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Review article

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

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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 economic (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 seem 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.

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

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destabilize the microalgal cells [8–10]. 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

2.1. 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)), economic 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 costs. 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 Eq. (1), which represents the total harvesting costs (P_c) for the production of 1 m^3 [15,16]:

$$Pc = \frac{((0.5*I/100 + M/100)*C*A) + C}{W*A*Qc} + \frac{Rc}{Qc} \tag{1}$$

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 CO_2 per required energy unit (g CO_2-eq MJ^{-1}). When no direct data were available, conversion was based on literature studies with the amounts CO_2 produced in relation to the distance (g CO_2-eq 100 km^{-1}), and converted using the formulae: $\frac{\frac{10\ \text{kg CO2}}{100\ \text{km}}}{\frac{1\ \text{MJ}}{0.39\ \text{km}} \cdot \frac{1\ \text{kg}}{1000\ \text{g}}}$ [17] or by dividing the reported amounts kg CO_2-eq ton^{-1} 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 CO_2 emissions per ton biodiesel for disk stack centrifugation [20–22], decanters [18,19], chamber filters [19,21], inorganic flocculation [23–25] and organic flocculation [24,26]. This approach allowed estimating 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.

2.2. 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.

2.2.1. Search queries

The first query comprised the keywords: (microalg+ OR algae OR algen+ OR algue+ 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.

2.2.2. 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