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Carbon Capture and Storage. A case study of mineral storage of CO_2 in Greece.

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Abstract

While the demand in reduction of CO_2 increases, the need for CO_2 sequestration processes is very high. One promising technology is the Carbon Capture and Storage (CCS). In this paper we refer to several papers which study the three main steps in CCS chain. CO_2 capture technologies, CO_2 transportation to the storage sites and the very critical step the CO_2 storage. Recently a novel method (mineral carbonation) for CO_2 sequestration has been proposed which is based in the reaction of CO_2 with calcium or magnesium oxides or hydroxides to form stable carbonate materials. Greece is a country that emits CO_2 mainly from the lignite fired power plant in Western Greece. After the study of the bibliographic references about the use of mineral carbonation process while injecting CO_2 in the appropriate geological forms we concluded that there are also these forms in our country and mainly in the area near to the power plant such as in sites Vourinos and Pindos. In these sites exist minerals rich in oxides and hydroxides of Ca, Mg and Fe representing the perfect materials for mineral carbonation.

Keywords: Carbon Capture and Storage, mineral carbonation, CO_2 sequestration, Greek Power Plants

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1. Introduction

Nowadays there is an increasing demand for energy resulting in the increase in the use of fuels, particularly the conventional fossil fuels (coal, oil and natural gas). Despite the fact that the fossil fuels are the key energy source since the industrial revolution they are causing the same time, through their combustion, a serious threat for the Environment emitting to the atmosphere high amounts of CO_2, a major anthropogenic greenhouse gas. It is clear that the human activities influence the climate system.[1] In 2016 the average concentration of CO_2 (403 ppm) was 40% than in the mid-1800s. It was estimated that the CO_2 concentration increased about 2 ppm/year in the last ten years.[2] In the light of global commitment achieved in Paris 2015, the rise of global temperature should be kept below 2 ^{o} C compared to pre-industrial levels and also the temperature increase should be limited to no more than 1.5 ^{o} C (UN Paris Agreement 2015). According to IEA the goal of Paris Agreement requires the storage of at least 1 gigatonne of CO_2 annually by 2030. A critical technology that could help in the fulfillment of the above goals is the Carbon Capture and Storage) CCS. The objective of CCS is to capture and store CO_2 in several manners. The CCS uses existing processes and technologies available in the oil and gas industries to capture the CO_2 and store it deep below the surface in appropriate geological formations for permanent storage.

The aim of this paper is to present the several capture, transportation and storage strategies according to bibliography. It also presents the CCS technologies around the Europe. Giving emphasis on the third part of the CCS chain (the storage) it concludes that the mineral carbonation could be a promising CO_2 storage technique. Taking into account that lignite combustion is the main industrial way to produce electricity in Greece and emits high amounts of CO_2, we tried to give some ideas in using CCS

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technology in Greece power plants. Based only on bibliographical references about the geology around the Greek Power Plant area, we concluded that mineral carbonation in sites Vourinos and Pindos under appropriate circumstances could be a potential way of sequestrating CO_2 in a safe and permanent way.

2. Materials and Methods

2.1 CO_2 Capture Technology

There are three technological routes for CO_2 capture from power plants: Pre-combustion capture where fuels are converted to H_2 and CO_2 and the CO_2 produced is separated before combustion, Post-combustion capture where CO_2 is separated from flue gas, which is produced by fuel combustion and Oxy-fuel, where pure oxygen is used instead of air during combustion, leading to a flue gas stream of nearly pure CO2. However, the application of this technology may reduce the efficiency of the plant by 14% and increase the cost of electricity by 30-70%).[3] The post combustion capture is of particular interest because it is a possible near–term CO_2 capture technology that can be used to existing power plant.[4] As a result we are going to focus mainly on post combustion technologies in this paper.

Chemical absorption is one of CO_2 Capture Technologies. According to [5-7]the classic CO_2 absorbent is aqueous monoethanolamine (MEA) especially for CO_2 separation in electricity generation. The first full-scale commercial post-combustion carbon capture and sequestration (CCS) project operated in coal fired power plant in Estevan, Saskatchewan, Canada used an amine based process reducing CO_2 emissions. There are also new absorbents [5] that has been studied for this purpose as single amine absorbents, amine blends, multi face absorbents e.g. the formulation of

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aqueous piperazine (PZ) and 2-amino-2-methyl-1-propanol (AMP), econamine FG+, KS-1 and Cansolv.

Adsorption is another technology for CO_2 capture. The use of adsorption process in electric power plants by [8-9], indicates that this technique can be used to power plants. Some classical adsorbents are carbons, aluminas, zeolites, silicas, metal organic frameworks, hydrotalcites, poliymers etc. More details about the adsorption in CO2 capture technologies and their development are indicated in[5].

There is another process, relatively new, proposed by Shimizu T. 1999[10] for CO_2 removal from flue gas released from air-blown combustion systems. The calcium looping process separates CO_2 using the reaction $CaO + CO_2 \rightarrow CaCO_3$ and regeneration of CaO using O_2 combustion. It is of interest the key advantages of this technique[11]: Large amount of high recoverable heat (600-900 ºC), Possible increase in power plant energy penalty (40-60%), no flue gas cooling and pretreatment (SOx) and finally it has low emissions and affordable price. A review of the calcium looping technology and its progress is indicated by [5].

Another technology, in an early stage of development, for capturing CO_2 from coal fired power plants is chemical looping [12-13] having the potentially of very low efficiency penalty and low CO_2 avoidance cost.[14] Details about the progress of this technology are shown by [5]. Membrane–based processes can be used in pre-combustion, oxy-combustion and post –combustion and are suitable for coal fired power plants. The development of this technology is described by Mai Bui 2018.[5] Ionic liquids (ILs) technology has attracted attention due to the energy and cost-efficient separation of CO_2 from post-combustion flue gas.[15] The progress of this technique is represented by [5].

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Finally, we should mention two methods of Carbon Dioxide Removal from the atmosphere. The first one is BECCS where oxidation or gasification of biomass extracts energy known as bioenergy, capturing at the same time the CO_2 from these two processes. However this approach has serious problems such as the need of the most arable land to be used for the food demand and not for biomass (National Academy of Sciences). The increase in electricity cost and decrease in energy security is another serious problem. [16]

The second method is Direct Air Capture and Sequestration (DAC). The capture is taking place directly through the atmosphere via absorption or adsorption separation processes. There is a DAC plant in Hilwil, Switzerland which filters CO_2 from the atmosphere and supply 900 tones of it annually to a nearby greenhouse helping the plants to grow (Grand opening of Climeworks commercial DAC plant, Gasword, 2017). DAC is a promising approach, however it can't replace the conventional CCS systems because the CO_2 concentration in air is 100 to 300 times lower than in the flue gas of gas or coal fired power plants. This results in a high cost of capturing CO_2 from air than from point sources and constrains the use of DAC. [17]

2.2. CO_2 Transportation

In CCS process, after the CO_2 capture and separation, the gas is transported to the storage site via pipeline when it is in dense phase or by trucks, rail and ships when is in the liquid phase. The efficacy of the methods depends on the distance of each point of storage. Ideally CO_2 would be stored where captured. According to (Zero emissions platform) for large distances >1500 Km, transportations via ships is

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preferable because of lower cost. Generally, the vast majority of transportations are expected to be via pipelines because they have a number of advantages [18] such as the continuous transport from the source to the storage site which is essential especially for power plants which operate continuously and also the more economic way of transportation than other ways like ships.[18-19] There are also some difficulties. The amount of CO_2 that is transported should be in dense phase, otherwise the system will have operational problems. For this purpose the appropriate temperature and pressure must be chosen so that the phase will remain the same along the length of the pipeline.[5, 18] Furthermore the impurities in the CO_2 stream are a vital subject and impact on the design and operation of the pipeline system.[18] Generally, it is considered that the cost of transporting CO_2 may be considerably reduced by using multiple diameter trunk lines which lower operating cost and ensure at the same time the right operating pressure throughout the whole pipeline.[5, 18, 20]

On the other hand, the CO_2 transportation via ships can be an effective cost solution for very long distances and for low quantities from small sources. [21] Details about the technology of CO_2 shipping can be found in (Dr Peter Brownshort 2015). [22]

2.3. CO_2 storage

CO_2 storage is the last step in the CCS chain. The CCS process comprises from ocean storage, geological storage and mineral carbonation. [23] Geological storage is considered to be the most viable option and includes depleted oil and gas reservoirs, coal formations, saline formations, basalt formation and Hydrate storage of CO_2 within the subsurface environment. Another option is deep ocean storage, however there is a constrain in this option (ocean acidifiacation and eutrophication) which

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limits this technology and mineral carbonation. Details about all these strategies and their progress can be found in numerous review.[5, 23-28]

2.3.1 Mineral Carbonation

Developing a method for secure sequestration of the CO_2 in geological formation is one of the most serious difficulties that the scientists have to overcome. The mineral carbonation is a method assembles many advantages. There are several features that make it unique among the other CO2 storage procedures. First of all the various minerals that may drive in carbonation reactions are very common all over the world contributing to a large storage capacity, secondly the permanence of storage of CO_2 in a stable solid form results in no CO_2 release from the storage site and finally the heat that released from the reactions theoretically could be used as power resources.[29-31]

As a result, in this study we are focused on this alternative storage option where the CO_2 gas is injected underground under optimized conditions and converted to stable carbonate minerals. During this method CO_2 is reacted chemically with calcium or magnesium oxides to form stable carbonate materials through the below reaction:

$$MO + CO_2 \rightarrow MCO_3 + heat$$

M is the divalent metal. The amount of heat depends on the metal and on the material containing the metal oxide.

The above reaction releases heat and this means that the mineralization thermodynamically is realized at low temperatures, otherwise the calcinations are taken place. The big challenge in this method is to accelerate the carbonation

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exploiting the appropriate amount of heat without causing problems to the environment.[32]

Mineral carbonation can be carried out in two ways. The first one is the *in-situ* method where the CO_2 is injected into a geologic formation for the production of stable carbonates as calcite $CaCO_3$, dolomite $Ca_{0.5}Mg_{0.5}CO_3$, Magnesite $MgCO_3$ and siderite $FeCO_3$. The formed products are thermodynamically stable as a result the sequestration is permanent and safe.[33] This method differs from the conventional geological storage because CO_2 is injected underground in the appropriate conditions so as to accelerate the natural process of mineral carbonization. The second one is the *ex-situ* method where the process takes place above the ground in a processing plant.[23, 34] The mineral carbonation process routes are described in details by Olajire 2013.[32]

The in situ mineralization is preferable because there is no need for additional facilities and mining, the CO_2 is injected directly into porous rocks in the subsurface and reacts directly with the rocks. Moreover there is no need for transportation of the reactants which could be a difficult process. Finally, the amount of the minerals is larger compared to minerals from industrial wastes. [27, 32] However there are also challenges in this way of mineralization as the critical choice of the rocks which should contain the metals and have the appropriate physical and chemical properties to accelerate the carbonation. Another challenge that the scientists have to overcome is to achieve the carbonation acceleration and to achieve the utilization of the heat released from the reactions.[32] Finally, the largest risk in this way of CO_2 storage is the leakage of the carbon[35-37], however this risk may be limited by dissolving the CO_2 into water prior to or when it is injected in the rocks, because this form is denser than

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CO_2 in gas or in supercritical phase.[38-40] Generally, the in situ method may be preferable for high volumes of CO_2.[41]

On the other hand, the main advantages of the ex-situ method are: the availability of minerals at low cost and also their high reactivity when compared to natural minerals.[32]

Minerals for potential CO_2 storage

Oxides and hydroxides of Ca and Mg are proposed as the appropriate materials for the mineral carbonation because they provide alkalinity. Although magnesia (MgO) and lime (CaO) are the most naturally common earth metal oxides, they usually bonded up as silicate such as olivine and serpentine (typically containing 30-60 wt% MgO) .[33] The carbonation of Ca is more effective however the MgO is more common in nature.[33] Basalts and ophiolite rocks are enriched in magnesium, calcium and iron silicates.[42] Among silicate rocks, mafic and ultramafic rocks contain high amounts of Mg, Ca and Fe and low content of sodium and potassium. Some of the main minerals in these rocks are olivines, serpentine, enstatite and wollastonite.[32] According to Coleman 1977[43] and Nicolas 1989[44] olivine, serpentine, peridotite and gabbro are mainly found in ophiolite belts geological zones. In Table 1 is indicated the composition of the most important minerals and their CO_2 sequestration characteristics.[32-33] R_{CO2} is the mass ratio of rock to CO_2 and R_c mass ratio of rock needed for CO_2 fixation to carbon burned. It can be seen that Basalt consists of a relatively small amount of MgO compared to Dunite and Serpentine however it's capacity is higher probably due to CaO and also it is required >1.8 ton of rock per ton of sequestered CO_2:

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Rock	MgO (wt%)	CaO (wt%)	Rc (Kg/Kg)	R_{CO2} (ton rock/ton CO_2)
Dunite (olivine)	49.5	0.3	6.8	1.8
Serpentine	40	0	8.4	2.3
Wollastonite	-	35	13	3.6
Talc	44	0	7.6	2.1
Basalt	6.2	9.4	26	7.1

Table 1. Composition of minerals and their CO_2 sequestration characteristics.[31,45]

There are several studies and projects have been conducted in natural minerals for CO_2 sequestration. First of all the CarbFix Pilot project which takes place in Iceland and used for permanent storage of CO_2 from geothermal power plant in the basaltic rocks and it was estimated that the 95% of the CO_2 injected in the basaltic rocks was mineralized to carbonate minerals in less than 2 years.[45-46] There are several studies that have been conducted for CO_2 storage in basalt rocks[39, 47-54] and they indicated that basalt rocks, under specific conditions, are the most capable and safe sites. Other possible minerals for storage are serpentine and harzburgite as indicated by Dichicco 2015 [29], Zevenhoven 2013[55] Veetil 2014 [56], Krevor 2011 [57], Turvey 2017[58], Klein 2011 [59] and Koukouzas 2009[30]). The improvement of reaction rates of olivine with CO_2 with several methods, could make this mineral appropriate for CO_2 storage.[60-62] Dunite can also be a possible mineral for CO_2 storage.[30, 63] Peridotite rocks can provide the mean for CO_2 storage through mineral carbonization. [63-65] Wollastonite could also be a mineral for carbonation under specific conditions.[66-68]

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Another deposit could be zeolite according to Vatalis 2012 [69] and also the sandstone formations according to Koukouzas 2018. [70] Finally, it was investigated the role of water carbonation of forsterite in $scCO_2$ and found that the reactions could be more extensive under specific conditions.[71]

3. Results and discussion

3.1 CCS technologies in Europe

There are 78 commercial scale projects around the Europe and they are in various stages of development (Table 2) (Carbon Capture and Storage Association).

Project	Location	Status/Started	Fuel	Storage
CarbFix	Near Hvergerdi, Iceland	Pilot/2012	other	Mineral carbonization
Snohvit	Melkoya, near Hammerfest, Norway	Operational/ 2008	Gas	Saline formation
Tiller CO2 Laboratory	Tiller, near Trondheim, Norway	Pilot / 2010	other	No storage
Industrikraft More CCS Project	Einesvagen, near Molde, Romsdal, Norway	Cancelled/ Dormant	Gas	EOR
Technology Centre Mongstad	near Bergen, Norway	Pilot/2012	Gas	No storage
Kollsness CO2 Storage Terminal	Rong, nr Bergen, Norway	In Design	other	Saline formation
Sargas Husnes	Husnes, Hardangerfjord, Norway	Cancelled/ Dormant	Coal	Unknown
Karsto	naer Haugesund, Rogaland, Norway	Cancelled/ Dormant	Gas	Saline formation
Klemetsrud	Klemetsrud, near Oslo, Norway	In Planning	other	Saline formation
Yara Porsgrunn	Heroya Industrial	Cancelled/	Gas	Saline formation

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Demonstration Project	Park, Porsgrunn, Norway	Dormant		
Norcem CCS Demonstration Project	Brevik, Norway	In Design	Unknown	Saline formation
Frevar capture plant	Fredrikstad, Norway	Speculative	Other	Saline formation
Stepwise Pilot Plant	Lulea, Sweden	Pilot/2017	Other	No storage
Karlshamn Field Pilot	Karlshamn, Sweden	Finished	Oil	No storage
Nordjyllandsvaerket	Nordjylland, Denmark	Cancelled/ Dormant	Coal	Saline Formation
Esbjerg Pilot Plant	Esbjerg, Denmark	finished	Coal	No storage
Meri Pori CCS Project	near Pori, Finland	Cancelled/ Dormant	Coal	Possibly EOR
Sleipner	Offshore Norwegian North Sea, Norway	Operational/ 1996	Gas	Saline Formation
Whitegate and Aghada CCS Project	Whitegate, Co. Cork, Republic of Ireland	Speculative	Gas	Depleted oil and Gas
Acorn Project	St Fergus, UK	In Planning	Gas	Unknown
Peterhead	Peterhead, Scotland, UK	Cancelled/ Dormant	Gas	Depleted Oil and Gas
Scottish Carbon Capture & Storage	Edinburgh, Scotland, UK	Pilot/	other	No storage
Caledonia Clean Energy Project	Grangemouth, Scotland, UK	In Planning	Gas	Unknown
Longannet	Fife, Scotland, UK	Cancelled/ Dormant	Coal	Depleted Oil and Gas
Oxycoal2	Renfrew, Scotland, UK	Pilot/2009	Coal	No storage
Hunterston	near Largs, North Ayrshire, UK	Cancelled/ Dormant	Coal	Depleted Oil and Gas
Alcan Lynemouth	Lynemouth, Northumberland, UK	Cancelled/ Dormant	Coal	Unknown
Blyth Power Station	Cambois, Blyth, UK	Cancelled/ Dormant	Coal	Unknown

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Teesside Collective	Teesside, UK	In Planning	unknown	Saline Formation
Lotte Chemicals CCUS Project	Wilton Site, Teesside, UK	In Design	Gas	Industrial Use
Teesside Low Carbon Project	Eston, Teeside, UK	Cancelled/ Dormant	Coal	Depleted Oil and Gas
Liverpool-Manchester Hydrogen Cluster	Ince Marshes, Merseyside, UK	Speculative	Gas	Depleted Oil and Gas
Pilot-scale Advanced Capture Technology	Beighton, near Sheffied, UK	Pilot	Other	No storage
Ferrybridge	West Yorkshire, UK	Finished	Coal	No storage
Millenium Generation Project	Stainforth, South Yorkshire, UK	Pilot	Gas	No storage
Killingholme	Immingham, North Lincolnshire, UK	In Planning	Coal	Saline formation
Aberthaw Pilot Plant	Aberthaw, near Barry, UK	Finished	Coal	No storage
Imperial College Carbon Capture Pilot Plant	South Kensigton Campus, London, UK	Pilot	Other	No storage
Tilbury Power Station	East Tilbury, UK	Cancelled/ Dormant	Coal	Unknown
Kingsnorth	Kent, UK	Cancelled/ Dormant	Coal	Depleted Oil and Gas
InfraStrata	Portland (exact loaction unknown), UK	Cancelled/ Dormant	Unknown	Unknown
Offshore Netherlands North Sea, Netherlands	GDF Suez	Operational/ 2004	Gas	Depleted Oil and Gas
Eemshaven	Groningen, Netherlands	Cancelled/ Dormant	Coal	Depleted Oil and Gas

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Buggenum Pilot Plant	Buggenum, near Roermond, Netherlands	Finished	Coal	No storage
Air Products Rotterdam	Botlek, Rotterdam, Netherlands	Cancelled/ Dormant	Oil	No storage
Pegasus Rotterdam	Port of Rotterdam, Netherlands	Cancelled/ Dormant	Gas	Depleted Oil and Gas
Barendrecht Project	Port of Rotterdam, Netherlands	Cancelled/ Dormant	Oil	Depleted Oil and Gas
Rotterdam Backbone Project	Rotterdam, Netherlands	In Planning	Other	Depleted Oil and Gas
Rotterdam Climate Initiative	Rotterdam, Netherlands	Cancelled /Dormant	Other	Depleted Oil and Gas
CO2 Smart Grid	Rotterdam, Netherlands	Speculative	Other	unknown
C.GEN Rotterdam	Europort, Rotterdam, Netherlands	Cancelled /Dormant	Coal	Unknown
ROAD	Maasvlakte, Rotterdam, Netherlands	Cancelled /Dormant	Coal	Depleted Oil and Gas
Antwerp CCS Feasibility Study	Port of Antwerp, Belgium	Speculative	Unknown	unknown
Leilac Pilot Plant	Lixhe, near Vise, Belguim	Pilot	Coal	No storage
Wilhelmshaven Pilot Plant	Wilhelmshaven, Germany	Pilot	Coal	No storage
Heyden Pilot Plant	near Minden, North Rhine-Westphalia, Germany	Pilot	Coal	No storage
Ketzin Pilot Injection Site	Ketzin, near Berlin, Germany	finished	Unknown	Saline formation
Herne Pilot Plant	Herne, North Rhine-Westphalia, Germany	Pilot	Coal	No storage
Hurth IGCC	Hurth, near Koln, Germany	Cancelled /Dormant	Coal	Unknown
Niederaussem, near Koln,	Niederaussem, near Koln, Germany	Pilot	Coal	No storage

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Germany				
Janschwalde	Brandenburg, Germany	Cancelled /Dormant	Coal	Saline formation
Staudinger Pilot Plant	Grosskrotzenburg, near Hannau, Germany	Pilot	Coal	No storage
EnBW Pilot Plant	Heilbronn, Germany	Pilot/2011	Coal	No storage
ArcelorMittal Florange	Florange, Moselle, France	In Planning	Coal	Saline Formation
C2A2 Field Pilot	Le Havre, Normandy, France	Pilot	Coal	No storage
Lacq CS Pilot	Lacq, Pyrenees-Atlantiques, France	Pilot	Gas	Depleted Oil and Gas
Compostilla Phase I	Cubillos del Sil, Ponferrada, Spain	Pilot	Coal	No storage
Puertollano	Puertollano, Ciudad Real, Spain	Finished	Coal	No storage
Belchatow	Lodz, Poland	Cancelled/ Dormant	Coal	Saline Formation
Kedzierzyn	Silesia, Poland	Cancelled/ Dormant	Coal	Saline Formation
CO2SEPPL	Durnrohr, near Tulln, Austria	Pilot/2010	Coal	No storage
Retznei Oxyfuel Demonstration	Retznei, near Graz, Austria	In Planning	Other	No storage
Porto Tolle	Porto Tolle, Veneto, Itlay	Cancelled/ Dormant	Coal	Saline Formation
Colleferro Oxyfuel Demonstration	Colleferro, near Rome, Italy	In Planning	Other	No storage
Brindisi, Puglia, Italy	Brindisi, Puglia, Italy	Pilot/2011	Coal	unknown
Delimara	Delimara, Marsaxlokk, Malta	In Design	Coal	Depleted Oil and Gas
Getica CCS Demonstration Project	Turceni, near Targu Jui, Gorj county, Romania	Cancelled/ Dormant	Coal	Saline formation
Maritsa	Stara Zagora Province, Bulgaria	Cancelled/ Dormant	Coal	Saline Formation

Table 2. The commercial scale projects of CCS technologies around the Europe

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36 (46%) of these projects are cancelled/dormant or finished, only 3 operate, 21 are in Pilot phase and 18 are in planning/speculative or in design (Table 2). The UK hosts most of these plants (22) followed by Norway (12), Netherlands (10) and Germany (9). The highest amount of these plants (35%) do not use storage site for the CO_2 but they follow the process of utilization, 23% store the CO_2 in saline formations and 15% in depleted oil and gas formations. The two of the three plants in operation (Snohvit in Norway and Sleipner in Norway) use as storage site saline formations and the Offshore Netherlands in Netherlands uses depleted oil and gas formations. It is of interest that all the pilot plants utilize the captured CO_2, except for the Lacq CS Pilot in France which stores it in depleted oil and gas formation and the Carbfix in Iceland which uses mineral carbonization technique.

3.1.1. The case of Carbfix (Iceland)

As we mentioned in previous part the mineral carbonization is a new, environmental safe and low cost technique. The Carbfix is a project in Iceland that is injecting solutions of mixed CO_2 and H_2S into basaltic rocks (basaltic lava flows and hyaloclastite) at 1000m. The field site is situated in SW Iceland close to a geothermal power plant which produces up to 30000 tones of CO_2 per year and it is estimated to be increased. The source of CO_2 is the geothermal gas which is a by product of the geothermal steam production. [51] The project started in 2007 and it's operation is since 2012. It has been estimated that in 2017 it was injected about 10000t CO_2. The percentage of CO_2 that is mineralized as carbonate in the basalt rocks is found to be almost complete (95%) in 2 years (Carbon Capture and Storage Association). The existence of large available area of basaltic rocks associated with the rapid carbonation reactions may result in a safe and permanent solution.

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3.2. CO_2 storage in Greece

The biggest source of CO_2 in Greece is the lignite fired power plants in western Macedonia. Greece ranks second in European Union and sixth worldwide in the terms of lignite production. Today, the 8 PPC lignite power plants represent 42% of the country's total installed capacity and generate nearly 56% of the country's electrical energy according to the website of the Public Power Corporation S.A. Hellas. The use of this important energy source is facing a challenge due to the vast amounts of CO2 emitted in the atmosphere during the lignite combustion. The CO_2 emissions from fuel combustions in Greece, was found 64.6 Mt [2] including a high amount from lignite fired power plants. The reduction of CO_2 emissions in the atmosphere is one of the highest challenges that the scientists have to face. The Paris agreement goal is keeping the global temperature rise below 2 ºC compared to pre industrial levels and also limit the temperature increase to no more than 1.5 ºC aiming to reduce the risks and impact in the climate change.[72] The CCS technologies in Europe as mentioned above are far away from the Greek Power Plants and the CO_2 transportation is a very difficult process. As a result, an appropriate CO_2 storage site in Greece would be the perfect solution.

There are several studies conducted about the CO_2 storage through the application of CCS technique in Greece. According to Tasianas 2016[73] one potential storage site in oil and gas fields lies in Prinos, Kavala in NE Greece. Furthermore, it was estimated through a model the potential storage capacity in the Pentalofos (Tsarnos and Kalloni members) and Eptahori reservoirs in NW Greece and found to be 728 Gt CO_2 for both storage sites. [73] In Prinos (Thassos- Kavala path) hydrocarbon field offshore in Northern Greece a monitoring system which simulated a potential CO_2 leakage from Prinos field was investigated and found that CO_2 reaches the

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seabed in approximately 13.7 years after the injection and it reaches it's peak after 32.9 years. The model results show that CO_2 will flow towards the Natura protected areas only in 5 days after the leakage and between this period the authorities should take the appropriate measures in order to avoid environmental problems. As a result a possible leakage would affect the environment.[74] However, according to Koukouzas 2016[74] the consequences of a CO_2 are considered limited and the ecosystem is capable of recovering. Finally it was calculated the amount for operating this system 0.38$/ton of CO_2 and 0.45$ /ton of CO_2 is found to be cost for EOR.

Vatalis 2012[69] proposed the CO_2 storage in the known deposite of Zeolite in Evros (Northern Greece). Koukouzas 2018[70] concluded that Pentalofos and Tsotyli sandstone formations could be a potential CO_2 storage site under specific conditions. However, this approach need more investigation.

Another promise technique for CO_2 storage without such an environmental risk, as we mentioned in previous part of this study, is the mineral carbonation. A study was conducted for the storage of captured CO_2 in magnesium silicates. For the experiment samples from ultramafic rocks from mountain Vourinos in Western Macedonia, Greece, were used. It was used the aqueous technique. The results indicated limited carbonation. However, this situation will probably change in different experimental conditions. For example, longer reaction time, the particle size and the discharge of impurities which poison the reaction, would probably improve the carbonation.[30]

Generally, mineral carbonation is a new CCS process which promises the permanent storage of CO_2. The most important aspect is that under specific conditions ensures that carbonates formed are environmentally benign and geologically stable.

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Taken into account the geological forms that are appropriate for CO_2 storage through mineral carbonation we concluded that Greece could be a potential site for CO_2 storage because throughout the continental part all of these geological forms can be found.

The most capable sites for CO_2 injections are the basaltic rocks. Several sites with basalts in Greece could be potential CO_2 storage sites for mineralization. Ultramafic lavas associated with high basaltic dykes are found in Othris Mountains in Central Greece.[75-79] In Othris ophiolite complex (Figure 1) was found olivine phyric lavas from the Agrillia area (about six Km NW from Lamia) and high MgO basaltic dykes from Pournari area (about 31 KM NW from Lamia). The major (in wt%) elements determined for ultramafic lavas from Agrillia area show the highest values for SiO_2, MgO, CaO and FeO in all sample cases and for high–Mg basalts from Pournari show the highest values for SiO_2 , FeO, MgO and CaO in all sample cases. [75] Furthermore, the lower unit of the Pindos ophiolitic belt is composed mainly by basaltic rocks.[80] Gabbroic and basaltic rocks are also found in Serbo-Macedonian (Volvi and Therma bodies) and western Rodopi (Rila mountains) massifs of Bulgaria and Greece.[81-82] Finally, basalts can be found in ophiolitic rocks of the Attic-Cycladic crystalline belt. According to Stouraiti 2017 [83] in Paros, western Samos (Kallithea), Naxos, central Samos, Skyros, Tinos and S. Evia are found basalts exhibiting high MgO concentrations. Moreover on Acrotiri Peninsula, Santorini, Greece are found basalts [84] and also in Kos-Nisyros.[85] However, the major factor that eliminates the potential CO_2 storage in the last areas is that they are islands with limited storage areas and the transportation of CO_2 in this case is a very difficult and high cost process.

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The ophiolites in Greece are widespread mostly exposed in central and northern Greece. Large ultramafic bodies are found in East Othris ophiolite belt (Figure 1). It was indicated[86-87] that in Vrinera ophiolitic unit the ultramafic rocks consist of serpentinized harzburgites and are below gabbros and diorites. The ophilithic units of Eretria, Aerino, Velestino consist mainly of serpentites the same case in the southern part of Aerino. Finally, serpentinites can be found in ophiolitic mélange of Ag. Giorgios but it is rather small (2Km^2). The ophiolite units of two Greek islands Evia and Lesvos comprise from amphibolites and below them underlie ultramafic masses which consist of serpentinized harzburgites , patches of dunites and serpentinized depleted iherzolites and harzburgites, respectively.[88] A study that has been conducted in the East part of Thessaly, Central Greece shows that the metaophiolites of this region consist mainly of serpentinites and metabasites.[89] The Pindos ophiolite complex in NW Greece is mainly comprised of large harzburgite-dunite masses > 1000 Km^2 in the mantle peridotites.[90-92] Among the Western Hellenic Ophiolites is Vourinos ophiolite complex in Western Macedonia, NW Greece, represents a mid-Jourassic complete lithospheric slab about 12 Km thick and 400 Km^2 and consist of depleted harzburgite mantle which hosts bodies of dunite ranging in size from several m to Km scale length.[30, 91, 93-95] There are several studies conducted in Vourinos and show that dunite surrounded by serpentinized harzburgites with some lenses of serpentinized dunite.[92] Furthermore, the Koziakas mountain ophiolite in western Thessaly, also belongs to West Greek ophiolite belt and comprises mantle peridoites with harzburgites and secondary plagioscale bearing Iherzolites.[92, 96]

As we can see there are several sites in Greece that could be potential CO_2 storage sites since their underground hosts rocks rich in olivine, serpentine, harzburgites, dunites, peridotites and basaltic glass which include high amounts of

Mg, Ca and Fe oxides and hydroxides. As we mentioned the islands could not be a part of these sites because the CO_2 transportation cost will be high. Greece has several industries that produce high amounts of CO_2 (the total CO_2 emissions from Greece in 2016 were 67870 kt according to world data atlas) and the mineral carbonation technology would be a sustainable solution in this problem taking into account that there are the appropriate geological forms capable for permanent and safe storage. We have already indicated that potential sites for CO_2 storage exist in continental part of our country as Orthis mountain in central Greece, Western Rodopi in Northern Greece, Pindos in NW Greece, Vourinos in Western Macedonia and also Koziakas in western Thessaly.

The Carbon Capture and Storage (CCS) is one of the most promising approaches to reduce the CO_2 emissions. The CO_2 storage being the third step of the CCS process chain is playing a very important role in this technology. The most suitable CO_2 storage site is established in basins where exists rocks containing the appropriate porosity and are near to power stations or industries in order to avoid transportation cost. The power stations in our country are placed mainly in Ptolemais-Amynteo lignite centre (Western Macedonia, Northern Greece). After a bibliographic research that has been conducted in the near region, it was indicated that the mountain Pindos ophiolite and mainly the Vourinos ophiolite complex (which extend SW of Kozani covering an area of 450 Km^2) is very close to the power station and they comprise of harzburgite-dunite masses in the mantle peridotites and dunite surrounded by serpentinized harzburgites with some lenses of serpentinized dunite, respectively. These natural minerals are rich in oxides and hydroxides of Ca, Mg and Fe representing the perfect materials for mineral carbonization. Mineral carbonization is a CO_2 storage permanent and environmental safe technology which do not incur

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longterm liability (avoiding the challenge of degrading the environment) or monitoring obligations. Taking in to account that these two areas is very close to the power stations limiting the CO_2 transportation cost this method could be a potential technique for reducing the CO_2 emissions fulfilling the goals of the Paris Agreement. However, it was fount also other potential capable for mineral carbonation place in continental Greece (e.g Orthis ophiolite belt) but it should be conducted an economical research in order to estimate the CO_2 transport cost and compared to the profit of the operation of such technology.

4. Conclusions

There are various potential methods for removing CO_2 from the atmosphere such as increase in energy efficiency, fuels with lower CO_2 emissions, energy sources with no CO_2 emissions and CO_2 capture and sequestration. In this study we refer to a number of bibliographical references about the three main strategies of CCS technology. The CO_2 capture technologies, the CO_2 transportation into the storage sites and finally the CO_2 storage processes. The method for secure sequestration of the CO_2 in geological formation is a very critical step and faces many difficulties. One potential environmental safe and low cost method with permanent results compared to the other technologies could be the mineral carbonation. During this method CO_2 is reacted chemically with calcium or magnesium oxides or hydroxides to form stable carbonate materials.

According to Carbon Capture and Storage Association there are 78 commercial scale CCS projects around the Europe and they are in various stages of development. Only 3 of them operate and 21 are in Pilot phase. However the only project uses

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mineral carbonation technique and has injected about 10000t CO_2 in basaltic rocks during 2017 is Carbfix in Iceland. It has been estimated that the percentage of CO_2 that is mineralized as carbonate in the basalt rocks in Carbic project is found to be almost complete (95%) in 2 years. As a result, this technique is very promising.

There are a few studies about the establishment of CCS technologies in Greece. Greece emits CO_2 in the atmosphere with biggest source the lignite fired power plants in western Macedonia. As a result we tried to give some ideas in using CCS technology in Greece in order to reduce the CO_2 emissions from power plants. After the research about the use of numerous geological forms as storage sites for mineral carbonation, we tried to find bibliographically such geological forms. We found that there are several sites with basalts in Greece that could be potential CO_2 storage sites for mineralization such as in Othris Mountains in Central Greece, in the lower unit of the Pindos ophiolitic belt in Serbo-Macedonian (Volvi and Therma bodies) and in western Rodopi (Rila mountains) massifs of Bulgaria and Greece, finally basalts are found in Greek islands but this could be an infeasible solution because of the difficulties and high cost in CO_2 transportation. Moreover there are large ultramafic bodies around the continental Greece which could be possible storage sites. In Vrinera, East Othris ophiolitic unit the ultramafic rocks consist of serpentinized harzburgites and are below gabbros and diorites. The ophilithic units of Eretria, Aerino, Velestino consist mainly of serpentites the same case in the southern part of Aerino. Furthermore, the Pindos ophiolite complex in NW Greece is mainly comprised of large harzburgite-dunite masses > 1000 Km^2 in the mantle peridotites. As we mentioned in previous part of this paper based on bibliography all these geological forms under specific conditions could be the appropriate areas for mineral carbonation.

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However, taking into account the large cost of CO_2 transportation and also that the CCS technology sites around Europe are away from Greece, we tried to find capable geological forms near the lignite fired power plant area in Western Macedonia, Greece. After the bibliographical research we concluded that the mountain Pindos ophiolite complex and mainly the Vourinos ophiolite complex which are very close to the power station consist of harzburgite-dunite masses in the mantle peridotites and dunite surrounded by serpentinized harzburgites with some lenses of serpentinized dunite, respectively. These natural minerals are rich in oxides and hydroxides of Ca, Mg and Fe and could be the perfect materials for mineral carbonation.

In any case, this is a potential proposal which is based only in bibliographic research about the geology of the surrounding area and is trying to give some ideas to scientific community. A detailed research about the geology, the chemical and hydrodynamic characteristic of the underground should be conducted compared to financial study in order to ensure that the proposed solution is economically and technologically viable. This could be a future research.

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