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Review article

Circularity in green chemical products, processes and services: Innovative routes based on integrated eco-design and solution systems

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A B S T R A C T

Understanding the current drivers, potentialities and challenges related to the role of chemical sciences in a circular economy is of fundamental importance when bioresources are taken into account. Particularly, after launching the European Commission action plan in 2015, the creation, development and use of green chemicals derived from renewable materials can be seen as more than simple opportunities in research and innovation. In this paper, the latest trends related to green chemical products, processes and services concerning eco-design and solution approaches will be focused on, using an orange waste biorefinery as a case study. Emphasis will be given to establishing new relationships with goods, materials, energy and, mostly, long-term cooperation and integration models among all partners involved.

1. Introduction

The circular economy is gaining momentum as a demand from various sectors towards global sustainability. Instead of a linear and take-use-disposal pattern, resources are used, recovered and renewed in internal cycles as long as possible, creating further maximum value [1]. When there is a broader closed loop resource circulation, as well as a contribution to reducing greenhouse gas emissions, natural systems can be understood as models to provide better systemic performance. The circular economy, a concept adopted in previous initiatives and other fields [2], can be described as: “an industrial system that is restorative or regenerative by intention and design. It replaces the end-of-life concept with restoration, shifts towards the use of renewable energy, eliminates the use of toxic chemicals, which impair reuse and return to the biosphere, and aims for the elimination of waste through the superior design of materials, products, systems and business models” [3,4].

The transition from the current linear to stronger circular economy models demands an ambitious transdisciplinary engagement, changing the systems based on “bigger-better-faster-safer” (and more competitive) and ownership through the selling of goods (or molecules) as services by rent, lease and share business models [5]. In an advanced scenario, manufacturers own the product, the process to obtain it and the resources that compose it (i.e., materials, energy, materialised knowledge, etc.), but also responsibility related to costs and risks associated to the so-called waste. This can promote profits from sufficiency on a larger scale involving several sectors of society [5]. An analogy for this model is known as Giotto's perfect circle. He proved the beauty of his - but also the human - capacity by drawing an absolute circle, which was a sufficient message for those knowing the sign of such achievement. As can be seen, the contemporary demands for alternative routes involving cycles of materials, energy, data processing/transferring, new collaborative consumption and production patterns, among other pressing requirements are becoming greater every single day [4,5].

2. Current drivers towards circular economy: implications for research and innovation

Based on prospective studies, it is estimated that the circular economy can offer an opportunity to save net material costs from USD 340 to 380 billion/year at EU level in a transition scenario and

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from USD 520 to 630 billion/year in an advanced context, just using reverse-cycle activities [6]. As a response, very recently the EU launched an action plan to help European businesses and people in general to make the transition towards an advanced circular economy aimed at contributing to "closing the loop" of product lifecycles, also boosting "competitiveness" and generating jobs. The EU package will receive substantial support from the European structural and investment funds (ESIFs) of USD 6.2 billion from structural funds for waste management and investments in the circular economy at a national level and, more importantly, USD 725 million from Horizon 2020, which is the EU funding programme for research and innovation [6–8]. Other key points, also including a timeline and a plan for concrete implementation and monitoring framework, involve:

- Promoting measures to minimise food waste, which include establishing common methods, e.g., to evaluate and improve data analysis;
- Developing quality patterns for secondary raw materials to gain confidence in the market;
- Planning actions in eco-design to improve repairability, durability, recyclability of products and energy efficiency;
- Establishing strategies considering plastics, addressing issues of recyclability, biodegradability and reducing the use of toxic substances in plastics, contributing to significantly minimising waste in marine environments;
- Promoting a number of actions concerning water reuse including a legislative proposal regarding minimum requirements for reusing wastewater;
- Revising regulations concerning fertilisers in order to promote the recognition of organic, bio- and waste-based fertilisers in the market, supporting the role of bio-nutrients.

Among the EU revised legislative proposals accompanying the series of actions for waste reduction, management and recycling, some are of particular concern in chemical sciences, such as establishing: harmonised definitions and calculation methods for recycling rates in the EU; routes to advance the re-use and stimulate industrial symbiosis by turning one industry's co- or by-product into another industry's feedstock; as well as economic incentives for producers to launch and maintain greener products on the market, supporting recovery and recycling systems (e.g. for medicines, food, packaging, electric and electronic devices, vehicles, aircrafts) [7]. However, until now, initiatives related to the circular economy have evolved primarily as research in waste management, raw material use and environmental impacts and, scarcely, in business and economic perspectives, showing the limitations related to its effective implementation since the advantages for mainstream industry and market are not completely explicit yet. It is important to emphasise that the circular transition has just started, offering good prospects for gradual improvement of the present production system [8,9].

3. An emergent scenario: contributions from chemical sciences to circular, bio-based and integrated contexts

Since the effective launch of the circular economy concept in 2010, an increasing number of papers correlating it to chemical transformations have been published [10]. From 2014 onwards, more than 215 publications were produced on circular economy, bioeconomy, renewable, green and sustainable chemistry and chemical processes. Furthermore, 74.4% of this total amount was published from January 2015 to September 2016 (ISIS Web of Knowledge; Thomson Reuters). Table 1 presents the main topics and related aspects described in these latest publications, ordered according to their number of citations.

Advances in the application of enzymes as a pretreatment strategy, mainly concerning laccase, and underlying directions for biofuel production are discussed in the most cited paper [11]. Bio-based processes can be advantageous due to the synergy between all the enzymes involved in the breakdown of lignocellulosic biomass, promoting more efficient processes, as is the case of cocktails containing laccases and hemicellulases to degrade the lignin and hemicellulose components (and possibly the benzyl ether, γ-ester and phenyl glycoside linkages between lignin and carbohydrates). Using new enzymes for the degradation of lignocellulosic waste and the enzymatic conversion of polysaccharides and related technologies continue to be one of the main subjects of interest in this field [12]. In general, a biological transformation is considered a greener alternative to the physical-chemical processes to increase the bio-based product recovery from waste improving saccharification and fermentation yields.

Case-studies based on d-Limonene have also been described in integrated chemical contexts, including a number of fundamental aspects such as waste prevention, design optimization, renewable feedstock and green processes [13,14] (Fig. 1). Dextrorotatory isomer (+)-limonene is the principal component of essential oils found in oranges, lemons, mandarins, grapefruits and limes. It is a renewable chemical with numerous applications such as flavoring, platform chemical, active agent and green solvent. Its worldwide generation exceeded 70,000 tons in 2013, and its potential production is expected to remain in the range of 100,000 M tons from now to 2030 [14]. Replacing toxic solvents such as toluene by d-limonene in many industrial processes seems to be most promising in Brazil, the USA, Spain, India and South Africa, since the toluene demand can be based on deploying their own domestic d-limonene extraction potential [14].

Various current and future possibilities playing a role in chemical and energy sustainable production based on CO_2 capture, conversion and use as an extractor agent were also identified [15–18]. In addition to phytomining of critical elements [19], the concept of sustainability to set a framework to develop the bioeconomy not only in the EU was also a relevant topic in these papers [20,21].

Some of the most recent and cited publications cover wider topics such as biomass hydrothermal fractionation [22], biosensors [23], bioenergy [24], other greener transformation technologies based on physical and chemical routes [25–28], frequently associated to the conversion of food waste into bioenergy and higher aggregate value chemicals [29–32], water treatment [33], green and sustainability metrics [34–36] and industrial symbioses towards new trends in biorefineries [37–40]. The eco-design of an integrated waste biorefinery can become real with the combined collaborative efforts of engineering and chemical sciences, biotechnology, environmental sciences including key players from industry, government and all sectors of our society, locally and globally [40,41].

4. Circularity from local to global perspectives: shades and shapes of a proposal based on an integrated orange biorefinery concept

The present and future upcoming state of art related to the transition of a stronger circular scenario needs to focus on the outlined hybrid integrated models to advance and provide critical solutions to promote sustainability from resource management to products, processes and services, which can prevent problems concerning pollution, scarcity and an unbalanced distribution of goods. This proposal can be inspired by a biorefinery superstructure-perfect circle system, viewed as a holistic way to

Table 1
Examples of the major topics, methods and applications in chemical science concerning circular economy or bioeconomy found in the most cited papers and recent correlated ones published from 2014 to 2016.

Topics (feedstock sources and themes)	Processes (transformation, separation, determination)	Products and applications
Lignocellulose (forestry waste; sugarcane bagasse; wheat or rice straw)	Enzymatic processes	First and second generation biofuels; green energy [11,12]
Citrus waste	Cold press, centrifugal separation; steam distillation; microwave treatment	d-limonene (platform chemical, green solvent, active agent for functionalised products) [13,14]
CO_2 conversion; extraction (supercritical CO_2)	Catalytic hydrogenation; electrochemical reduction, biotechnology routes; extraction, fractionation and reactions involving lipids and particle formation	Methane, methanol, light olefin, polymers, polypropylene carbonate; removal of bioactive compounds [15–18]
Phytomining of inorganic trace elements	Uptake and accumulation of e.g. Cu, Zn, Cr, Pb, Cd by plants; thermochemical transformation	Phytomanagement to remediate impacted environments; recovery of elements from *Miscanthus* species [19]
Biomass supply and demand survey	Pretreatment, chemical, thermochemical and biochemical processes	Framework for sustainable bioeconomy deployment until 2020 [20,21]
Cellulose, hemicellulose, and lignin	Biomass hydrothermal fractionation (supercritical water)	Lactic acid, glycolaldehyde, 5-HMF, pyruvaldehyde, xylulose, furfural [22]
Genetically encoded biosensors	Metabolic engineering	Commodities and high value chemicals, including pharmaceuticals [23]
Bioenergy	Modelling	Risk assessment (quantifying and mitigating future risks for bioenergy crops) [24]

move biofuels and other valorised co-products onto much greener footprints, with green and sustainable metrics providing the rigor needed to make the best decisions regarding feedstock (re)use, up scaling processes and value chain formats [41].

One fundamental aspect to remember is the availability of the non-food bioresources in quantity, quality and for a long period, associated to reliable and robust technological flexibility to incorporate changes when necessary, for instance, considering the agricultural seasonality on biomass production [42]. Among cases derived from food residue, orange waste biorefinery has been extensively studied over the last two years [43–46], allowing more than a proof of concept in some countries, for instance, Brazil. A number of chemicals can be extracted and manufactured from citrus peel and bagasse, such as d-limonene, pectin, dietary fibres, soluble solids, proteins, enzymes, acids, sugars and flavonoids [13,14,42]. Currently, green formulations have been prepared from waste by complexing flavonoids with plant-essential metals, which were also removed from landfill sites [47], in a symbiotic production chain (Fig. 2).

An illustration showing zero waste can be seen in Fig. 3, correlating the main fluxes and circulation of resources and emphasising the (re)use of materials composing functional products. The collaborative and symbiotic routes as a new dimension were also included.

Recirculation here can be dictated by the order of preference or

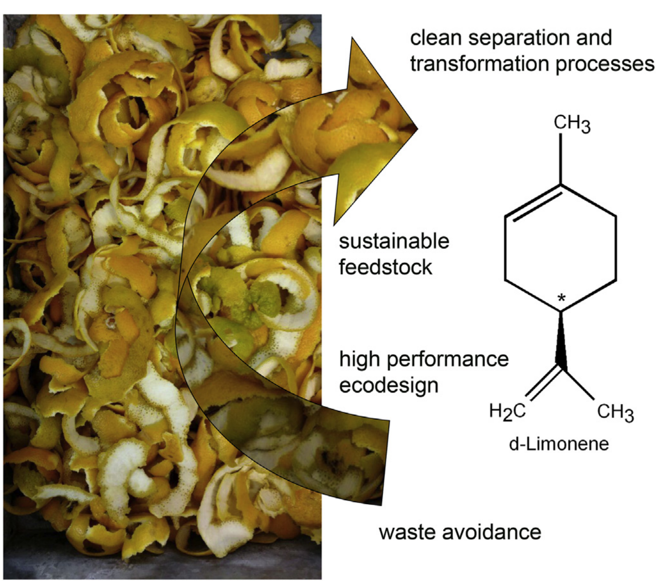

Fig. 1. d-limonene life cycle chemical aspects towards circular and bioeconomy models [10,13].

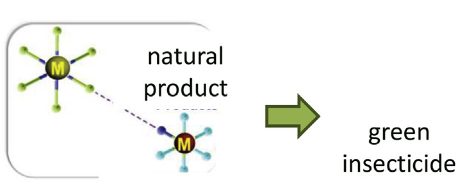

Fig. 2. Bioactive natural products complexed with inorganic ions showed potent and effective insecticide effects [47].

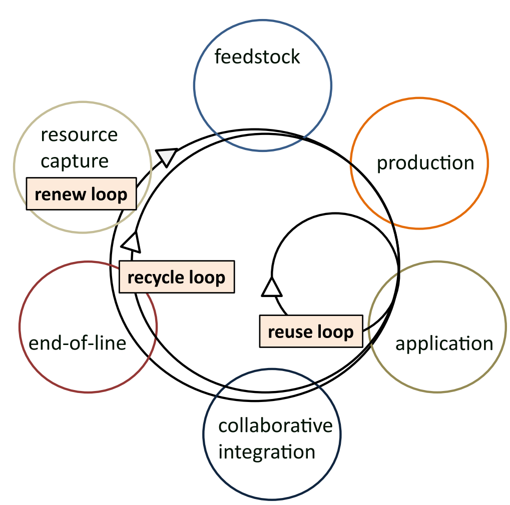

Fig. 3. Integrated biorefinery system for multi-purpose applications based on circular economy and bieconomy models [10,41].

need stated by a determined value to a given material, process or service, governed by its form and available function and use [43].

5. Conclusion

The emergence and establishment of an innovative concept based on circular and bioeconomy approaches creating a new generation of methods, products and services has been transforming not only research in a wider sense, but also industries and markets, globally speaking. From a chemical science perspective, the continued (re)use of co- or by-products is considered of topical importance, generating designed renewable materials or energy whose function and durability are decided previously, before producing them. The idea of product is not based on ownership, but on rent, lease and sharing business. This new circular model is a great opportunity to integrate a web of systems using biological resources and maximising value creation supported by transdisciplinary research teams. This novelty will also demand a new route of collaborative thinking and acting aiming to understand the opportunities and challenges associated with chemistry and its continuously changing interfaces [48,49].

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