GREEN CHEMISTRY AND PROCESS INTENSIFICATION: MILESTONES ON A SUSTAINABLE DEVELOPMENT

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ABSTRACT

As a philosophical support, green chemistry (GC) becomes at present well-combined into the scientific system to assist scientists and engineers consider how to decrease or remove waste and avert the employment and formation of hazardous substances in the design phase of chemicals. Such design attempts in turn affect the full life-cycle of the chemical, from getting the starting materials until the end-use product is recycled or disposed of. There is a considerable advance noted in such direction during the last three decades, this review focuses on GC research. As a comparatively fresh technique, revision in process intensification (PI) is fast and investigation could rapidly lead to outstanding outcomes. However, numerous features of PI could take more time to be fact. Much of the study stays in academic and industrial laboratories, even if large-scale implementations of micro-reactors are actuality. Fields of PI enterprise that have progressed quickly are the expansion of carbon capture techniques, an increasing interest in GC, and the beginning of momentous study into connecting solar energy to intensified methods like chemical reactions. Electric fields (e.g., microwaves and ultrasound) are observed in larger usages, and the application of electrokinetic forces at the micro- and nanoscale persist to fascinate. Huge investigations are working for the sake of ideas like the perfect reactor. PI remains a motif leading to attain a sustainable society. This work may be an orientation in the investigation of product development and design, production and application, in a constructive and stimulating way.

Contribution/Originality: This study documents green chemistry (GC) and process intensification (PI) as milestones on a sustainable development. GC becomes well-combined into the scientific system to assist engineers in decreasing waste and avert the formation of hazardous substances. PI investigation could lead to outstanding outcomes especially in product development and design.

1. INTRODUCTION

During the last quarter century, green chemistry (GC) has furnished a background for chemical engineers to participate in the large field of global sustainability [1-3]. Thousands of researches and publications were dedicated to furnish a rich data to the chemical engineers and others for recognizing present troubles and proposing solutions
The present work constitutes an appropriate domain to emphasize several of the most important axes on the wide topic of GC [1, 2].

There is an increasing interest towards GC [7, 8]. As an example, recently (June 14–June 18, 2021) a Green Chemistry & Engineering Conference took place virtually on the theme “Sustainable Production to Advance the Circular Economy” [1, 2]. It called attention to the fact that contributions should consider a systems approach to decreasing ecological effect via intentional design of chemicals, not just considering how raw materials are sourced and in the fabrication and employment of industrial and consumer goods but also how such materials and goods could be reused, recycled, or upcycled [2, 9, 10]. Adopting life-cycle thinking as a target among the GC community emerges at the backdrop of the recognition of restricted resources and a climate crisis [11–13]. The following axes seem to be determining steps on the route to progress closed-loop economies while still working as models for innovation at a basic scale within their respective chemistry sub-disciplines [2, 14, 15].

The motivation for efficacy in organic synthesis combines the best of the idealisms of the Enlightenment and the Renaissance [2]. Surely, there is a premium on rationalism, with an aspiration of mechanistically sound reaction design and process development [16–18]. However, simultaneously, the aesthetic appeal of the fresh thoughts that peak in the progresses we presently touch usually is evident [19–21]. There is no limitation on curiosity when we examine the boundary conditions of efficacious, environmentally benign techniques [22–24]. Conversely, such reflections generated novel concepts and approaches, ranging from postmodern expansions of photochemistry [12, 25, 26] reconsideration of seminal thinking about solvation, importation of physical and mechanical phenomena [27–29] to reaction development - the creativity born of efficiency considerations now drives major technology innovation in chemistry [2, 30, 31].

2. REDUCING DEPENDENCE ON FOSSIL FUELS

Designing techniques for circularity requires multidisciplinary methods and accurate description of the trouble being handled [32]. One of the pivotal dares encountered remains to be diminishing humankind dependence on fossil fuels for chemical and fuel production [2]. This necessitates both the efficacious and clean transformation of renewable (biobased) raw materials into functional chemicals and fuels, as well as efficacious CO\textsubscript{2} capture and conversion into fuels and chemicals Ganesh, et al. [2]. Wang, et al. [33] examined the expansion and usage of engineered multifunctional nanohybrids of carbon nanomaterials and metal/metal oxide nanoparticles, which exhibit promising multifunctionalities for addressing the critical energy–water–environment nexus [2].

More precisely in terms of CO\textsubscript{2} capture and use, Halliday and Hatton [34] defined a pivotal dare for carbon capture and storage (CCS) - the quest for net-negative emissions. They noted that bioenergy with carbon capture and storage (BECCS) with molten sorbents present a distinctive chance to obtain net-negative emissions with minimal indirect emissions and low-cost separation of CO\textsubscript{2} from other gases [2]. BECCS can eliminate 300–850 kg of CO\textsubscript{2} equivalents from the atmosphere per megawatt-hour of electrical output (kg/MWh\textsubscript{e}), rendering it superior to other low carbon techniques and allowing the offset of emissions from hard-to-abate industries. Focusing on efficacious materials for carbon capture, Jiang, et al. [35] noticed the expansion of light-responsive metal–organic frameworks (LMOFs) that possess tunable structures and performances. Whilst traditional amines could not attain controllable adsorption separation, such LMOFs possess tunable amine-based active sites that ameliorate CO\textsubscript{2} capture and control adsorption and separation. Some progresses as well call attention to the enhancement of techniques for decrease of CO\textsubscript{2} founded on electrocatalytic techniques, comprising the employment of Ru and Re catalysts [36] and the usage of ionic liquids for such implementation [37] as well as investigation of Earth-abundant iron catalysts [38]. Complementary to electrochemical processes, enzymatic [39] and photocatalytic [35] techniques are as well proved to possess a position in the toolbox for conversion and usage of CO\textsubscript{2} [2].

The efficacious and clean transformation of renewable raw materials into functional chemicals and fuels has emerged from a theoretical construct to an interdisciplinary field that is fueled by fundamental innovations in
synthetic chemistry and guided by practical applications of biorefineries [2]. The needs of biorefineries for flexibility of feedstocks, comparatively mild running circumstances, and low ecological effect have conducted to highly innovative techniques for defunctionalizing and refunctionalizing biomass feedstocks to value-added chemicals and materials. Relating to the defunctionalization front, Yang, et al. [40] observed an organocatalytic technique for photochemical C–O bond cleavage of the β-O-4 linkage in lignin scaffolds. Such process furnishes a metal-free strategy to prior reports and is applicable to continuous flow processing. Cellulose-derived platform chemicals could as well be obtained more efficiently from using a redox-switchable biocatalyst for controllable oxidation or reduction of 5-hydroxymethylfurural into high-value derivatives [41].

Extending the range of new platform chemicals that could be obtained from biomass, Del Río, et al. [42] defined the occurrence of valuable phenolic compounds embodied into lignins (like flavonoids, hydroxystilbenes, and hydroxycinnamic amides) that conduct as authentic lignin monomers and are considerably obtainable in big quantities from the abundant in waste products from processing of agricultural or forest biomass. Nakagawa, et al. [43] explored the utility of erythritol as a C4 platform in biomass refinery, derived from fermentation of sugars and glycerol [2].

The direct valorization of lignin to functional chemicals constitutes possible novel approaches for efficaciously transforming lignin to chemicals Ganesh, et al. [2]. Dong, et al. [44] noticed a lignin-carbohydrate complex that is an effective antioxidant for scavenging reactive oxygen species in vitro and zebrafish in vivo. Further, lignin was employed in the fabrication of polyurethane generated by oxidative liquefaction [45]. Several different conversions of biomass to functional chemicals and materials are examined in two deep discussions on biomass-derived carbonaceous materials [46] and bacterial cellulose-based composite scaffolds for biomedical applications [47]. Relating to transforming our polymer platform to renewable materials, consistent with the circular economy, Zhao, et al. [48] examined how we could close the gap for bioplastic usage in single-use food packaging, focusing on the most recent development successes in bioplastic materials and highlighting the “gaps” between bioplastics and their conventional counterparts with respect to their properties [2, 49].

3. REDUCING THE EFFECT OF CHEMICAL SYNTHESIS AND MANUFACTURING

For a circular economy, in promoting industrialization infrastructure, one more pivotal dare stays the reduction of the overall environmental footprint [2]. Such reduction must decrease both the environmental effect of the industrialization techniques of chemicals as well as their likely risks and persistence of the commodity chemicals produced [50-52]. While the former has been a cornerstone of research in GC from its inception in the early 1990s, it is still a fertile area of research [53-55]. The latter will be addressed in further detail later in this work.

In organic chemistry (particularly in organic synthesis), sustainability continues to a driving innovation for many years [2]. With the quantity of waste formed in numerous synthetic chemistry approaches, we are faced with not only an ethical imperative to promote more sustainable chemical methods and products but also a financial imperative [56-58]. Metrics to gauge such advance, comprising process mass intensity, have been suggested that let all aspects of a technique to be contrasted. As an illustration, controlling a reaction in water may not necessarily be an amelioration if several volumes of an organic solvent are required to extract/purify the product. A fresh perspective calls attention to the necessity to explicitly comprise the evaluation of sustainability employing GC metrics [59].

Several researchers focused on progress in catalysis and reaction engineering that conducted to waste and energy reduction for techniques that remain constitute important dares [2]. Some scientists concentrated on usage of less toxic reagents, catalysts, or routes [60-62]. In terms of catalysis front, there is an augmentation in the field of synthetic transformations assisted by supported catalysts [63, 64]. As an illustration, Wang, et al. [65] defined the fabrication of a reusable magnetic Ag–Fe3O4 catalyst supported on cellulose microspheres for reduction of nitrophenols. Further, there is an augmentation in examination of the usefulness of base metals in catalysis and
organocatalytic methods being realized under mild conditions and with low catalyst loading. Indeed, for highly enantioselective epoxidation of α,β-unsaturated ketones, Majdecki, et al. [61] used an amide-based Cinchona alkaloid at loadings as low as 0.5 mol % as a hybrid phase-transfer organocatalyst.

In terms of application, the demand for minimizing typically elevated E-factors in the pharmaceutical sector persists Ganesh, et al. [2], Andrews, et al. [66] and Isidro-Llobet, et al. [67] focused on the necessity to suggest less uneconomical and poisonous techniques for peptide and oligonucleotide synthesis and purification approaches, in light of the growing utility of biological peptide-based pharmaceuticals. Rossen [68] underlined insights from process chemistry that could diminish the effect of synthetic chemistry at smaller scales, and Hayler, et al. [69] presented a perspective from the pharmaceutical industry on the role of catalysis with Earth-abundant metals.

Catalysis using Earth-abundant rather than costly metals persists to be an active domain of experimentation and crucial component of sustainable chemistry testing [2]. Researchers worked on cobalt [70] and iron [71] catalysts for hydrosilylation and nickel precursors and heterogeneous catalysts for Suzuki–Miyaura coupling, one of the most largely employed processes for C–C bond generation. Further, Van Putten, et al. [72] observed the role of manganese in mediating C–C bond generation from organoboranes.

Further, waste lowering persists to be tackled in terms of solvent in suggesting alternative solvents, like less poisonous task-specific ionic liquids, deep eutectic solvents, and augmenting the field and efficacy of reactions in water (e.g., amidation, amine synthesis, and transfer hydrogenation [2]. There is as well the expansion of novel biomass-derived solvents with greener profiles like glycerol-derived 1,2,3-triethoxypropane and systematic solvent selection protocols for “greener” solvents [2].

4. DESIGNING CHEMICAL PRODUCTS WITH LOWER DANGER

Whilst reducing the influence of chemical agents out of usage of renewable resources and cleaner techniques stays crucial, the significance of the toxicological profile of the chemical products being circulated becomes yet more and more being realized [2, 73, 74]. The dare of efficaciously estimating the toxicological hazards of thousands of chemical products at the design step and for chemical agents already in usage needs interdisciplinary research efforts [75–77]. Several toxicological pathways of interaction stay defectively comprehended and thus unregulated Ghernaout and Elboughdiri [78]; Saiba, et al. [79]; Boucherit, et al. [80]. Chen, et al. [81] defined how in silico techniques of molecular dynamic simulations will assist to describe molecular initiation events and identify toxicological pathways, authorizing rapid and efficient screening of a broader collection of chemical products. Moreover, Kwiatkowski, et al. [82] suggested that individual testing of thousands of chemical products is impractical, and chemical “classes” have to be screened employing large subclasses as needed. Blum, et al. [83] furnished outstanding illustrations of the regrettable substitution of polybrominated diphenyl ethers with organophosphate ester flame retardants, conducting to unwanted toxicological consequences, and Kwiatkowski, et al. [82] adopted the scientific bases for dealing with perfluoroalkyl chemicals as a class.

The design of plastics to be both biodegradable and sustainably generated from non-petrochemical sources is one more crucial circular design domain requesting pressing research input Ganesh, et al. [2]. Napper and Thompson [84] announced a shortage of biodegradable plastic formulations utilized for carrier bags that displayed rapid rates of degradation juxtaposed to traditional plastic bags. Relating to ecological worries, Grbic, et al. [85] suggested a new and easy procedure to evaluate the amounts of microplastic fragments in environmental matrices, employing hydrophobic iron nanoparticles to extract microplastics from soil, sediments, and water magnetically [19, 21] and can be utilized as a sustainable remediation tool.

5. SUSTAINABLE WATER RESOURCES

The sustainable supply of drinking water remains a universal obligation Ganesh, et al. [2]; Ghernaout [87]; Ghernaout [88]. Ghimire, et al. [89] presented a crucial estimation and perspective of two main pathways for
promoting more sustainable and circular-economy-founded wastewater treatment and depicted that combining both concepts may lead to superior energy and resource efficiency for wastewater treatment systems Ghernaout, et al. [90]; Ghernaout and Elboughdiri [91]; Ghernaout and Elboughdiri [92]. Eggensperger, et al. [93] mentioned the usage of self-healing bacterially generated cellulose fiber networks producing sustainable biological membranes apt to filter water for potable water purposes. Bentel, et al. [94] announced that alternative energy-efficient and sustainable remediation methods comprising the implementation of ultraviolet (UV)/sulfite water treatments could furnish efficient decay of persistent with perfluoroalkyl chemicals in raw water for onward potable water use.

Further, environmentally sustainable and energy-efficient nanotechnology membranes possess water purification usages [95-97] frequently in integration with additional environmental usages, comprising energy harvesting, environmental sensing, and remediation [2, 98, 99]. Desalination techniques with superior high efficiency are founded on selective desalination membranes [11, 31, 100] and reduced graphene oxide membranes with minimal impact of nanowrinkles [101]. In addition to desalination, the decomposition of persistent organic pollutants stays as well a fundamental concern for water treatment. Pan, et al. [102] worked on the usage of a porous, coral-like nanostructure photoelectrode for such objective whilst jointly generating electricity. Shao, et al. [103] defined routes of carbo-catalysis in carbon nanotubes with persulfate oxidation for water treatment implementations.

6. CLOSING THE LOOP

The environmentally sustainable recuperation of resources from wastes is as well a flourishing domain of study Ganesh, et al. [2]. Chamas, et al. [104] described benchmarks for decay rates of usual plastics that will assist in prioritizing future study attempts on chemical decay to close the loop. Parker, et al. [105] investigated the depolymerization of ethylene with palladium catalysts to comprehend techniques to remediation of polyolefins. Liu, et al. [106] focused on the recuperation of lithium from spent batteries employing an acid-free mecanochemical method, which uses salt and sodium carbonate as the sole reagents. Wu, et al. [107] discussed the usage of selective sorbents for the elimination and recuperation of phosphates from wastewaters. Researchers [108] depicted the recuperation of nitrogen from sewage sludge and livestock manure as offering some circularity to the high global demand for nitrogenous fertilizers. Jun, et al. [39] suggested the capture and transformation of carbon dioxide to bicarbonate employing an environmentally easy setup of stabilized carbonic anhydrases loaded onto electrospun polymer nanofibers.

7. LIFE-CYCLE AND SYSTEMS THINKING

The possibility to implement life-cycle thinking to prioritize research and develop truly long-term, sustainable solutions remains crucial for defining potential routes to deal with circularity [2]. There are two frequent life-cycle assessment (LCA) methods (i.e., process LCA and economic input–output LCA), both of them are subjected to various limitations. Therefore, numerous combined methods are utilized to ameliorate the total accuracy. Luo and Ierapetritou [109] examined numerous integrated LCA methods over a case study of two biomass-based p-xylene fabrication techniques and suggested essential insights appropriate to future LCA. Moreover, LCAs of the production of broadly employed titanium dioxide nanoparticles [110] and butyl acetate [111] furnished conceptual directing for more sustainable synthesis of both. This procedure was used in suggesting the conversion of agricultural land in groundwater-stressed areas from irrigated crop types to rain-fed crop types combined with diversification to solar harvesting, so-called “agrivoltaics” [112].

In this work, the cited references typify the varied and interdisciplinary contributions relating to the topic discussed here [2]. They as well permit us to define fields that will demand more priority to greatly progress the domain toward implementations that smooth circularity [2]. Numerous of such fields are listed in Table 1.
8. PROCESS INTENSIFICATION (PI)

8.1. Definition

In the chemical engineering field, process intensification (PI) is described mostly in two principal manners: (1) PI greatly ameliorates transport rates, (2) it furnishes every molecule the identical processing chance [116, 117]. Such description could be viewed as being a process development implying greatly smaller tools that conducts to: (i) enhanced dominance of reaction kinetics furnishing bigger selectivity/reduced waste products; (ii) bigger energy efficiency; (iii) decreased capital costs; (iv) decreased inventory/improved intrinsic safety/fast response times [116].

In the heat transfer engineering, ‘intensification’ is similar to ‘enhancement’, and intensification is founded largely on active enhancement procedures, which are employed broadly in heat and mass transfer (HMT). Implementing PI successfully depends on the numerous techniques that may be employed to intensify unit operations and as well of many effective implementations [116].

The most frequently identifiable characteristic of an intensified process is that it is smaller, sometimes by orders of magnitude (ideally between 100- and 1000-fold [117]), than that it supersedes [116]. The structure distinctive to intensified processes (e.g., the pocket-sized nitric acid plant) runs to offer the decrease in scale possible, employing very great HMT enhancement. From such compactness of factory, cleanliness and energy-efficiency have tendency to be perceptible, especially in chemical processes and unit operations, at the same level with safety that is authorized by the implicit smaller inventories of what may be toxic chemical products that are passing through the intensified unit operations [116].

Throughout the 1970s, PI primarily caught remarkable interest in the chemicals domain, where it is most broadly known until now. Several PI techniques had been expanded before the phrase itself even occurred. As an illustration, any continuous process constitutes an intensification juxtaposed to the batch process [117] and the Podbielniak centrifugal contactor was suggested in the 1930s [116]. On the other hand, the benefits of PI are not usable only to the chemical industry but also in most process industries such as the food and pharmaceutical industries [116].

8.2. Applying Process Intensification (PI) For Environment

PI techniques possess very interesting ecological applications [117-119]. Indeed, a small, compact, greatly intensified factory is more possibly to be below the tree line, making it far less of an eyesore for the general public than the unsightly and massive steel works characterizing our current chemical factories [120-122]. Moreover, new reactor designs founded on the PI principle will authorize clean technique to be employed via letting waste reduction at origin [123-125]. In other words, high-selectivity operation in intensified reactors will diminish or banish completely the generation of undesirable by-products that if not eliminated from the effluent before discharge could provoke irreparable harm to the nature [126-128]. Thus, high-purity product will be acquired without enduring huge downstream purification prices [117, 129, 130].

The enhanced energy efficacy expected in intensified unit operations forms even an additional greatly interesting advantage of PI [117, 131, 132]. Indeed, PI is a favorable point for the chemical engineering [133-135].

Table 1. Future research needs in developing a circular economy [2].

| Field #1 | Description |
| --- | --- |
| Field #2 | Depolymerization and defunctionalization processes for present chemical products that could permit circularity, particularly for plastics. |
| Field #3 | Design of circular systems taking into account human health and ecotoxicity, ideally through rational design of benign commodity chemicals. |
| Field #4 | Systematic implementation of LCA, or thinking, and process metrics in promoting fresh manufacturing/synthetic procedures. |
| Field #4 | Usage of machine learning and other big-data techniques [113-115] to direct innovation toward novel paradigms of circularity. |
As mentioned above, considerable ameliorations in HMT could be reached in intensified units [136-138]. These ameliorations imply that process periods and so energy consumption could be greatly decreased for a given operation [139-141]. In addition, alternative energy sources (e.g., microwave and light energy [17]) that can be selected to wanted process chemistries, may conduct to less energy waste than is frequently faced with traditional thermal energy sources Ghernaout, et al. [142]; Ghernaout, et al. [143]; Ghernaout and Elboughdiri [144]. Boodhoo and Harvey [117] presented a case study focusing on the energy-saving capacity of a PI technique.

8.3. Dares of Applying Process Intensification (PI)

As a consistent philosophy, PI was primarily suggested since the 1970s; this is why it has not had more influence until these days [117]. For the shortage of large application at the industrial level, there are many causes. Table 2 lists most of them.

| Reason | Description |
|--------|-------------|
| Reason #1 | Perception of danger |
| Reason #2 | Shortage of 'champions' within industry |
| Reason #3 | Control/ Monitoring |
| Reason #4 | PI’s restrictions |

Table 2. Reasons for the lack of broad usage of process intensification (PI) [117].

Numerous of such dares are well known to anyone who has had a fresh technique to develop. The fresh technique should possess an established and considerable economic merit. Such economic merit is not only reached via diminishing the size of a piece of process equipment (or, preferably, the entire plant), but can be realized through some additional routes as listed in Table 3.

| Route | Description |
|-------|-------------|
| Safer operation | Enhanced product quality decreases the charge on downstream separations, possibly permitting a decrease in size, or molar techniques to be employed, or process stages to be avoided completely, that way diminishing capital and running prices in other areas of the plant Figure 1 [147]. The higher product quality generated in intensified methods thanks to the improved effectiveness of HMT diminishes the quantity of waste formed, herewith reducing the quantity that must be disposed of. In many cases, where the wastes are greatly poisonous, for example, treatment is one of the biggest running prices. |
| Product quality | The utmost performance of intensified factories diminishes energy prices. In a fossil fuel-founded energy supply system, as most currently are, this equates directly to decreased CO₂ emissions. These problems must be more and more serious since carbon trading becomes more standard. |

Table 3. Routes for bringing economic advantage besides reducing the size of the plant via applying process intensification (PI) [117].
9. CONCLUSION

To permit future contributions to underline the feasible implementation to circular economies, numerous interrogations could be viewed. First, think about the source of the materials being utilized, and if it is fossil-fuel-derived, see if there is a bio-founded alternative that may furnish the identical chemical insights. If elements are utilized that are encountering restricted supply, examine whether the synthetic techniques may aid in diminishing the usage of such materials. Second, consider whether the procedure defined may touch various steps of a chemical product’s life-cycle. As an illustration, is it a sage that is possibly reversible founded on kinetics and thermodynamics? If it forms by-products, could those be a worthy resource? And finally, think about whether the process or product forms a safe product that could be either recycled, reused, or remanufactured. Dealing with such interrogations suggests the engineers to use life-cycle thinking and to enlarge their attention to the prior art most closely related to the progresses depicted. Next fruitful contributions to such domain from the innovative specialists are more than wanted.

2. During the previous three decades, PI has attained noteworthy activity as a radical program to chemical processing. As a result, considerable advances have been noticed in the research and development of PI techniques. With major stress being put on sustainable development, PI could be a crucial component in making future chemical and pharmaceutical industries greener. This is because it could be employed to importantly decrease the size of numerous unit operations, and in doing so diminish not only the energy needed to run them but as well the surrounding infrastructure and the energy and materials utilized in their fabrication. Bigger selectivity and yields may lead to ameliorated atom efficiency and waste minimization at source, conducting to the decrease of downstream processing. Certainly, PI will remain and persist to participate in the expansion of the chemical and processing industry. However, some dares stay needing a planned labor from both engineers and industry partners before PI is more largely applied. How effective PI is in its implementation to a selected process depends greatly on the features of that process. An inherently slow process will not be made any faster via intensifying HMT properties. Therefore, it remains substantial to visibly sense the basis of a process prior any trial at intensifying it is performed. On the other hand, there are more process-related restrictions to the universal usage of PI like: (i) many processes run at too small a scale for PI to be economically worthwhile, (ii) several factories need a flexibility of operation that is not easy to attain in a continuous factory, which is usually purpose-designed. Regardless of such implementation restrictions and dares, the potency for PI stays considerably not yet exploited. The economic
recompenses for those plants that do introduce PI are likely to be substantial, and this will have ecological merits for a more sustainable future [117].

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