess-plastic-packaging

[E] Press release by the nu company (27 February 2020) https://news.unipack.ru/eng/79830/

[F] Press release by Samova (April 2020) https://press.unipack.ru/eng/79522/

[G] Press release by Selfridges (June 2019).

[H] Article in Bioplastics News, 'Selfridges Goes for Futamura NatureFlex' (April 2020) https://bioplasticsnews.com/2020/04/27/selfridges-bioplastic-compostable-natureflex-futamura/