Technology Overview of Biogas production in anaerobic digestion plants: A European Evaluation of Research and Development

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Abstract

Anaerobic digestion (AD) technology is used commercially around the world, especially in Europe, which has set some challenging targets to diversify its energy mix with more renewable energy. This study intends to demonstrate, through technology prospecting, the relation between academic research (published articles) and technology development (patent applications) evolved from 1990 to 2015. Published articles were classified under the topics and wastes they cover, which include manure, agricultural and food waste, wastewater, sewage sludge and the organic fraction of municipal solid waste, with the last of these often being associated with co-digestion processes. Meanwhile, the patents in the area are mostly for equipment of the AD process and new methods or means of purifying the biogas obtained. It was found that the patents filed in Europe tend to protect their innovations only occasionally in countries outside the EU. Germany is the clear leader in all the areas of research and the commercial applications of the technologies, followed by Italy, Spain and Sweden. This study also demonstrates the immense potential of biogas throughout Europe, not just for energy generation, but also as a fuel and a by-product of the treatment of different kinds of waste.

Key words: Biogas; Technology Prospection; Anaerobic Digestion; Organic Waste; Renewable energy.

Highlights

- The European Union demonstrates a global leadership role in the biogas production field
- Frequent feedstock are wastewater, manure, agricultural and industrial waste
- Germany is the country with more development in all analysed aspects

1. Introduction

In 1630, Jan Baptist van Helmont (1580-1644), pointed that organic material in decomposition produced flammable gases. Some years later (1776), Alessandro Volta (1745-1827) discovered methane by collecting gas emerging from Lake Maggiore (Italy) (Abbasi *et al.*, 2011) and in 1804, John Dalton (1766-1844) established the chemical constitution of methane. The concept of anaerobic digestion has been introduced around 1870 with the development of the septic tank system by Jean-Louis Mouras. It was Louis Pasteur (1822-1895) who reported that biogas could be used for heating and lighting. Indeed, in 1895 Donald Cameron design led to light up the streets of Exeter (England). Biogas development presented an inflection point in the energy shortages of the Second World War and during petroleum crisis in 1970. From then to now, anaerobic digestion has been studied, microbiologically identified and converted into a technology that, nowadays, is being used either for the treatment of wastewaters and solid wastes. In this sense, anaerobic digestion (AD) has become an interesting alternative for energy production, not only for the environmental advantages, of using waste as a raw material to produce biogas and a high-quality fertilizer (digested material) as its main products, but also for their relative low cost when compared to other techniques (Horváth *et al.*, 2016). In fact, any kind of biomass has potential to be a substrate for biogas production as long as they contain carbohydrates, proteins, fats, cellulose and hemicelluloses as main components (Braun, 2007).

Briefly, anaerobic digestion is a biological process running under anaerobic conditions (strict absence of oxygen) in which a consortium of microorganisms breaks down complex biodegradable organic matter to methane (50 to 80%) and carbon dioxide (30 to 50%): biogas. Consequently, biogas can be used as a valuable energy source (5.5 to 7 kWh/m^3 of biogas).

According to Barre and Mattheeuws (2010), AD is not a new technology. It was already known in the 17th century, but it was only in the 1980s that it started to be used more widely for treating industrial and municipal wastewater, sewage sludge or municipal solid waste. As it is now a mature technology, it could be key to reducing organic waste, recovering the energy contained in biomass, and generating biofuels and energy (Nardin and Mazzett, 2014).

This process is therefore seen as a profitable treatment procedure, and it is increasingly being used, especially in Europe, to the detriment of other methods like incineration and disposal in landfill sites. Also, the use biogas promotes a sustainable society, reducing the dependence on oil, reducing the pollution and supplies energy with less impact on the environment (International Gas Union Office, 2015). Anaerobic digestion technology for biogas production, constitute today the most sustainable way of using the energy present in biomass and other wastes, because it also increases nutrient recovery and reduces greenhouse gas emissions. According to this potential, countries such as Germany, The United Kingdom and The USA established new legislation introducing alternative sources of energy including biogas (Edwards *et al.*, 2015). Some biogas applications are reported in literature, for example, biogas has been used to efficiently heat a greenhouse during the typical winter conditions in eastern Turkey (Esen *et al.*, 2013) or for heating households in India (Lewis *et al.*, 2016) or been used in a biogas-powered train (Sweden).

In parallel with these environmental advantages, AD could be also an instrument in helping countries to meet the new targets set by the European Union (EU), whose members have the overall goal of generating 20% of all their energy from renewable sources and of having 10% of their transport sector run on renewable energy by 2020 (Havukainen *et al.*, 2014). The state members must also cut their disposal of municipal solid waste by 50% by 2020 (European Union Council, 2012). Reinforcing the importance of biogas in Europe, in 2013 the biogas production was comparable approximately to 13.4 M tons oil equivalent, 15.5 Mrd m^3 methane and 3% of natural gas consumption, also a production of 39.5 Mtoe is estimated for 2020, which corresponds to approximately 10% of EU natural gas consumption (European Biogas Association, 2015). Finally, one extra advantage of using this technology has to do with the versatility of biogas, which can be used to generate electricity as well as a vehicle fuel. Consequently, it is attracting increasing interest on the part of researchers from academia and the public and private sectors.

By 2030, it is estimated that Europe's biogas production capacity will have reached around 18 to 20 billion m^3, which corresponds to around 3% of European current natural gas consumption (Kovacs, 2013). Reducing global warming, enhancing its energy grid, and diversifying its power generation capacity constitute the main priorities of the European Parliament's environmental policies. Europe's targets for renewable energy production, greenhouse gas emission reductions and the sustainable management of waste can all be attained by using AD technology. It is one of the few processes that has the capacity to fulfil these three main European priorities (Seadi *et al.*, 2008).

According to the Global Intelligence Alliance (2010), the potential sources of biogas at world level are: 75% in agricultural crops, by-products, and manure; 17% in municipal and industrial organic waste and 8% in sewage wastewater treatment facilities. Figure S1 (supplementary material) shows the general biogas production chain. The first link is the entrance of substrates (with a high biodegradable organic load) that can be degraded by AD. In theory, all biodegradable materials that are not composed exclusively of lignin (e.g. wood), which, because of their molecular structure, have to be pre-treated to expose the biodegradable material to microorganisms, can serve as a substrate for AD processes and direct biogas production. Agricultural waste, manure, municipal solid waste, food waste, sewage, wastewater, and different industrial effluents with a high organic load can also be used in biogas production plants. Sometimes, they need to go through a pre-treatment stage, or to be co-digested, when one material is combined to improve process efficiency and synergic effects (Luostarinen *et al.*, 2011). The logistic stage of collection, storage and transportation should be kept as simple as possible to keep overall process costs down. In some cases, especially when agricultural wastes are subject to seasonality, a logistic stage is required where these materials are transported to AD units (Ericsson *et al.,* 2013).

The production stage is the most complex and involves two sub-stages: pre-treatment of the raw material and the AD reaction *per se*. Pre-treatment is needed to remove any impurities from the feedstock, like metals, plastics, or stones, and also to adjust the physical and chemical process conditions. More specific pre-treatments may be required to break down materials with a high lignocellulosic content and improve their bioavailability to the microorganisms involved in the process (Zhang *et al.,* 2014). Currently, the literature breaks the types of pre-treatment down into three groups: chemical, physical and biological. They can be used individually or in combination.

As the biogas constitutes a mixture of gases, a purification stage is also required. There are different ways to separate out the constituent gases, but physical and chemical absorption are the most efficient and less complicated (Weiland, 2010). The biogas purification stage is a crucial part of the process as a whole. Generally, biogas must be purified to avoid problems in the subsequent heat and power units. These purifications processes remove unwanted components, like H_2S, particulate matter, dust and water (Nordic Energy Research, 2010). However, when biogas is intended to be used as a fuel for vehicles, injected into the natural gas distribution network or used in fuel cells, high purification grade must be achieved and CO_2 must also be removed. Purification costs are very high, and this is currently the stage that is the most challenging in the whole process. According to Jeon and Lee (2015), using membrane for biogas purification has the advantages to be simple technique and easy to scaling up.

The objective of this study is to evaluate the potential innovation and commercial applications of biogas production through anaerobic digesters in the European Union by investigating academic papers published and patent applications in the area. Using a technology prospection and collecting data from patents and articles for a long period (1990-2015), the correlation between Research, Development and Production is presented. It is not the objective of this study to provide a review on the process parameters of anaerobic digestion, as the biogas yields, organic matter removals or quality of the biogas. This study, intends to demonstrate, through technology prospecting, the relation between research institutions (throughout analysing published articles), and the relation between research institutions and technology development (throughout analysing patent applications). Apart from providing and extended overview of the research carried out on the production of biogas in anaerobic digesters, the expected impact of this study is the diffusion of bibliometric studies as a tool to expand and consolidate new knowledge about biogas and anaerobic digestion research.

2. Methodology

This study is a foresight analysis involving the empirical study of academic publications and patents documents. Reference literature was used to give a picture of the state of the art in biogas production and research in terms of the leading country (production and articles publication), main assignees and main research institutions in biogas production. To perform this analysis indexed articles and patents have been evaluated between 1990-september 2015.

For articles evaluation, a search in the Web of Science database for academic papers indexed between 1990 and September 2015 was made using the following keywords (*biogas* or "*green gas*" or *greengas* or *biomethane* or "*digestated gas*" or "*renewable natural gas*"). Note that these keywords do not include the keyword "*Anaerobic Digestion"*. This is due to the focus of this research is focused on the product "biogas", our search assumed that if the objective of the research was the production of biogas, keywords related to biogas should be present in the title or the abstract of the articles. Of course, papers related to the biogas purification step are also recovered, thus complementing our search on the use of the product "biogas".

After this, the data mining processes was started with the software Vantage Point®. This software allows organizing the documents and sorting them out by country, priority, year, author, etc. showing correlations between research institutions or companies that joins its efforts for research or patent inscription. As previously mentioned, for this study just papers or patents, published or filed in Europe were considered for analysis. After importing to Vantage Point®, articles were separated by country, retaining only those where the first author was from European institutions. This country selection shows that 47% of the total articles retrieved had the first author associated with a European country. Duplicates and papers that had no abstract were also eliminated. Finally, those articles that contained words related with methane generation from other means than AD (such as gasification, landfilling, pyrolysis, etc.) were identified, read and discarded if not applicable to this study.

To identify key words and to give a general idea of the subjects covered in the academic publications, a word cloud using the TagCrowd online tool (http://tagcrowd.com) was generated, which presents words in a cloud, highlighting them according to the word frequency in the texts analyzed.

Regarding patent evaluation, it must be noted that utility models were not considered in this search. Utility models are used to protect new industrial applications, like new formats, which result in improved uses or manufacturing conditions. Derwent Innovations Index was used to retrieve a patent document, retrieving only priority patents filed since 1990 to September 2015. Patent documents identification by the Derwent Manual Code #D05-C14 (methane fermentation) and three International Patent Classification (IPC) subclasses: C02F-011/04 (anaerobic treatment; production of methane by such processes), C02F-003/00 (biological treatment of water, wastewater or sewage), and C02F-003/28 (anaerobic digestion process) was carried out. The patent documents retrieved were imported into Vantage Point®, where they were separated into their respective priority countries. Only the patents whose priority country was from Europe were considered. Duplicates were removed and considered only those documents that contained at least one of the following words in the abstract: *biogas*, *biomethane*, *green gas*, *anaerobic digestion*, *anaerobic treatment*, *production of methane*, *digestate*, *methane*.

3. Results and Discussion

3.1 General evaluation of academic publications from Europe

Using the strategy described above, 4,682 articles were retrieved from the time period 1990-2015. It was found that biogas research is motivated by many different factors, but there are some points that recur in several of the papers: (i) analyses of case studies in order to better meet a country's needs (e.g. Raven *et al.*, 2008); (ii) publications associated with technological challenges designed to meet some requirement, such as comparing the efficiency of two feedstock, evaluate co-digestion or two biogas purification methods (e.g. Agyeman *et al.*, 2014); (iii) microbiological studies and ways of optimizing the degradation of the raw material and (iv) analyses of the carbon cycle, life cycle assessment and footprint (e.g. Evangelisti *et al.*, 2014).

The word cloud produced from the titles of the publications presented in Figure 2S (supplementary material) allows extracting important information about the focus of the academic studies involving AD technology and biogas production. Afterwards, the word cloud is qualitatively analysed, to obtain the general information of the retrieved articles. As mentioned previously, this study is a technology prospection to evaluate the biogas production chain and to identify the relation between research institutions and technology development.

Regarding the raw materials used as substrate for AD process, it was found that the ones receiving most attention by researchers are *sludge*, *sewage*, *waste*, *wastewater*, *agricultural biomass*, *crops*, *manure*, *municipal waste* and *other residues*. After examining the abstracts in detail, also publications that use algae as raw material for biogas production were identified. Of the total number of publications that mention its use in their abstracts, 55% deal specifically with microalgae. As municipal solid waste, industrial wastes were also mentioned in the abstracts, but in a lower number.

Substrates such as sludge, sewage, wastewater or manure are in many cases treated to effectively eliminate pathogens (Bruns *et al.,* 2011; Sutherland; *et al.*, 2015) and to allow the use of the digestate in agriculture (Gissén *et al.*, 2014). Other important feedstock includes agricultural biomass, crops, municipal waste, and lignocellulosic waste. To quantify information from word cloud, Figure 1 details number of publications for the main used substrates. It can be seen that sludge is the substrate with a higher number of publications along the studied period. Also, it can be noted that the use of algae and food waste as substrates for anaerobic digestion rose since the year 2011, and is still rising. At real scale, Forest (2012) reported that the most used wastes in Europe are manure, harvest residues and energy crops, being maize the first choice in the majority of existing co-digestion biogas plants.

The word cloud also shows the term pre-treatment. Indeed, its use in anaerobic digestion research is important as it expands the range of potential feedstock for this process. Pre-treatment methods reported include biological, mechanical or physicochemical processes to increase the anaerobic biodegradability of substrates. For example, Cesaro and Belgiorno (2014) reported different pre-treatments for improving food waste anaerobic digestion and Michalska *et al.* (2015) studied energy crops pre-treatment to increase biogas production. Such pre-treatment methods aim to accelerate the initial hydrolysis stage, which is traditionally the rate-limiting step in anaerobic processes dealing with high solid content; and caused by the presence of lignocellulosic and fatty fractions in various organic substrates. The use of pre-treatments allows that biogas production from the agricultural wastes has become a very fast growing market in Europe presenting an increased interest in many parts of the world in the last decades (Weiland, 2009).

It was also found many publications describing models for evaluating the environmental impact and the kinetics of the process. For example, Muha *et al.* (2015) modelled kinetics of anaerobic digestion or Marvuglia *et al.* (2013) presented a critical review on Life Cycle Assessment of biogas production. This is corroborated by the presence in the word cloud (supplementary material, Figure 2S) of the words *model, evaluation, environmental* or *impact.* There are also several studies that investigate specific aspects of the AD process, which is corroborated by the presence of words like *yield*, *optimization*, *assessment*, *community*, *thermophilic* and *co-digestion*.

Even though biogas is the mean goal of this paper, some interesting aspects of the process could be also investigated. Around 5% of all the publications investigate co-digestion, and around 6% use some kind of pre-treatment in the raw material. This result shows that the simultaneous digestion of two or more substrates, co-digestion, is increasing as an important way to improve the biogas yield. Pre-treatment is becoming an important step especially to residues that present high content of lignocellulosic materials. This is due to the fact that this strategy allows nutrients balance and control acidogenesis in the anaerobic digestion process (Esposito *et al.*, 2011). It is reported that simultaneous anaerobic digestion of different substrates can be a way of overcoming obstacles like adjusting the C/N ratio or the pH or regulating the nutrient content. For instance, manure has a low C/N ratio that will lead to high ammonia concentration in the digester, which could inhibit methanogenic bacteria. Meanwhile, municipal solid waste could contain toxic materials and a high concentration of heavy metals, inhibiting microbial growth. There is also the issue of the seasonality of agricultural and farm waste, which can hamper a continuous biogas generation process (Mata-Alvarez *et al.*, 2014). All these points make co-digestion a potentially advantageous option. Mata-Alvarez *et al.* (2014) also noted that 50% of all the articles retrieved about co-digestion were published in 2012 and 2013, and that 75% were published between 2009 and 2013. Also, for biogas purification, around 3% of the total studies is about cleaning and upgrading the biogas.

Continuing with the biogas chain (supplementary material, Figure 1S) and regarding the purification step, there is a recurrence of publications that address this topic directly or indirectly. Around 5.8% of the articles deal with biogas cleaning. In agreement with our results, Bauer *et al.* (2013), show that there is a growth in the studies related to the biogas purification. The authors suggested that the mean biogas upgrading technologies are: gas separation membranes, organic solvent scrubbing, amine scrubbing, water scrubbing and pressure swing adsorption (PSA). Also, future applications of upgraded liquefied biogas are increasing. This result also suggests that there is an increase in the use of purified biogas as observed in some European countries. According to Sustainable Energy Authority of Ireland (2012), in 2010 more than 1.4 million vehicles were using natural gas like fuel, and Sweden is the leading country in the conversion of biogas into biomethane.

In most of the articles it is not clear the final use of biogas, and some studies deal specifically with biogas production. In those papers where biogas is used, it was found that the main application of biogas (9%) is for electricity production. A small part, 3.9%, deals with the use of biogas for fuel cells. In agreement with Rathod *et al.* (2014), more efforts are required, especially in the reforming of the biogas.

3.1.1. Distribution of publications per year and per EU country

To demonstrate in which European countries these research articles have been published over the years Figure 3S (supplementary material), shows that although since the 1990s some studies had been published on biogas, it was only from 2000 that such research started to take off. This temporal evolution with high and low periods of productivity can be explained by historical factors. According to Chanakya and Malayil (2012), for example, in 1970 the biogas studies decreased, probably because the petrol boom, and the research about biogas only rising again in 1973 with the oil crisis. Nonetheless, it was only in the middle of the first decade of the 2000s, after the second energy crisis, that alternative technologies, including AD, were again investigated as potential sources of renewable energy. Since then, there has been a steady rise in the number of studies, and everything indicates that this high level of interest in this topic will be maintained in Europe, especially given the growing investments in renewable energy and the ambitious targets set.

When these data are considered for each country (Figure 2), Germany stands out as the leading publisher of articles, accounting for 42% more articles than the second-placed country, Italy.

Germany's leadership in the development of technologies for alternative and renewable energy sources, including biogas, is nothing new. It could be partly associated with the country's strong rejection of nuclear energy, but also its tradition of using AD technologies and biogas production. Back in 1906, Karl Imhoff developed an anaerobic wastewater treatment unit (the Imhoff tank) with separate spaces for settling and digesting the waste matter (Deublein and Steinhaiser, 2008). This and other developments have made Germany a pioneer in this area of research. Budzianowski (2016) pointed out that government financial support reinforces this leadership and the search for alternative energy sources, promoting in Germany the use of various types of organic material as substrates for biogas production, including dedicated energy crops.

Italy is the country with the second number of publications on the topic. Carrosio (2013) reported that the country's interest in this area has been heightened by some successful government incentives in recent years for the research and development of renewable energy and also with a system of obligations and incentives (Carroisio, 2014). The country has also an abundance of biomass and waste materials, which could be processed using AD, resulting in the production of biogas as one of the main energy sources. In fact, the number of facilities has increased in the last years, and also the number of them related to dedicated biomass produced specifically for energetic purposes (2015).

In Spain, like in other European countries, some important legislative marks have had a positive influence on the rising of biogas production in the country. One example is the Landfill Directive 99/31 of 26 April 1999. This legislation obliges to have a reduction of biodegradable waste that is deposited in landfills. This Directive scheduled the reduction of the amount of biodegradable wastes, gradual and mandatory, with values of reduction of 25% at 5 years 50% at 8 years and 65% at 15 years. This implies that in 2009, the reduction reached was 50%, while in 2016 should reach 65%. According to this Directive, it is expected that more potentially biodegradable waste usable for biogas production should be used in the next years (European Union law Council, 1999). However, this European legislation and the increase in biogas plants have not been supported by an adequate regulation (Sedigas, 2015).

According to a report published by Nordic Energy Research (2010), in Nordic countries (Denmark, Finland, Iceland, Norway and Sweden), the use of biogas is still limited, with Denmark and Sweden taking the leadership in its production. Although these countries have many features in common, the use of the biogas generated is very different. In Denmark, biogas is used in cogeneration plants, while in Sweden it is normally converted into vehicle fuel. Møller and Martinsen (2013) reported that Denmark's legislation supports the use of biogas, as it has set the target of having 50% of all the country's manure being processed to generate renewable energy.

In the United Kingdom, the biogas industry has had an increasing development, especially in the last five years. This crescent importance, as in other European countries, comes from government policy that is offering funding for new biogas projects (Morton, 2015).

Finally, regarding Poland (in the seventh place), Igliński, *et al.* (2015) reported that 39.44 pentaJoules (PJ) of energy could be obtained from Poland biogas production and if all this potential were harnessed, it would be able to cover 7.5% of the country's energy needs. These authors also stress that although there are many challenges associated with energy generation, biogas use has grown significantly in recent years and the trend is set to continue. At the same time, Bielski *et al.* (2015) reported that Poland has a large potential capacity for biogas generation, which could cover around 47% of the domestic demand for natural gas.

3.1.2. Main institutions of academic publications

Research articles have been analysed from the point of view of the research institutions. The institution with the highest number of articles in Europe is the Technical University of Denmark, accounting for 51.2% of its country's publications, which makes it a real benchmark for biogas and AD research. Likewise, in Sweden, just one institution, Lund University, has produced 26.2% of all the country's publications on the subject, while in France, the Institut National de la Recherche Agronomique (INRA) accounts for around 31% of this country's publications. It is important to mention that this institute is unique, but the research centres are spread all over France. In these three countries: Denmark, Sweden and France, there is a clear concentration of research efforts in a single institution that has a steady number of publications about biogas or AD processes.

In contrast, the institution responsible for most publications in Germany, the University of Hohenheim, has produced 8.6% of all the country's articles on the subject. Meanwhile, there are some intermediate cases like Spain, where research efforts seem to be spread between its research institutions, although five of these institutions account for 38.7% of the country's total academic output in this area.

When these publications were analysed to correlate the institutions with the countries (Figure 3), it was found that, when research is undertaken under partnership, this is generally done by institutions from the same country. Even so, several institutions have fluent relationships that led to the production of join articles, being Sweden, England and Austria the countries with the highest international collaborations.

There is also a strong association between Spain's institutions, such as Spanish National Research Council, the Autonomous University of Barcelona and the University of Valladolid, being this last institution the one that demonstrates, through publications, its international collaborations with Southampton University (UK), the Vienna University of Technology and the University of Innsbruck (Austria) or the University of Milan (Italy).

Meanwhile, Ghent University in Belgium and Institut National de la Recherche Agronomique in France have not co-authored any publications with other institutions from their own or other countries.

3.2 General evaluation of patent applications filed in Europe

Using the strategy described earlier, patent applications with priority in a European country from 1990 to 2015 were analysed. It was found that those documents represent 12% of the world (2.963) patent applications. Other countries like China (41% of the total), Japan (21% of all the documents), the United States (10%) and South Korea (7%) are the leading producers of patents of technologies for AD and biogas generation.

As for the content the retrieved documents, it was found that most of the patent applications are for devices, equipment, tanks, experimental apparatus, reactors and containers. It was demonstrated that the focus of most patents is on protecting new types of equipment for AD processes. In fact, 48% of patent applications are related with equipment for AD. This is highlighted in the word cloud (Figure 4S) (supplementary material), based on the titles of all the patent documents retrieved. The focus of most of these documents is on the production methods and processes: *producing*, *process*, *production, container, device, tank, system, unit* or *apparatus*. The main process is normally organic waste treatment. Sludge (15%), wastewater (10%) and agricultural waste (7%) are the main substrates claimed in patent applications; they are like those used in academic publications.

There is also a focus on biogas purification, accounting for 12% of patent applications in Europe. Compared with 48% of those related to equipment it suggests that the most part of the innovative activity is focused on the development of new equipment for the process of AD rather than biogas purification.

The word cloud also highlights the final applications of biogas suggested in some of these patent documents: *generating*, *heat*, *hydrogen*. This finding is also corroborated by Alves *et al.* (2013), which defends the use of biogas for the production of hydrogen because it is a versatile gas from alternative raw materials and an excellent source of methane. There are also words that are associated with the purification of biogas and the separation of methane from its other components. This could be associated with the necessity to cover the biggest scope as possible. Around 41% of the claims cover subjects related to the use of biogas, the most frequent is energy generation (23%) of the total patents that mention the use. The step of purification or cleaning the biogas was mentioned in 12% of the claims, suggesting that innovation in this step is still possible.

An analysis using the International Patent Classification (IPC) codes was made. Most of the patents identified were classified in section C, which covers the areas of chemistry and metallurgy. The subclass under which the highest number of patent applications in this area was classified was C02F, for the treatment of water, wastewater, sewage or sludge. The next most frequent categories were apparatus for enzymology or microbiology (C12M), fermentation or enzyme-using processes to synthesize a chemical compound (C12P), and separation of solids or liquids (B01D).

Another interesting feature is that biogas production is not always the target of the protection granted by the patents. Often, it is a method or process designed to treat a solid or liquid waste, and biogas production is merely a welcome side effect. This is interesting as waste is reported as a source of new products different from biogas (Sánchez *et al.*, 2015).

Considering the purification step, more interest was detected in the patent documents compared to research articles. This is probably justified by the greater easiness of protecting a method or equipment through a patent. It was found that around 12% of the patents have at least one claim about biogas purification. In spite of that, about 2.2% of the patent applications mentioned the use of biogas in fuel cells and 5.8% mentioned the biogas to be used to generate electricity. This result is in agreement with Warlick (2015) that carried out a study about patents of anaerobic digestion for the production of fuel cells. In this study, the author show that 70% of the deposits have been published in the last five years which although demonstrating a great advance is based on anaerobic digestion improvements of the technology, and only a few patent applications are directed to the less known technologies: as cells of microbial fuel. Correlating the information of articles and patents for use of biogas it was found that fuel cell is an important application that is developing fast. The analysis of the claims of the patent documents allows to identify the technological solution, which is already commercially in application and industrial use. Differently from the analysis of the articles that allow us only indicative of the emergence of new concepts and ideas still at the stage of development.

3.2.1. Distribution of patent applications per year and per EU country

Figure 5S (supplementary material) shows how the patenting of technology for this area has developed over the period analysed (1990-2015). It must be considered that due to the delay in indexing the base and/or confidentiality period of a patent (18 moths), the number of documents with priority in 2014 and 2015 may not be complete. The number of applications started to grow in 2007, and continues to remain high. However, new filings have been made consistently throughout the period under study, confirming that this is not a new technology and that it has always been studied. However, it is gaining interest now probably because of environmental concerns.

Indeed, part of the rise in the number of patent applications could be due to the need to comply with increasingly stringent legislation and the opportunity that these technologies represent for companies to obtain economic and environmental gains. Figure 4 shows a view to demonstrate which European countries are the leading patent producers in the field of biogas and AD technology, also shows the strong predominance of patents whose priority country is Germany. Around 47% of all the patents filed in Europe are from this country.

USSR filed 54 patent applications between 1990 and 1993; after this date, it split into different countries and Russia inherited the technologies it had developed. Given the large number of patent applications, this is a country with a strong history in the field. The graphic also shows that some patents are designed to provide Europe-wide protection by being filed at the European Patent Office.

In Figure 4 it is also shown that Germany is the leader country as priority patent country, in all the years shown (1990-2015); it was found that always at least 38% of the documents have priority in this country. Besides, it was found that Switzerland is starting to become more relevant in the last year with available data (2015). This increase is largely due to biogas plants combining farms waste with animal husbandry.

3.2.2. Main protected markets

An analysis of the countries where the patent families are distributed was made to identify which markets these patents with priority in Europe are seeking protection in. A patent family is a group of inventions which, like a family, are all interrelated, in this case via the priority patent. It can be seen from Figure 6S (supplementary material) that these patent applications are designed to provide protection inside the EU. This is proven by the overwhelming presence of European countries in the pie chart, together with the high number of patents filed regionally at the European Patent Office. The non-EU countries where the European inventions are protected by patent are China, Australia, Canada, the United States and India.

3.2.3 Main institutional assignees

Figure 5 shows a map correlating the leading assignees and the technologies their patents are designed to protect. The technologies were correlated using their respective IPC subclasses. It shows some groups with common interests, like the association between four German companies, UTS Biotechnick, Schmack Biogas, Bekon Energy Technologies and Agraferm Technology, which all have patents classified under IPC subclass C12M, which is for apparatus for enzymology or microbiology. In other words, they have to do with the development of fermentation reactors and AD tanks and chambers.

Another major group is constituted by the patents whose technologies are classified under IPC subclass C02F, the assignees in this group are Agriculture Electricity Research Institute (Russia), Paques (Netherlands), Degremont (Spain), Voith Paper (Germany), Council Science & Industrial Research (India) and Fraunhofer Ges Foerderung Angewandten Ev (Germany). The Indian research centre that is part of this group has filed priority patents in Europe, showing that there is also an interest on the part of non-residents to protect their technologies in the EU. Two Germany companies were also identified; DGE Guenther Enge and Evonik Degussa, which both have technologies classified under IPC subclass BO1D, which covers the separation of solids and liquids. The technological solutions are associated with the development of equipment to improve the process and the main companies engaged in this stage provide turnkey engineering solutions for AD plants.

It is clear from Figure 5 that biogas is not sought as a source of energy, but as a secondary benefit for industry players when they treat their effluents and solid waste. Likewise, it was found that biogas production is an offshoot of waste management and a method for producing renewable energy, and it is also important for nutrient recovery

(Huttunen *et al.*, 2014). In the early 1990s, there were few companies working in any of the stages of the biogas production chain, but since 2000 the number has grown exponentially. In 2015, there are over 700 companies around the world, most of which are construction companies and plant operators (Martin, 2014). In view of this trend, it is expected that the main actors involved in this field will become increasingly diverse, including chemicals companies, equipment manufacturers, and gas companies (Global Intelligence Alliance, 2010).

3.3 From research to market

Patents and research articles are associated with two independent processes; however, science and technology or research and innovation are strength correlated. In this study, this correlation could be measure by using a patentability ratio (number of patents/number of scientific articles) per each country. In this study, it can be found that this ratio varies from 0 to 2. As closest to 0, the lower value, the average interconnection shows a weak degree between academic research and scientific development, demonstrating that there is still no effective conversion of studies in business solutions (patents). In the same way, if this ratio is closer to 2, it means that there is a strong degree between the academic research (articles) and technological development (patents). In Figure 6, the patentability ratio show that most European countries have produced more academic papers than patents in this area (ratio lower than 1). This reflects the logical fact that not all research constitutes knowledge that can be necessarily transformed into patentable intellectual property. However, it was found that Germany and Sweden have more patent applications than academic publications. Also, Russia (data not show in Figure 10) presents a ratio of 11, which due to the studied period includes some patents coming from Union of Soviet Socialist Republics (URSS). It is shown in Figure 6 that Germany is the country that has the highest ratio (1.79), being also the European country with highest number of plants and biogas leader producer. Also, Switzerland has a good ratio of R&D (1.68). This could be because in Europe a patent application must describe an innovation that has never been described before. Greece and Ireland are countries with the lowest patentability ratio.

According to the European Biogas Association (2014), there are over 14,500 biogas plants installed in Europe, and the number is still rising. The clear leaders are Germany and Sweden. In 2013, the number of biogas plants in Hungary, the Czech Republic, Slovakia and Poland increased by 18%. Recently, Italy approved feed-in tariffs for biomethane to be injected into the natural gas grid, which will certainly help in diversifying the country's energy profile.

Meanwhile, in Germany there is a considerable amount of public funding for renewable energy producers. In 1991, a new law was passed (Stromeinspeisungsgesetz - StromEinspG) that introduced feed-in tariffs, providing a minimum of compensation for any electricity generated from renewable energy sources and exported back into the grid, including biogas (Sutherland *et al.*, 2015). Germany sees R&D as strategic, since this country integrates technology with its business strategy, applying research to effective results. R&D is a way of helping businesses expand their activities, find new opportunities, and expand their technological capacity. Also, Germany is clearly the most important country of the European biogas sector, in terms of installed production and capacity although it prioritizes the construction of small capacity plants.

Raboni and Urbini (2015) reinforced that not only Germany but also other countries such as Austria, Czech Republic and the Netherlands prioritize plants of small scale. On the other hand, countries like France choose to adopt higher capacity plants. In this sense, there is the advantage of economy of scale, but generally larger plants are more complex to operate.

Overall, 90% of the plant capacity built in the EU is in the range of 15.000 to 80.000 tons y^{-1} and the average values of capacity are 38,000 tons y^{-1}. It is worth noting that some plants with a capacity exceeding 200,000 tons y^{-1} are present (Raboni and Urbini, 2015).

The new legislation and funding demonstrate that companies are leading in the R&D of AD technology. It is worthwhile for companies to protect their technology via patenting, because they can charge royalties for its use or license it to third parties. In recent years, the number of biogas plants has been on the rise, especially since governments have brought in higher subsidies for the installation of new facilities (Deublein and Steinhaiser, 2008).

4 Conclusions

With Europe's increasingly stringent regulations geared towards sustainable energy use, anaerobic digestion has come to be an opportunity for the continent's countries to treat their waste more effectively and generate energy or fuel from biogas. The application of technology prospecting has allowed establishing a relation between academic research (published articles) and technology development (patent applications) evolved from 1990 to 2015. Our findings show that from the 2000s, patenting in this area has increased considerably, and it is likely that the current high numbers will continue in the next years.

The country in the continent with the most priority patents is Germany, which is also the country where most academic papers are published, followed by Italy and Spain. Other EU countries tend to protect their technologies regionally. It was found that Germany leadership is probably associated with the time and resources invested in the AD technology to make the biogas a useful source of energy.

The biogas industry is dynamic and multidisciplinary, and associated with multiple factors, especially the countries' expertise acquired over the years, and the incentives supplied by their governments through feed-in tariff policies or projects designed to boost biogas production. Biogas generation is not only linked to energy generation, but is intrinsically related to other factors like waste treatment, reducing greenhouse gas emissions and decreasing the demand for non-renewable resources.

There are different aspects of biogas production chain where policy decisions can still promote the improvement of the process: i) as new materials are intended for anaerobic digestion, more investigation on pre-treatment processes will be needed in order to maximize biogas production; ii) use of additives to increase biogas production or iii) in parallel with the new uses of biogas (injection into the natural gas distribution network, used in fuel cells or as fuel for vehicles) new research efforts must be done to reduce costs of biogas upgrading.

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