

Life cycle assessment of nature-based coagulant production: Light and dark sides of the freeze-drying process

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ABSTRACT

Different review articles published in the last several years highlighted an importance and benefits of bio-coagulants/biofloculants utilisation in water and wastewater treatment as an eco-friendly and non-toxic alternative to currently used commercial ones. Although there is many research on various types of bio-coagulants/biofloculants (animal-, microorganism- and plant-based ones), there is still a significant knowledge gap which limits their utilisation at large scale. In order to achieve optimised solution several production options should be evaluated and compared in terms of their simplicity, economic viability, ecological impacts and their efficiency. When compared, these characteristics could lead to easier decision making and developing bio-coagulant/biofloculants with optimal *eco-design*. In order to compare 4 coagulant production processes for novel biocoagulant production (from common bean seed – *Phaseolus vulgaris*), life cycle assessment, cost analysis and performance within wastewater treatment were conducted in this study, while simplicity of each design is indicated as well. 4 distinct powdered coagulants were denoted as: KNO (conventional solid/liquid extraction with NaCl solution, spray dried), UNO (ultrasound extraction with NaCl solution, spray dried), GA (ultrasound extraction with NaCl solution, spray dried with gum Arabic as a carrier) and UVO (ultrasound extraction with distilled water, freeze dried). Based on the defined functional unit, KNO coagulant was the most eco-friendly and cost-beneficial coagulant, while UVO coagulant showed the highest coagulation ability. However, UVO coagulant has the most challenges within environmental deterioration, due to high electricity demand for freeze drying process. Although spray drying process showed ecological and economic benefits, high quality of freeze drying process should not be neglected and optimisation and comparison at full scale might be subject of future study. The present study also indicated at which points production process could be optimised by scale-up.

1. Introduction

Deterioration of environment caused by poor wastewater quality calls for significant activities and improvements in the wastewater management sector (Abdessemed et al., 2003; Radovic et al., 2023a). High turbidity, suspended solids and COD (Chemical Oxygen Demand) in wastewater contribute to environmental pollution directly (Shak and Wu, 2015) or they can interfere another wastewater treatment process and deteriorate other micropollutant removal (Dhangar and Kumar,

2020), but those parameters could be drastically reduced by simple coagulation/flocculation (CF) technology.

CF was firstly used over 100 years ago and could serve as an adequate treatment or pre-treatment which can help develop advanced strategies for sustainable wastewater treatment (Jiang, 2015; Kurniawan et al., 2022). The main mechanisms involved in CF are charge neutralisation, polymer bridging, sweep flocculation and double-layer compression (Owodunni and Ismail, 2021). Those mechanisms are triggered by the addition of coagulants/floculants, which in industry are still of the

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chemical origin. Commercial coagulants, are cheap and efficient, but could cause serious health issues and negative impacts on the environment (Teh et al., 2016). Those could be overcome by development of novel bio-based coagulants (biocoagulants) of animal, microorganism or plant origin (Kurniawan et al., 2022). They are beneficial in comparison to commercial ones, due to their nontoxicity, biodegradability, sustainability, availability of the feedstock, less sludge formation and other (Ahmad et al., 2022; Ang and Mohammad, 2020). However, certain limitations for their commercial/industrial use still exist (Ang and Mohammad, 2020; Kurniawan et al., 2022). One of limitations is their stability and durability in liquid state, which could be overcome by conversion into powdered form. Hence, it could be said that making powdered form of biocoagulants is necessity. Conversion could be achieved by several drying techniques such as spray drying, freeze-drying and spray freeze drying (Karthik and Anandharamakrishnan, 2013). Although there is a lot of examples, especially of plant-based coagulants (Othmani et al., 2020; Owodunni and Ismail, 2021), there is still limited research on their powdered phase production. Most of the research was conducted on *Moringa oleifera* biocoagulants (Katayon et al., 2006; Mohamed et al., 2015; Mohammad et al., 2013; Noor et al., 2015, 2013; Silva et al., 2019). As it can be seen from literature review spray and freeze-drying are far the most used drying technologies in the segment of powdered biocoagulant production.

Freeze-drying process has been abundantly used for conditioning of materials containing thermo labile components (Shukla, 2011), which is why this drying process was assumed better for common bean seed biocoagulant production, comparing to spray drying process which is working at high temperatures (over 100 °C) which can cause denaturation proteins (Katayon et al., 2006) that are the main biocoagulant component with coagulation ability. As it works on low temperatures under vacuum pressure it has numerous advantages such as preservation of material thermo labile components (such as peptides, proteins and complex synthetic organic molecules) and production of high-quality product (Franks, 1998). The main concerns about freeze-drying process is energy demand, which should be estimated for each utilisation and optimised if possible. However, energy consumption was highlighted as process hot spot even in situations where another techniques were used for biocoagulant/biofloculant production (Carlqvist et al., 2020). Biocoagulant production by freeze-drying process has not been tested yet for environmental impacts.

An importance of future research approach including production optimisation and production and application at industrial-scale was also highlighted as a step toward biocoagulant widespread utilisation (Kurniawan et al., 2022). Additionally, production should be optimised in order to go along with sustainable development and initiatives of the eco-design. Eco-design is defined as an approach which aims to reduce environmental impacts of products or services through integration of environmental aspects into products or service development (Vallet et al., 2013). In order to do so, life cycle analysis (LCA) has been widely applied in wastewater treatment systems (Corominas et al., 2013). LCA could quantitatively assess environmental impacts providing support in making decision on alternative operational scenarios, hence could be useful in optimisation of different processes within the water sector (Corominas et al., 2020), including optimisation of biocoagulant production conditions emphasised as future perspective in biocoagulant research (Kurniawan et al., 2022). Production of raw material (common bean seeds) was previously investigated by Abeliotis et al. (2013). Environmental impacts of the biocoagulant production process and utilisation were investigated in detail herein, while end phase of the product life cycle was theoretically discussed. Disposal or utilisation of generated sludge after CF process presents the end of the biocoagulants life cycle. It is already emphasised that the sludge could be converted in a product with added value and used in agriculture (Alnawajha et al., 2022). As different sludge can have different physical-chemical, nutritional and hazardous/toxic properties (Feria-Díaz et al., 2016), future studies should include evaluation of the possibility to use biocoagulant

from common bean seeds as a fertilizer or soil improver in agriculture by thorough investigation of listed sludge characteristics.

To sum up, the main novelties of this study include: representation of the overall eco-design of the novel biocoagulants made from common bean seeds (environmental, cost and performance analysis of four biocoagulant production processes) as well as thorough comparison of four production processes based on the same functional unit with highlighted comparison between spray and freeze-drying technology utilised for this purpose. Although the study is conducted at lab-scale, with all inputs (ecological, economical and performance aspects) and suggestions for optimisation at higher scale, it could serve as valuable decision making tool and base for thorough investigation at pilot or industrial level. Benefits and limitations, especially environmental hot spots of the production process are highlighted indicating future perspective and need for improvements in this area of research. This research/case study covers geographic region of Serbia and the experiments were carried out from 2019 to 2023 in the laboratories at the Faculty of Technical Sciences and the Faculty of Technology Novi Sad, University of Novi Sad, Serbia.

2. Materials and methods

2.1. Life cycle assessment (Goal and scope)

The main objectives of the LCA of freeze-dried biocoagulant production were:

- To identify production process hot spots;
- To compare environmental impact of freeze-drying and spray drying process used for the same purpose and help in future decision making;
- To investigate possibility and extend of process optimisation.

Thorough evaluation based on the set objectives of LCA, would serve as tools the optimisation of the biocoagulant production process, leading to more environmentally-friendly products. Furthermore, comparison of different types of the biocoagulant production would serve as valuable direction in utilisation of more eco-justified production process.

100 m³ of treated wastewater (ww), whose turbidity was reduced for 55.2–71 %, has been chosen as functional unit (FU). WW volume has been selected as FU in order to enable comparison between obtained coagulants and their production processes. 100 m³ of wastewater represents volume that can be treated during 2 shifts in one working day if we assume that wastewater treatment facility has hydraulic capacity of 5–6 m³/h. Referent flows (Table 1), indicating starting points for building LCA models were calculated for each coagulant based on the

Table 1
LCI Background processes.

Flow	Flow property	Unit	LCI Dataset	Geography
Electricity	Energy	kWh	market for electricity, low voltage	RS
Maize starch	Mass	kg	market for maize starch	GLO
Sodium chloride	Mass	kg	market for sodium chloride, powder	GLO
Tap water	Mass	kg	market for tap water	Europe without Switzerland
Tissue paper	Mass	kg	market for tissue paper	GLO
Deionised water	Mass	kg	market for water, deionised	Europe without Switzerland
Waste graphical paper	Mass	kg	market group for waste graphical paper	Europe without Switzerland
Wastewater	Volume	m ³	market for wastewater, average	Europe without Switzerland

selected FU. The following equation was used for the calculation of the referent flows:

$$\text{Referent flow(kg)} = OD \times DM \times 100000 \text{ m}^3 \quad (1)$$

Where OD represents optimal coagulant dosage used in jar tests (mL/L) and DM is dry matter of the coagulant (g/mL). DM for KNO, UNO, GA and UVO coagulants are 47.23, 46.14, 46.14 and 17.16 g/L, respectively.

The system boundaries (Figs. 1 and 2) for the production of 3 coagulant types by spray drying process have already been explained in details in previously published work of (Radovic et al., 2023b). The only change was the addition of the preparation phase which includes return of the powdered coagulant to its liquid state before jar test experiments (details explained in Section 2.2 performance study). On the other hand, system boundaries for LCA of freeze-dried coagulant production have been presented for the first time in present work. As it can be seen from the schematic view (Figs. 1 and 2), freeze-drying process includes less instruments and material and energy flows, than spray-drying process.

Fig. 3 shows simplified schematic diagram of freeze-drying process.

Due to the negligible environmental impacts, the production of raw material (common bean seeds) was not considered within system boundaries (production of 1 kg of bean generates 0.127–0.438 kg CO₂ eq. (Abeliotis et al., 2013)). Three step production process includes: mechanical processing (grinding and screening), extraction and freeze-drying/spray drying phase. Preparation phase is the same as

explained for spray drying.

First two production stages and spray drying have been previously explained in detail in our previous work (Radovic et al., 2023b). Briefly, raw material (common bean seeds – *Phaseolus vulgaris*) has been purchased from the local market, milled and screened. The need for the extraction and further processing of the biomaterial comes from the fact that biocoagulant in its raw form (only grinded common bean seeds) is not as effective as processed one. The proteins are the main component of the raw material that have coagulation ability, hence, by extraction we concentrate those making biocoagulants with higher performances. Fraction below 0.4 mm in diameter was used for further extraction process (with ratio of 1:20, material to extraction agent). Fraction above 0.4 mm (appx. 50 % of former material) could be further used as fertiliser or feed, which is why this material flow was not considered in the study (Ali et al., 2010). The main compounds with coagulation activity are proteins and polysaccharides (Ang and Mohammad, 2020). In order to extract those compounds different extraction agents could be used, such as distilled water or salt solutions. Liquid coagulants dried by spray drying process were obtained by either conventional solid-liquid either ultrasound extraction, but all with 0.5 M NaCl solution, instead of distilled water. During conventional liquid/solid extraction the smaller bean fraction was suspended in extraction agent solution and stirred for 10 min with the magnetic stirrer (Antov et al., 2010). In the case of ultrasound extraction, liquid suspensions were treated in ultrasonic bath with constant frequency of 40 kHz for 50 min at 25 °C. After extraction,

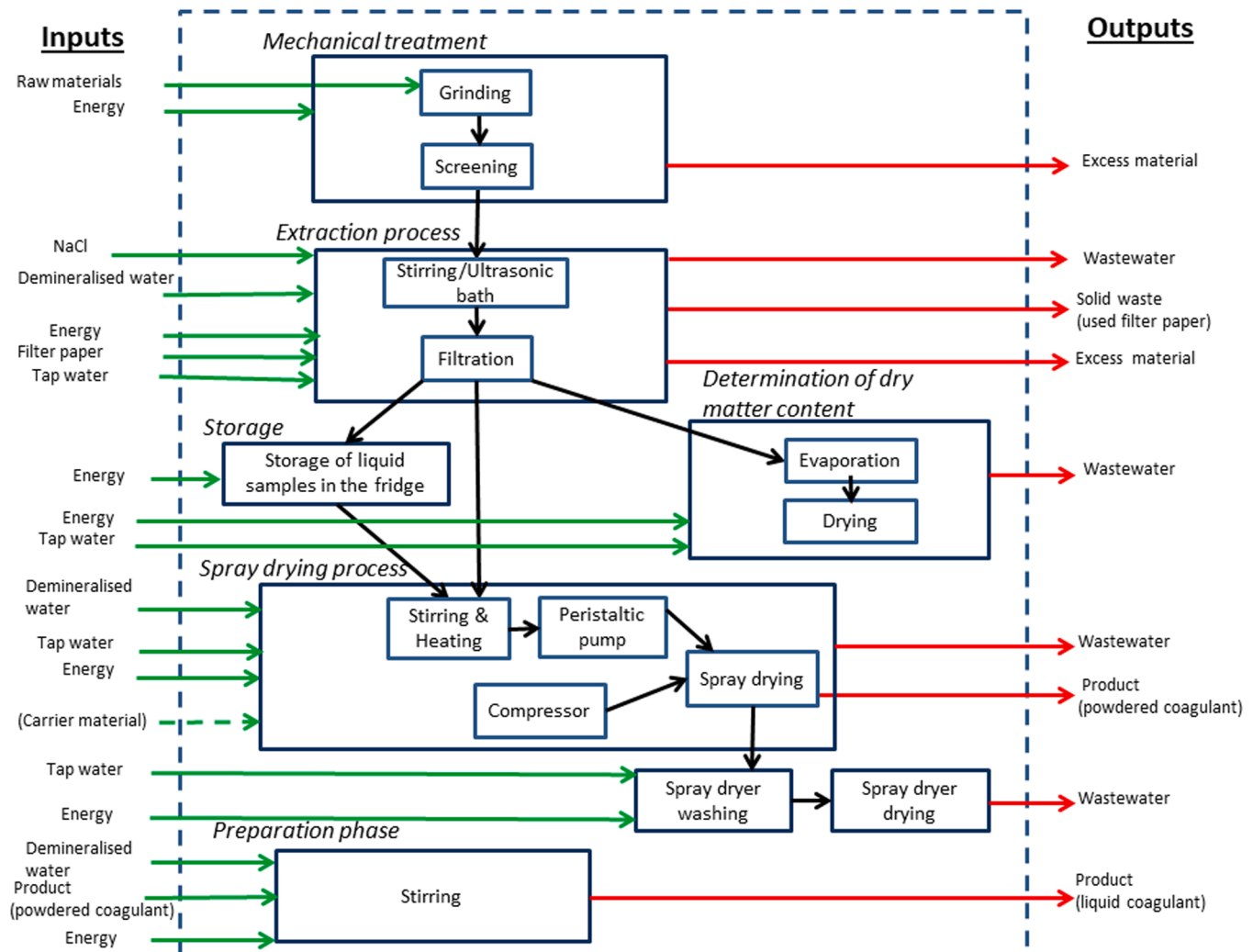


Fig. 1. System boundaries for the three phases of coagulant preparation by spray drying process (Radovic et al., 2023b), partially changed.

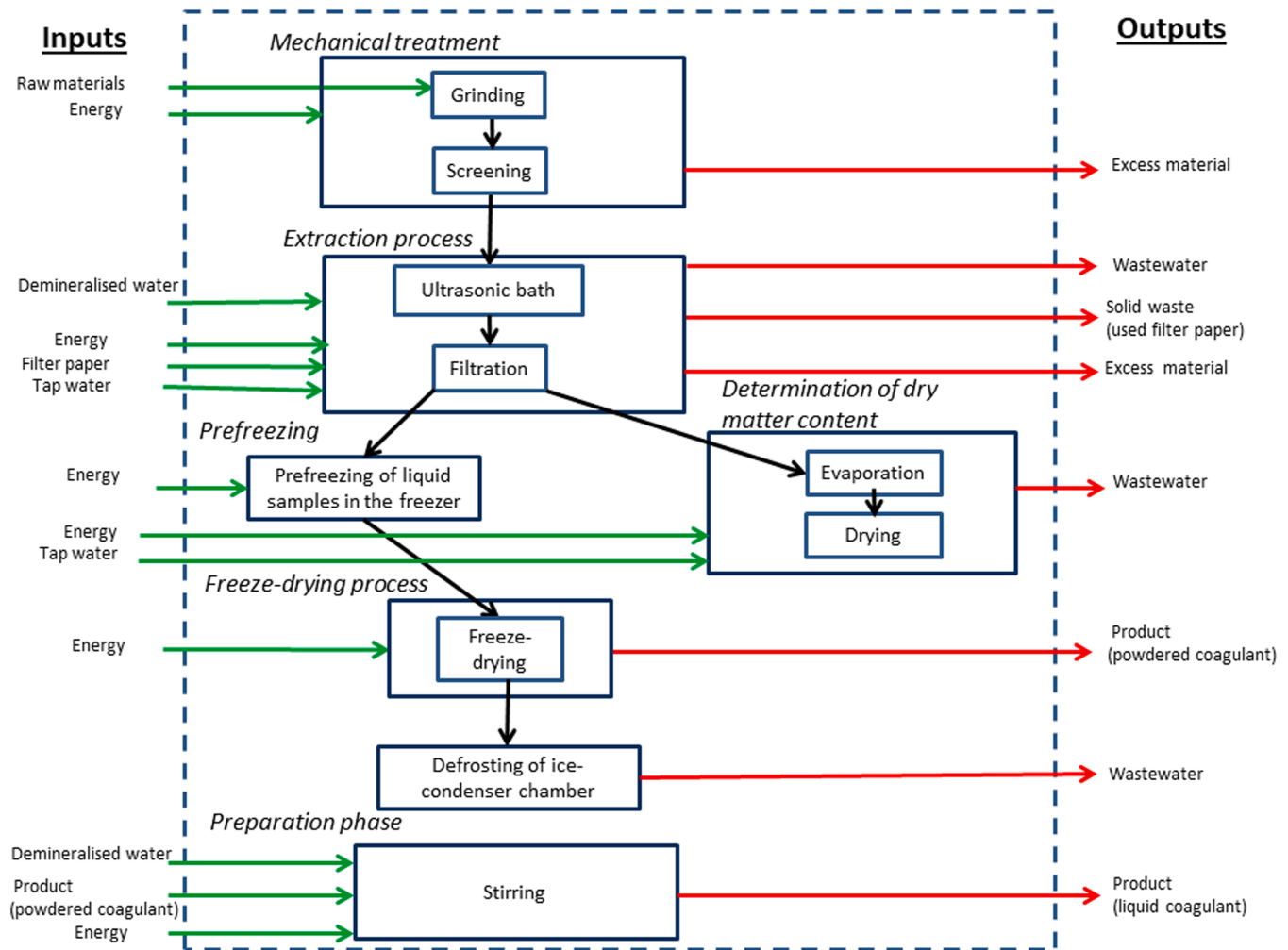


Fig. 2. System boundaries for the three phases of coagulant preparation by freeze-drying process.

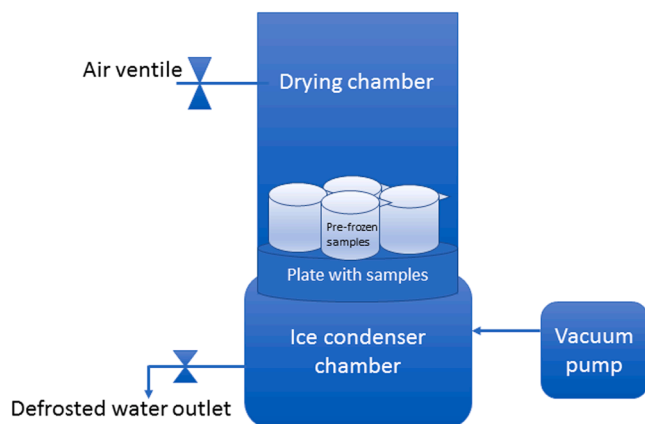


Fig. 3. Simplified schematic diagram of freeze-drying process.

suspensions were filtered through rugged Macherey – Nagel MN 651/120 filter paper. Filtration was done in Bihner's funnel connected with a vacuum pump. When filtered, all three types of liquid extracts were dried under same conditions by spray drying process, in Anhidro laboratory spray drier at 120–125 °C and pressure 3.2 bar (Radovic et al., 2023b).

Considering coagulants made by freeze-drying process, the only difference in preparation of liquid extracts is that the extraction agent

used within ultrasound extraction process was distilled water, not the salt solution such as in previously explained case. Hence, ultrasound extraction has different operation conditions. It lasted for 45 min at 42 °C, under the same frequency of 40 kHz. Filtration was also done, as previously described. However, further processing and production of powdered coagulants is completely different from spray drying. Before drying extract by freeze-drying, there is an additional step that samples have to undergo. Before the third phase, liquid extracts have to be frozen. During freezing step, it is important to cool the samples below the material's eutectic point (the lowest temperature that allows the coexistence of both, solid and liquid material phase) in order to ensure sublimation instead of melting in further drying steps (Shukla, 2011). Adequate freezing leads to optimised drying process. Freezing could be done in the freeze drier directly, however, that would lead to additional costs as freeze drier consummates a lot of energy for its working and the exact freezing time has to be previously determined for all the materials independently (Karthik and Anandharamakrishnan, 2013). Hence, freezing was, in this case, done in Gorenje freezer. 600 mL glass bakors were filled with sample up to 1/6 of their volume and put in the freezer overnight. Frozen extracts were then freeze-dried in Alpha 1–2 LDplus freeze-drier. Extracts were dried under vacuum pressure of 0.07 Pa and temperature –45 °C for 48 h. Selected drying conditions were selected based on the previous experience and research (not published data).

Based on the type of extraction, type of drying process (freeze- or spray drying) and type of spray drying process (with or without carrier), powdered coagulants obtained, evaluated and compared within present study were denoted as UVO (powdered coagulant produced by

ultrasound extraction, distilled water as extraction agent and dried in freeze-dryer), KNO (powdered coagulant produced by conventional liquid/solid extraction and dried without carrier), UNO (powdered coagulant produced by ultrasound extraction and dried without carrier) and GA (powdered coagulant produced by ultrasound extraction and dried with 100 % gum Arabic as a carrier). The benefit of produced biocoagulants is that they trigger both, coagulation and flocculation mechanism and do not call for additional chemical addition.

2.2. Performance study

CF procedure was conducted in jar tester. For experimental purposes, model water (200 NTU and pH 6) was prepared by 1 % kaolin suspension diluted with tap water and pH was set with analytical grade HCl. The preparation of 1 % kaolin suspension meant addition of 10 of kaolin to 1 L of tap water, followed by 1 h of stirring in order to achieve uniform dispersion of kaolin particles. The suspension was finally left for 24 h so as to kaolin particles completely hydrate (Antov et al., 2012). Although dynamic of jar test was previously described in the work of Radovic et al. (2023b), the procedure could be briefly showed as: different biocoagulant dosage (0 (blank sample), 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 1, and 1.5 mL/L) were added to model water (200 mL) and stirred fast (200 rpm) for 1 min and then slowly (60 rpm) for another 30 min, after which they were left to settle for the next 1 hour. 50 mL of upper clarified liquids were collected from each sample and those samples were tested for turbidity levels. In order to inject coagulants into the system (wastewater), coagulants have to be returned from their powdered into liquid form, which was done by simple suspension of certain amount (determined by dry matter content) of powdered coagulant into distilled water followed by moderate stirring during 10 min. The amount of powdered coagulant suspended/dissolved in 10 mL of distilled water, used for experiments conducted on the same day were 0.4723, 0.4614 and 0.1716 g for KNO, UNO, GA and UVO coagulant.

In order to calculate coagulation activity (%) expressed as turbidity removal, the following equation was used:

$$\text{Coagulation activity} = \frac{(C_b - C_s)}{C_b} \times 100 (\%) \quad (2)$$

Where C_b and C_s (mg/L) represent turbidity of the blank and the samples, respectively.

2.3. Cost analysis

In order to compare 4 evaluated coagulants production from economical aspect, the cost of each production process has been calculated at lab-scale basis. It is important to note that those estimation serve as indication of the most economically favourable coagulant from 4 tested ones, but the exact numbers could be lower by up-scaling production process to industrial level. Material and process flows which figured in cost estimation were: cost of raw material (common bean seed), NaCl, distilled water, tap water (according to the cost of 1 m³ of water for commercial use for legal entities, estimated by JKP Vodovod i kanalizacija (<https://www.vikns.rs/cenovnik/>, 13.11.2023.)), filter paper, electric energy (based on the price of electric energy for commercial use, <https://www.eps.rs/lat/snabdevanje/Stranice/objasnjenje-racuna-ks.aspx>, 10.11.2023.) and cost of carrier material (gum Arabic) for the production of GA coagulant.

2.4. Life cycle inventory (LCI)

Table 1 shows the background processes, applied datasets from Ecoinvent LCI database with cut off system modelling (version 3.7).

According to FU and coagulant activity, the following referent flows were calculated for each coagulant, using Eq. 1 (Table 2).

Table 2

Referent flows according to FU.

Coagulant	Referent flow (kg)
UNO	1.024
KNO	0.264
GA	5.562
UVO	1.716

Tables 3–6 show life cycle inventory for each coagulant, including three steps of the coagulant production (grinding and screening, extraction and spray drying) as well as preparation of the powdered coagulant for the utilisation (returning to liquid phase).

2.5. Life cycle impact assessment

For calculation and modelling of LCA in this research, openLCA software version 1.10.3 (openLCA 2024) and Ecoinvent database version 3.7 (Wernet et al., 2016) were used. For life cycle impact assessment (LCIA) CML method was used (Guinée, 2002) with updated characterisation factors from year 2016 (CML 2016). Results from LCIA were calculated for eleven impact categories: abiotic depletion (kg Sb eq), abiotic depletion (fossil fuels) (MJ), acidification (kg SO₂ eq), eutrophication (kg PO₄ eq), freshwater aquatic ecotoxicity (kg 1,4-DB eq), global warming (GWP100a) (kg CO₂ eq), human toxicity (kg 1,4 DB eq), marine aquatic ecotoxicity (kg 1,4 DB eq), ozone layer depletion (kg CFC-11 eq), photochemical oxidation (kg C₂H₄ eq), and terrestrial ecotoxicity (kg 1,4-DB eq).

3. Results and discussion

3.1. LCIA results

LCIA results for four coagulants (KNO, GA, UNO and UVO) and four processes within coagulants production and preparation for use are provided in Fig. 4. Drying phase in production process of all obtained coagulants (KNO, GA, UNO and UVO) had the highest environmental impact through all impact categories. Between 60 % and 80 % of impact through most of impact categories for KNO and UNO coagulant originated from spray drying phase. In case of ozone layer depletion and terrestrial ecotoxicity impact categories drying process contributed with approximately 40 % of all environmental impact from the production of those two coagulants. Returning of KNO and UNO powdered coagulants to their liquid state before conducting jar test experiments (preparation for use), contributed with around 20 % to environmental deterioration concerning all impact categories. Extraction phase had less influence on the environment. The only two impact categories, which were more notably influenced by extraction phase were ozone layer depletion (almost 40 %) and terrestrial ecotoxicity (around 30 %), while grinding

Table 3

LCI for KNO coagulant.

	Grinding and screening	Extraction	Spray drying	Preparation for jar test	Total
Electricity	0.079	3	69.59	23.1	95.769
Maize starch	0	0	0	0	0
Sodium chloride	0	0.3	0	0	0.3
Tap water	0	26	120	0	146
Tissue paper	0	1.4	0	0	1.4
Wastewater	0	0.03	0.12	0	0.15
Deionised water	0	14	10	5.6	29.6
Waste graphical paper	0	1.4	0	0	1.4

Table 4

LCI for GA coagulant.

	Grinding and screening	Extraction	Spray drying	Preparation for jar test	Total
Electricity	0.079	34.9	692.56	498.3	1225.839
Maize starch	0	0	5	0	5
Sodium chloride	0	3	0	0	3
Tap water	0	620	1020	0	1640
Tissue paper	0	14	0	0	14
Wastewater	0	0.665	1.02	0	1.685
Deionised water	0	145	120	120.55	385.55
Waste graphical paper	0	14	0	0	14

Table 5

LCI for UNO coagulant.

	Grinding and screening	Extraction	Spray drying	Preparation for jar test	Total
Electricity	0.32	13.27	275.13	91.76	380.48
Maize starch	0	0	0	0	0
Sodium chloride	0	1.17	0	0	1.17
Tap water	0	160	420	0	580
Tissue paper	0	5.6	0	0	5.6
Wastewater	0	0.175	0.42	0	0.595
Deionised water	0	55	40	22.2	117.2
Waste graphical paper	0	5.6	0	0	5.6

Table 6

LCI for UVO coagulant.

	Grinding and screening	Extraction	Freeze drying	Preparation for jar test	Total
Electricity	0.79	36.75	12690.6	413.3	13141.44
Maize starch	0	0	0	0	0
Sodium chloride	0	0	0	0	0
Tap water	0	665	20	0	685
Tissue paper	0	14	0	0	14
Wastewater	0	0.765	0.112	0	0.877
Deionised water	0	200	0	100	300
Waste graphical paper	0	14	0	0	14

and screening phase had negligible impact to environmental deterioration in comparison to other production phases. Similar situation was observed for GA coagulant. Spray drying impact was between 40 % and 60 %, impact from preparation phase was between 25 % and 40 %, while extraction phase had less environmental influence and grinding and screening affection was negligible. Within UVO coagulant production, the environmental impact of the drying phase (freeze drying) was the most dominant, more than 90 % for all impact categories. Electricity consumption in spray drying process (KNO, UNO and GA coagulant) and freeze drying process (UVO coagulant) was the main cause of high impacts of drying phase on the environment.

Fig. 5 and Tables S1-S11 of Supplementary Material provide comparative LCIA results for tested coagulants by each impact category. Herein the impact of electricity consumption is more obvious. The

highest effect of electricity consumption was observed for marine aquatic ecotoxicity impact category. Namely, $3.10 \times 10^{+02}$, $4.82 \times 10^{+06}$, $1.50 \times 10^{+06}$ and $5.16 \times 10^{+07}$ kg 1,4-DB eq of impact originated from KNO, GA, UNO, and UVO coagulant, respectively. Another impact category highly affected by electricity consumption was abiotic depletion (fossil fuels), where 8.24×10^{-1} , $1.28 \times 10^{+04}$, $3.97 \times 10^{+03}$ and $1.37 \times 10^{+05}$ MJ of impact originated from KNO, GA, UNO and UVO coagulant, respectively. On the other hand, the lowest impact was observed for ozone layer depletion impact category, where 4.04×10^{-10} , 6.26×10^{-06} , 1.94×10^{-06} and 6.71×10^{-05} kg CFC-11 eq correspond to KNO, GA, UNO and UVO coagulant, respectively.

Beside electricity consumption for spray and freeze drying processes, wastewater disposal caused the second highest impact on the environment. Namely, to eutrophication ($2.04 \times 10^{+00}$, $2.29 \times 10^{+01}$, $8.07 \times 10^{+00}$ and $1.19 \times 10^{+01}$ kg PO₄ eq originating from the production of KNO, GA, UNO and UVO coagulants, respectively), abiotic depletion (fossil fuels) ($6.66 \times 10^{+02}$, $7.48 \times 10^{+03}$, $2.64 \times 10^{+03}$ and $3.89 \times 10^{+03}$ MJ originating from the production of KNO, GA, UNO and UVO coagulants, respectively), marine aquatic ecotoxicity ($1.16 \times 10^{+02}$, $1.30 \times 10^{+03}$, $4.59 \times 10^{+02}$ and $6.76 \times 10^{+02}$ kg 1,4-DB eq originating from the production of KNO, GA, UNO and UVO coagulants, respectively) and global warming potential impact categories ($7.13 \times 10^{+01}$, $8.01 \times 10^{+02}$, $2.83 \times 10^{+02}$ and $4.17 \times 10^{+02}$ kg CO₂ eq originating from the production of KNO, GA, UNO and UVO coagulants, respectively). On the other hand, wastewater disposal had the lowest impact to ozone layer depletion (5.08×10^{-09} , 5.70×10^{-08} , 2.01×10^{-08} and 2.97×10^{-08} kg CFC-11 eq impact from KNO, GA, UNO and UVO coagulant, respectively).

3.2. Cost analysis

Coast analysis (Table 7) of all 4 types of coagulant production revealed that the most cost-beneficial production, based on the FU, is one of the KNO coagulant (5778.1 RSD/FU), while the most cost-beneficial production based on 1 kg of the obtained product is GA coagulant (16762.8 RSD/kg).

3.3. Performance study

To fully compare four obtained natural coagulants, it is important to evaluate their performance. Fig. 6 shows coagulation activity of KNO, UNO, GA and UVO coagulants, in terms of turbidity reduction from model water. Maximum coagulation activities with standard deviations (and corresponding optimal coagulant dosages) were 55.2 ± 3.8 % (0.1 mL/L), 68.3 ± 0.32 % (0.4 mL/L), 61.5 ± 0.5 % (1 mL/L) (Radovic et al., 2023b) and 71 ± 2.1 % (1 mL/L) for KNO, UNO, GA and UVO coagulant, respectively.

3.4. Discussion

All four coagulants made from common bean seeds have double role in coagulation process as once added to the water, they serve as both, coagulants and flocculants. This valuable behaviour should be highlighted as it positively affects cost of water treatment and need for chemicals / natural materials. Hence, the comparison with other studies concerning both coagulants and flocculants could be made to the certain extent. As the literature is sparse with LCA of coagulant/flocculants, this study's goal was not only to identify production hot spots for each coagulant, but also to expend investigation and compare different drying processes used for the same purpose. As already stated in our previous work (Radovic et al., 2023b), electricity consumption, especially for drying process within coagulant production stood out as the most influential part of the production process on the environment. As it can be seen from the current results, this trend was observed for both types of drying, spray and freeze drying. However, freeze drying (UVO coagulant production) showed noteworthy higher environmental impact

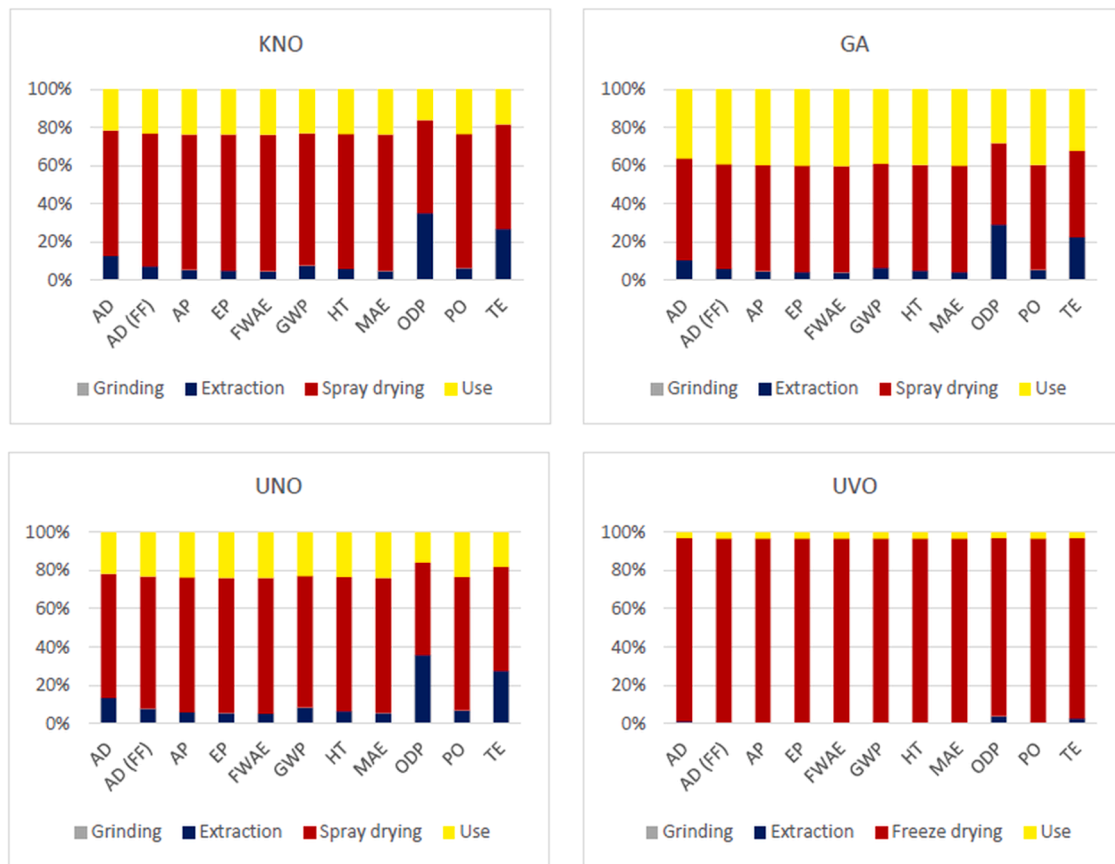


Fig. 4. LCIA results for KNO, GA, UNO and UVO coagulants (AD abiotic depletion, AD (FF) abiotic depletion (fossil fuels), AC acidification, ET eutrophication, FE fresh water aquatic ecotoxicity, GWP global warming potential, HT human toxicity, ME marine aquatic ecotoxicity, ODP ozone layer depletion, PO photochemical oxidation, TE terrestrial ecotoxicity).

(around 90 % of all impact originated from freeze drying process) in comparison to spray drying process used for all other coagulants production (KNO, GA and UNO) where 40–80 % of all impact originated from drying phase and through all impact categories. The lowest impact was observed for GA coagulant due to the utilisation of carrier materials within spray drying phase, which has different benefits explained in our previous work (Radovic et al., 2023b). Although freeze drying is common energy demanding process it is abundantly used in different industries due to its high performance in conservation of heat-labile products and their components. In the present study, those components are proteins (the main active constituents of coagulants made from common bean seeds). Hence, it is important to further investigate possibility to optimise freeze drying at large scale. It is important to mention that in current study, spray dryer was of half-industrial scale, while freeze dryer was of laboratory scale, hence, lower amounts of coagulant extracts could be dried in already time consuming freeze drying process (48 h for 0.5 L) than during spray drying (40 min for 0.5 L). In larger freeze dryers, for the same time, much higher amounts of extracts could be dried. High environmental impacts from the electricity use within production of coagulant/flocculant was also emphasised in the study of Carlqvist et al. (2020) indicating that high energy consumption does not necessarily comes from the drying stage of production process, but can also come from production of chemicals needed for the production process.

Impact categories mostly affected by electricity consumption in our study were marine aquatic ecotoxicity and abiotic depletion (fossil fuels) which are both connected with extraction and utilisation of fossil fuels for energy production. On the other hand, negligible effect on ozone layer depletion comes from the fact that there is no noteworthy emissions caused by two drying processes. Also, chemical utilisation within

production process is very poor, which also goes in favour of low influence on atmospheric and deterioration of the environment, in global.

Wastewater disposal mostly affected water bodies, promoting eutrophication and marine aquatic ecotoxicity due to the uncontrolled discharge of water containing different waste materials. Global warming potential and abiotic depletion (fossil fuels) were also affected. The most obvious impact through all four mentioned impact categories originated from the GA coagulant, hence, it could be assumed that the utilisation of carrier material mostly influenced wastewater characteristics and its influence on the environment.

Cost analysis showed that the highest price (per FU and kg of obtained coagulant) is connected with UVO coagulant production and it predominantly comes from the extensive use of electricity (Table 7). It is worth mentioning that cost analysis is done at lab-scale; to roughly compare production processes of the 4 coagulants from the economic aspect and it is just an indication of the real price, which could be optimised at real scale production. On the other hand, KNO was the most cost-beneficial production due to the lower electricity demand required by simple conventional solid-liquid extraction and most importantly due to low referent flow, which indicates that the lowest amount of coagulant (in comparison to UNO, GA and UVO) is needed for treating the same amount of wastewater (1 FU). However, it is important to mention that KNO coagulant showed lower turbidity removal in comparison to other coagulants. For instance, there is a 15.8 % increment in coagulation activity when UVO coagulant was applied (UVO coagulant showed the best coagulation activity which indicates benefits of drying at lower temperatures and conservation of heat-labile, active components of natural coagulants).

We have recently published some of the steps toward practical application of the biocoagulants produced by freeze drying process

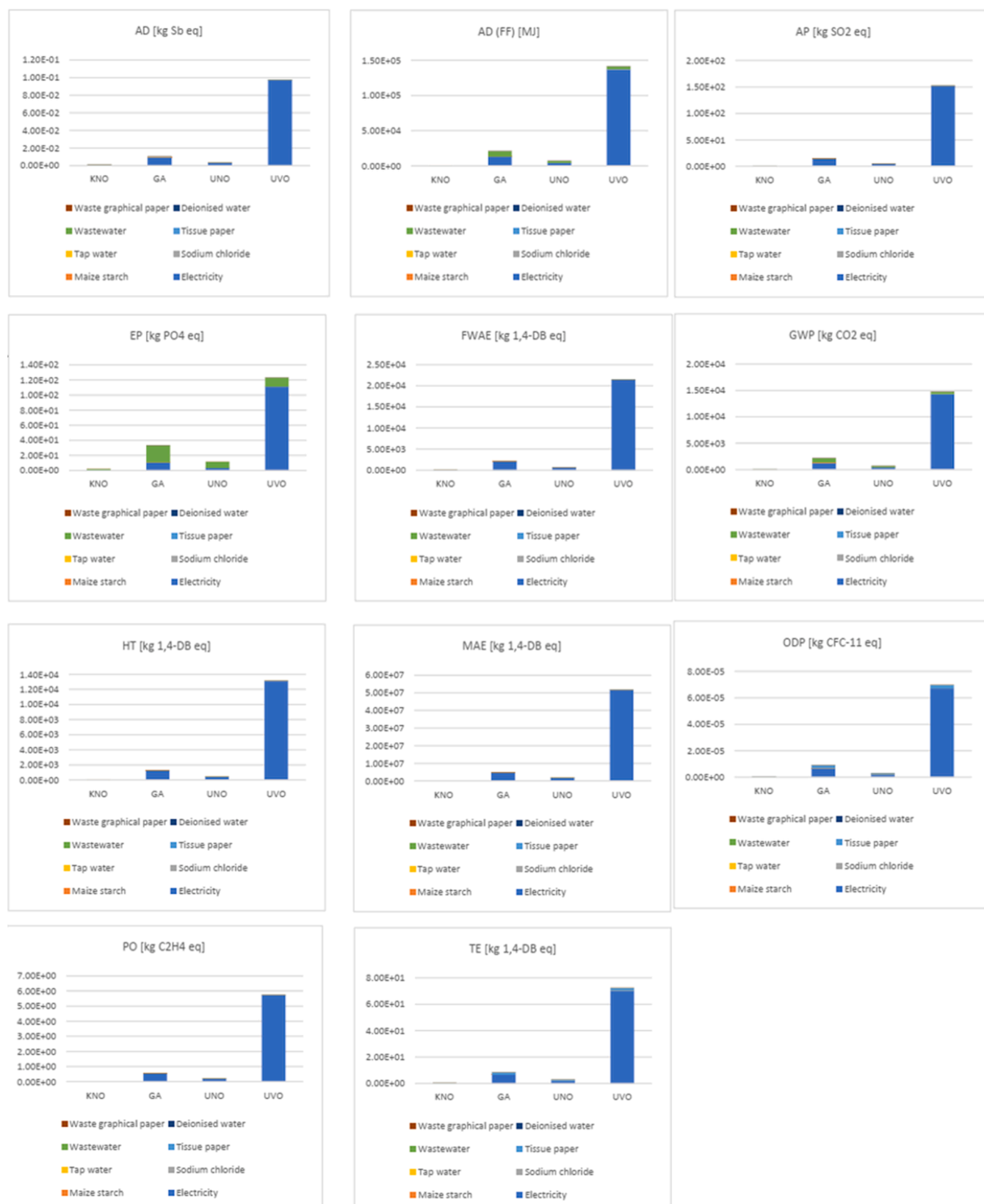
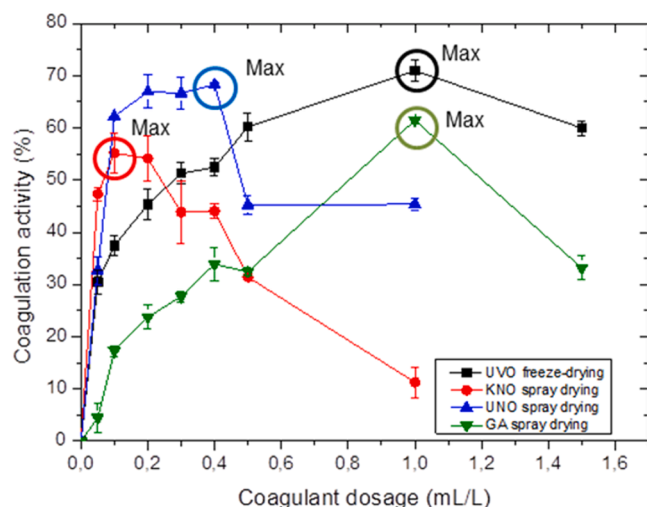


Fig. 5. LCIA results for KNO, GA, UNO and UVO coagulants (AD abiotic depletion, AD (FF) abiotic depletion (fossil fuels), AC acidification, ET eutrophication, FE fresh water aquatic ecotoxicity, GWP global warming potential, HT human toxicity, ME marine aquatic ecotoxicity, ODP ozone layer depletion, PO photochemical oxidation, TE terrestrial ecotoxicity).

Table 7

The price of coagulant production per FU and per kg of obtained coagulant.

Coagulant	UVO	KNO	UNO	GA
Material				
Common bean seeds (RSD)	3000	300	1200	3000
NaCl (RSD)	0	57	222.3	570
Distilled water (RSD)	28000	3360	13300	37100
Filter paper (RSD)	7872	787.2	3148.8	7872
Water (RSD)	180.8	38.5	153.1	432.8
Electric energy (RSD)	216378.4	1235.4	4908.2	12380.2
Carrier material (RSD)				31879.7
Sum (RSD/FU)	255431.2	5778.1	22932.4	93234.7
Sum (RSD/kg)	148852.7	21886.7	22394.9	16762.8

**Fig. 6.** Coagulation activity (turbidity removal) of UVO (freeze-drying), KNO (spray drying), UNO (spray drying) and GA (spray drying) biocoagulant in model water (200 NTU, pH 6).

(Cojbasic et al., 2024), while some results considering spray dried biocoagulants are yet to be published. Mentioned article also possess some of the more detailed properties of the biocoagulants such as their stability and durability, however, those properties were not of specific relevance or influence on production process. On the other hand, one of the important properties which could influence maintenance of the equipment for the biocoagulant production is the addition of carrier material. Liquid coagulant extracts that are enriched with carrier material are less sticky and could prevent clogging of different equipment parts (pipes for extract inflow to spray drier, atomiser etc.). Another difference between coagulant properties is whether they are produced using salt solution or distilled water as an extraction agent. However, there was no indication that NaCl from the salt solution influenced production process. More detailed analysis could be one of the tasks for the future studies.

This study is another contribution which express viability of this concept. However, scale-up of the production process should be done in future studies. There is a justified assumption that transferring the production process from lab to large-scale would lead to certain changes in coagulant production preferences. For instance, freeze drying process could be optimised by drying in large freeze driers, due to the fact that for the same time greater amount of coagulant could be dried/produced. Hence, electricity consumption could be reduced. Some of the ideas for further investigation include thorough technology research and comparison (different freeze driers or spray driers comparison for large scale production) as well as inclusion of different electricity sources (using another electric grid or some more eco-friendly solutions – green energy).

4. Conclusion

Based on all three evaluated aspects of coagulant production process (ecological, economical and performance) it could be seen that there is no unique solution and sole coagulant which is the most beneficial. Hence, it is important to evaluate what are the main purposes of the coagulants. Either way, the results from current study are important first step for determination of the most beneficial technology for coagulant production. It is confirmed that the freeze drying process is energy demanding and causes higher environmental impacts than spray drying process used within production of novel powdered coagulant from common bean seeds. Energy demands consequently affected cost of coagulant production. However, coagulant produced by this drying technique (UVO) had the highest turbidity removal ability, which should not be neglected.

Main strength of this research is that it provides new information on LCA of biocoagulant production process by freeze drying process. Furthermore, presented research includes comparison of four biocoagulant production process from the environmental, cost, and performance perspectives. On the other hand, major limitation of this research is that it is performed on the laboratory scale. In order to obtain the most realistic picture of coagulant preferences, it is important to transfer current study to the larger scale.

Ethical approval

This article does not contain any studies with human participants or animals performed by any of the authors.

CRediT authorship contribution statement

Sanja Cojbasic: Writing – review & editing, Writing – original draft, Visualization, Investigation, Data curation, Conceptualization. **Boris Agarski:** Writing – review & editing, Validation, Software, Resources, Methodology. **Djordje Vukelic:** Writing – review & editing, Software, Resources. **Maja Turk Sekulic:** Writing – review & editing, Supervision, Resources, Project administration. **Sabolc Pap:** Writing – review & editing, Validation, Supervision. **Marija Perovic:** Writing – review & editing. **Jelena Prodanovic:** Writing – review & editing, Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.indcrop.2025.120699](https://doi.org/10.1016/j.indcrop.2025.120699).

Data availability

Data will be made available on request.

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