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Feasibility of lactic acid production from cereal milling residues in the UK

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List of abbreviations

- cPET: Crystalline Polyethylene Terephthalate
- DLA: D- lactic acid
- LA: Lactic acid
- LLA: L-lactic acid
- PET: Polypropylene
- PDLA: Polylactic acid made of DLA monomers
- PLA: Polylactic acid
- PLLA: Polylactic acid made of LLA monomers
- PP: Polyethylene Terephthalate
- sc: Stereo complexed
- tpa: Tonnes per annum
- 1G: First generation
- 2G: Second generation
- 3G: Third generation

Aim and scope of this study

The aim of the study was to determine if UK derived cereal residues and associated products provide an economic and technically feasible route to produce lactic acid (LA) at sufficient scale to provide the raw material for a UK world-scale (30,000 tpa) polylactic acid (PLA) plant.

Attention is given to the current state of the EU bioplastics and PLA market, processes required for conversion of cellulosic feedstocks to LA, availability of residues from wheat, oat and barley milling operations and value chain development prospects. Many of the study parameters were based on earlier results (Reineck, 2008), for which feasibility of a 30,000tpa UK PLA plant using starch-based feedstocks was assessed.

Executive Summary

The production of poly-lactic acid (PLA), a biodegradable and bio-based polymer, from cereal milling residues (wheat, barley, oat, starch) at large scale represents a market opportunity in the UK provided five crucial parameters are in place: 1) a supportive political framework, 2) market attractiveness, 3) technological feasibility, 4) feedstock availability, and 5) economic viability. In consideration of these parameters, this study assessed the potential for developing a UK lactic acid (LA) plant capable of providing sufficient LA for a 30,000 tonnes per year (tpa) PLA plant, utilising cereal milling residues as feedstock.

A review of the EU political framework for bio-based products and evaluation of the existing and near-future PLA market demonstrated that a supportive environment exists in the UK for large-scale production of LA. Brand owners are continually seeking to demonstrate their products are environmentally sustainable through introducing bio-based ingredients and biobased packaging, while consumers are increasingly keen to support environmentally-friendly products. In accordance, the PLA market in the EU is expected to grow 13% by 2025, with demand expected to outstrip supply. Meanwhile, growing competition between food markets and industry for traditional agricultural crops have meant that utilisation of non-food cellulosic feedstock is becoming increasingly attractive. As a consequence, there can be expected to be high future demand in the EU for LA derived from cellulosic biomass.

To date, there has been little activity in developing LA from milling by-products. However, by analysing the technologies and production pathways needed for production of LA and PLA from cereal residues, we found numerous technologies that have been commercialised for LA and PLA production from cellulosic biomass and several processes that have focused on the conversion of milling residues to fermentable sugars, thus indicating technical feasibility. In consideration of these processes, a potential production pathway for conversion of milling residues to LA was constructed.

This study further demonstrates that there is good potential in the UK for developing value chains based on production of LA from cereal milling residues – notably wheat bran/middlings and oat husks/hulls – in sufficient volumes to supply a 37,500 tpa LA plant (equivalent to the feedstock requirement of a 30,000 tpa PLA). Most encouragingly, it was discovered that a single oat milling facility exists in the UK which alone produces almost sufficient residue volumes to supply a facility of this scale.

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The main findings of the study are summarised in Summary Table 1.

Summary Table 1. Cumulative results scheme from feasibility analysis for production of PLA from cereal milling residues in UK

	Waste wheat milled grain	Wheat bran	Wheat germ	Wheat middlings (wheatfeed)	Residual starch	Oat bran	Oat husks/ hulls
Abundance/ availability	High	High	Low	High	Low	Low	High
Total carbohydrate content (wet basis)	80% (endosperm)	63%	47%	63%	Up to 90%	83%	67%
Competitive uses	Industrial (periodic)	Food/ feed	Food/ feed	Feed	Food/ industrial	Food/ feed	Feed/ energy
Technological feasibility/ maturity	1G/ C6 Fermentation	2G/ C6+C5 Ferm	1G/ C6+C5 Ferm	2G/ C6+C5 Ferm	1G/ C6 Ferm	2G/ C6+C5 Ferm	2G/ C6+C5 Ferm
Economic competitiveness	Good	Good	N/A	Good	Good	N/A	Good
Water content (transportation/ storage)	20%	10%	14%	10%	Very low	12%	5%
Overall attractiveness	Low	High	Low	High	Low	Low	High

Ultimately, this study shows that there is a strong and growing market for LA and PLA, the production pathways from cellulosic feedstocks are becoming commercialised and therein lies potential to develop value chains from cereal milling residues in the UK. However, little work has been done to date regarding the manufacture of LA from milling residues, and therefore this opportunity can be expected to have significant risks, especially in regards to technical feasibility.

1. Bio-based plastics market

1.1 The EU Bioeconomy

The oil-based economy provides the foundation for modern society; with fossil resources meeting much of our energy, chemical and material needs. However, growing concerns over climate change and oil price volatility has stimulated great interest within Europe of facilitating a transition to a bioeconomy. According to the European Commission, the bioeconomy *"integrates the full range of natural and renewable biological resources - land and sea resources, biodiversity and biological materials (plant, animal and microbial), through to the processing and the consumption of these bio-resources⁷¹. The bioeconomy has the potential to provide sustainable and renewable replacements for all commodities and products currently derived from fossil fuels.*

The European Union has already taken an important step towards establishing an era of low-carbon, resource efficient, sustainable and inclusive economic growth in Europe with the decision to formulate a bioeconomy strategy and action plan². As the European Commission rightly acknowledges, the bioeconomy has the potential to contribute not only to tackle global problems such as climate change, CO₂ emissions reduction and sustainable economic growth, but also to furthering research and innovation excellence in Europe, and more sustainable agricultural policies linked to regional and rural development.

The commitment of the EU to developing a sustainable bioeconomy is further evidenced by:

- A reform of the European Common Agricultural Policy (CAP). The reform will
 result in an investment of over €100 billion between 2014 and 2020 for ensuring that
 EU agriculture 'meets the challenges presented by soil and water quality, biodiversity
 and climate change'³.
- Development of the Horizon 2020 research and innovation program. The initiative will provide nearly €80 million funding between 2014 and 2020 to drive economic growth and create jobs within the EU. A bio-based industry PPP (BRIDGE 2020)⁴ has been established as part of the program and will receive investments of

¹ <u>http://ec.europa.eu/research/consultations/bioeconomy/introductory_paper.pdf</u>

² European Commission. 2012. Innovating for Sustainable Growth: A Bioeconomy for Europe.

³ <u>http://europa.eu/rapid/press-release_IP-13-613_en.htm</u>

⁴ <u>Bio-based Industries Consortium. 2013. Bio-based and Renewable Industries for Development and Growth in</u> <u>Europe.</u>

€1 billion from the Commission and €2.8 billion from industry to develop new value chains, optimise feedstock usage and bring novel technologies to maturity.

Within the bioeconomy, bio-based plastics have the potential to deliver many of the proposed benefits of the bioeconomy. Bio-based plastics are simply plastics derived from biomass instead of oil. Some bio-based plastics are structurally and functionally identical to fossil-based plastics (e.g. Bio-PET), while other bio-based plastics may only be derived from biological resources (e.g. polylactic acid). Bio-based plastics have many advantages over traditional plastics including the potential to have a lower carbon footprint, reducing dependency on fossil resources and opportunities for developing products with novel characteristics (e.g. compostability).

1.2 Market drivers for bio-based plastics

Whilst there is a realisation from policy-makers that the economy will be dependent on fossilderived products for some considerable time, there is also a drive to ensure that replacement technologies are incentivised in order to close this time frame and protect the market from the increasing volatility of fossil prices.

There are many other benefits of bio-based plastics over fossil-plastics. The branding opportunity afforded by "green" and "environmentally-friendly" products is one of the most attractive benefits of bio-based plastics. A 2013 survey undertaken by the European Commission of 25,000 citizens across the EU-27 assessed the attitudes of European consumers towards green markets (European Commission, 2013). The survey identified that more than 80% of EU citizens surveyed currently buy environmentally-friendly products while 77% of citizens are willing to pay a premium for such products, highlighting the importance of green marketing on consumer behaviour.

Moreover, the main brands regularly seek extra functionality in their material chain in order to make product improvements. Bio-based plastics offer opportunity for developing such novel properties. The most notable development to date is the development of biodegradable and compostable bio-based plastics. While the majority of fossil-based plastics will persist in the environment almost indefinitely, many bio-based plastics (such as PLA) will decompose naturally through hydrolysis or photodecomposition. Such bio-based plastics, therefore, have more advantageous end-of-life options than fossil plastics; biobased plastics can be used in industrial composting facilities or potentially anaerobic digesters. Bio-based plastics may, in the future, be valorised higher than fossil plastics on account of the incentives that exist for the use of biomass in energy generation. However, a

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greater degree of market penetration and improved methods for measuring the biogenic content of waste streams are required before this benefit can be fully realised.

A comprehensive list of the existing key market drivers for sustainable plastics is shown in Table 1, with those most relevant to the Polylactic acid (PLA) market in bold

Table 1. Key Market Drivers for Sustainable Plastics

Positive Influencing Factors (+)	Negative Influencing Factors (-)		
Sustainability of I	Feedstock supply		
+ Recycled (conserves fossil and bio resources)	- Fossil (supply available for plastics long term)		
+ Renewable (conserves fossil resources)			
+ Renewable (photosynthesis consumes CO ₂)			
+ Carbon sink (conversion to plastic)			
Sustainability of E	nd-of-Life Options		
Biodegradability	(compostability)		
+ Diversion from landfill	- Generates greenhouse gases (destroys carbon sink)		
+ Conversion to fertiliser and fuel	- Option unavailable to many end users		
Recycl	ability		
+ Diversion from landfill	- Limited to ~ 50% of total (back to plastic)		
+ Carbon sink (back to plastic)	- Generates greenhouse gases ("energy recovery /		
+ Option available to most end users	recycling" destroys carbon sink)		
	- Recycling technology unavailable, or facilities		
	unavailable or inaccessible		
	- Contaminates other recycling streams		
Sustainability of P	roduction Process		
+ Recycled polymer (conserves energy / reduces greenhouse gas emissions)	 Virgin fossil polymer (consumes energy / generates greenhouse gases) 		
+ (Bio)Renewable polymer (use of renewable energy reduces greenhouse gas emissions)	 Virgin (Bio)Renewable polymer (consumes energy / generates greenhouse gases) 		
Polymer Performance (properties)			
+ Improving performance based on polymer design and	- General stakeholder belief bio-based polymers have		
additive packages	inherent performance issues		
Price (influenced b	y production cost)		
+ Greater independence from crude oil price increases	- Rising oil price driving up cereal price		
and fluctuations	- Cost of fertiliser and farm fuel linked to oil price		
	- Rising cereal price drives up bio-based polymer prices		
Regulation (Legislation)		
+ Bans on plastic bags (see Section 3.2. B. b.) with	- Bans which apply to plastic bags of all kinds.		
requirement to use biodegradable plastic	- Bag charges which apply to all plastic bags		
+ In Germany, biodegradable packaging products do			
not have to pay the Green Dot (recycling tax.)			
+ Others that favour recycled or bio-based plastic			
Commercial	l availability		
Available in commercial quantities	(i.e. not pilot plant or development)		
+ Production capacity keeps pace with demand	- Production capacity lags far behind demand		
	- No announcement of new capacity		
Location of production plant			
+ non-GM source (i.e. European)	- GM is an issue for polymers such as PLA produced		
	from starch in USA		

1.3 Market forecasts

European plastics demand is growing in line with gross domestic product (GDP) whilst packaging – a significant market for plastics – is growing faster than GDP.

According to *European Bioplastics*, global production capacity of bio-based plastics was around 1.15 million tonnes in 2011 (Figure 1) with Europe responsible for 18.5% of capacity⁵ (Figure 2). *European Bioplastics* further predict global production capacity for bio-based plastics to increase significantly over the next few years, increasing to around 6 million tonnes by 2016 and for Europe's market share to drop over this period to 4.9% (although capacity is still expected to increase from around 200,000 tonnes to just over 280,000 tonnes).



Figure 1. Current and projected global production capacity of bio-based plastics⁵

⁵ European bioplastics. Bioplastics: Facts and Figures.





Demand for bio-based plastics in Europe is expected to grow significantly in the long term on account of the positive branding opportunities afforded by environmentally-friendly products and the expected increase in price of comparative fossil-derived materials. Additional opportunities may be created through regulatory interventions such as compostable bags being exempt of a ban or charges placed on plastic bags. Assuming an average GDP growth of 2.5% annually for the next 15 years, the European plastics market is projected to grow from around 48 million tons in 2010 to over 70 million tons by 2025 (Reineck, 2011). By further assuming that 20% of the plastic supply chain will be bio-based by 2030 (the industry consensus view is 20–30%), European demand for bio-based plastics is estimated to be 9.25 million tons in 2025, representing a total market share of 13%. To meet this demand will require a substantial increase in existing production capacity.

1.4 Summary

Due to climate change and oil price volatility concerns, EU policies and regulations have been developed to incentivise the use of bio-based feedstocks in the fuel, chemical and energy markets. Supported by strategies and regulations that prioritise bio-based products, research and development funding schemes and investment incentives have supported the commercialisation of innovative technologies making use of biological technologies and facilitated market penetration of bio-based plastics.

Meanwhile, the importance of environmental impacts (such as carbon footprinting) in influencing consumer choices continues to grow and has increased demand for sustainable products. Brand owners are, therefore, gradually seeking to improve the sustainability of

their products by introducing bio-based ingredients in their supply chains. As a result, the EU bio-based plastics market is expected to grow significantly in the near future.

2. Lactic acid production and markets

2.1 Lactic acid

Lactic acid (LA) is a platform chemical that is commercially manufactured from sugars through large-scale fermentation. LA is most commonly produced from glucose, derived from the processing of starch from cereal crops such as corn or wheat. However, there are increasing concerns over the utilisation of traditional agriculture crops in non-food applications due to potential indirect impacts on food prices and land use change (e.g. deforestation). This is particularly evident in the biofuels industry, where investment in the EU has stalled due to uncertainties over how such impacts should be addressed in policy. Given the growing importance of sustainability in brand development, there is significant interest in the utilisation of lignocellulosic feedstocks such as straw, wood and crop residues for production of renewable chemicals in order to avoid potential negative interactions with food markets. As it is feasible to produce LA by fermentation of both C6 and C5 sugars, therein lies potential to utilise lignocellulosic biomass for commercial production of LA.

According to Bozell and Petersen (2010), LA is one of the top ten most attractive bio-based chemicals for production by sugar biorefineries. LA can be converted into a multitude of chemicals and materials such as PLA, ethylene glycol, lactate esters, and acrylic acid/esters (Figure 3).





Historically, the major markets for LA have been food, pharmaceuticals and cosmetics. Approximately 70% of current production is used in the food industry, much of which is used in the production of cheese and yoghurt (Martinez *et al.*, 2013). LA can further be used to provide acidity in food and beverages while lactate derivatives are often used as flavourings. In the pharmaceutical industry, LA can be used in a variety of applications including blood coagulants, topical wart preparations and anti-inflammatory drugs (Taskila and Ojamo, 2013). However, it is the polylactic acid (PLA) market that can be expected to be the primary driver of LA market growth in the future.

2.2 European PLA market

PLA is a 100% bio-based polymer produced through the polycondensation of LA that offers unique functionality (the chemistry of PLA synthesis is described in Appendix 1). PLA was first discovered in the 1930s but was only commercialised as an industrial polymer in the last 15 years.

PLA has emerged as the world's leading niche bioplastic, with global sales of over 100,000 tpa, and is expected to remain so for at least the next 10 to 15 years. The Nova Institute has indicated that some 25 companies have developed a cumulative production capacity of more than 180,000 tpa of PLA⁶. The largest producer is NatureWorks, in the US, with a capacity of 140,000 tpa. Nova has further estimated that global production capacity will increase to 800,000 tpa by 2020. By this time, Europe could be responsible for between 140,000 tpa and 180,000 tpa of supply.

An earlier market study commissioned by the NNFCC, forecasted total EU PLA demand to grow from the current ~30 ktpa to 184 ktpa in 2015, reaching 650 ktpa in 2025 based on forecast annual growth from 2015 to 2025 of 13%⁷. Accounting for supply and demand estimates, this results in a market gap of between 166 and 206 thousand tpa PLA by 2020 (Table 2).

⁶ <u>Nova Institute. 2012. Bio-based Polymers in the World - Capacities, Production and Applications: Status Quo</u> and Trends towards 2020.

⁷NNFCC. 2008. Techno-Economic Assessment of Potential PLA Manufacturing Plant in UK.

Table 2. Forecasted supply and demand of PLA in Europe

EU PLA Market (thousand tpa)	2010	2015	2020	2025
Estimated Demand	30	184	346	650
Estimated Supply	30	45-60	140-180	280-420
Market Gap	-	124-139	166-206	230-370

The largest opportunity for PLA market development in the near future is in **thermoformed sheet**, mainly used for rigid food packaging trays and yoghurt pots etc., which is projected to account for over 40% of demand for PLA in the EU in 2015. The oven-ready meal tray is a target application, to replace crystalline polyethylene terephthalate (cPET).

PLA demand for **injection moulded rigid packaging** is projected at 16 thousand tons (kt). On this basis, total demand for PLA for rigid packaging is forecast to be 91 kt or close to 50% of the market in 2015. A target application is the microwavable ready-made soup pot, replacing PP.

Clear Film (including biaxially oriented "BOPLA" film) is projected to account for 18 kt PLA in 2015, mainly for wrapping fresh produce, convenience foods and short shelf-life items.

Co-Blended Biopolymer (CBP) film is a major market opportunity for PLA, projected to consume over 18% of PLA in Europe in 2015. CBP utilises hard, rigid PLA to provide stiffness and strength in a blend with soft, flexible starch polymer (TSP) or polyester (PBAT). The main applications for CBP films are in the non-food area, including shrink-wrap for secondary packaging, compostable waste bags and biodegradable shopping bags.

General purpose (GP) injection moulding for non-packaging applications is projected to become significant, as several compounders are developing formulations.

Fibre is expected to grow strongly from a small base, notably in carpet and textiles, as is Paperboard Coating, with hot drinks cups as the main application.

3D Printing is only a nascent industry but with potential to grow substantially in the future. PLA is one of the most suitable products to use as a print material on account that it sets quickly and is resistant to warping.

The rigid packaging applications are expected to require formulated PLA. In the remaining segments, customers buy polymer which they then formulate to produce film, CBP and injection moulding compounds.

As a result of the presence of leading retailers and major packaging producers in this sophisticated marketplace, the UK is expected to account for 10–20% of the EU total consumption and there is an excellent opportunity for a UK plant to capture 20% of the EU market.

2.3 PLA classifications

PLA is a polymer produced by the polycondensation of lactic acid (LA). LA is a hydroxycarboxylic acid that can react with itself to form a dimer (known as a lactide) and a molecule of water. The chemistry of PLA production is described in more detail in Appendix 1.

Due to the chiral nature of lactic acid, several forms of PLA exist, each one having distinctive characteristics, properties, potential market applications, and potential market demand and price. PLA can be produced as homopolymers using single enantiomers of lactic acid i.e. PLLA contains only L-lactic acid and PDLA contains only D-lactic acid. PLA can also be produced as a random polymer of L and D lactic acid.

Commercially Available PLA

Specialised grades of PLA are currently produced in very small volumes and are generally targeted at the high value biomedical engineering sector. Most of the PLA currently available on the market is produced by Natureworks at its Blair Nebraska plant, and can be characterised as first generation PLA.

First Generation PLA is a random copolymer of LLA with typically 3–6% DLA, which for practical purposes does not crystallise. As this type of PLA is practically amorphous, it has properties typical of an amorphous material: glassy, brittle, etc. Fibre and film (biaxial oriented poly(lactic acid)) based on drawing and stretching, are natural applications for this material. First general PLA is, however, limited in its application areas.

Second Generation PLA is PLLA, which provides improved properties when used alone or in blends with minor amounts of PDLA to form stereo complex-PLA. PDLA and PLLA form a highly regular stereocomplex with increased crystallinity, melting temperatures and heat deflection temperature which can be increased by physically blending PLLA with low concentrations of 3–10% of PDLA.

Third Generation PLA is stereo complex-PLA based on blends of equal quantities of PLLA and PDLA. With 50:50 blends the temperature stability is maximised, providing superior

polymer properties. Commercialisation of these products on a large scale would depend on development of the market and large scale production of DLA.

All PLA produced by Natureworks is shipped in the form of beads (pellets) to customers which convert the beads into film, sheet, fibre, injection mouldings, etc. In this respect, Natureworks and PLA are typical of the plastics industry, whereby resin (polymer) producers do not produce rods or sheets, as this is their customer's business.

The characteristics of PLA and potential for developing new PLA grades are key to understanding the market potential for PLA. These are described in detail in Appendix 2. The polymer is biorenewable, offers high tensile strength and low density, is biodegradable, demonstrates compostability in accordance with EN13432 and is easily embossed (or printed onto). PLA also offers attractive end-of-life options when considered against the 'waste hierarchy' introduced by the EU Waste Framework Directive⁸. Many waste PLA streams can be readily processed through mechanical and chemical recycling. Meanwhile, PLA film and fibre that is not easily recycled can be composted in accordance with EN13432 or can be converted to renewable energy by straight combustion, gasification or pyrolysis.

Second generation PLA has the properties required to replace oil-based plastics such as polypropylene (PP) and polyethylene terephthalate (PET) in high value packaging applications. Growth of demand for second generation PLA as a replacement for oil-based plastics will be driven by PLA's combination of 100% bio-based content and its unique functionality.

Market discussions in the UK and EU confirm strong interest in PLA with improved functionality – especially high temperature resistance properties offered by 2G and 3G PLA – and that would be available in commercial quantities from an EU (non-GM) source, as a replacement for oil-based plastics.

2.4 Competitor analysis

There are approximately 25 companies worldwide involved with the production and sales of PLA. An overview of some of the major players in the industry is shown in Table 3, indicating existing assets and interest in technologies that utilise lignocellulosic feedstocks.

⁸ European Commission. 2008. Directive 2008/98/EC. Waste Framework Directive.

There are also a number of other companies involved in the manufacture of the upstream LA for both food and industrial markets. These include Archer Daniels Midland, Galactic and several Chinese companies such as B&G and CCA Biochemical Company.

NatureWorks is by far the largest manufacturer of PLA in the world, with its commercial facility in Nebraska, USA responsible for around 75% of global capacity. However, many PLA pilot and demonstration plants have been, or are being, developed around Europe. This includes Plaxica's and TMO's facilities in the UK.

Cellulac has made notable progress with the development of LA from non-food sources such as straw, spent brewer's grains, DDGS and lactose. Corbion-purac and Plaxica have also demonstrated an ambition to optimise processes for utilising cellulosic feedstocks.

Company	Existing assets	Interest in lignocellulosic technologies
Corbion-Purac	 Developed technology for production of LLA and DLA using "GMO-free" feedstocks exclusively 10 lactic acid production plants across USA, the Netherlands, Spain and Brazil 	 Actively involved in various development programs to develop PLA from non-food, cellulosic feedstocks. Committed to developing a PLA pilot facility that utilises by-product streams as feedstock
Uhde Inventa- Fischer	 Developed the PLA<i>neo®</i> process for developing PLA from polymer grade LLA Mini-plant in Berlin, Germany and 500 tpa pilot plant in Guben, Germany 	
Plaxica	 Developed the Versalac process for producing LLA and DLA from sugars using a low cost chemical route Developed Optipure process for converting LLA to DLA Demonstration plant in Wilton, UK 	 Versalac technology well suited to cellulosic sugars – converts both C5 and C6 sugars and tolerant to impurities
Cellulac	 Developed proprietary technology for converting cellulosic biomass to LLA, DLA, sodium lactate and ethyl lactate using the 'SoniqueFlo' steam explosion process, enzyme pretreatment and bacterial fermentation 137 patents granted and pending Acquired brewery in Dundalk, Ireland to produce pure LLA and DLA from agricultural and brewery waste; Phase 1 – 20,000 tpa Phase 2 – 100.000 tpa 	All activities pertain to cellulosic technologies

Table 3.	LA	competitor	comparison
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Company	Existing assets	Interest in lignocellulosic
		technologies
Myriant	Developed technology to produce optically pure DLA and LLA using a one-step biosynthetic process that has been licensed to Purac	
NatureWorks	 Produces a range of Ingeo[™] PLA grades from plant sugars using fermentation for LA synthesis Operating a140,000 tpa capacity PLA plant in Nebraska, USA and planning a new facility in Thailand 	Ambitions to develop Ingeo process to use cellulosic sugars in future
TMO Renewables	 Patented bacterial (<i>Geobacillus</i>) platform that converts sugars into LA Upgrading a demonstration unit in Guildford, UK to produce 3,000 tpa non-GM PLA 	 Experience in engineering bacteria for fermentation of cellulosic sugars and ambition to produce PLA from non-food feedstocks

2.5 Summary

Significant growth is expected in both the global and EU PLA market in the near future with new varieties of PLA with improved properties undergoing development. This can be expected to increase the number of available applications of the polymer, and thus the overall PLA market, while also creating further opportunities for developing improved value chains. Meanwhile, the growing importance for brand owners of applying sustainable practices in product development can be expected to further increase the attractiveness of utilising PLA over functionally similar fossil-based plastics.

The consequence is that there is a projected gap in supply and demand of PLA and in the EU, indicating a promising investment landscape for a LA plant in the UK. Previous efforts have demonstrated the viability of such a plant, utilising starch as feedstock (Reineck, 2008)

3. UK Grain Milling industry

3.1 Background

The UK grain milling industry is well established in the UK, with the flour milling sector alone consisting of over 30 companies – operating around 50 sites – with a combined annual turnover of around £1 billion⁹. The principle product of the industry is flour, utilised for food markets. However, the milling process often results in a wide variety of by-products/residues

⁹ http://www.nabim.org.uk/content/1/36/the-flour-milling-industry.html

that, while often used in low value markets, may also be used as a source of cellulosic sugar in industrial applications.

The milling industry uses a variety of different milling technologies that can broadly be split into two different processes: wet milling and dry milling.

Wet milling

Wet milling is the traditional process used for production of starch from wheat (or maize). Wet milling involves steeping the grain in water to soften the seed kernel followed by grinding and separation of the starch, gluten and fibre by filtration. Wet milling for starch production generates large quantities of co-products – primarily fibre (bran), sugar syrup (sweeteners) and gluten protein (germ) – the revenues of which are often critical to the economic viability of starch mills.

Dry milling

In the dry milling process, the entire grain is ground into a fine powder, known as meal, before being slurried through the addition of water.

Dry milling is a less versatile process than wet milling and results in the production of a lower amount of by-products. However, the process is frequently used in modern grain ethanol refineries where whole grain is milled before fermenting to ethanol as it results in higher efficiency with lower capital and operational costs. This process can be adjusted so that LA is the fermentation product instead of ethanol. The single by-product of an ethanol dry mill is distillers' dried grains with solubles (DDGS) which is typically sold as a high protein animal feed.

A variety of different technology options for milling also exist, including hammer milling, roller milling and pearling.

• **Hammer milling** is used in grains with low husk content. Hammer milling operates via grinding and friction, and is supported by a sieving device, that separates the product from the recycling stream. The process is capable of either wet or dry grinding.

- Roller milling is often used when grain has high bran content¹⁰. This process is typically used for flour production and results in the production of various side-product streams. In a typical dry mill, 70–80% of product is flour, with the remaining 20–30% consisting of bran, germ and a mixed stream of bran/flour/germ (middlings)¹¹.
- **Pearling** is an advanced cereal fractionation technology commercialised in the 1990s. In the process, the grain is de-branned by gradually separating outer layers (e.g. pericarp, aleurone etc.) from the endosperm. Each bran layer has unique composition and properties, and thus market value.

In addition to the various different processing options used by millers, a variety of different grains are also utilised. Three different cereal grains are commonly milled in the UK: wheat, oats and barley.

3.2 Wheat

UK production

Wheat is by far the most abundant cereal crop in the UK with yearly production volumes often exceeding 15 million tonnes. However, annual fluctuations in crop yield can often be large on account of weather conditions. For instance, 2012 crop yields were almost 15% lower than in 2011 due to extended periods of wet weather early in the year (Figure 4). This also had a negative impact on grain quality with 152,000 tonnes of wheat grain classed as 'out of specification' or supplied to ethanol and starch producers in 2012 (DEFRA, 2013).

¹⁰ The ethanol textbook. Chapter 2. D.R. Kelsall and T.P. Lyons

¹¹ <u>http://www.nabim.org.uk/content/1/60/flour-production---consumption.html</u>



Figure 4. Historical wheat yields and harvest volumes in the UK¹²

Broadly speaking, wheat grain can be split into two different varieties: milling wheat and feed wheat.

Milling wheat is high in protein e.g. >13% dry matter and is most frequently used for making bread flour in the UK. Approximately 6 million tonnes of milling wheat is processed by the UK milling industry every year.

Feed wheat is a low grade wheat variety that is most often used as animal feed. The grain is also used as a feedstock for ethanol production in the UK. Feed wheat is not regularly milled on account of its lower protein content.

Milling products and residues

Products

Flour

Flour is the principle product of wheat grain milling and contains the wheat endosperm. Approximately five million tonnes of flour is produced in the UK on an annual basis with the majority (around 80%) used in the food market for production of breads, cakes and other baked goods (Table 4). The remainder is used in industrial applications for production of starch and ethanol.

¹² FAOSTAT

Wheat milling product	Annual production ('000 tonnes		
Wheat mining product	2011	2012	
Total milled wheat	6123	6305	
Total flour produced (of which)	4858	5006	
- Total bread making flour (of which)	2846	2988	
- White bread flour	2416	2560	
- Brown bread flour	107	98	
- Wholemeal bread flour	323	330	
- Biscuit flour	597	567	
- Cake flour	115	115	
- Household flour	123	137	
- Food ingredients flour	172	190	
- Other flour (for starch and ethanol production)	1004	1009	

Table 4. Annual production of wheat flour in the UK^{13,14}

Starch

Wheat starch can be produced either directly via wet milling or from flour through dry milling. The process of starch production from wheat flour is presented in Figure 5.



Figure 5. Starch production from flour (Kamm et al., 2010)

¹³ <u>https://www.gov.uk/government/publications/cereal-usage</u>

¹⁴ https://www.gov.uk/government/publications/animal-feed-production

There are three varieties of starch produced in the flour making process: slurry, dry powder, and glucose syrup:

- 1. Starch slurry has only 30% starch content and is the poorest quality grade. On account of its low purity, it is expensive to store and transport.
- Dry starch is that which is most frequently manufactured and usually contains around 90% starch content.
- 3. Glucose syrup is a niche product that is produced via starch hydrolysis. It requires further saccharification steps (breaking the polymer down into smaller units) as well as concentration (removal of water).

Total consumption of starch in the EU was estimated at 10 million tonnes in 2011¹⁵. From the total volume consumed, 57% was used as a starch based sweetener in food applications, 23% as native starch and 20% as modified starch. By far the largest non-food application in the EU is within paper, board and corrugating industries, although the use of starch and starch derivatives for production of biopolymers like PLA is gaining interest.

While well-established markets for starch already exist in the EU, bio-based polymers offer large potential for developing improved value chains. However, utilisation of starch for production of LA in the UK has previously been assessed in Reineck (2008) and is therefore not considered any further in this study.

Residues

Wheat bran

Wheat bran is a cellulosic material containing moderately high amounts of C5 and C6 sugars, accounting for around 15% of the grain weight¹⁶. Almost 1 million tonnes of bran was produced in the UK in 2012. It has a low, but variable, selling price of between £56 and £185 per tonne (figures sourced from personal communication with an industry representative) and is often used either as an additive in wholegrain bread production or as an animal feed in wheat middlings.

Wheat germ

Wheat germ accounts for approximately 3% of the wheat grain and contains starch, proteins, vitamins and oils (Reineck, 2008). Germ is often mixed with bran during pearling and used in

¹⁵ <u>http://www.aaf-eu.org/european-starch-industry/</u>

¹⁶ <u>http://www.hgca.com/publications/documents/cropresearch/PR425_Final_Project_Report.pdf</u>

wholegrain bread production. The product is rather expensive at a selling price of around £560 per tonne, largely on account of its high protein content.

Wheat middlings

Wheat middlings are a by-product of the milling process and consist of a mixed stream of milling wheat residues composed of roughly 60% wheat bran, 25% wheat milling residues, and 15% screenings (figures sourced from personal communication with an industry representative). Middlings are a low value product with a selling price of approximately £142 per tonne. More than 800,000 tonnes are produced annually in the UK, most of which is used as an animal feed additive.

A summary of residue production from the UK milling of wheat is shown in Table 5.

Table 5. Annual production of wheat milling by-products for 2012^{17,18}

Wheat milling residue	Annual production ('000 tonnes)
Total wheat milled	6,305
Bran	946
Germ	189
Middlings	813*

*N.B. this value is should not be considered as additional to bran production, on account that much of the bran produced will enter middlings

3.3 Oats

UK production

Oat grains are highly nutritious, containing a very high protein and lipid content. The grain is typically used in porridge and muesli cereals, although it can also be ground into flour for production of oat bread, cakes and biscuits. A minor fraction is also used in cosmetics manufacturing. Production of oats in the UK is low in comparison to wheat, with annual yields usually between 600,000 and 800,000 tonnes (Figure 6).

¹⁷ <u>https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/263471/cerealuse-statsnotice-05dec13.pdf</u>

¹⁸ https://www.gov.uk/government/publications/animal-feed-production



Figure 6. Historical oat yields and harvest volumes in the UK¹⁹

Milling products and residues

Products

Oat flakes (rolled oats)

Oat flakes are the predominant product of oat milling in the UK. Cleaned groats, produced from de-hulling of the raw oat grain, are processed to produce flakes using large rollers. Flakes are less refined than flours and consist of the endosperm, germ and bran of the oat. Oat flakes are regularly used in food applications, notably breakfast cereals such as porridge, muesli and granola. Annual production in the UK is approximately 200,000 tonnes every year (Table 6).

Oat flour

Oat flour can be produced by de-branning of cleaned groats. The flour itself is usually free of bran, containing just the endosperm of the oat. Oat flour can be used in similar applications to wheat flour although is differentiated by its absence of gluten. Oat flour can therefore be marketed towards niche food markets such as gluten-free applications. Oat flour production in the UK is relatively low at just under 100,000 tonnes per annum (Table 6).

¹⁹ FAOSTAT

Table 6. Annual production of oat products in the UK²⁰

Oat milling products and by products	Annual production ('000 tonnes)		
	2011	2012	
Total milled Oat	472	475	
Gross production (of which)	304	296	
- Oat flakes and rolled flakes	200	202	
- Oat flour and other cuts (including animal feed and groats)	104	94	

Residues

Oat germ

Oat bran is a very nutritious milling co-product very rich in cellulose and hemicellulose. It is produced via de-branning of oat grains during the production of oat flour. On account of the low volumes of wheat flour produced in the UK, oat bran production is similarly low. Much like wheat bran, oat bran is used in food and feed applications.

Oat husks/hulls

Oat grains contain an outer material known as the husk (or hull). It is a cellulosic material rich in C6 and C5 sugars that comprises up to 40% of the grain by mass. During the milling of oats, this outer layer is removed by the de-hulling process. The most common use for oat husks is an animal feed. However, alternative uses include animal bedding, bioenergy (via combustion) and biofuel production. An estimated 182,000 tonnes of oat husks are produced in the UK every year.

A summary of residue production from the UK milling of oats is shown in Table 7.

Table 7. Annual production of oat milling by-products for 2012²¹

Wheat milling residue	Annual production ('000 tonnes)
Total oats milled	475
Bran	87
Germ	9
Husks/hulls	182

²⁰ <u>https://www.gov.uk/government/publications/cereal-usage</u>

²¹ <u>https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/263471/cerealuse-statsnotice-</u>05dec13.pdf

3.4 Barley

Barley is a typical monocotyledon grass species widely cultivated in the UK for its grain. Around 5 million tonnes of barley is grown in the UK every year, much of which is used either by the brewing industry for production of beer or as an animal feed. While barley is regularly milled as part of the brewing process, it is not milled for flour production; as a consequence there are no milling residues produced from barley in the UK, aside from postdistillation by-products. On account of this fact, barley is not considered further in this study.

3.5 Summary

Grain milling is a mature industry in the UK although margins are often small. Consequently, the value of by-products is often an important factor in ensuring economic viability. However, these residues often serve low value markets such as animal feed. Therefore, any opportunity to increase profitability by adding value to by-products can be expected to be an attractive prospect to the grain milling industry.

There are several milling by-products produced in significant volumes in the UK, namely wheat bran/middlings, wheat germ, oat bran and oat husks/hulls, where availability could potentially be sufficient to provide feedstock for large-scale industrial applications such as LA production (Table 8).

	Annual production
	('000 tonnes)
Wheat milled	6,305
Bran	946
Germ	189
Oat milled	474.6
Bran	86.9
Germ	8.7
Husks/hulls	182.2
Barley milled	0

Table 8. Annual production of cereal milling by-products for 2012²²

²² <u>https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/263471/cerealuse-statsnotice-05dec13.pdf</u>

4. Lactic acid production from milling residues

4.1 Utilisation of milling co-products for chemical production

Over recent years, interest in developing chemicals and fuels from cereal crop residues has increased substantially. However, commercial operations have largely been limited to the production of bioethanol from crop harvest residues (e.g. stover, straw etc.) in respect of biofuel mandates in the EU and US and the relative availability of these feedstocks.

For instance, several cellulosic ethanol biorefineries utilising corn stover (leaves, stalks and empty cobs of maize) as feedstock are nearing completion in the US; Du Pont are developing a 30 million gallon plant in Nevada, Iowa²³ while Poet-DSM are constructing a 25 million gallon facility in Emmetsburg, Iowa²⁴. Abegoa and Quad County Corn are also in the process of developing similar facilities. More advanced biorefineries can be expected to come online in the near future in the US as the nation aims to meet its cellulosic biofuel target under the Renewable Fuel Standard (RFS) of 16 billion gallons by 2022. Meanwhile, Beta Renewables has constructed Europe's first commercial advanced bioethanol plant in Crescentino, Italy, with 3 further cellulosic refineries to follow. The Crescentino facility produces approximately 40,000 tonnes of bioethanol per year employing wheat straw, rice straw and arundo donax as feedstock– a fast growing lignocellulosic crop²⁵. The development of EU legislation to provide greater support to advanced biofuels can be expected to stimulate further deployment of such facilities in the future.

To date utilisation of grain milling by-products for production of renewable fuels and chemicals remains largely in the research & development stage, although interest is growing as potential opportunities for developing cereal-based biorefineries become established (10; 11). Such facilities could enable the production of a range of different valuable product streams, utilising all the various chemical components of the biomass, including lignocellulosic materials.

Latterly, significant efforts have been made to assess the potential for processing wheat bran into platform chemicals. A strategy for the production and valorisation of succinic acid from whole-crop wheat biorefining has recently been developed. The strategy outlines a comprehensive processing pathway involving front-end milling, enzyme hydrolysis and a combination of different fermentation technologies (conventional fermentation for flour and

²³http://biofuels.dupont.com/fileadmin/user_upload/live/biofuels/Feedstock_Program_2014.pdf

²⁴ http://poet.com/cellulosic

²⁵ http://www.betarenewables.com/press-release-detail/2/crescentinos-biorefinery-grand-opening

solid-state fermentation for wheat bran/middlings) (Lin *et al.*,2011). However, the commercial feasibility of such an approach would still require verification. Further efforts have focused on the potential to use wheat bran as a substrate for ethanol production (Palmarola-Adrados *et al.*, 2005; Okamoto *et al.*, 2011). One study demonstrated efficient enzymatic hydrolysis of non-starch carbohydrates following dilute acid pretreatment, with sugar production as high as 80% of the theoretical yield (Palmarola-Adrados *et al.*, 2005). Meanwhile, a separate study showed efficient ethanol production from direct fermentation of wheat bran using a novel fungal strain, with yields as high as 78.8% of the theoretical yield (Okamoto *et al.*, 2013)

There has also been interest in utilising wheat bran for LA production. However, much of the research has concentrated on using it as a nutrient source, rather than a carbon substrate (Li *et al.*, 2010; John *et al.*, 2006). It has been demonstrated that use of acid-pretreated bran as a nutrient source for LA fermentation can enable conversion efficiency close to theoretical yields (Li *et al.*, 2010). In using wheat bran over yeast extract, nutrient supplement cost was calculated to be reduced by approximately 9-fold. Similar results were also found for use of protease treated wheat bran as a nutrient source (John *et al.*, 2006). In the event that wheat bran is also used as a carbon substrate for LA production, this research demonstrates that fewer nutrient sources would be required than for fermentation of many other substrates, thus offering potential to reduce comparable operational costs.

Efforts have also focused on using wheat bran as a substrate for LA production. Efficient fermentation of wheat bran starch to LA (0.78g LA per gram of starch in wheat bran) using solid-state fermentation has previously been demonstrated (Naveena *et al.*, 2004) with a further study affording similar results (Naveena *et al.*, 2003). While there is clear potential to use wheat bran as a substrate for commercial LA production on account of its high carbohydrate content, it is clear that more research would be required to optimise the parameters and demonstrate commercial process feasibility.

Research has also been conducted into the potential for using oat residues as a feedstock for producing renewable platform chemicals. One study has assessed a range of different pretreatment options for production of fermentable sugars from oat hulls with a view to produce cellulosic ethanol (Perruzza, 2010). The study demonstrated that high xylose yields (85% of theoretical) and glucose yields (89% of theoretical) could be achieved from hydrolysis of oat hulls using 'disc refining' and 'mercerisation' pretreatments. It was further shown that incorporation of a solid-liquid separation step was effective at removing fermentation inhibitors and improved ethanol yields.

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Finally, research has been conducted on the potential of lactic acid bacteria to ferment sugars present in oat bran (Kontula *et al.*, 1998). While two of the three strains assessed were only capable of fermenting polymers containing C6 sugars, a *Lactobacillus* strain was shown to be capable of fermenting both the C6- and C5-containg carbohydrates present in oat bran.

Ultimately, little commercially oriented work has been done to date regarding the potential for using cereal milling by-products for LA production. However, clear progress has been made in commercialising technologies for LA production from cellulosic feedstocks, with Corbion-Purac, Cellulac, Plaxica and Natureworks all attempting to scale production processes. Cellulac's proposed facility could be of notable relevance to this project on account that it plans to utilise spent brewers grain from a distillery effluent, which can be expected to be of similar chemical composition to milling by-products (although higher levels of lignin can be expected).

4.2 Process overview for the production of lactic acid from cereal residues

LA production from cereal residues would most likely include the following steps: 1) advanced pretreatment of grain fractions; 2) saccharification of carbohydrates to fermentable sugars; 3) fermentation of fermentable sugars to lactate salts; 4) acidification of lactate to LA; 5) purification of LA; and 6) concentration of LA formulations. The purpose of each process is described in Table 9.

The pathway described is regarded as the most commercially attractive process for the production of LA from cereal residues, although alternative pathways do exist, e.g. via chemical conversion (as developed by Plaxica).

Table 9. Processes for lactic acid production from cereal residues (modified from Reineck(2008))

Process	Purpose
Pretreatment/liquefaction	Thermal- and/or chemical-assisted modification of cereal grain structures
	and carbohydrate-rich pockets
Saccharification	Solubilisation and extraction of fermentable sugars from carbohydrate
	polymers
Fermentation	Microbial conversion of fermentable sugars to LA, addition of neutralising
	agent to maintain pH in fermentor, and make lactate salts
Acidification	Convert lactate salts to LA
Purification	Separation of cells and nutrients, and residual sugars, to obtain LA

Concentration

Pretreatment

The pretreatment step facilitates disruption of the cellulosic fibres to enable downstream processing of the bound sugars. A wide variety of different pretreatment technologies exist, although, frequently, the process will involve cooking the biomass in water at elevated temperature and pressure (either with or without use of chemical agents).

Cellulosic raw materials often have specific requirements that have to be achieved before pretreatments can be effective; depending on the chemical and structural composition of the material. Cellulose tends to be more recalcitrant to physical, thermochemical and biological pretreatment methods than starch-rich materials. For instance, arabinoxylans can be solubilised and extracted by medium temperature water cooking, while cellulose requires cooking in temperatures as high as 195°C and the use of harsh chemicals.

Consequently, the pretreatment technology of choice would depend upon the chemical and structural composition of the milling residue.

Saccharification

After pretreatment, the sugars are mainly in the form of oligomers (shorter chain length polymers) and therefore require further processing to form monomeric, fermentable sugars. This process is known as saccharification and there are a wide variety of enzyme catalysts tailored for this purpose. Saccharification is usually performed at temperatures in the 40–80 °C range, and it is often combined with the subsequent fermentation stage.

LA fermentation

Fermentation is the process by which microorganisms metabolise sugars into a variety of different chemical compounds under a controlled environment. Traditionally, industrial fermentation technologies have focused around the production of ethanol using yeast. However, developments in synthetic biology and industrial scale-up procedures have enabled the production of a wide variety of products using a broad range of different yeast, bacteria and algal species. LA is one such product that has been successfully manufactured by this process.

The technology behind LA fermentation from glucose is mature at commercial scales. Globally, 95% of LA production is produced through fermentation using LA-producing bacteria (LAB) such as *Lactobacillus* spp., a process used for centuries to produce yogurt. Other bacteria and yeast can also be used to convert fermentable sugars to LA. Fermentable sugars traditionally originate from materials such as sugar cane, wheat, corn, or potato that have a high content of C6-sugars. However, new routes have been developed utilising lignocellulosic biomass for the production of fermentable sugars. The carbohydrate content of this 'biomass' generally contains:

- cellulose and β-glucans, composed of C6 sugars
- arabinoxylans rich in C5 sugars (sugars with 5 carbon atoms, e.g. xylose and arabinose).

There is a broad range of LAB strains that convert either C6 or C5 sugars (or both) into LA. Known species include *Lactobacillus, Bacillus,* and *Rhizopus* spp. Strain selection depends on many parameters including productivity, product yield, by-product formation, operating temperature and pH, co-production of enzyme for starch saccharification and chiral purity.

A crucial parameter for LA fermentation is the composition of the fermentable sugars. This is because a different set of reactions occur in the production of LA, depending on whether it is C6 or C5 sugars being fermented. While C6 sugars are converted to LA entirely, with no coproducts, fermentation of C5 sugars results in the production of LA and acetic acid in a 3:2 stoichiometric ratio. Consequently, fermentation of sugars from lignocellulosic biomass will result in a poorer LA yield than use of starch feedstock on account of its higher C5 sugar content. However, the additional acetic acid produced can be utilised to develop further valuable co-products that can be sold to improve the viability of the process.

Acidification

LA fermentation products are typically in lactate salt form, which needs to be converted to LA via acidification. Acidification requires strong acids with (in the case of sulphuric acid) gypsum being produced as the by-product which can be used as a fertiliser or component of plaster.

Purification

Purification is one of the most expensive processes in LA production and therefore impacts strongly on overall process viability. It is a multi-step process which involves removal of

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microorganism cells and chemical impurities. In order to avoid excessively high purification costs, addition of chemicals in upstream processing needs to be very selective.

In the initial stage of the process, microbial cells are removed from the product mix using filtration or flocculation, depending on the size of the cells. LA solvent extraction or distillation is then used to purify the product. LA can be purified further by use of activated carbon, and ion-exchange resins. Other purification strategies include electrodialysis or purification via intermediate ester formation.

Concentration

During concentration water is removed and LA is concentrated using standard technologies. LA in the final product often has an optical purity of >98% and a concentration of about 60-70% (Reineck, 2008). The LA produced is a valuable commodity which can be sold directly into a variety of end-user markets used for further processing, such as production of PLA.

4.3 Summary

All cereal milling residues contain high levels of fermentable sugars that may feasibly be converted to lactic acid (LA). However, these sugars are predominantly found in complex polymer structures (cellulose, arabinoxylans, β -glucans etc.) and thus require intensive processing over and above what is needed for starch-based feedstocks. The decomposition of carbohydrate polymers prior to fermentation is generally performed through pretreatment and saccharification. These steps are essential in achieving LA production from cellulosic sugars.

Unfortunately, pretreatment is notoriously expensive and, consequently, this process impacts significantly on process viability. This is largely due to the complex chemistries involved in deconstructing the cellulosic polymer chains. Moreover, many of the more promising pretreatment technologies remain in the development / demonstration stage. Despite these facts, raw material cost often forms the greatest share of the overall production costs for chemical commodities. Consequently, there is strong potential for using undesirable and inexpensive lignocellulosic biomass over conventional starch-based feedstocks for commercial manufacture of renewable fuels and chemicals, including LA. There are further, less tangible, benefits encouraging this transition, such as the importance of supply chain sustainability in brand development. This could assist by providing a premium for the product, further improving the economic viability of production.

5. Feasibility assessment

5.1 Feedstock requirements

In Section 3 it was demonstrated that the UK grain milling industry produces a range of byproducts, namely wheat bran, wheat germ, oat bran and oat husks/hulls, that could potentially be utilised as feedstock for a lactic acid (LA) plant. Other potential feedstocks produced from the grain industry also exist, such as starch and out-of-specification grains. However, starch was discounted from further analysis on account that PLA production from this feedstock has already been undertaken (Reineck, 2008). Meanwhile, production of outof specification grains is highly weather-dependent, with wet weather usually producing a poorer quality harvest. This annual variability is expected to introduce too much uncertainty into supply chain security and therefore, out-of-specification grains were also discounted from further evaluation²⁶.

In order to determine the feasibility of using UK cereal milling by-products as feedstock for a LA plant, it is essential to establish feedstock volumes required for development of a facility of viable scale. It has previously been demonstrated that a 30,000 tpa PLA plant in the UK is of appropriate scale for economic viability (Reineck, 2008). We have therefore provided estimates in this study based upon a LA plant capable of supplying a 30,000 tpa PLA plant (i.e. 37,500 tpa LA plant).

To determine required feedstock volumes, the amount of fermentable sugar in each milling by-product was determined (Table 11) and conversion rates for the process outlined in section 4.2, established²⁷.

²⁶ It should be noted that therein lies potential to use out-of-specification grains as an alternative to existing feedstock for a LA plant –however, this fact should not be used for justification of a business case on account of the high level of risk involved.

²⁷ Conversion factors:

^{1.1} gram glucose is theoretically yielded from hydrolysis of 1 gram starch/cellulose/β-glucan; glucose yield from pretreatment/hydrolysis of starch/sugar of 100%; glucose yield from pretreatment/hydrolysis of cellulose/β-glucan of 80% for wheat residues (Palmarola-Adrados et al.,2005) and 89% for oat residues (Perruzza, 2010); 1 gram glucose yields 0.90 grams lactic acid (in literature: 0.74-0.99 gram lactic acid(Martinez et al., 2013)) 1.136 gram xylose/arabinose is theoretically yielded from hydrolysis of 1 gram arabinoxylan; xylose/arabinose yield from pretreatment/hydrolysis of arabinoxylan of 80% for wheat residues (Palmarola-Adrados et al., 2005) and 84% for oat residues (Perruzza, 2010); 1 gram xylose/arabinose yields 0.61 gram of lactic acid and 0.41 gram acetic acid (theoretically 0.682 gram lactic acid and 0.455g acetic acid)(Martinez et al., 2013; Taskila and Ojamo, 2013)

^{1.2511} gram of LA is needed to produce 1 gram PLA.

Table 10. Chemical composition of cereal milling residues

	Wheat		Oat		
	Bran²⁸ (Saunders and Walker, 1968)	Germ (Šramková <i>et</i> <i>al.</i> , 2009)	Bran (Kamm <i>et</i> <i>al.,</i> 2010)	Husks/hulls (Kamm et al., 2010; Perruzza, 2010)	
Cellulose, %	22		37	34	
Arabinoxylan, %	27		46	33	
β-glucan, %		10			
Sugars, %	5	17			
Starch, %	9	20			
Non-sugars (proteins, fats, ash, lignin), %	27	40	9	28	
Moisture content, %	10	14	12	5	

The selection of feedstock has a strong impact on the design and the size of the LA production plant. As a rule of thumb, the higher the sugar content of the feedstock, and the greater the ratio of C6 to C5 sugars, the smaller the fermentation capacity of the LA production plant needs to be. It should be further considered that while LA product yield is lower for C5 sugar fermentation, acetic acid is formed as a co-product which could be sold as a platform chemical – or used to produce other valuable products streams – to generate further revenues for the plant.

Of all cereal milling residues assessed, the greatest volumes of wheat germ would be required to provide 37,500 tpa LA (Table 11), estimated at 84,200 tpa. This is largely on account of the comparatively low carbohydrate content of wheat germ. This required volume would represent almost half of total UK production, suggesting that use of this feedstock would unlikely be feasible given the distributed nature of wheat milling in the UK and consequential challenges in regards to transport logistics.

Meanwhile, 78,200 tonnes of wheat bran would be required to supply sufficient LA for a 30,000 tpa PLA plant, a figure close to that of wheat germ. However, almost 1 million tonnes of wheat bran is produced in the UK every year. Wheat bran requirement would therefore represent only 8% of UK production, suggesting that sufficient volumes of this feedstock would be available to supply a UK LA plant.

²⁸ Chemical composition of wheat middlings assumed to be equivalent to that of bran.

Slightly lower volumes of oat bran and oat hulls/husks would be required than wheat milling by-products to supply a LA plant, with estimates at 73,000 tpa and 60,000 tpa, respectively (Table 11). However, availability of oat bran is low with UK production at less than 90,000 tpa, suggesting that use of this feedstock would encounter logistical difficulties in supplying a UK based world-scale LA plant. Availability of oat husks/hulls is higher at 182,000 tpa. A LA plant based on oat husks/hulls would therefore require just over a third of UK production. While this might appear ambitious, a representative from industry confirmed the existence of at least one facility in the UK producing 60,000 tpa of husks/hulls from oat milling, a quantity that alone is almost sufficient to supply a 37,500 tpa LA plant.

Table 11. Carbon mass balances for converting fermentable sugar constituents of cereal milling residues to LA (results are given on an 'as received' basis)

For 30,000 Tonnes PLA production capacity, the plant requires as feedstock:					
either	78,200	Tonnes milling wheat bran			
or	84,200	Tonnes milling wheat germ			
or	60,000	Tonnes milling oat bran			
or	73,000	Tonnes milling oat husks/hulls			

On account of these estimates of relative availabilities, wheat bran and oat husks/hulls can be considered the most feasible milling by-products for use as feedstock for a UK LA plant. Therefore, it is these that were taken forward for assessment of economic viability.

5.2 Economic assessment

To determine the potential of developing effective values chain from conversion of cereal milling residues to LA, an economic assessment of LA production from wheat bran, wheat middlings and oat husks/hulls was undertaken, utilising a range of scenarios to account for varying cost prices for each feedstock. Again, the analysis has assumed production of 37,500 tpa LA, sufficient to supply a 30,000 tpa PLA.

A full techno-economic analysis of each feedstock is outside the scope of this study. Instead, the exercise concentrated on determining value added to feedstock, based on the buying price of the feedstock and the selling price of the LA, to determine whether an attractive value chain could be developed i.e. the analysis did not include feedstock logistics, capital costs, other purchasing costs, manpower, or operational expenditure.

Selling price ranges for wheat and oat milling by-products (as gathered from literary references and communication with industry representatives) are shown in Table 12.

Table 12. UK prices of cereal milling by-products

Prices (£/T)	Low	Medium	High
Wheat bran	56**		185♦
Pelleted wheat middlings*♦	141	143	151
Pelleted oat milling residuals (husks/hulls, bran, middlings)♦		150	

*Wheatfeed consisting of wheat bran, wheat germ, and wheat flour tails.

**Shipped from Ukraine

▲ Figure from Alibaba.com

• Figures sourced from personal communication with an industry representative

For the purpose of this analysis, it is assumed that LLA, rather than DLA, is manufactured by the plant. This is because the technology production of LLA is more mature and, consequently, the vast majority of LA available on the market is LLA (with pricings more readily available).

In 2007, the price of LLA in the market ranged from \$1.38/kg for 50% purity to \$1.54/kg for 88% purity (£856–£955 per tonne) (Martinez *et al.*,2013). However, recent discussions with industry indicate that current price for LLA to be between €1.30 and €2.30 per tonne (£1005–£1860 per tonne) (figures from communication with an industry representative). For the purpose of this study we used a mid-price estimate of £1400 per tonne LLA. Potential sales of co-products of the LA production process, e.g. acetic acid derivatives, were not included in the analysis.

When accounting for these price assumptions it is evident that effective value chains can be developed from production of LLA from cereal milling residues; value added estimates for all by-products ranged from £38 million for wheat bran under a high cost price scenario to £48.1 million for wheat bran under a low cost price scenario (Table 13).

Table 13. Value analysis of scenarios comparing purchasing costs of required feedstocks per year with product revenues from sales of 37,500 tpa LLA

		Low price	Medium price	High price
Wheat	Feedstock cost (£/MT)	141	143	151
middlings	Feedstock cost (£/year)	11,021,038	11,177,365	11,802,672
(Wheatfeed) Wheat bran	Revenues from LA (£/year)	52,500,000	52,500,000	52,500,000
	Value added (£/year)	41,478,962	41,322,635	40,697,328
	Feedstock cost (£/MT)	56		185
	Feedstock cost (£/year)	4,377,150		14,460,227
	Revenues from LA (£/year)	52,500,000		52,500,000

		Low price	Medium price	High price
	Value added (£/year)	48,122,850		38,039,773
Oat hulls/husks	Feedstock cost (£/MT)		150	
	Feedstock cost (£/year)		10,946,071	
	Revenues from LA (£/year)		52,500,000	
	Value added (£/year)		41,553,929	

In the event that production pathways for DLA become better established, therein lies potential for improving these value chains further. As discussed in Section 2, new varieties of PLA containing high DLA content often demonstrate improved properties over conventional PLA. A higher price can therefore be demanded for these new polymer varieties, and likewise for the DLA itself; industry representatives suggest DLA could command a price of between £3,000 and £4,000 per tonne. However, due to the nascence of the DLA market there will inevitably be technical challenges involved in large-scale DLA manufacture, such as the scaling-up of fermentation processes that involve engineered microbial strains. Therefore, while the potential rewards of DLA production can be expected to be higher than for LLA manufacture, the level of risk involved is higher as well.

This analysis demonstrates that therein lies potential for developing effective value chains in the UK based on production of LA from cereal milling residues. However, it should be reminded that the information provided in this analysis is alone insufficient to give confirmation of feasibility. A wide range of other factors require further consideration, such as transport logistics, capital and operational costs etc.

6. Conclusions

On account of climate change and oil price volatility concerns, current EU policies and regulations strongly incentivise the development of bio-based alternatives to fossil materials, chemicals and fuels. As a consequence, the market for biopolymers can be expected to grow significantly in the near future, this includes PLA. New varieties of PLA with improved properties (e.g. with high heat distortion temperatures) are undergoing development. This can be expected to increase the number of available applications of the polymer, and thus the overall PLA and LA markets, while also creating further opportunities for developing improved value chains.

By analysing the technologies and production pathways needed for production of LA from cereal residues, we found little prior research or activity in the utilisation of milling coproducts. However, we did establish several relevant technologies that would be suitable for such processes and consequently demonstrated a theoretical processing pathway for conversion of milling by-products to LA.

This study also demonstrated that there is good potential in the UK for developing value chains based on production of LLA from cereal milling residues, notably wheat bran/middlings and oat husks/hulls, in sufficient volumes to supply a 37,500 tpa LA plant (equivalent to the feedstock requirement of a 30,000 tpa PLA). Most encouragingly, it was discovered that a single oat milling facility exists in the UK which alone produces almost sufficient residue volumes to supply a facility of this scale. Improvements upon the value chains discussed here could be realised in the event that DLA is provided as the output to the LA plant. However, this can be expected to increase the level of risk involved.

The main observations from the analyses are summarised in Table 14.

	Waste wheat milled grain	Wheat bran	Wheat germ	Wheat middlings (wheatfeed)	Residual starch	Oat bran	Oat husks/ hulls
Abundance/ availability	High	High	Low	High	Low	Low	High
Total carbohydrate content (wet basis)	80% (endosperm)	63%	47%	63%	Up to 90%	83%	67%
Competitive uses	Industrial (periodic)	Food/ feed	Food/ feed	Feed	Food/ industrial	Food/ feed	Feed/ energy
Technological feasibility/ maturity	1G/ C6 Fermentation	2G/ C6+C5 Ferm	1G/ C6+C5 Ferm	2G/ C6+C5 Ferm	1G/ C6 Ferm	2G/ C6+C5 Ferm	2G/ C6+C5 Ferm
Economic competitiveness	Good	Good	N/A	Good	Good	N/A	Good
Water content (transportation/ storage)	20%	10%	14%	10%	Very low	12%	5%
Overall attractiveness	Low	High	Low	High	Low	Low	High

Table 14. Cumulative results scheme from feasibility analysis for production of PLA from cereal milling residues in UK

Ultimately, this study demonstrates that there is a strong and growing market for LA, the production pathways from cellulosic feedstocks are becoming commercialised and therein lies potential to develop value chains from cereal milling residues in the UK. However, little work has been done to date regarding the manufacture of LA from milling residues, and

therefore this opportunity can be expected to have significant risks, especially in regards to technical feasibility without further research.

7. Further recommendations

The use of wheat bran or oat husks need further investigation to be verified as competitive feedstocks for LA production in UK. First, further research and development is needed from a scientific point of view to confirm: 1) practical suitability of the suggested processing pathway for conversion of milling by-products to LA; 2) scalability of the processes through experimental investigation from bench to pilot scale; and 3) LCA analysis of PLA production from wheat bran and oat hulls/husks in UK. Moreover, system integration and a full techno-economic analysis is also essential for further investigation for confirming feasibility (EI Mekawy *et al.*,2013; Kazi *et al.*,2010)

Other feedstocks that were out of scope for investigation in this report, but represent promising feedstock for a UK LA plant, include rapeseed press cake, wheat straw, barley straw, and rape straw (Martinez *et al.*,2013) These feedstocks are regarded as attractive opportunities because they have high sugar content, are abundant and available at low cost in the UK, are non-food residues and processing technologies are already available to convert them to LA. It is therefore recommended that these feedstocks are also investigated in the future due to their competitive value and characteristics.

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Appendix 1 – Chemistry of PLA

PLA is a polymer produced by polycondensation of lactic acid (LA.) LA is a hydroxycarboxylic acid that can react with itself to form a dimer and a molecule of water. This dimer dehydrates to form a cyclic lactide and a second molecule of water. The lactide can then be polymerised by ring opening (much like caprolactam forms Nylon 6) to form a polylactide.



LA is chiral, existing as D- and L- optical isomers (enantiomers), termed DLA and LLA, respectively. The L- enantiomer is produced by enzymes in animals and microorganisms, such as yeasts, produce (racemic) mixtures of LLA and DLA. However, CORBION uses proprietary microorganisms to produce LLA essentially free of DLA and can also produce DLA essentially free of LLA.

There are three forms of lactide: D-/D-, L-/L- and D-/L-.

As described in the text, there are also three forms of PLA. Most of the PLA which is commercially available currently is the polymer of L-/L- lactide and mesolactide. Enantiomeric PLLA (the polymer of L-/L- lactide) and PDLA (the polymer of D-/D- lactide) have been developed and are expected to be commercially available in the near future. The molecular structure of PLA determines its properties as a thermoplastic.

As a random copolymer, the PLA which is currently available in the marketplace is essentially amorphous. However, enantiomeric PLLA and PDLA are homopolymers and as such are essentially crystalline.

Appendix 2 – PLA characteristics and considerations

Physical Properties

Physical properties which are inherent to all grades of PLA are as follows:

- Low density of 1.25 g/cm³ (meaning lower part weight compared to PET and PVC >1.35)
- High tensile strength (meaning thickness can be reduced)
- Stiffness (similar to polystyrene)
- High transparency (haze of 2.1%, only slightly less transparent than PS or PET; although not immediately visible, PLA has a marked yellow colouring and a slight milkiness)
- High moisture vapour permeability (MVP) (an issue for most packages whose function is to keep water out)
- High permeability to gases
- Biorenewability
- Biodegradable in industrial composting or anaerobic digestion (but not home compostable)
- Low UV absorbance (meaning that package would not protect contents from certain wavelengths of UV radiation)
- Good printability (meaning excellent results can be obtained with various ink blends and printing systems)
- Easily embossable (which is of great interest for thermoformed sheet applications)
- Excellent dead-fold (and twist effect ~ 25% better than that of Cellophane) (meaning it is possible to produce transparent packages by folding PLA film or sheet, much as with paperboard)
- Good crease resistance (same as with Cellophane, but approx. 20% better than all other films currently on the market)
- Flavour and odour barrier (excellent resistance vis-à-vis most of the oils and fats found in foodstuffs)
- Heat sealability (processing temperature: 80° C, heat seal strength: > 0.13 bar) (however this might be considered low versus processing temperatures for other filmic materials)

For First Generation PLA:

- Impact resistance of 20–25 J/m is far too low for engineering applications
- Heat distortion temperature (HDT) of 50–55 °C means poor resistance to heat which shows up as deformation of items such as trays and cups during transport and use. Also, lumping of pellets during transport, storage and processing has been an issue.
- Gas barrier properties are an issue for many types of packaging (not only carbonated drinks)

For **Second Generation PLA** (polymers are based on PLLA and PDLA as an additive):

- Straight PLLA has HDT = ~60°C
- Crystalline sc-PLA produced from blend of 5% PDLA (as 10% of 50% PDLA stereocomplex) with PLLA has HDT = ~100°C. Incorporation of sc-PLA crystallites increase the rate of crystallisation of PLLA, providing advantages such as faster throughput, lower energy consumption, lower shrinkage and higher degree of crystallinity in injection moulding applications.

For **Third Generation PLA** (polymers are sc-PLA based on 1:1 PLLA-PDLA blends, with HDT = >180°C):

- Target applications for sc-PLA would require HDT 80–160°C and Izod impact strength up to 140 J/m, replacing standard and engineering plastics such as polyethylene terephthalate (PET) and polycarbonate (PC)

Targeting these development markets will require plastics technology including alloys, additives, fillers, compounds and nano-composites

PLA can be solid stated (similar to PET) to increase molecular weight and crystallinity: this is done now in biomedical applications.

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