Citation	Nico Deconinck, Koenraad Muylaert, Wilfried Ivens, Dries Vandamme (2018), Innovative harvesting processes for microalgae biomass production: a perspective from patent literature Algal Research, vol 31, 469-477
Archived version	Author manuscript: the content is identical to the content of the published paper, but without the final typesetting by the publisher
Published version	http://dx.doi.org/10.1016/j.algal.2018.01.016
Journal homepage	https://www.journals.elsevier.com/algal-research
Author contact	dries.vandamme@uhasselt.be ; koenraad.muylaert@kuleuven.be
IR	https://lirias.kuleuven.be/handle/123456789/614186

(article begins on next page)

Innovative harvesting processes for microalgae biomass production: a perspective from patent literature

Nico Deconinck¹, Koenraad Muylaert², Wilfried Ivens³, Dries Vandamme^{2,4*}

¹Belgian Patent Office, Patent Information dep., Vooruitgangstraat 50, 1210 Brussels, Belgium

²KU Leuven Campus Kulak, Laboratory for Aquatic Biology, E. Sabbelaan 53, 8500 Kortrijk, Belgium

³Open Universiteit, Faculty of Management, Science and Technology, 6401 DL, Heerlen, The Netherlands

⁴UHasselt – Hasselt University, Institute for Materials Research (IMO-IMOMEC), Applied and Analytical Chemistry, Agoralaan-gebouw D, B-3590 Diepenbeek, Belgium

* Corresponding author: Email: dries.vandamme@uhasselt.be

Tel: +32 11 26 8318

Abstract

The harvesting of microalgae for biofuel production consists of a primary concentration step, followed by a separation step to isolate the microalgal biomass from its aquatic environment. Recent research focussed mainly on the technological feasibility of various separation processes. However, to what extent these innovative harvesting strategies have been commercialized and therefore have led to actual innovation in the current microalgae biotech industry by the creation of intellectual property, has remained unexplored. This study reviews the scientific literature based on technological, economical and environmental criteria of 13 primary and 8 secondary harvesting methods. Commercial deployment was evaluated via patent analysis. Auto- and co-flocculation, as well as sedimentation, overall scored best for economical (CAPEX and OPEX) and environmental (energy and GHG) criteria, while belt filters scored the highest on the technological criteria (TSS). Hence, only 4 patents based on auto-/co-flocculation, sedimentation and only two for belt filtration are still in force. Technologies based on organic, electrolytic and magnetic flocculation seems to be more successfully patented. Since patenting involves making the technology freely available for others, small but sometimes crucial improvements in low-tech systems may be often kept as a company secret instead. So far, no single harvesting process with superior feasibility has emerged for application on a large commercial scale. This is mainly due to the difference in relative importance of technological, economical and environmental criteria for each harvesting process dependent on the used strain and the final products.

Keywords: microalgae, harvesting, dewatering, concentration, IP, biofuels

1. Introduction

Microalgae biomass production has gained increasing global interest in the search for renewable resources for a sustainable, bio-based economy. Microalgae are considered as the most promising feedstock for biofuels, but it will still take several years to develop production processes that are both sustainable and economical [1]. Meanwhile, alternative high-value products derived from multiple microalgal components, are further explored ([2]; [3]). Microalgae can be grown onto non-fertile soils in ponds or photo-bioreactors, in marine or brackish waters using N and P from wastewater resources. Over the last decade, substantial research efforts have resulted in increased microalgal biomass productivity. However, because of ineffective water and nutrient recycling combined with energy-intensive harvesting, the production of microalgal biofuels is currently not competitive with fossil fuels ([4]; [5]). Because microalgae are small and grow at low concentration in culture, biomass harvesting by conventional separation processes is expensive, which hampers economical microalgal biomass production on a commodity scale ([6]; [7]).

Microalgal harvesting consists of a concentration and a separation process to produce an algal cake, paste or sludge of 15 to 25% or more dry solids from a dilute biomass of 0.02–0.06% dry solids. Harvesting is often divided in primary and secondary concentration steps. Primary concentration methods assist in thickening of the microalgal biomass slurry up to 1–5% in order to facilitate the separation from their culture medium. Further dewatering of the biomass requires an additional step, generally referred to as secondary concentration. This concentration step can

produce a microalgal sludge with an average concentration of about 200 g L⁻¹. Generally, concentration techniques are based on physical, chemical or biological processes. Physical concentration techniques apply mechanical or electrical forces to concentrate the microalgal biomass. Ultrasonic waves and electrolysis are used to destabilize the microalgal cells ([8]; [9]; [10]). Chemical techniques make use of inorganic or organic additives to enhance coagulation, or for example (nano)particles with magnetic properties to neutralize the microalgal negative charge for coagulation [11]. Finally, concentration techniques that are based on biological processes to induce spontaneous or natural flocculation, are generally referred to as bioflocculation [12]. These methods do not require additional chemicals but rely on interactions with bacteria, fungi or even with other microalgae species for co-flocculation. Usually, these processes are followed by a secondary dewatering step based on filtration, enhanced sedimentation, centrifugation or flotation [6]. The technological feasibility of most of these separation processes for harvesting microalgae has been experimentally validated in several original studies, and reviewed extensively in technical overviews and techno-economic analyses ([7]; [13]; [14]). However, it is currently not well documented to what extent these novel harvesting strategies have been commercialized and therefore have led to actual innovation in the current microalgae biotech industry by the creation and maintenance of intellectual property.

The aim of this study was to provide an overview of harvesting technologies for microalgae biomass production that have been patented worldwide over the last years. First, a detailed overview of several technological and economical parameters for several harvesting methodologies is given based on scientific literature, followed by a detailed patent analysis to overview currently expired and protected microalgal harvesting processes.

2. Materials and Methods

A. Literature review

The 21 most studied harvesting techniques were selected and compared, i.e. 13 primary concentration/separation and 8 secondary concentration/separation techniques based on technological criteria (strain limitation and final total suspended solids concentration (TSS)), economical criteria (capital expenditures (CAPEX) and operational expenditures (OPEX)), and environmental criteria (energy demand and greenhouse gas emission (GHG)).

The scientific literature was screened in order to obtain information about the final biomass concentration after separation, expressed as total solid suspension (TSS). A high TSS means that the harvesting technology is efficient in terms of concentration, consequently leading to high water removal. The capital expenditure (CAPEX) is the capital investment in equipment while the operational expenditure (OPEX) represents the operational cost. These costs were (a) obtained directly from scientific studies or (b) determined by a relative comparison with other harvesting techniques. For harvesting by disk stack centrifugation, dissolved air flotation, electrolytic flocculation, bioflocculation and sedimentation (c) both CAPEX and OPEX were calculated based on the following equation (1), which represents the total harvesting costs (P_c) for the production of 1 m³ ([15]; [16]):

$$P_c = \frac{((0.5 \cdot I/100 + M/100) \cdot C \cdot A) + C}{W \cdot A \cdot Q_c} + \frac{R_c}{Q_c} \quad (1).$$

Wherein I = interest rate (% of investment): 6%, M = maintenance cost (% of investment): 2%, C = investment cost (EUR), A = amortization (years); which is the number of years that someone has to pay off for the investment: 10, W = working hours in a year (h): 8400, Q_c = capacity ($m^3 h^{-1}$), R_c = running cost of the system (raw material + energy consumption (EUR h^{-1})). CAPEX was calculated by eliminating M and R_c/Q_c in the equation.

The energy consumption (energy) is the energy in kWh required to achieve the given biomass concentration per m^3 . The amount of greenhouse gas emissions (GHG) for each method was expressed as the amount of produced CO₂ per required energy unit (g CO₂-eq MJ⁻¹). When no direct data were available, conversion was based on literature studies with the amounts CO₂ produced in relation to the distance (g CO₂-eq 100 km⁻¹), and converted using the formulae:

$$\frac{\frac{10 \text{ kg CO}_2}{100 \text{ km}}}{\frac{1 \text{ MJ}}{0.39 \text{ km}} \cdot \frac{1 \text{ kg}}{1000 \text{ g}}} [17] \text{ or by dividing the reported amounts kg CO}_2\text{-eq ton}^{-1} \text{ algae by the average}$$

microalgal net calorific value (18.5 MJ kg⁻¹) ([18]; [19]). These types of conversions were applied for decanters ([18]; [19]) and belt filters ([18]; [20]). Other conversions were based on the reported amount of CO₂ emissions per ton biodiesel for disk stack centrifugation ([21]; [20]; [22]), decanters ([18]; [19]), chamber filters ([21]; [19]), inorganic flocculation ([23]; [24]; [25]) and organic flocculation ([26]; [24]). This approach allowed to estimate the amount of greenhouse gas emissions based on literature data across several studies and report it as an interval between minimum and maximum reported values.

B. Patent analysis

Patents were retrieved from the EPODOC database of the European Patent Office (EPO). Only

European (EP), Patent Cooperation Treaty (PCT) and United States (US) patents or patent applications for harvesting techniques, published from 2000 onwards, were retained. The patent search strategy was based on a combination of the International Patent Classifications (IPC), Cooperative Patent Classifications (CPC) or European Patent Classifications (EC) (which is no longer in use) and English, French or German keywords with the boolean operators “OR” and “AND” in full-text EP, PCT or US patent documents. The selected patents were analyzed by six quality indicators, using Espacenet (<http://worldwide.espacenet.com>), the European Patent Register (<https://www.epo.org/searching/free/register.html>) and PAIR (Patent Application Information Retrieval) (<http://portal.uspto.gov/pair/PublicPair>) of the USPTO. Patents that met the quality indicators were discussed in further detail.

i) Search queries

The first query comprised the keywords: (microalg+ OR algae OR algen+ OR algae+ OR phytoplankton+ OR cyanobacter+ OR algal+ OR biomass).

The second query consisted of: (microorgan+ OR mikroorgan+ OR (mi?ro W organ+) OR cell? OR zell? OR cellule?) AND (biodiesel? OR biofuel? OR biobrennstoff+ OR biokraftstoff+ OR biocarburant+ OR biocombust+).

Both queries were introduced in combination with 91 different IPC, CPC or EC (actually replaced by CPC), representing the 21 harvesting techniques.

ii) Patent quality indicators

Patent selection was based on the following quality indicators: (1) grant of a patent application, (2) payment of renewal fees, (3) patent family size, (4) number of International Patent Classifications (IPC), (5) number of backwards citations cited in the international search reports and (6) number of claims. These indicators [27] were adapted to measure the relative impact of the retrieved patents or applications.

A granted patent (1) means that the application met the patentability conditions, i.e. novelty, inventive step and industrial applicability. However, a patent application that is not yet granted, but still under examination and which is thus not abandoned or withdrawn, will also be taken in account. Secondly, patents for which the renewal fees were paid for at least 5 years or at least the first annual fee (= in the 3.5th year) for the US (2) were also retained. The family size (3) is the number of equivalents filed for an invention in different countries, based on one or more earlier priority applications. ‘Many family members’ means that multiple patents are filed in several countries. Patents with at least one other family member were selected.

Patents with at least 3 IPC (4) were subsequently retained. A large IPC number means that the invention can have a wide number of technical applications. The number of backward citations (5) is another indicator that relates to the number of prior art documents. A small number of backward citations means that the technology could be a pioneer for that technical field. The number of “X” (=novelty) or “Y” (=inventive step) documents cited in the search report are indicators of the importance of the invention against the background prior art. A high number for “X” or “Y” means that the patent application has relevant background citations. Only those patent applications with fewer than 3 X or Y documents were retained. At last, (6) the number of claims determines the scope of the invention. ‘Many claims’ means that several features can be

protected by the patent. Patents with at least 10 claims were retained. Forward citations are not considered here because most of the patents were very recent. Only those patents that strictly referred to concentration and separation techniques in their abstracts were retained.

3. Results and discussion

A. Literature overview

A 1-ha-scale open-pond microalgae biomass production facility with a minimum productivity of 35 ton biomass ha⁻¹ year⁻¹ would need to process 200–300m³ of microalgae culture daily. This means that 99.95% of that volume is water that needs to be separated from the biomass, assuming a biomass concentration of 0.5 g L⁻¹. Unfortunately, most of the reported harvesting techniques are only tested on a lab, bench or pilot scale (Table 1). Pilot scale data are available for centrifugation and filtration-based methods. Spiral plate rotor technology (SPT) is based on centrifugation optimized for microalgae separation by the Dutch company Evodos. Thin films of microalgal suspensions are subjected to centrifugal forces between plates. A high throughput has been reported [28], but the discharge is discontinuous with a maximum throughput of only 4 m³ h⁻¹. Decanter centrifugation is another centrifugal separation technique that consists of two concentric rotating elements that operate continuously [29]. Both centrifugal techniques achieve a high TSS, but published data for decanters has been limited to bench scale setups [30].

Filtering techniques generally consume less energy than centrifugal techniques. This is particularly true for belt filtration and microstrainers. Belt filtration consists of two belts that squeeze the liquid from the solids, resulting generally in a high TSS. Moreover, it has a high flow throughput up to 200 m³ h⁻¹ for large algal species [31]. Unfortunately, the performance of filtering techniques generally depends on the microalgal species. Filtration is only sustainable for long-length microalgae or those which form large colonies, but cultures with low microalgal

cells concentration can also be harvested [7]. Microstrainers consist of a rotating drum with a belt and a backwash spray, while vacuum belts make use of a continuous belt with a suction force. A chamber filter press uses plates with filter medium that build up a cake that must be regularly removed. Tangential flow filtration (TFF) is a separation technique that uses cross-flow filtering membranes. The energy consumption for this particular technique varies strongly and depends on the operating pressure ([32]; [33]; [34]). However, screen clogging and membrane fouling remain important limiting factors for filtration techniques, as they increase operational costs [33]. The CAPEX for belt filter presses was generally lower than for centrifugation techniques and some authors concluded that microstrainers were more cost effective than centrifuges, i.e. disc centrifuges [35].

Table 1: Microalgal harvesting methods overview: comparison based on strain limitation, final total suspended solids concentration (TSS), capital expenditures (CAPEX), operational expenditures (OPEX), energy demand and greenhouse gas emission (GHG)

Harvesting method	Strain limitation	TSS (%)	CAPEX (\$ m ⁻³)	OPEX (\$ m ⁻³)	Energy (kWh m ⁻³)	GHG (gCO ₂ -eq MJ ⁻¹)	Scale ⁴	REFs
Primary concentration/separation								
Sedimentation	high	0.5–3	0.03	0.05–0.39	0.05–0.1	2.11–28	pilot	[36] [37] [38] [39] [29] [15] [40] [16] [41] [42] [43] [44] [45] [22] [46] [47]
Auto/Co-flocculation/biofilms¹	medium	1.4–5	0.03	0.06–1.5	0.02–0.2	10	pilot	[48] [36] [15] [40] [49] [50] [46] [51] [52] [53]
Inorganic flocculation	low	1.2–7	0.36	0.53–2.26	0.00084–2.85	1.26–36	pilot	[54] [31] [18] [39] [29] [23] [24] [55] [25] [50] [51]
Organic flocculation	medium	0.6–15	0.26	0.1–21.45	0.1–14.81	8.88–56	lab	[26] [56] [32] [18] [39] [57] [29] [24] [16] [49] [50] [58]
Electrolytic flocculation	low	3–5	0.05–6.03	0.11–1.45	0.04–9.5	47.9	bench	[54] [15] [34] [44] [45] [50] [46] [62] [26] [60] [61] [62]
Magnetic flocculation	medium	4.4	1.02	0.62	6.5	65	lab	[63] [41] [64] [65]
Hydrocyclone	high	0.4	4.32	1.87	0.3	160	bench	[39] [29] [16]
Dissolved air flotation (DAF)	medium	1–8	1.46	0.26–1.80	0.6–20	57.8–80	pilot	[36] [26] [66] [67] [15] [16] [42] [44] [35] [45] [50] [46] [51] [52]
Electrolytic flotation	medium	3–5	1.07	0.65	0.3–2	47.9	bench	[43] [44] [45] [46] [62]
Suspended air flotation (SAF)²	medium	1.4–5	1.04	0.65	0.003–0.015	70–90	pilot	[66] [68] [43] [50] [46]
Microstrainer	high	1.5–3	0.05	0.02	0.02–0.5	50	pilot	[69] [16] [41] [35] [43] [52]

filtering								
Acoustic aggregation	low	7.6	2.6	0.65	16–40	47.4	lab	[8] [26] [70]
Secondary concentration/separation								
Decanter	medium	12–30	0.58– 1.75	0.39– 1.13	3.6–10.8	39–80	bench	[18] [29] [16] [22] [51] [19]
Disk stack centrifugation	medium	10–22	0.48– 0.55	0.2– 1.63	0.7–1.4	53– 398.48	bench	[39] [29] [15] [71] [16] [72] [28] [73] [21] [34] [20] [45] [22] [51]
Spiral plate rotor (SPR)	low	31.5	0.41– 0.78	0.2– 0.39	0.42–1.94	242	pilot	[74] [38] [75] [71] [28] [76] [77] [50] [62]
Membrane filtration/TFF³	medium	2–27	1.87	0.35	0.012–10	40–45.4	bench	[78] [79] [80] [26] [66] [56] [32] [71] [41] [73] [33] [34] [52]
Belt filtering	high	12–50	0.29– 0.88	0.18– 0.57	0.16–0.88	20–79	pilot	[69] [81] [82] [18] [83] [29] [16] [41] [20] [46]
Chamber filtering	high	5–27	0.19	0.07	0.88	1.65– 241.87	no data	[66] [39] [29] [16] [21] [45] [19]
Vacuum belt filtering	high	9.5–18	0.42	0.18	0.1–5.9	92.5	pilot	[69] [56] [39] [29] [84] [16] [34]
Vibrating screen filtering	medium	1–10	0.62	0.19	0.4–3	70.5	pilot	[85] [41] [45] [42] [44] [34]

¹Auto- and co-flocculation are taken together; biofilms on substrata are included in this category

²Hydrophobic layer separation included

³Micro-and ultra-filtration and Tangential Flow Filtration (TFF) are combined

⁴Lab: volumes <10 gallons, bench: volumes 10–1000 gallons, pilot: volumes >1000 gallons [30]

Coagulation-flocculation-sedimentation is a separation technology that has been established for decades in different industries like drinking water production or wastewater treatment. Inorganic flocculation uses inorganic chemicals as flocculants, such as metallic salts, to neutralize the negative charge of the microalgae and allow floc formation. The energy requirements are acceptable compared with other flocculation techniques, but these flocculants can contaminate the biomass and limit water recovery. Moreover, inorganic flocculants are required in higher doses than organic flocculants, but are less species dependent [57]. Bioflocculation is a concentration technique that covers spontaneous (auto-flocculation) or natural flocculation (co-flocculation). Auto-flocculation is similar to sedimentation, but only requires pH regulation ([72]; [43]; [45]; [6]) or stress conditions ([86]; [87]) that promote flocculation. Both techniques have been applied on a pilot scale [30]. The costs for both sedimentation and bioflocculation were very competitive. However, the OPEX for bioflocculation strongly varies depending on the kind of additives used for pH adjustment, the kind of flocculating organisms or the potential application of genetic engineering [88]. Sedimentation and bioflocculation have a low energy consumption and low GHG emissions ([40]; [52]), however, the energy consumption for the growth of micro-organisms for co-flocculation should be taken into account.

Flotation can be seen as inverted sedimentation with the additional benefits of high TSS, a small areal footprint and lower operation time. Dissolved air flotation (DAF) uses air bubbles that are released after pressurization. The air bubbles attach to microalgae and carry them to the water surface. DAF can be used on a large scale ($>1000 \text{ m}^3 \text{ d}^{-1}$) and a flow throughput of $25 \text{ m}^3 \text{ h}^{-1}$ has been reported ([35]; [42]). However, air compression comes with a significant increase in energy demand, which will in turn increase CAPEX and OPEX to higher levels compared to, for

example, bioflocculation followed by sedimentation. Suspended air flotation (SAF) makes use of dispersed air bubbles, eliminating the compressing step. This usually results in better techno-economics despite the lower TSS.

In any case, the biological properties of microalgae species should be taken into account as well, as they might affect the overall efficiency and general applicability of microalgal harvesting methodology. The cell size, shape, density, surface charge, robustness and overall lipid content of the specific microalgal species or strain generally affect significantly the performance of most of the harvesting technologies [89]. Sedimentation for example requires large (spherical) microalgae (>100 nm diameter) [72] or microalgal flocs with a high density [90] to increase the settling velocity. On the other hand, for flotation-based separation, the size of the cells and flocs needs to be smaller to increase buoyancy and bubble attachment [45]. Secondly, the chemical composition of the growth medium will affect harvesting efficiency (pH, nutrients, salinity, temperature, density). This is particularly true for flocculation and bioflocculation techniques [12]. A high salinity inhibits organic flocculation as well [29] but could improve electrolytic flocculation [41]. Finally, the addition of toxic compounds, like heavy metals that are used as flocculants or released from electrodes, but also some synthetic polymers, can restrict the downstream use of biomass or suppress the growth of the microalgae when the medium is recycled [70]. This demonstrates that an optimal microalgal harvesting method will depend on the targeted strain and its cultivation conditions.

This literature overview shows that auto- and co-flocculation, as well as sedimentation, overall

scored best for the economic (CAPEX and OPEX) and environmental (energy and GHG) criteria, while belt filters scored the highest on the technological criterion (TSS) (Table 1). Some authors have proposed to combine bioflocculation with gravity sedimentation for microalgal biofuel production, because it seems to be the most cost-effective harvesting method ([7] ; [89]). Another recent review [90], based on a comparative analysis of harvesting methods for the industrial production of biodiesel, suggested a combination of organic flocculation and one of the following techniques: disk stack centrifuges, cross flow filtration or decanters. So far, no harvesting technique was superior on all criteria. However, the relative importance of technological (e.g. strain sensitivity, TSS), economic (e.g. CAPEX and OPEX) and environmental (e.g. energy and GHG) criteria will determine which combination of harvesting technology is most promising. For high value products it is very likely that environmental and economical criteria are less important than the technological criteria. For low value products on the other hand, the relative importance of economical and environmental criteria might be higher.

B. Patent analysis

This patent search and analysis retrieved 79 EP, PCT (WO) or US patent publications since 2000. Of those, 36 were published worldwide (WO), 36 for the United States (US) and seven for Europe (EP) (Table 2). Most of the patent applications were filed for organic flocculation (nine applications), auto-flocculation (eight applications), electrolytic and magnetic flocculation (eight applications) and membrane filtration (seven applications), but magnetic flocculation and belt filtering are the technologies which represent the highest number of granted patents. In total, 36

patents are still in force, which represents 46% of the total number of applications. From these 79 patent applications, 27 were granted (20 in the US and 7 via the EP procedure), whereof 22 (5 EP and 17 US patents) are currently active. Sixteen of the 22 granted patents fall under the scope of the quality indicators, while this is only the case for six (and one under appeal) of the other 16 applications. The remaining 43 applications were lapsed, abandoned, rejected or withdrawn.

Table 2: General overview of patent analysis: patent applications/granted in Europe (EP), USA (US) and worldwide (WO) and the total number valid or expired

Harvesting method	EP		US		WO	TOTAL	
	Appl.	Grant.	Appl.	Grant.		Valid	Exp.
Primary concentration/separation							
Sedimentation			2		1		3
Auto-flocculation		2	1	1	4	2	6
Co-flocculation				1	2	2	2
Biofilms			2		1		3
Inorganic flocculation			1	1	1	2	1
Organic flocculation		1	2	2	4	4	5
Electrolytic flocculation				1	7	4	4
Magnetic flocculation		1	3	4		5	3
DAF		1	1		3	2	3
Electrolytic flotation					1		1
SAF					1		1
Acoustic aggregation			1	2	1	3	1
Secondary concentration/separation							
Spiral Plate Rotor		2				2	
Membrane filtration				2	5	4	3
TFF			1		2		3
Belt filtering				4		2	2
Chamber filtering				1	1	2	
Vibrating screen filtering					1	1	
Hydrocyclone			1	1	1	1	2
TOTAL		7	16	20	36	36	43

In total, eight patent applications were filed for auto-flocculation, four for co-flocculation, three for sedimentation and four for belt filters. However, only three of the granted patents ([93]; [94]; [95]) and one of the valid applications [96] fall under the scope of the quality indicators (Table 3).

First, the patented auto-flocculation techniques require the culturing of genetically engineered microorganisms, which is currently only allowed in closed photo-bioreactors. This technology is described in EP2294179B1 by using microalgae from the genus *Chlamydomonas*, *Dunaliella*, *Scenedesmus* or *Haematococcus* [93]. An alternative could be the use of naturally occurring organisms or info-chemicals, that cause a defense reaction by the microalgae to induce flocculation [97]. More research is required to improve the technical feasibility of this method.

Secondly, co-flocculation relates to the use of other micro-organisms, like bacteria [94] or fungi [96]. US8574887B2 covers microalgae from *Chlorella*, *Scenedesmus*, *Nannochloris*, *Chlamydomonas*, *Chlorococcum*, *Euglena*, *Cryptomonas* and *Ellipsoidion* [94] and in US2012282651A1, the microalgae are selected from a very large group [96]. However, the co-production of these species also requires additional energy and costs.

Thirdly, sedimentation techniques are in general not frequently patented. Low flow throughput and species dependency are the major barriers [98]. As a result, none of the three retrieved patent applications is still in force. Lastly, belt filters seem to be applicable on a large scale with a reduced energy consumption and GHG. However, only one patent [95] for belt filters, that falls within the scope of the quality indicators, is still valid. In that patent US8092691B2, species from the genus *Botryococcus*, *Chlorella*, *Euglena* and *Nannochloropsis* were used to test the filter screens [95].

From 2014 onwards, a number of patent applications have been filed for membrane filtration and electrolytic techniques. One of the claimed improvements to prevent filter clogging is the use of a hydrogel to increase the effective diameter of the algal cells [99]. However, despite an increasing number of patent applications, this analysis reveals that not a single patented technology is able to meet all of the requirements to be a dominant harvesting solution for microalgae biomass.

C. A comparison of the literature overview versus the patent analysis

The different economical and environmental criteria that were used to compare the harvesting technologies in the literature overview are generally not specified and described in the analyzed patents. The economical and environmental advantages are only occasionally mentioned in the patent applications, but are generally not discussed in detail. It is therefore difficult to compare the technological potential of a patented system based on the criteria defined in scientific literature. It is even more difficult to interpret the provided empirical data in a patent application to determine its applicability on a large scale.

The information provided in scientific literature is based on empirical data usually collected for a single or only a few microalgae species for comparison. This means that conclusions for the studied harvesting technology cannot always be generalized. While such studies provide new knowledge and insights, they are not always directly targeted to commercial applications. The aim of a patent application, on the contrary, is to protect and legally cover crucial information required for the commercial development of a new technology. In the case of harvesting technology, this kind of protection can be applied for a harvesting device, a separation method or novel use of a known compound. The patent granting procedure requires considerable costs and investments, and it is therefore often strategized to make claims in a broad scope, for example by covering a large number of microalgal and/or cyanobacterial species. All claims should be covered by the description for consideration in the granting process, so including a large list of

microalgal species in the patent claims could raise doubts about its enablement because all of the claimed species should have been tested for the invention.

Since patenting involves making the technology freely available for others to reproduce, small but sometimes crucial improvements may not be patented and instead kept as a company secret. To minimize this risk, small but sometimes crucial improvements are therefore often not patented and kept as a company secret [100]. The publication of a defensive patent publication can on the other hand be novelty destroying.

Future research and development should be conducted into the sequential combination of low-cost concentration and separation techniques. One of the most promising routes from a techno-economic point of view is the use of auto-flocculation techniques in for example wastewater, that serves as microalgal growth medium, in combination with filtration or centrifugation.

Furthermore, the use of CO₂ as pH regulator should be further examined because it can be used to release the microalgal cells from their precipitants ([101]; [102]; [103]). Belt filters with a low differential pressure, based on the patented technology, could be applied after auto-flocculation to increase the TSS of the microalgal biomass. Finally, the integration of subsequent processes, such as controlled cell-disruption, component extraction, catalytic conversion, etc. into a single step harvesting process might be interesting future focus areas both from a techno-economical as from an intellectual property point of view.

Table 3. Patent analysis for microalgal harvesting technologies: EP and US patent and PCT applications (WO) with the patent numbers (publication numbers), the technology type, the number of family members, the number of IPC, the number of claims, the number of backward citations and the number of X or Y documents in the search report, the number of paid renewal fees and the maximum validity date or the current legal status.

Priority (year)	Harvesting method	Patent Number (or publication)	Inventor(s)	Claims	Families	IPC	X/Y	Granted (year)	Renewals paid	Valid till or legal status
1999	Magnetic flocculation	EP1097905	[104]	6	4	16	0	2005	15	Lapsed
2002	TFF	US2008213868	[105]	20	4	3	2	-	-	Abandoned
2007	SPR	EP2178617	[106]	15	18	2	3	2011	9	2028
2007	Inorganic flocculation	WO2009082696	[107]	30	2	2	2	-	-	Abandoned
2007	Biofilms	WO2009037355	[108]	10	10	2	5	-	4	Withdrawn
2007	DAF	EP2167431	[109]	9	29	7	0	2015	9	2028
2007	Hydrocyclone	WO2008140307		13	6	3	1	-	-	Withdrawn
2008	Hydrocyclone	US2010031561	[110]	19	1	1	2	-	-	Abandoned
2008	Belt filtering	US8372631	[111]	9	2	2	0	2013	-	Lapsed
2008	Belt filtering	US8377687	[112]	7	2	2	0	2013	-	Lapsed
2008	Auto-flocculation (genetic)	EP2294179	[93]	15	14	4	4	2014	7	2029
2008	Co-flocculation (stress)	WO2010036334	[113]	24	2	1	4	-	-	Withdrawn
2009	SPR	EP2475461	[114]	13	2	3	6	2013	7	2030
2009	Membrane filtration	WO2010085619	[115]	22	5	2	1	-	3	Withdrawn

2009	TFF	WO2010120992	[116]	28	2	3	4	-	-	Withdrawn
2009	Auto-flocculation (genetic)	US8404473	[117]	4	1	2	18	2013	1	2029
2009	Hydrocyclone	US8434626	[118]	20	2	2	0	2013	1	2030
2009	Sedimentation	US2010314323	[119]	21	3	1	1	-	-	Abandoned
2009	Electrolytic flocculation	US8772004	[120]	10	2	6	1	2014	-	2030
2009	Auto-flocculation (pH)	WO2011040955	[101]	12	2	2	2	-	-	Withdrawn
2009	Sedimentation	US2010264094	[121]	20	1	1	1	-	-	Abandoned
2009	Co-flocculation (bacteria)	US8574887	[94]	1	3	4	0	2013	-	2030
2009	SAF	WO2011008784	[122]	17	2	4	2	-	-	Withdrawn
2009	Membrane filtration	WO2011026482	[123]	20	3	2	2	-	-	Withdrawn
2009	Belt filtering	US8092691	[95]	3	9	1	3	2012	1	2029
2009	Chamber filtration	US8518132	[124]	23	3	1	0	2013	-	2030
2009	Organic flocculation	US8281515	[125]	33	4	2		2012	1	Expired
2009	Electrolytic flocculation	WO2010136195	[126]	18	3	1	3	-	5	Withdrawn
2010	Biofilms	US2011217764	[127]	26	2	1	5	-	-	Abandoned
2010	Auto-flocculation (genetic)	EP2441828	[128]	14	4	3	1	2015	5	Lapsed
2010	Organic flocculation	EP2397541	[129]	14	5	6	0	2015	7	2030
2010	DAF	WO2012000056	[130]	16	7	2	3	-	-	Withdrawn
2010	Belt filtering	US9095808	[131]	22	1	1	2	2015	-	2031
2010	Inorganic	US8790425	[132]	9	2	2	2	2014	-	2031

	flocculation									
2010	Membrane filtration	WO2012085210	[133]	15	4	2	4	-	-	Withdrawn
2010	Acoustic aggregation	US8889388	[134]	21	1	4	3	2014	-	2031
2010	Magnetic flocculation	US8399239	[102]	9	2	5	0	2013	-	2031
2010	Magnetic flocculation	US9464268	[135]	20	3	5	3	2014	-	2034
2010	Magnetic flocculation	US8828705	[136]	19	1	1	3	2014	-	2031
2010	DAF	WO2012047680	[137]	33	5	1	4	-	-	Withdrawn
2010	Organic flocculation	WO2011123970	[138]	23	6	2	2	-	4	Withdrawn
2010	Electrolytic flocculation	WO2012054404	[139]	43	2	3	7	-	-	Withdrawn
2011	Organic flocculation	WO2013063605	[140]	16	2	3	6	--	-	Withdrawn
2011	Electrolytic flotation	WO2013010252	[141]	14	3	2	0	-	-	Withdrawn
2011	Magnetic flocculation	US2012238003	[142]	12	3	2	1	-	-	Abandoned
2011	DAF	WO2012097981	[143]	10	2	1	2	-	-	Withdrawn
2011	Co-flocculation (fungus)	WO2013055887	[144]	34	1	2	4	-	-	Withdrawn
2011	Sedimentation	WO2012150390	[145]	10	5	1	3	-	4	Withdrawn
2011	Inorganic flocculation	US2014273173	[146]	15	4	2	0	-	-	-
2011	Auto-flocculation	WO2013059754	[147]	44	2	12	7	-	-	Withdrawn

	(pH)									
2011	Organic flocculation	WO2013076072	[148]	26	3	5	4	-	3	Withdrawn
2011	Organic flocculation	US2013026106	[149]	12	1	1	3	-	-	Abandoned
2011	Auto-flocculation (genetic)	WO2012139086	[150]	54	5	4	3	-	-	Withdrawn
2011	Magnetic flocculation (genetic)	US2013210064	[151]	20	1	4	0	-	-	Abandoned
2011	Organic flocculation	WO2013049553	[152]	23	5	1	2	-	5	-
2011	Co-flocculation (fungus)	US2012282651	[96]	20	2	4	5	-	-	Appeal
2011	Auto-flocculation (genetic)	US2014234904	[153]	30	2	2	3	-	-	Final rejection
2011	Acoustic aggregation	US2013116459	[154]	29	1	2	5	-	-	Abandoned
2011	Acoustic aggregation	WO2013028727	[155]	20	4	1	3	-	5	-
2011	Electrolytic flocculation	WO2012129031	[156]	28	3	3	2	-	4	Withdrawn
2012	Organic flocculation	US9267105	[157]	14	4	4	0	2016	-	2032
2012	TFF (microfiltration)	WO2014003988	[158]	19	2	2	6	-	-	Withdrawn
2012	Biofilms	US2014011246	[159]	13	1	6	6	-	-	Abandoned
2012	Auto-flocculation (stress)	WO2014003530	[160]	15	2	2	8	-	-	Withdrawn

2012	Organic flocculation	US2015284673	[161]	19	2	2	6	-	-	-
2012	Membrane filtration (pressure filtration)	US8980618	[162]	20	1	1	0	2015	-	2033
2012	Membrane filtration (pressure filtration)	US9051554	[163]	20	1	2	5	2015	-	2033
2012	Acoustic aggregation	US8668827	[164]	5	8	2	4	2014	1	2033
2012	Electrolytic flocculation	WO2014074790	[165]	20	23	7	3	-	-	Withdrawn
2012	Chamber filtration	WO2014041063	[166]	14	3	9	3	-	4	-
2012	Electrolytic flocculation	WO2013116357	[167]	20	35	2	5	-	4	-
2013	DAF	US2015128838	[168]	20	1	5	2	-	-	-
2013	Magnetic flocculation	US2014248680	[169]	20	2	4	1	-	-	-
2013	Magnetic flocculation	US9322013	[170]	6	4	3	4	2016	-	2034
2014	Vibrating screen filtering	WO2016052174	[171]	13	4	3	0	-	-	-
2014	Electrolytic flocculation	WO2016088057	[103]	19	1	9	2	-	-	-
2014	Electrolytic flocculation	WO2015196241	[172]	71	2	3	2	-	-	-
2014	Membrane filtration (hydrogel)	WO2015121618	[99]	38	2	4	1	-	3	-

2015	Membrane filtration	WO2016168871	[173]	16	2	2	-	-	-	-
-------------	------------------------	--------------	-------	----	---	---	---	---	---	---

Acknowledgements

D. Vandamme is a postdoctoral researcher funded by the Research Foundation – Flanders Belgium (FWO) (12D8917N). We would like to thank Prof. Dr. Carolien Kroeze and Dr. Jan Lutgerink for their support and guidance.

References

- [1] R. H. Wijffels and M. J. Barbosa, "An outlook on microalgal biofuels," *Science*, vol. 329 (5993), pp. 796-799, 2010.
- [2] L. Zhu, "Biorefinery as a promising approach to promote microalgae industry: An innovative framework," *Renewable and Sustainable Energy*, vol. 41, pp. 1376-1384, 2015.
- [3] J. Ruiz Gonzalez, G. Olivieri, J. de Vree, R. Bosma, P. Willems, H. Reith, M. Eppink, D. Kleinegris, R. Wijffels and M. Barbosa, "Towards industrial products from microalgae," *Energy Environ.Sci.*, vol. 9, pp. 3036-3043, 2016.
- [4] N.-H. Norsker, M. J. Barbosa, M. H. Vermuë and R. H. Wijffels, "Microalgal production: a close look at the economics," *Biotechnology Advances*, vol. 29 (1), pp. 24-27, 2011.
- [5] H. C. Greenwell, L. M. Laurens, R. J. Shields, R. W. Lovitt and K. J. Flynn, "Placing microalgae on the biofuels priority list: a review of the technological challenges," *J. R. Soc. Interface*, vol. 7 (46), pp. 703-726, 2010.
- [6] D. Vandamme, I. Foubert and K. Muylaert, "Flocculation as a low-cost method for harvesting microalgae for bulk biomass production," *Trends in Biotechnology*, vol. 31 (4), pp. 233-239, 2012.
- [7] A. Barros, A. Gonçalves, M. Simões and J. Pires, "Harvesting techniques applied to microalgae: A review," *Renewable and Sustainable Energy Reviews*, vol. 41, pp. 1489-1500, 2015.
- [8] R. Bosma, W. A. van Spronsen, J. Tramper and R. H. Wijffels, "Ultrasound, a new separation technique to harvest microalgae," *Journal of Applied Phycology*, vol. 15, pp. 143-153, 2003.
- [9] C. Chung, J. Popovics and L. Struble, "Flocculation and sedimentation in suspensions using ultrasonic wave reflection.," *The Journal of the Acoustical Society of America*, vol. 129, pp. 2944-251, 2011.
- [10] G. E. Hincapie and A. Marchese, "An Ultrasonically Enhanced Inclined Settler for Microalgae Harvesting," *Biotechnology Progress*, vol. 31 (2), pp. 1-10, 2014.
- [11] Y. C. Lee, K. Lee and Y. K. Oh, "Recent nanoparticle engineering advances in microalgal cultivation and harvesting processes of biodiesel production: A review," *Bioresource Technology*, vol. 184, pp. 63-72, 2015.
- [12] M. Alam, D. Vandamme, W. Chun, X. Zhao, I. Foubert, Z. Wang, K. Muylaert and Z. Yuan, "Bioflocculation as an innovative harvesting strategy for microalgae," *Reviews in Environmental Sciences and Bio/Technology*, vol. 15, pp. 573-583, 2016.
- [13] J. Richardson, M. Johnson, R. Lacey, J. Oyler and S. Capareda, "Harvesting and extraction technology contributions to algae biofuels economic viability," *Algal Research*, vol. 5, pp. 70-78, 2014.
- [14] J. Yuan, A. Kendall and Y. Zhang, "Mass balance and life cycle assessment of biodiesel from microalgae incorporated with nutrient recycling options and technology uncertainties," *GCB Bioenergy*, vol. 7, pp. 1245-1259, 2015.
- [15] A. K. Lee , D. M. Lewis and P. J. Ashman, "Energy requirements and economic analysis of a full-scale microbial flocculation system for microalgal harvesting,"

- Chemical Engineering Research and Design*, vol. 88 (8), pp. 988-996, 2010.
- [16] F. Mohn, "Experiences and strategies in the recovery of biomass from mass cultures of microalgae," *Algae biomass*, pp. 548-571, 1980.
 - [17] X. Liu , A. F. Clarens and L. M. Colosi, "Algae biofuels have potential despite inconclusive results to date," *Bioresource Technology*, vol. 104, pp. 803-806, 2012.
 - [18] F. Delrue, P.-A. Setier, C. Sahut, L. Cournac, A. Roubaud, G. Peltier and A.-K. Froment, "An economic, sustainability, and energetic model of biodiesel production from microalgae," *Bioresource Technology*, vol. 111, pp. 191-200, 2012.
 - [19] G. G. Zaimes and V. Khanna, "Microalgal biomass production pathways: evaluation of life cycle environmental impacts," *Biotechnology for Biofuels*, vol. 6 (88), p. 11, 2013.
 - [20] D. L. Sills, V. Paramita, M. J. Franke, M. C. Johnson, T. M. Akabas, C. H. Greene and J. W. Tester, "Quantitative uncertainty analysis of life cycle assessment for algal biofuel production," *Environ. Sci. Technol.*, vol. 47 (2), pp. 687-694, 2012.
 - [21] K. Sander and G. S. Murthy, "Life cycle analysis of algae biodiesel," *Int J Life Cycle Assess*, vol. 15 (7), pp. 704-714, 2010.
 - [22] V. Vasudevan, R. W. Stratton, M. N. Pearlson, G. R. Jersey, A. G. Beyene, J. C. Weissman, M. Rubino and J. I. Hileman, "Environmental performance of algal biofuel technology options," *Environ. Sci. Technol.*, vol. 46 (4), pp. 2451-2459, 2012.
 - [23] R. M. Handler, C. E. Canter, T. N. Kalnes, F. S. Lupton, O. Kholiqov, D. R. Shonnard and P. Blowers, "Evaluation of environmental impacts from microalgae cultivation in open-air raceway ponds: Analysis of the prior literature and investigation of wide variance in predicted impacts," *Algal Research*, vol. 1 (1), pp. 83-92, 2012.
 - [24] E. Lee and B. H. Zaribaf, "Applications of LCA to algae production systems," in *BioWet Summer School*, 2012.
 - [25] R. W. Stratton, H. M. Wong and J. I. Hileman, "Life cycle greenhouse gas emissions from alternative jet fuels," 2010.
 - [26] C. E. Canter, "The sustainability of biofuels produced from microalgae," 2013.
 - [27] M. Squicciarini, H. Dernis and C. Criscuolo, "Measuring patent quality: indicators of technological and economic value," OECD, 2013.
 - [28] D. O'Connell, "Life cycle assessment of dewatering routes for algae-derived biodiesel processes," 2013.
 - [29] E. M. Grima, E.-H. Belarbi, F. A. Fernandez, A. R. Medina and Y. Chisti, "Recovery of microalgal biomass and metabolites: process options and economics," *Biotechnology Advances*, Vols. 20 (7-8), pp. 491-515, 2003.
 - [30] L. Christenson and R. Sims, "Production and harvesting of microalgae for wastewater treatment, biofuels, and bioproducts," *Biotechnology Advances*, vol. 29 (6), pp. 686-702, 2011.
 - [31] R. T. Davis, "Characterizing microalgae (*Nannochloris oculata*) harvesting by aluminum flocculation," 2011.
 - [32] M. K. Danquah, L. Ang, N. Uduman, N. Moheimani and G. M. Forde, "Dewatering of microalgal culture for biodiesel production: exploring flocculation and tangential flow filtration," *J Chem Technol Biotechnol*, vol. 84 (7), pp. 1078-1083, 2009.
 - [33] N. Rossignol, L. Vandanon, P. Jaouen and F. Quéméneur, "Membrane technology for the continuous separation microalgae/culture medium: compared performances of cross-flow microfiltration and ultrafiltration," *Aquacultural Engineering*, vol. 20 (3), pp. 191-208, 1999.
 - [34] S. Sathe, "Culturing and harvesting marine microalgae for the large-scale production of biodiesel," 2010.

- [35] T.-S. Sim, A. Goh and E. W. Becker, "Comparison of centrifugation, dissolved air flotation and drum filtration techniques for harvesting sewage-grown algae," *Biomass*, vol. 16 (1), pp. 51-62, 1988.
- [36] J. R. Benemann and W. J. Oswald, "Systems and economic analysis of microalgae ponds for conversion of CO₂ to biomass," 1996.
- [37] J. Benemann, I. Woertz and T. Lundquist, "Life cycle assessment for microalgae oil production," *Disruptive Science and Technology*, vol. 1 (2), pp. 68-78, 2012.
- [38] P. Collet, A. Hélias, L. Lardon, M. Ras, R.-A. Goy and J.-P. Steyer, "Life-cycle assessment of microalgae culture coupled to biogas production," *Bioresource Technology*, vol. 102 (1), pp. 207-214, 2011.
- [39] R. Gori, G. Munz, C. Lubello, D. Daddi, N. Biondi and M. Tredici, "Energy demand and economic evaluation of solid-liquid separation systems used for harvesting and concentration of cultivated microalgae," in *Third International Symposium on Energy from Biomass and Waste*, Venice, 2010.
- [40] T. J. Lundquist, I. C. Woertz, N. W. Quinn and J. R. Benemann, "A Realistic Technology and Engineering Assessment of Algae Biofuel Production.," Berkeley, 2010.
- [41] S. L. Pahl, A. K. Lee, T. Kalaitzidis, P. J. Ashman, S. Sathe and D. M. Lewis, "Harvesting, thickening and dewatering microalgae biomass," in *Algae for biofuels and energy*, Springer Science, 2013, pp. 165-184.
- [42] I. Rawat, R. R. Kumar, T. Mutanda and F. Bux, "Biodiesel from microalgae: a critical evaluation from laboratory to large scale production," *Applied Energy*, vol. 103, pp. 444-467, 2013.
- [43] G. Shelef, A. Sukenik and M. Green, "Microalgae harvesting and processing: a literature review.," National Technical Information Service, Haifa, 1984.
- [44] K. Y. Show, D. J. Lee and J. S. Chang, "Algal biomass dehydration," *Bioresource Technology*, vol. 135, pp. 720-729, 2013.
- [45] N. Uduman, Y. Qi, M. K. Danquah, G. M. Forde and A. Hoadley, "Dewatering of microalgal cultures: A major bottleneck to algae-based fuels," *Journal of renewable and sustainable energy*, vol. 2 (1), pp. 012701-1-012701-15, 2010.
- [46] P. E. Wiley, J. E. Campbell and B. McKuin, "Production of biodiesel and biogas from algae: a review of process train options," *Water Environment Research*, vol. 83 (4), pp. 326-338, 2011.
- [47] C. Zamalloa, E. Vulsteke, J. Albrecht and W. Verstraete, "The techno-economic potential of renewable energy through the anaerobic digestion of microalgae," *Bioresource Technology*, vol. 102 (2), pp. 1149-1158, 2011.
- [48] V. Andersson, S. Broberg and R. Hackl, "Integrated algae cultivation for biofuels production in industrial clusters," 2011.
- [49] E. Suali and R. Sarbatly, "Conversion of microalgae to biofuel," *Renewable and Sustainable Energy Reviews*, vol. 16 (6), pp. 4316-4342, 2012.
- [50] D. Vandamme, "Flocculation based harvesting processes for microalgae biomass production," 2013.
- [51] P. J. I. B. Williams and L. M. Laurens, "Microalgae as biodiesel & biomass feedstocks: Review & analysis of the biochemistry, energetics & economics," *Energy & Environmental Science*, vol. 3, pp. 554-590, 2010.
- [52] I. Woertz, "Life cycle assessment of competing algae harvesting techniques," 2012. [Online]. Available: algaeconnect.crowdvine.com. [Accessed 24 06 2013].
- [53] Z. Wu, Y. Zhu, W. Huang, C. Zhang, T. Li, Y. Zhang and A. Li, "Evaluation of

- flocculation induced by pH increase for harvesting microalgae and reuse of flocculated medium," *Bioresource Technology*, vol. 110, pp. 496-502, 2012.
- [54] X. Cui, "Impact factors of harvesting chlorella autotrophica with electro-flocculation (EF)," 2012.
 - [55] A. Schlesinger, D. Eisenstadt, A. Bar-Gil, H. Carmely, S. Einbinder and J. Gressel, "Inexpensive non-toxic flocculation of microalgae contradicts theories: overcoming a major hurdle to bulk algal production," *Biotechnology Advances*, vol. 30 (5), pp. 1023-1030, 2012.
 - [56] M. K. Danquah, B. Gladman, N. Moheimani and G. M. Forde, "Microalgal growth characteristics and subsequent influence on dewatering efficiency," *Chemical Engineering Journal*, Vols. 151 (1-3), pp. 73-78, 2009.
 - [57] M. R. Granados, F. G. Ación, C. Gomez, J. M. Fernandez-Sevilla and E. Molina Grima, "Evaluation of flocculants for the recovery of freshwater microalgae," *Bioresource Technology*, vol. 118, pp. 102-110, 2012.
 - [58] H. Zheng, Z. Gao, J. Yin, X. Tang, X. Ji and H. Huang, "Harvesting of microalgae by flocculation with poly (γ -glutamic acid)," *Bioresource Technology*, vol. 112, pp. 212-220, 2012.
 - [59] T. L. Morrison, "Electrolytic methods as a cost and energy effective alternative of harvesting algae for biofuel," 2012.
 - [60] A. K. Sharma, A. K. Chopra and V. Kumar, "Overview of electrolytic treatment: an alternative technology for purification of wastewater," *Archives of Applied Science Research*, vol. 3 (5), pp. 191-206, 2011.
 - [61] K. Tumsri and O. Chavalparit, "Optimizing electrocoagulation-electroflotation process for algae removal," in *2nd International Conference on Environmental Science and Technology*, 2011.
 - [62] B. Yu, "CFD modeling of two-stage parallel plate sedimentation centrifuge for microalgae dewatering," 2012.
 - [63] H. Isogami, N. Saho, M. Morita and T. Takagi, "Sewage treatment performance of a continuous superconducting-magnetic separator," *Int J SER B*, vol. 44 (4), pp. 675-679, 2001.
 - [64] G. Prochazkova, I. Safarik and T. Branyik, "Harvesting microalgae with microwave synthesized magnetic microparticles," *Bioresource Technology*, vol. 130, pp. 472-477, 2013.
 - [65] L. Xu, C. Guo, F. Wang, S. Zheng and C. Z. Liu, "A simple and rapid harvesting method for microalgae by in situ magnetic separation," *Bioresource Technology*, vol. 102 (21), pp. 10047-10051, 2011.
 - [66] T. Coward, J. G. Lee and G. S. Caldwell, "Development of a foam flotation system for harvesting microalgae biomass," *Algal Research*, vol. 2 (2), pp. 135-144, 2013.
 - [67] J. Hanotu, H. Hemaka Bandulasena and W. B. Zimmerman, "Microflotation performance for algal separation," *Biotechnology and Bioengineering*, vol. 109 (7), pp. 1663-1673, 2012.
 - [68] V. Preston, "'Phyto-Phuel' Algae into energy creation research," Anne Arundel County, Maryland, 2011.
 - [69] A. O. Alabi, M. Tampier and E. Bibeau, "Microalgae technologies & processes for biofuels/bioenergy production in British Colombia: current technology, suitability & barriers to implementation," 2009.
 - [70] P. Chen, M. Min, Y. Chen, L. Wang, Y. Li, Q. Chen, C. Wang, Y. Wan, X. Wang, Y. Cheng, S. Deng, K. Hennessy, X. Lin, Y. Liu, Y. Wang, B. Martinez and R. Ruan,

- "Review of the biological and engineering aspects of algae to fuels approach," *Int J Agric & Biol Eng*, vol. 2 (4), pp. 1-30, 2009.
- [71] J. J. Milledge and S. Heaven, "Disc stack centrifugation separation and cell disruption of microalgae: a technical note," *Environment and Natural Resources Research*, vol. 1 (1), pp. 17-24, 2011.
 - [72] D. D. R. F. Montes, "Chlorella sp. coagulation-flocculation by inducing a modification on the pH broth medium," Lissabon, 2009.
 - [73] M. Rahbari, "Physical characteristics of pleurochrysis carterae in relation to harvesting potential for biodiesel production," 2009.
 - [74] M. Brocken, "Low cost & low energy harvesting," in *AquaFUEls Roundtable - conference review*, Brussels, 2010.
 - [75] H. Lyko, "A novel centrifuge concept: rotates slowly and saves energy," *F & S International Edition*, vol. 12, p. 3, 2012.
 - [76] C. Ryan, "Cultivating clean energy: the promise of algae biofuels," National Resources Defense Council, 2009.
 - [77] P. Schlagermann, G. Göttlicher, R. Dillschneider, R. Rosello-Sastre and C. Posten, "Composition of algal oil and its potential as biofuel," *Journal of Combustion*, p. 14, 2012.
 - [78] M. R. Bilad, D. Vandamme, I. Foubert, K. Muylaert and I. F. Vankelecom, "Harvesting microalgal biomass using submerged microfiltration membranes," *Bioresource Technology*, vol. 111, pp. 343-352, 2012.
 - [79] E. S. Bejor, C. Mota, N. M. Ogarekpe, K. U. Emerson and J. Ukpata, "Low-cost harvesting of microalgae biomass from water," *International Journal of Development and Sustainability*, vol. 2 (1), p. 11, 2013.
 - [80] R. Bhave, T. Kuritz, L. Powell and D. Adcock, "Membrane-based energy efficient dewatering of microalgae in biofuels production and recovery of value added co-products," *Environ Sci Technol*, vol. 46 (10), pp. 5599-5606, 2012.
 - [81] N. A. Carter, "Environmental and economic assessment of microalgae-derived jet fuel," 2012.
 - [82] R. Davis, A. Aden and P. T. Pienkos, "Techno-economic analysis of autotrophic microalgae for fuel production," *Applied Energy*, vol. 88 (10), pp. 3524-3531, 2011.
 - [83] E. D. Frank, J. Han, I. Palou-Rivera, A. Elgowiny and M. Q. Wang, "Life-cycle analysis of algal lipid fuels with the GREET model," 2011.
 - [84] V. Kothandaraman and R. L. Evans, "Removal of algae from waste stabilization pond effluents - a state of the art," State of Illinois, Urbana, 1972.
 - [85] M. A. Borowitzka, "Fats, oils and hydrocarbons," *Micro-algal Biotechnology*, pp. 257-287, 1988.
 - [86] D. C. Manheim, "Improved microalgal biomass harvesting using optimized environmental conditions and bacterial bioflocculants," 2012.
 - [87] A. Vlaski, "Mycrocystis aeruginosa removal by dissolved air flotation (DAF) - options for enhanced process operation and kinetic modelling," A.A. Balkema Rotterdam, 1998.
 - [88] A. K. Lee, D. M. Lewis and P. J. Ashman, "Microbial flocculation, a potentially low-cost harvesting technique for marine microalgae for the production of biodiesel," *J Appl Phycol*, vol. 21 (5), pp. 559-567, 2009.
 - [89] R. R. Soomro, T. Ndikubwimana, X. Zeng, Y. Lu, L. Lin and M. K. Danquah, "Development of a two-stage microalgae dewatering process - a life cycle assessment approach," *Frontiers in Plant Science*, vol. 7 (113), p. 12, 2016.

- [90] M. Al Hattab, A. Ghaly and A. Hammouda, "Microalgae harvesting methods for industrial production of biodiesel: critical review and comparative analysis," *J.Fundam Renewable Energy*, vol. 5 (2), p. 26 , 2015.
- [91] S. Lama, K. Muylaert, T. Karki, I. Foubert, R. Henderson and D. Vandamme, "Flocculation properties of several microalgae and a cyanobacterium species during ferric chloride, chitosan and alkaline flocculation.," *Bioresource Technology*, no. 220, pp. 464-470, 2016.
- [92] M. J. Griffiths, R. G. Dicks, C. Richardson and S. T. Harrison, "Advantages and challenges of microalgae as a source of oil for biodiesel," in *Biodiesel - feedstocks and processing technologies*, InTech, 2011, pp. 177-200.
- [93] M. Mendez, G. Behnke, Y. Poon and P. Lee, "Induction of flocculation in photosynthetic organisms". European patent Patent 2294179, 2009.
- [94] S. G. Stepenoff and K. L. Hastings, "Methods and compositions to aggregate algae". United States patent Patent 8574887, 2010.
- [95] R. O. Youngs and J. R. Cook, "Method and apparatus for separating particles from a liquid". United States patent Patent 8092691, 2010.
- [96] S. J. Yuan and S. Xie, "System and method of co-cultivating microalgae with fungus". United States patent application Patent 2012282651, 2012.
- [97] C. Gonzalez-Fernandez and M. Ballesteros, "Microalgae autoflocculation: an alternative to high-energy consuming harvesting methods," *J Appl Phycol*, vol. 25 (4), pp. 991-999, 2012.
- [98] J. J. Milledge and S. Heaven, "A review of the harvesting of micro-algae for biofuel production," *Reviews in Environmental Science and Biotechnology*, vol. 12 (2), pp. 165-178, 2012.
- [99] M. Pearce, "Processing of microalgae". Patent Cooperation Treaty application Patent 2015121618, 2015.
- [100] M.-Y. Wang, S.-C. Fang and Y.-H. Chang, "Exploring technological opportunities by mining the gaps between science and technology: microalgal biofuels," *Technological Forecasting & Social Change*, vol. 92, pp. 182-195, 2015.
- [101] A. Schlesinger, D. Eisenstadt, S. Einbinder and J. Gressel, "Method and system for efficient harvesting of microalgae and cyanobacteria". Patent Cooperation Treaty application Patent 2011040955, 2010.
- [102] T. Zhang and R. Crowell, "Compositions and methods for continuous harvesting of suspension growth cultures". United States patent Patent 8399239, 2011.
- [103] S. Avinash, M. Sairam, P. G. Ninad and G. G. Ashwin, "A method for separating solid particles from a waterbody". Patent Cooperation Treaty application Patent 2016088057, 2015.
- [104] N. Saho, H. Isogami, M. Morita and T. Sano, "Membrane magnetic separating apparatus". European patent Patent 1097905, 2000.
- [105] R. Fournier, "Concentrated aqueous suspensions of microalgae.". United States application Patent 2008213868, 2008.
- [106] H. Boele , "Separating device and method". European patent Patent 2178617, 2008.
- [107] G. Radaelli, D. Fleischer, B. Vick, M. Caspari, J. Weismann and D. Rice, "Methods for concentrating microalgae.". Patent Cooperation Treaty application Patent 2009082696, 2008.
- [108] K. Vanhoutte and J. Vanhoutte, "Method for harvesting algae or plants and device used thereby". Patent Cooperation Treaty application Patent 2009037355, 2008.
- [109] R. L. Clayton, S. N. Falling, J. S. Kanel and C. C. Churn, "Process and apparatus for

- adsorptive bubble separation". European patent Patent 2167431, 2008.
- [110] P. G. Hatcher, E. Salmon and A. Stubbins, "Raceway for cultivating algae in liquid medium i.e. water, has cyclone precipitators algal biomass produced in raceway is precipitated from liquid medium and concentrated.". United States application Patent 2010031561, 2009.
 - [111] S. Shepherd , "System for harvesting algae in continuous fermentation". United States patent Patent 8372631, 2009.
 - [112] S. Shepherd, "System for harvesting algae in continuous fermentation". United States patent Patent 8377687, 2012.
 - [113] B. C.-P. Wu, D. Stephen, G. E. Morgenthaler and D. V. Jones, "Systems and methods for producing biofuels from algae". Patent Cooperation Treaty application Patent 2010036334, 2009.
 - [114] H. Boele, "Centrifugal separator, method for separation". European patent Patent 2475461, 2010.
 - [115] L. Dong, K. J. Drury and A. G. Fadeev, "Methods for harvesting biological materials using membrane filters". Patent Cooperation Treaty application Patent 2010085619, 2010.
 - [116] Q. Hu, M. Sommerfeld, Y. Chen and X. Zhang, "Method of separation of algal biomass from aqueous or marine culture". Patent Cooperation Treaty application Patent 2010120992, 2010.
 - [117] O. Kilian and B. Vick, "Cyanobacterial isolates having auto-flocculation and settling properties". United States patent Patent 8404473, 2009.
 - [118] J. D. King and K. D. Willis, "System and related method for concentrating biological culture and circulating biological culture and process fluid.". United States patent Patent 8434626, 2010.
 - [119] M. H. Lean, D. K. Fork, J. Seo, J. S. Fitch and A. R. Volkel, "Method and apparatus for continuous flow membrane-less algae dewatering.". United States application Patent 2010314323, 2009.
 - [120] G. C. Schafran, J. F. Kolb, A. Stubbins and K. H. Schoenbach, "System and method for high-voltage pulse assisted aggregation of algae". United States patent Patent 8772004, 2010.
 - [121] G. Schwartz, M. Massingill, J. Van Olst and J. Carlberg, "Method of developing a rapidly settling algal floc". United States application Patent 20100264094, 2010.
 - [122] M. Tegen, W. W. Berry and W. R. Sutterlin, "Method for harvesting microalgae suspended in an aqueous solution using a hydrophobic chemical". Patent Cooperation Treaty application Patent 2011008784, 2010.
 - [123] C. Thomsen, "Method for harvesting algae from an algae suspension.". Patent Cooperation Treaty application Patent 2011026482, 2010.
 - [124] A. C. Rettenmaier, "Methods of algae harvesting utilizing a filtering substance and uses therefor". United States patent Patent 8518132, 2010.
 - [125] E. J. Nichols and J. R. Scott, "Methods for growing and harvesting algae and methods of use". United States patent Patent 8281515, 2010.
 - [126] H. Junge and H. Franke, "Electrochemical separation of algae". Patent Cooperation Treaty application Patent 2010136195, 2010.
 - [127] L. Christenson and R. Sims, "Rotating bioreactor and spool harvester apparatus for biomass production". United States application Patent 20110217764, 2011.
 - [128] A. Falciatore, R. Raniello and C. Bowler, "Algal bio-flocculation by inactivation of photoreceptors". European patent Patent 2441828, 2010.

- [129] A. Malm and R. Tanner , "A method for harvesting algae". European patent Patent 2397541, 2010.
- [130] L. Sirmans, B. Beard, W. Drusko and L. Pochat-Pochatoux, "Harvesting microorganisms". Patent Cooperation Treaty application Patent 2012000056, 2011.
- [131] J. E. Taylor, M. Inman, J. Kell, H. Mccrabb, A. Ferrante, R. Youngs, N. J. Meister and R. J. Cook, "Electrolytic system and method for filtering and aqueous particulate suspension". United States patent Patent 9095808, 2011.
- [132] H. Urayama, M. Kurata and H. Fukuda, "Method for aggregating and separating algae". United States patent Patent 8790425, 2011.
- [133] H. Van Kaathoven, J. P. Haan, W. M. Bond, J. Leendert and W. C. Den Boestert, "Process for separation of a mixture containing a microbial substance and a liquid". Patent Cooperation Treaty application Patent 2012085210, 2011.
- [134] Z. Wang, D. Feke and J. Belovich, "Acoustic device and methods thereof for separation and concentration". United States patent Patent 8889388, 2011.
- [135] H. Liang, "Harvesting micro algae". United States patent Patent 9464268, 2014.
- [136] V. S.-Y. Lin, B. G. Trewyn, K. Kandel and I. I. Slowing, "Magnetic mesoporous material for the sequestration of algae". United States patent Patent 8828705, 2011.
- [137] K. Booth, M. Cosby, M. Fosshage, S. Poe and D. Schlegel, "Floated solids separation". Patent Cooperation Treaty application Patent 2012047680, 2011.
- [138] S. Wang, Q. Zhao and G. Zhang, "Non-destructive method for algae contaminated water treatment and algae harvest or removal". Patent Cooperation Treaty application Patent 2011123970, 2010.
- [139] M. Green, N. Eckelberry, S. Fraser and B. Goodall, "Systems, methods and apparatuses for dewatering, flocculating and harvesting algae cells". Patent Cooperation Treaty application Patent 2012054404, 2011.
- [140] R. Anthony and R. Sims, "Methods for harvesting biomass.". Patent Cooperation Treaty application Patent 2013063605, 2012.
- [141] F. Bensebaa, "Photobioreactor". Patent Cooperation Treaty application Patent 2013010252, 2012.
- [142] T. Fukaya, K. Tsutsumi, A. Yamazaki, I. Yamanashi and S. Seki, "Microorganism concentrating apparatus and method". United States application Patent 2012238003, 2011.
- [143] R. Gloeckler, "Method of harvesting algae, and device for harvesting algae for carrying out the method of harvesting algae". Patent Cooperation Treaty application Patent 2012097981, 2012.
- [144] B. Hu, R. R. Ruan, J. Zhang and W. Zhou, "Microalgae culture and harvest". Patent Cooperation Treaty application Patent 2013055887, 2012.
- [145] E. Kabakian, "Method for harvesting microalgae, and device for implementing said method". Patent Cooperation Treaty application Patent 2012150390, 2012.
- [146] H. Kaku, "Method for separating out and recovering microalgae.". United States application Patent 2014273173, 2012.
- [147] L. E. Katz, K. A. Kinney, J. Choi and E. Chen, "Continuous flocculation deflocculation process for efficient harvesting of microalgae from aqueous solutions". Patent Cooperation Treaty application Patent 2013059754, 2012.
- [148] G. Krueger and R. Dallwig, "Method for separating algae, in particular microalgae, from an aqueous phase, and a device for carrying out this method". Patent Cooperation Treaty application Patent 2013076072, 2012.
- [149] M. Liberatore, "Optimized flocculation algae using cationic polymers". United States

- application Patent 2013026106, 2012.
- [150] G. A. Oyler, J. N. Rosenberg and D. P. Weeks, "Single chain antibodies for photosynthetic microorganisms and methods of use". Patent Cooperation Treaty application Patent 2012139086, 2012.
 - [151] P. Nath and S. Twary, "Magnetotactic algae and methods of use". United States application Patent 2013210064, 2012.
 - [152] C. J. G. Walterick, D. Whitt and J. Juchinski, "Method for flocculation algae using polymers including tannin". Patent Cooperation Treaty application Patent 2013049553, 2012.
 - [153] S. Herbert and L. G. Lowder, "Method for harvesting photosynthetic unicells using genitically induced flotation". United States application Patent 2014234904, 2014.
 - [154] B. L. Marrone, D. M. Kalb, J. E. Coons and T. Dale, "Method and apparatus for acoustically manipulating biological particles". United States application Patent 2013116459, 2012.
 - [155] J. Dionne, J. King, B. Lipkens and E. A. Rietman, "Ultrasonic agglomeration of microalgae". Patent Cooperation Treaty application Patent 2013028727, 2012.
 - [156] M. P. Green, S. A. Fraser, N. D. Eckelberry and J. L. S. Pina, "Enhancing algae growth by reducing competing microorganisms in a growth medium". Patent Cooperation Treaty application Patent 2012129031, 2012.
 - [157] W. Farooq, Y.-C. Lee and J.-W. Yang, "Method for rapidly and efficiently harvesting green algae using cationic organoclay". United States patent Patent 9267105, 2012.
 - [158] S. Gormly, M. Atwood, A. Septimus and J. P. Barlow, "Dewatering systems and methods for biomass concentration". Patent Cooperation Treaty application Patent 2014003988, 2013.
 - [159] R. Sims, C. Miller, J. T. Ellis, A. Sathish, R. Anthony and A. Rahman, "Methods for harvesting and processing biomass". United States application Patent 2014011246, 2013.
 - [160] I. Wahby, I. Bennis, H. Arroussi and R. Benhima, "Method for increasing the potential for biofuel production from microalgae by using bio-modulators". Patent Cooperation Treaty application Patent 2014003530, 2013.
 - [161] T. Langer, A. Aravanis and S. Matsumoto, "Harvesting algae from saline water using flocculants". United States application Patent 2015284673, 2013.
 - [162] R. Varadaraj, "Algae aggregation and harvesting". United States patent Patent 8980618, 2013.
 - [163] R. Varadaraj, "Harvesting algae by foam aggregation and filtration". United States patent Patent 9051554, 2013.
 - [164] J. S. Kniep and A. Kale, "Rectangular channel electro-acoustic aggregation device". United States patent Patent 8668827, 2013.
 - [165] A. Kale, A. Galvez and S. Kuhlman, "Reducing concentration of contamination with electro-coagulation". Patent Cooperation Treaty application Patent 2014074790, 2013.
 - [166] J. Grossmann, F. Cotta, M. Matschke and C. Griehl, "Method for the solid/liquid separation of a suspension containing microalgae". Patent Cooperation Treaty application Patent 2014041063, 2013.
 - [167] N. Eckelberry, "Systems and methods for harvesting and dewatering algae". Patent Cooperation Treaty application Patent 2013116357, 2013.
 - [168] K. Bryan, "Apparatus for harvesting algae from open body of water". United States application Patent 2015128838, 2014.

- [169] R. J. Powell and R. T. Hill, "Compositions and methods for collecting algae". United States application Patent 2014248680, 2014.
- [170] P. Nath and S. N. Twary, "Magnetic separation of algae". United States patent Patent 9322013, 2014.
- [171] T. Kusakibaru and M. Suzuki, "Cultured algae water concentration system, method for operating cultured algae water concentration system, and method for concentrating algae water". Patent Cooperation Treaty application Patent 2016052174, 2015.
- [172] L. D. Milton, L. A. Kwong, T. Kalaitzidis and A. Isdepsky, "Methods and systems for separating particulates from a liquid medium". Patent Cooperation Treaty application Patent 2015196241, 2015.
- [173] F. Emminger, "Method for obtaining, in particular for harvesting, algae and microorganisms". Patent Cooperation Treaty application Patent 2016168871, 2016.
- [174] I. Udom, B. H. Zaribaf, T. Halfhide, B. Gillie, O. Dalrymple, Q. Zhang and S. J. Ergas, "Harvesting microalgae grown on wastewater," *Bioresource Technology*, vol. 139, pp. 101-106, 2013.