New concept for energy storage: microwave-induced carbon gasification with CO₂

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Abstract

Energy storage is a topic of great importance for the development of renewable energy, since it appears to be the only solution to the problem of intermittency of production, inherent to such technologies. In this paper, a new technology for energy storage, based on microwave-induced CO₂ gasification of carbon materials is proposed. The tests carried out in this study on different carbon materials showed that charcoal consumes the least amount of energy. Two microwave heating mechanisms, a single-mode oven and a multimode device, were evaluated with the latter proving itself to be the more efficient in terms of energy consumption and recovery. The initial results obtained showed that this technology is able to achieve energy efficiencies of 45% at laboratory scale with every indication that these results can be improved upon to make this approach highly competitive against other energy storage technologies.

Keywords

CO₂ gasification; Microwave heating; Energy storage; Energy efficiency

1. Introduction

Energy storage is one of the main concerns of the energy industry [1, 2]. Apart from the electronics or transport sectors where energy must be stored before it can be used in mobile applications (e.g. mobile phones, computers, cars), the peculiar characteristics of renewable energy generation have promoted research into the use of energy storage in static devices on a high scale [1, 2].

Renewable energy has an important drawback that stems from its intermittent production [2-6]. The generation of electricity by means of renewable technology gives rise to peaks (e.g. when there are strong gusts of wind or in the central hours of the day when solar radiation is higher) and valleys in production (e.g. when the wind stops or during the night). These peaks and valleys do not always coincide with the peaks and/or valleys of demand. This situation leads to important supply problems [3, 4]. For this reason, it is essential to develop systems that allow the storage of excess energy produced in the peak hours, so that it can be subsequently used in periods of low production and high demand [3, 4].

Traditionally, large-scale energy storage has been achieved by means of dams. However, such facilities cause considerable environmental and socio-economic problems [1, 7]. For this reason, new solutions are being proposed to reduce the negative environmental impact and to avoid unwanted socio-economic side effects. Technologies commonly applied to mobile systems (fuel cells, batteries, capacitors) have been studied, but they are designed for use on a smaller scale or for different purposes [8-11]. Another possible solution could be to use chemical energy

transmission systems (CETS). These systems were initially intended for use in nuclear plants [12, 13]. The idea underlying these systems is to use a reversible reaction. In the first step of the process, the energy is consumed in an endothermic reaction. Then, in the next step, the energy is liberated in an exothermic reaction. Figure 1 illustrates this process [12].

FIGURE 1

To date, the reactions that have been proposed for this kind of system are heterogeneous catalytic reactions, the most frequently employed being the CO₂ reforming of CH₄ [13-17].

However, the difficulties involved in implementing this on an industrial scale (especially that of the rapid deactivation of the catalyst) and the extreme conditions needed to achieve high conversions in the exothermic step have obstructed research in this area. For this reason, it is necessary to find alternative solutions. Gasification of carbon materials could be an attractive way to modify these systems. This solution is based on expending energy by gasifying a carbonaceous material and then recovering the energy spent by combusting the gases produced in the previous gasification steps. Of the different gasifying agents (steam, O₂, CO₂), CO₂ is the best option (reaction 2), since it is a product of combustion and can be recycled for use in the gasification step (see Figure 2).

FIGURE 2

CETS have been proposed for use in nuclear plants, since the high temperatures achieved in these facilities can be used for the endothermic reactions. However, in the case of renewable energy, the electricity is normally produced directly, which means that there are no high temperatures that can be used in the gasification step. Microwave heating avoids this problem, since it transforms electricity directly into heat. Carbon materials are, in general, good microwave absorbers and can facilitate gasification [18]. Moreover, microwave heating has been proved to be an excellent technology for enhancing catalytic heterogeneous reactions, since it is able to increase conversions and selectivities in heterogeneous processes, such as NO_x reduction [19, 20], SO_2 reduction [21] or the CO_2 reforming of methane [22]. These results are mainly due to a “pseudo-catalytic” effect resulting from the appearance of microplasmas during microwave heating [23]. However, only a few works have obliquely dealt with the microwave-induced CO_2 gasification of carbon materials [24, 25].

The main objective of the present work is to study the microwave-induced gasification of several carbonaceous materials and to evaluate the extent of energy recovery through the subsequent combustion of the gas produced in the gasification, and thereby assess this concept as an alternative for storing energy in intermittent production technologies based on renewables.

2. Materials and methods

2.1. Materials

Four carbon materials were evaluated for gasification with CO₂: anthracite (A), activated carbon (AC), charcoal (CC) and metallurgical coke (CK). All the materials proved to be good microwave absorbers as received with the exception of charcoal. This material needed to be devolatilized at 600 °C before being able to absorb microwaves.

2.2. Reactivity of the materials

The CO₂ reactivity profiles of the four carbon materials were obtained using a thermogravimetric analyser (TGA) SDTQ600 from TA Instruments. The samples were heated from r.t. to 1000 °C at a rate of 10 °C·min⁻¹ under a flow rate of 25 mL_{STP}·min⁻¹ of CO₂.

2.3. Microwave-induced gasification procedure

The experiments were carried out in two different microwave ovens: a multimode oven and a single-mode oven (see Figure 3).

FIGURE 3

Essentially both ovens consist of a magnetron where microwaves are generated and a waveguide which conducts them to the reactor. In the case of the multimode oven, the

waves are conducted to a multimode cavity where the reactor is located. The reactor is a quartz tube with an internal diameter of 26 mm. Part of the tube is filled with Raschig rings and quartz wool, on top of which approximately 6 g of carbon material is placed. The height of the filling is adjusted so that the sample is on the same level as the window of the waveguide. In the case of the single-mode oven, the reactor is situated inside the waveguide. The sample is introduced using the same method as in the multimode system. Both ovens have tuning screws which prevent reflected power from being sent back to the magnetron and damaging it. The temperature is controlled by means of a thermocouple placed inside the reactor. This parameter was limited to 1200 °C. The main difference between the two heating devices is in the way the waves reach the sample. In the single-mode device the waves propagate in a single direction, reaching the sample from this direction. In the case of the multimode oven, the reactor is placed in a resonant cavity where the waves propagate in several directions and reach the sample from all of these directions.

Before the experiment, the reactor was purged with N₂ at a flow rate of 100 ml/min for 30 minutes. The microwave power was set to operate between 200 and 650 W before being switched on. Once the sample temperature had stabilized, the flow of N₂ was stopped and the CO₂ flow was switched on. The CO₂ flow rate varied from 25 to 650 ml·min⁻¹. The duration of the experiments was 30 minutes. The outlet gases were collected in Tedlar® bags with a polypropylene fitting for sampling and then analysed on a Varian CP-3800 gas-chromatograph equipped with a thermal conductivity detector (TCD) and two columns (an 80/100 Hayesep Q and an 80/100 Molesieve 13X) connected in series. The second column was bypassed by a six-port valve for the

analysis of CO₂ and hydrocarbons ($\geq C_2$). The conversion of CO₂ was calculated by means of the following equation:

$$\text{CO}_2 \text{ conversion, \%} = 100 \times [(\text{CO})_{\text{out}}/2]/[(\text{CO}_2)_{\text{out}} + (\text{CO})_{\text{out}}/2] \quad (\text{equation 1})$$

A pre-established condition for studying energy consumption was that the conversion of CO₂ should be higher than 95%. After the established amount of carbon material (6 g) had been introduced and the CO₂ flow rate set (25-650 ml·min⁻¹), the microwave-power was selected. If under these conditions the conversion of CO₂ was higher than 95% (at a temperature of < 1200°C) the experiment was considered valid for this study. Otherwise the experiment was discarded and repeated at a higher microwave power. This procedure was repeated until the conversion had reached at least 95%, at < 1200°C. This parameter was selected after preliminary tests had revealed differences in energy efficiency to be negligible between 80 and 95% of conversion (see Supporting Information, Fig. S1) due to the compensation of the energy spent in the gasification by the energy recovered in the combustion. Table 1 shows the experiments that satisfied this condition. None of the experiments with anthracite or coke are included in the table since the highest CO₂ conversions achieved were 50% and 70% respectively.

TABLE 1

3. Results and discussion

3.1. CO₂ reactivity

Figure 4 shows the CO₂ reactivity profiles of the four materials.

FIGURE 4

All of the materials exhibit different behaviours. Anthracite started to react at temperatures of around 450°C and showed a very low reactivity over the entire range of temperatures studied (see TableS1 in the Supporting Information for further details). Coke started to react at a slightly higher temperature than anthracite. However, it showed quicker reaction rates, especially at temperatures higher than 650 °C. As both of these materials exhibited a low reactivity they were discarded for this study. The decision to discard anthracite and coke was reinforced by the poor results obtained in the first microwave-induced CO₂ gasification tests, since at 1200°C (the limit chosen for the process), under a very high microwave power, the conversions were only 50% for anthracite and 70% for coke, whereas charcoal and activated carbon reached conversions of 95% under considerably milder conditions.

Activated carbon and charcoal showed higher reactivities than anthracite and coal. They exhibited an initial mass loss that can be attributed to adsorbed water. They then start to react with CO₂ at 350 °C in the case of charcoal and at 475 °C in the case of activated carbon, showing in both cases high reaction rates. At 630 °C, all the charcoal has reacted, whereas in the case of activated carbon the reaction was not completed until a temperature of 800 °C had been reached. Anthracite and coke were not able to react completely even at 1000 °C. The reactivity profiles show that charcoal is the material that has the highest reaction rates with CO₂ and that charcoal and activated carbon are the most suitable materials for the purpose of this study, whereas anthracite and coke can be discarded because of their low reactivities. Although charcoal presented the best

reactivity, activated carbon was also selected for purposes of comparison since it is known to be as an excellent microwave absorber [22, 26].

3.2. Microwave-induced gasification with CO₂

Once coke and anthracite had been discarded, the study of the influence microwave-induced gasification with CO₂ was performed only with activated carbon and charcoal. The influence of the materials, particle size and heating device was studied. Only the experiments that fulfilled the pre-established criterion of 95% of conversion were considered valid for the study.

3.2.1. Influence of the material used

Fig. 5 shows the specific power (expressed in kW per kg of carbon material) needed for the CO₂ gasification of charcoal and activated carbon at different volumetric hourly space velocities (VHSV). In the same figure, the approximate temperatures achieved in the process (not the exact ones due to the difficulties involved in measuring temperature in microwave systems [27]) are also shown. The gasification process was carried out in batch. Therefore it was not possible to work at steady state because the mass of the sample was constantly diminishing. The figure shows the mean values obtained after 30 minutes of reaction for each experiment. The energy measured was the energy spent during these 30 minutes. The energy value was then divided by the mean value between the initial mass of sample (6 g) and the final amount of sample (slightly less than 6 g).

FIGURE 5

The results indicate that charcoal is the material that functions with the lowest specific power and therefore consumes the least amount of energy. At low VHSV_s, both materials present similar consumptions. However, the gap between the consumption of each of these materials increases as the VHSV increases. For this reason, the rest of the study was performed only with charcoal.

In the case of charcoal, at low VHSV_s, the variation in specific power needed to achieve a conversion of CO₂ higher than 95% was considerable. However, at high VHSV_s, the specific power required varied only slightly, although the increase in the amount of CO₂ reacted was substantial and, therefore, the increase in the energy consumed due to the reaction was higher than at low VHSV_s. This suggests that the heating of the material contributes more to the consumption of energy than the energy demanded by the reaction. At low VHSV_s, lower temperatures are needed and the energy required to heat the material is lower. At this point the energy required by the reaction could have a greater influence on the total amount of energy consumed. However, at higher VHSV_s there is a corresponding increase in temperature which is probably the main contributor to the energy consumed.

3.2.2. Influence of the particle size

It is well known that particle size plays a key role in processes of this type [28-30]. The lower the particle size is, the higher the surface area is and therefore, more area is available for the gasification reaction with CO₂. In order to assess the influence of this parameter on the consumption of energy, experiments were performed using different

particle sizes: 1-2 mm, 2-3 mm and 3-5 mm. Figure 6 shows the results of the specific power needed for the CO₂ gasification of charcoal using different particle sizes. As was expected, as the mean particle size increased, the energy needed to obtain 95% of CO₂ conversion increased. This is due to the lower specific area available for the reaction of gasification with the CO₂. This suggests that it is better to work with the smallest possible particle size. However, if the particle size decreases, the pressure drop along the reactor augments, which makes the process more costly.

FIGURE 6

3.2.3. Influence of the heating device

Few studies are devoted to comparing different microwave-heating mechanisms. However, since multimode and single-mode mechanisms are different, differences in energy consumption could arise from the type of device being used. For this reason, the results obtained in the multimode oven were compared with those of a single-mode oven. The results of this comparison are shown in Figure 7.

FIGURE 7

At low VHSV's, the single-mode oven appears to need lower specific powers than the multimode oven. However, these differences are quite small and show a similar tendency, whereas at high VHSV's, the results are completely different. Whereas the energy consumption in the multimode oven is stable, the consumption in the single mode oven soars. This can be attributed to the transmitted power. When the power

supplied in the single-mode oven to the sample is high, a considerable amount of this power cannot be absorbed by the sample and is transmitted through it until it reaches a water circulator where it is dissipated. In the case of the multimode oven, the power not absorbed by the sample is transmitted through it, reflected against the walls of the cavity and then back again to the sample continuously. In this way, part of the power not absorbed initially by the sample is gradually dissipated with each reflection against the walls, further absorption occurring with each subsequent rebound. Although the multimode oven is better insulated than the single-mode oven, the influence of the loss of energy through the walls of the oven can be ignored, because of the low values of the difference in energy loss between both devices, estimated to be in the order of 2-4 kW·kg⁻¹.

3.3. Energy efficiency of the process

Once the consumptions have been determined, it is important to establish the energy efficiencies of the systems by comparing the energy spent in the microwave-induced CO₂ gasification of the charcoal and the energy that can be recovered by means of the combustion of the CO produced. Figure 8 shows the results of the energy recoveries of the single mode and the multimode systems. The results for the energy efficiency shown in this figure have been calculated as follows:

$$\begin{aligned} \text{Energy recovery (\%)} &= \frac{\text{MW Power (kW)}}{\text{Power CO combustion (kW)}} \cdot 100 \\ &= \frac{\text{Power measured in the device (kW)}}{\Delta H_{CO \text{ comb}} \left(\frac{\text{kWh}}{\text{m}^3} \right) \cdot Q_{CO} \left(\frac{\text{m}^3}{\text{h}} \right)} \cdot 100 \end{aligned}$$

FIGURE 8

In the range of VHSVs studied the energy recoveries in both systems increases to maximum values of 30% in the single-mode device and 44% in the multimode device. If these initial results are compared with those of other storage technologies (see Figure 9), it can be seen that these data are almost as good as the recovery values achieved by systems like batteries, which have similar autonomy periods and power rates to those of microwave-induced CO₂ gasification systems. This is one of the most important advantages of the proposed technology. Due to its versatility, it is able to cover a wide range of autonomy periods and achieve high power rates. Comparison with fuel cells is also very favourable to the system proposed here. Fuel cells show good efficiencies (60-80%) [10], although these efficiencies are dramatically compromised by the previous generation of H₂ [31, 32], which diminishes the global efficiency of fuel cells to values of around 20-45%, lower than those achieved by the system proposed in this study. This represents a significant achievement given that the values reported in this work were obtained at lab-scale with devices that were not optimized for energy efficiency. Other alternatives like water reservoirs (pumped hydro storage) or flywheels provide higher efficiencies, but these technologies have serious drawbacks, e.g. the environmental and socio-economic impact in the case of water reservoirs or the loss of efficiency with storage time in the case of flywheels [33, 34]. Only supercapacitors are far superior to the technology we propose, but they are just in their infancy and their characteristics (high power density and high charge/discharge rate) [35, 36] make them more appropriate for other uses, in hybrid electric vehicles or portable electronic applications where the rapid storage and release of energy at high power rates are required. It should also be pointed out that, in the system proposed in this work, high quality energy (the

electricity supplied to the microwave oven) is recovered as low quality energy (the heat obtained in the combustion of CO). Moreover, not all the energy from the combustion of CO can be recovered, since this process suffers from its own efficiency. Nevertheless, in this study the results were obtained at lab-scale and it has been shown that the energy efficiency of microwave heating dramatically improves at greater scale [37].

FIGURE 9

Moreover, from Figure 8, it can be seen that, in the single-mode device, the rate of increase of energy efficiency starts to decrease at the highest VHSV, whereas in the multimode device the rate of increase remains constant. This indicates that the energy recovery achieved in this preliminary study can be improved upon in both devices, but especially so in the multimode oven. For example, the initial devolatilization (see Materials and Methods section) needed in the charcoal could provide an interesting solution for increasing energy efficiency. The hot gases leaving the reactor could be used to devolatize the charcoal before it is fed to the reactor. This would lead to an increase in the calorific power of these gases thanks to the volatiles produced and incorporated to the gases from gasification. Further studies are currently being undertaken to improve upon these initial results by upgrading the design of the microwave devices.

4. Conclusions

A new system for energy storage based on the microwave-induced CO₂ gasification of carbon materials has been proposed. Four different materials were studied. Anthracite and coke showed very low reactivities with CO₂, whereas activated carbon and charcoal exhibited excellent reactivities. Charcoal yielded the best results in terms of energy consumption, especially at high VHSVs.

Particle size has an important influence on energy consumption, since as the size increases, the power needed for the process also increases. Therefore, a compromise solution between pressure drop and energy consumption must be found.

A comparison between multimode and single-mode microwave heating was carried out. It was found that the multimode heating involves less energy expenditure and is more energy efficient.

The process proposed in this study offers higher energy efficiencies than H₂-based fuel cells exhibiting values similar to those of batteries. Other technologies like water reservoirs and flywheels show higher energy efficiencies, but give rise to environmental and socio-economic problems. Only supercapacitors seem to be superior to the microwave-induced CO₂ gasification of charcoal.

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Figure Captions

Fig. 1. Scheme of a CETS.

Fig. 2. Scheme of the modified CETS proposed.

Fig. 3. Microwave devices employed in the study: a) Multimode (1. Control panel; 2. Power meter; 3. Waveguide; 4. Tuner; 5. Microwave cavity; 6. Reactor; 7. Insulation); b) Single mode (1. Power regulator; 2. Thermocouple; 3. Magnetron; 4. Waveguide; 5. Tuner).

Fig. 4. Profiles of reactivity of activated carbon (AC), anthracite (A), coke (CK) and charcoal (CC) with CO₂.

Fig. 5. Variation in the specific power needed to obtain CO₂ conversions higher than 95% versus the volumetric hourly space velocity of activated carbon and charcoal in the multimode oven. The approximate temperatures reached in the process are shown in parenthesis.

Fig. 6. Variation in the specific power needed to obtain CO₂ conversions higher than 95% versus the particle size of the charcoal in the multimode oven.

Fig. 7. Specific power consumptions required to achieve 95% of conversion in the CO₂ gasification of charcoal in a single-mode and a multimode microwave device.

Fig. 8. Energy recovery through the microwave-induced CO₂ gasification of charcoal in a single-mode and a multimode microwave device.

Fig. 9. Autonomy period, power rate and energy efficiency covered by different energy storage technologies.

Table 1. Experimental conditions in the different assays that fulfilled the pre-established criterion of 95% of conversion

Material	Oven	Size (mm)	VHSV (m ³ /kg·h)
AC	Multi	1-2	0.25
AC	Multi	1-2	0.61
AC	Multi	1-2	1.84
AC	Multi	1-2	3.26
AC	Multi	1-2	5.26
CC	Multi	1-2	0.25
CC	Multi	2-3	0.25
CC	Multi	3-5	0.25
CC	Multi	1-2	0.67
CC	Multi	1-2	1.45
CC	Multi	1-2	3.37
CC	Multi	1-2	5.38
CC	Multi	1-2	9.58
CC	Single	1-2	1.05
CC	Single	1-2	2.36
CC	Single	1-2	4.22
CC	Single	1-2	6.63

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Supporting Information

Table S1. Temperatures for 50% of conversion and total conversion corresponding to the CO₂ reactivities of the carbon materials studied.

Parameter	Anthracite	Coke	Activated carbon	Charcoal
T _{50%} (°C) ^a	>1000	820	650	550
T _{100%} (°C) ^b	>1000	>1000	800	620

^a T_{50%} represents the temperature at which the material has lost 50% of its initial mass

^b T_{100%} represents the temperature at which the material has lost all the volatile matter

Fig. S1. Influence of the conversion achieved in the microwave-assisted gasification of charcoal upon the energy recovery of the system (unimode device, VHSV = 7 m³ kg⁻¹ h⁻¹).