Bulgarian Chemical Communications, Volume 51, Special Issue B (pp. 119 – 123) 2019 DOI:10.34049/bcc.51.B.017

On the way to circular economy: a systems approach towards sustainable *Arctium Lappa* valorization via valuable medicinal products extraction

D. Sengupta[1], M. El-Halwagi[1*], R.P. Stateva[2], S. Santzouk[3], M. Papadaki[4]

[1] *TEES Gas & Fuels Research Center, The Artie McFerrin Department of Chemical Engineering, Texas A&M University College Station, Texas 77843-3122*

[2]*Institute of Chemical Engineering, Bulgarian Academy of Sciences, 1113 Sofia, Bulgaria*

[3]*Santzouk Samir and Co. General Partnership, PANAX, Chrissostomou Smirnis 14, Agios Konstantinos, Aetoloacarnania, GR30100, Greece*

[4]*Department of Environmental Engineering, Seferi 2, Agrinio, GR30100, University of Patras, GREECE*

Received: July 15, 2019; revised: July 23, 2019

In our work for the first time is outlined the application of a novel methodology to examine whether the roots of *Arctium Lappa*, commonly known as burdock, can be considered a potential feedstock for a biorefinery targeted to producing valuable medicinal products with a wide spectrum of applications. The biorefinery should implement predominantly mild, advanced green processes and techniques that will allow obtaining from the burdock roots the high value added products without damaging one or more of the extracts, particularly those that are heat sensitive. The biorefinery sustainability will be determined using life cycle and techno-economic assessment analyses. The residual biomass will be available for the production of further products of the same or potentially lower added value than the primary extracts from the roots. Via further steps, the entire mass of the plant can be used so that zero waste is possible.

Key words: *Arctium lappa*, pharmacological activity, biorefineries, life cycle assessment

INTRODUCTION

Arctium lappa is a traditional Chinese medicinal herb and an edible perennial plant of the *Asteraceae* family. Popularly known as burdock, this plant is native to Europe and Asia and was rapidly spread across North America by the early European settlers.

The different parts of the plant possess antioxidant [1-5], antibacterial [1, 5-7] and anti-inflammatory biological activities [5, 8-9] and are commonly used to treat various illnesses, such as throat infections, intoxications, skin infections and to relieve rheumatic pain and fever.

Recently, it was confirmed that *A. lappa* is capable of improving mucus protection in the stomach and intestines, besides preventing mucous injuries caused by alcohol [10]. Some burdock components have been reported to possess pharmacological activities like antioxidant, anti-inflammatory, antiproliferative and antiviral [11-15].

Burdock thus, can be a potential feedstock for a wide variety of chemicals. However, as an option for extraction of all of these components from the different parts of a single plant, the economic viability and environmental impacts for cultivation and conversion need to be studied for the plant species. This plant is known to originate from the northern parts of Europe and Asia, but also can be found around the world; because of its easy and rapid acclimatization. It is classified as an invasive weed in the majority of countries outside its native range, which makes it very suitable as a feedstock for the production of useful products as its utilisation will simultaneously form a method of control of its spread. At the same time, it develops to a rather large bush which allows the collection of substantial biomass from a single plant.

In our work for the first time, the application of a novel methodology for multi-objective optimization and systems integration to monetarizing burdock into added value products is sketched. It involves an outline of plant collection and the presentation of a simplified process for the production of added value products from the most valuable part of the plant, its roots. Added value products of this kind can greatly improve the viability of the further exploration of the respective biomass. Alternative/additional processes can be used for the extraction of added value products from other parts of the plant. The remaining biomass can be valorised towards the production of additional products. The valorisation of this biomass forms an unprecedented opportunity for the implementation and development of the-top-to-bottom, bottom-to-top coupled method, described in detail by Pham and El Halwagi [16]. The method can be also coupled with the atomic targeting and design as presented by El-Halwagi in [17]. Furthermore, the work of Sengupta and Pike [18] focusing on the

* To whom all correspondence should be sent:
E-mail: el-halwagi@tamu.edu

© 2019 Bulgarian Academy of Sciences, Union of Chemists in Bulgaria

production of different chemicals including biofuels from waste biomass provides the foundation for the further exploitation of this biomass and a methodology to follow towards that objective.

METHODOLOGY

However, as an option for extraction of all of these components from the different parts of a single plant, the economic viability and environmental impacts for cultivation and conversion need to be studied. Figure 1 displays a macroscopic, overall procedure, which can form a starting point for the development of a network of options, the viability of which needs subsequently to be examined.

The selection of the feedstock is the first step. Feedstock can be collected either as a weed from abandoned uncultivated land or obtained as a cultivated product. In the former case, the cost of the land and its cultivation can be considered negligible; however, the control over the desired quantity and probably quality of the feedstock may be questionable and it is greatly condition depended. Cultivated on-purpose burdock is not subject to all of the above drawbacks. However, the costs of cultivation (including irrigation, fertilizing of land, other weed and insects controls, potential weather protection means) add to the costs of its production. Of course a combination of the above two sources of origin could potentially form a much better solution.

The collection of the feedstock is another important aspect. Burdock roots are the most important part of the plant. Thus, deep digging may be necessary rendering the accessibility of the land and the capacity to dig out the burdock plants - an issue of key importance. The cultivated crop fields can be designed in advance, in a way which makes crop collection easier, while the selection of the land for the cultivation can be such, so as to minimalise the transportation costs. Of course, freely grown burdock plants may also be accessible and Figure 1.

Feedstock selection, collection and pretreatment scheme be encountered in locations where transportation is easy.

Depending on the timeline and the procedures followed, the collection of the feedstock and the conditions of its transportation may impose that preservation and pretreatment of the feedstock is necessary, either as steps immediately following collection of the plants or as steps subsequent to their transportation activity.

The best time for the feedstock collection is in the summer, when most of the above ground sections of the plant are almost dry, but the roots are still containing a high amount of moisture. The plants can be dug out and left to dry outside, exploiting the solar energy and contributing to the viability of the process.

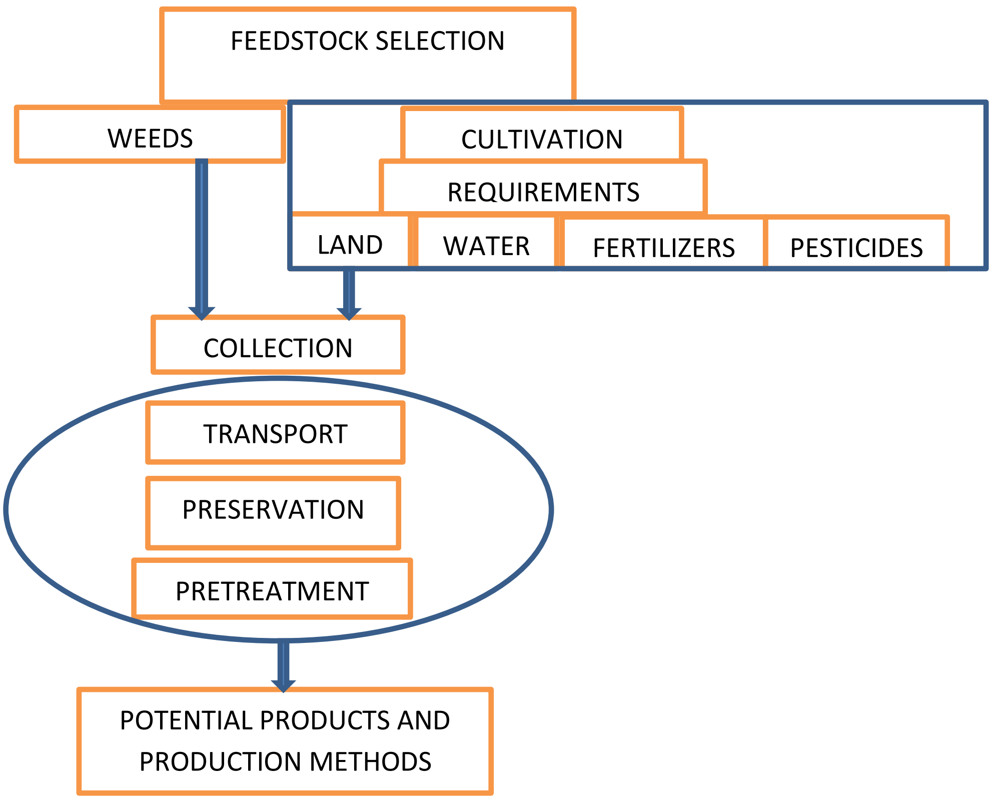

Fig.1: Feedstock selection, collection and pretreatment scheme

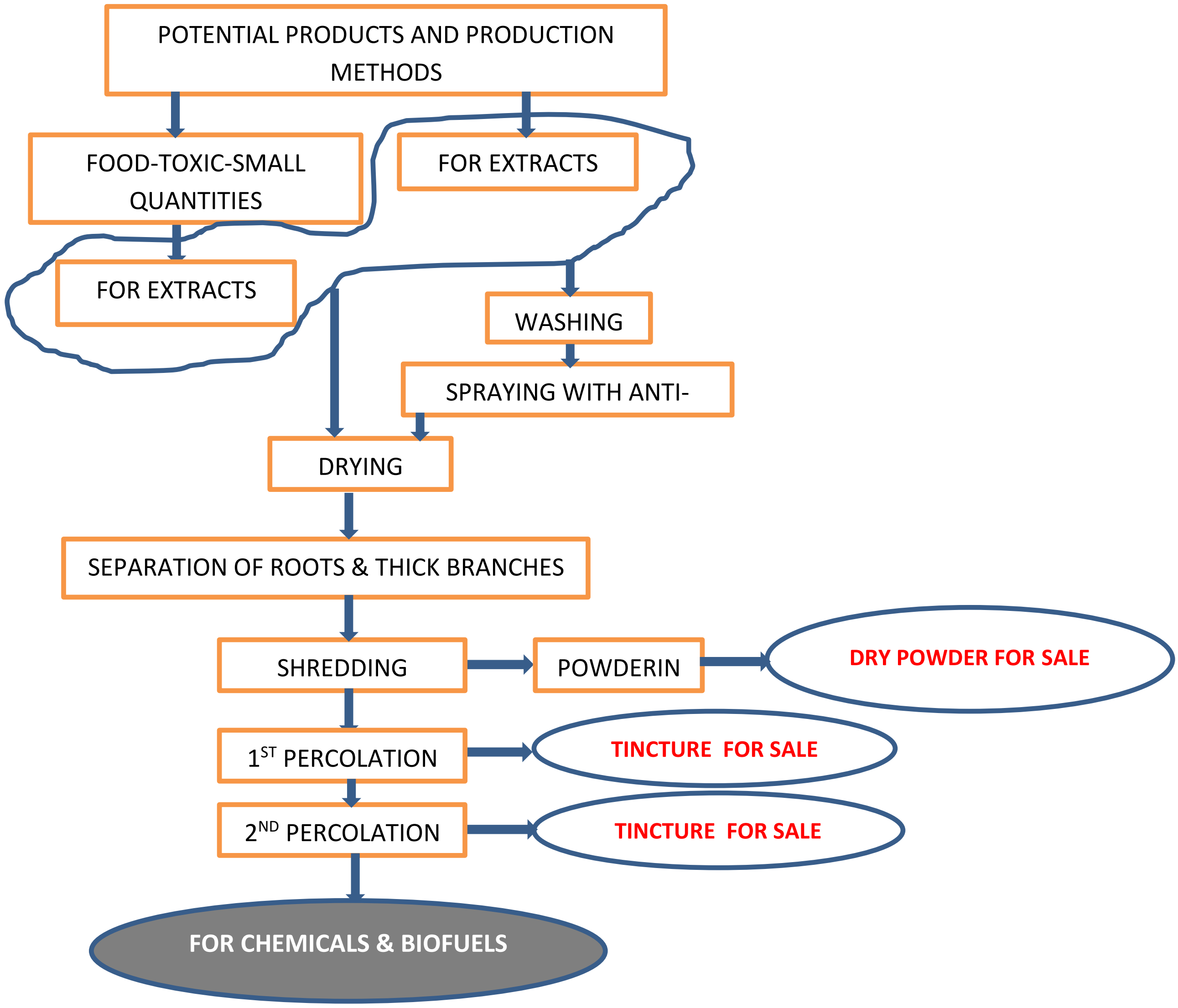

Fig. 2: Valuable natural medicines production steps

Moreover, mobile units can be employed for the treatment of parts of the plant, so that only part of it needs to be transported to a central unit for further treatment.

Figure 2 shows the necessary subsequent steps for the production of added value products.

The plants can be dried as collected, or washed first and then dried. In the latter case, spraying of the plants with a mold preventing solution is necessary. In the former, the need of such mold prevention pretreatment depends on the conditions *i.e.* the ambient moisture and whether a possibility of fungi growth on the plant exists. For high quality medicinal products and providing that the measures to prevent mold development are in place, the drying of the plants is better to be done in an open air-shaded place. The use of furnaces is not recommended.

The dried plants are then separated into leaves and roots; thick branches are collected with the roots. They are consequently shredded. The shredded parts can be either milled further to form a powder which can be sold or used for the preparation of formulations, or can be used for making tinctures *of arctium lappa*. The latter is done through percolation employing 60° ethanol in water solution. After removing the liquid extract, a second percolation can follow using a 1:10 by weight mixture of ethanol:wet solid. The tinctures and powders obtained via this procedure possess excellent, powerful, but at the same time gentle for the body medicinal properties. Being very valuable products can have a significant contribution towards the viability of an integrated process which subsequently valorizes the remaining biomass from this plant.

At present, burdock is underused and there are just a few studies in the literature devoted to the obtainment, characterization and application of its valuable extracts. Burdock root extractions with supercritical carbon dioxide ($scCO_2$) and compressed propane, characterization and biological activities of the extracts, influence of process conditions (pressure and temperature) and solvent effects on the chemical composition, total phenolic content and antioxidant activity were reported in [1,2].

In order to examine a multitude of alternative techniques to the same feedstock (burdock leaves), de Souza *et al.* [19] studied the capabilities of pressurized liquid extraction with hydroalcoholic solutions with different mass fractions of water, and compared those to the results obtained by: i) a sequential six step supercritical extraction with ethanol as a co-solvent and ii) a supercritical extraction procedure where distilled water was added to the ethanol as an auxiliary solvent, referred to as $scCO_2$+Aq. EtOH procedure. Furthermore, the PLE burdock leaves extracts contained higher concentrations of chlorogenic acid and rutin and exhibited considerably high DPPH free radical scavenging activity [19].

Major compounds found in the burdock root extracts were diisooctyl phthalate (DIOP) and 2,3-Dihydro-3,5-dihydroxy-6-methyl-4H-pyran-4-one (DDMP), glycerol, methyl oleate, butanoic acid and pentadecanal. Analogous experiments were performed on the areal parts (leaves) of the plant by de Souza *et al.* [2]. A number of important phenolic compounds like lupeol acetate, amyrin acetate, diisooctyl phthalate and phytol were identified in the extracts obtained and it was concluded that supercritical extraction with ethanol as a co-solvent has a potential as a viable technique.

The promise to replace fossil sources employing the remaining waste biomass of burdock as a feedstock allows a step towards circular economy, and expands the potential to establish an alternative supply chain which can be further studied in a wholesome mode through life cycle assessment.

The feasibility of a biorefinery including decisions on scale, processes, products, water use, and waste management through the entire plant, needs to be studied by means of a techno-economic assessment. Finally, the environmental impacts and the safety of the processes need to be quantified for the processes that convert burdock to products, including transportation of the species to the biorefinery.

Therefore, burdock can be a potential feedstock for a wide variety of chemicals. However, as an option for extraction of all of the aforementioned components from a single plant, the economic viability and environmental impacts for cultivation and conversion need to be studied for the plant species. The multiobjective framework will be applied, where potential tradeoffs are considered. The production of high added value products via the exploitation of its roots and thick stems as shown schematically in Figures 1 and 2 should be the first priority. However, for the production of high value pharmaceuticals, organic, good quality plants are an obvious necessity. Less qualitative plants together with the leftover biomass from the aforementioned extraction process can be used for the production of valuable chemicals. For that purpose, the valuable compounds identified to be contained in burdock biomass by previous research have to be quantified. The waste biomass after those processes can be further used for biofuel production via anaerobic digestion for instance, or to they can be further treated for the formation of adsorbents, compost, etc.

CONCLUSION

The ultimate goal of this work is to study whether burdock can be considered as a potential feedstock and to determine the viability using life cycle assessment and techno-economic assessment analyses.

Using process integration, the traditional approaches can be modified if it is infeasible, or to identify product and production pathways which will improve its feasibility.

For that purpose, a systems-based “forward-backward” approach as outlined by Pham & El Halwagi [16] will be followed. The forward part aims at identifying the possible intermediates while the reverse synthesis starts with desired products and moves “backwards” for the identification of pathways leading to them. The optimization process will involve the formation of sub-problems. For each of them, the available technologies and the best policy will be examined. Subsequently, appropriate programming algorithms will be developed to determine the optimal pathways.

Acknowledgements: *This project has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 778168*

REFERENCES

1. A.R.C. de Souza, A.R. Guedes, J.M. Folador Rodriguez, M.C.M. Bombardelli, M.L. Corazza. *J. Supercrit. Fluids,* **140**, 137 (2018).
2. J.M.F. Rodriguez, A.C. de Souza, R.L. Krüger, M.C.M. Bombardelli, M.L. Corazza, *J. Supercrit. Fluids*, **135**, 25, (2018).
3. W. Liu, J. Wang, Z. Zhang, J. Xu, Z. Xie, M. Slavin, X. Gao, *Int. J. Biol. Macromol,*. **65,** 446 (2014).
4. Y. Jiang, J. Yu, Y. Li, L. Wang, L. Hu, L. Zhang, Y.

Zhou, *Int. J. Biol. Macromol.,* **123,** 531 (2019)

5. R. Claudiu, M.I. Georgiev, I. Fierascu, C. Ungureanu, S. Marius, A. Ortan, M. Ioana, A. Nicoleta, A. Zan, C.E. Dinu-pirvu, B. Stefan, V. Anuta, *Food Chem. Toxicol.,* **111**, 44 (2018).
6. R. Oliveira, R.B. de A. Almeida, P. das G.F. Vilela, F.E. Oliveira, R.F. Rocha, A.O.C. Jorge, L.D. Oliveira, . *Oral Biol.,* **9**, 3 (2014).
7. Z. Lou, H. Wang, W. Lv, C. Ma, Z. Wang, S. Chen, *Food Control.,* **21**, 1272 (2010).
8. A.B.A. de Almeida, M. Sánchez-Hidalgo, A.R. Martín, A. Luiz-Ferreira, J.R. Trigo, W. Vilegas, L.C. Dos Santos, A.R.M. Souza-Brito, C.A. De La Lastra, *J. Ethnopharmacol*., **146**, 300 (2013).
9. X. Wu, Y. Yang, Y. Dou, J. Ye, D. Bian, Z. Wei, B. Tong, L. Kong, Y. Xia, Y. Dai, *Int. Immunopharmacol.,* **23**, 505 (2014).
10. Y.C. Wu, L.F. Lin, C.S. Yeh, Y.L. Lin, H.J. Chang, S.R. Lin, M.Y. Chang, C.P. Hsiao, S.C. Lee, *Fooyin J. Health Sci*., **2**, 26 (2010).
11. A. Li, J. Wang, M. Wu, X. Zhang, H. Zhang, *Eur. J. Pharmacol*., **747**, 71(2015).
12. M.K. Cho, J.W. Park, Y.P. Jang, Y.C. Kim, S.G. Kim, *Int. Immunopharmacol*., **2**, 105 (2002).
13. Y.P. Jang, S.R. Kim, Y.H. Choi, J.Kim, S.G. Kim, G.J. Markelonis, T.H. Oh, Y.C. Kim, *J. Neurosci. Res.,* **68**, 233 (2002).
14. Y. Jeong, J. Park, D. Kim, H. Kim, *Biomol. Ther.*, **22**, 497 (2014).
15. H. Liu, Y. Yang, X. Cai, Y. Gao, J. Du, S. Chen, *Pharm. Biol.*, **5**, 1 (2015).
16. V. Pham, M. El-Halwagi. *AIChE Journal*, **58**, 1212 (2012).
17. M. El Halwagi, *Process Integration and Optimization for Sustainability,* **1**, 3 (2017).
18. D. Sengupta, R. W Pike. *Chemicals from Biomass: Integrating Bioprocesses into Chemical Production Complexes for Sustainable Development, CRC Press* (2012)
19. A.R.C. de Souza, S. Stefanov, M.C.M. Bombardelli, M.L. Corazza, R.P. Stateva, *The Journalof Supercritical Fluid*s, in press, https://doi.org/10.1016/j.supflu.2019.104573