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EXERGETIC AND ENVIRONMENTAL ASSESSMENT OF MINERAL SEQUESTRATION PROCESSES IN BRAZIL

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Abstract. *International agreements on global warming identify an urgent need for a technology that captures and sequesters carbon dioxide (CO_2) in large scale, in order to reduce anthropogenic CO_2 emissions into the atmosphere. Among all CCS technologies (Carbon Capture and Storage), mineral sequestration is the most promising. This technology converts CO_2 into an environmentally stable solid carbonate. Thus, many researches are being elaborated using Life Cycle Assessment (LCA) in order to assess the energy requirement and the CO_2 emissions derived from all stages involved in the mineral sequestration process, starting from the mining of raw materials until the final disposition of the products. In this work, the quantity and quality of raw materials available in Brazil to perform a mineral sequestration process were determined. Two scenarios were analyzed, assessing two types of mineral sequestration process: by strong acids and ammonium salts, respectively, oriented to capture the CO_2 emissions from power plants fueled with coal. The energy requirement was based on exergetic analyses while the environmental analyses were based on the Life Cycle Assessment (LCA). The results show that the best mineral raw material is the serpentine (1.9 $t_{serpentine}$ and 2.3 $t_{serpentine}$ were used for strong acid and ammonium salts, respectively). The mineral sequestration stage is the most critical one because it takes larger quantities of energy, principally for the regeneration of chemical inputs (i.e. 3.4 MW and 1.2 MW for strong acid and ammonium salts, respectively). However, the mineral sequestration by ammonium salts is the most promising, thus it can produce around 3.14 t and 0.91 t of carbonates and SiO_2 per each tCO_2 fixed, respectively. Moreover, the quantity of CO_2 avoided by ammonium salts process was 0.54 t of CO_2.*

*Keywords****: carbon capture, carbon dioxide, mineral carbonation, exergetic assessment.***

1. 1. INTRODUCTION

Large amounts of carbon dioxide (CO_2) are emitted into the atmosphere as a result of combustion process of fossil fuels (coal, oil and natural gas) for power generation. If the current trend of worldwide energy consumption keeps increasing in upcoming years, it will cause an increase in atmospheric CO_2 concentration at an approximate rate of 2 ppm per year, causing instability in the carbon cycle and intensifying the effects of global warming (Park A. A. and Fan L., 2004; IPCC, 2005; Arce et. al, 2014).

Considering the importance of fossil fuels for the global economy, international agreements about global warming identify the urgent need to capture and sequester the *anthropogenic* CO_2 emissions on a large scale. Thereby, technologies for carbon sequestration (Carbon Capture and Storage - CCS) have been proposed for this purpose such as: geological, oceanic sequestration, biological fixation and mineral sequestration (Park A. A. and Fan L., 2004)

As pointed out by Machado et al (2013), Brazil acknowledges the importance of implementing CCS technologies as alternatives to reduce the anthropogenic CO_2 emissions as part of the national strategy for climate change.

In this work, it is developed an energy requirement and environmental analysis based on exergy concepts and Life Cycle Assesment (LCA) respectively, for two mineral sequestration processes being by strong acid (SA) and aqueous ammonium salts (AAS), considering the emission of CO_2 from coal power plants, in order to develop new projects and get benefits in the voluntary carbon market and/or to produce value – added products, such as: hydromagnesite and nesquehonite. These products can be used as build materials, soil recovery, as well as, silicon in the electronic industry.

2. METHODOLOGY

2.1 LCA Process and boundary control

Mineral sequestration processes need two main raw materials: mineral silicates and CO_2 (i.e. from coal power plant). In this study, it was considered that mineral silicate source are located in the state of "*Goiás*" and the CO_2

emission source is from the state of "*Rio Grande do Sul*" (Table 1 and Figure 1). Both raw materials sources are located in opposed sites, as presented in Figure 1. Thus, it is assumed that the mineral sequestration unit will be sited in state of São Paulo, due to its location, which is in a strategic and intermediate distance between mineral silicate and CO_2 sources.

Table 1 - Elemental analysis of mineral silicate[Ferreira O.B. and Linares W.B, 2009].

Mineral	Content (%)										
	MgO	CaO	MnO	FeO_{tot}	Cr_2O_3	Na_2O	K_2O	P_2O_5	SiO_2	TiO_2	Al_2O_3
Serpentine (Goiás)	41.1	0.11	----	8.5	8.4	0.004	0.05	0.05	40.1	0.05	0.99

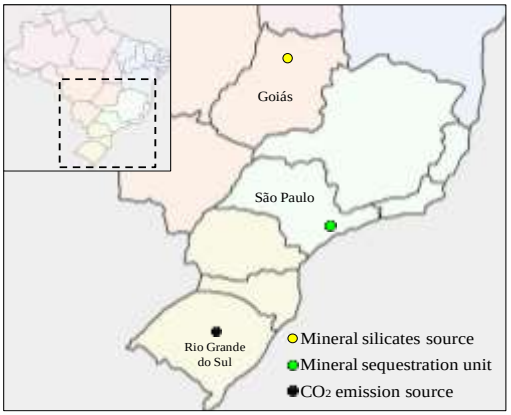

Figure 1 - Raw materials sources for mineral sequestration process: CO_2 emission (Power plant in the state of Rio Grande do Sul) and mineral silicates (state of Goiás).

The first step of the LCA is to identify and quantify the energy inputs and outputs, CO_2 emissions, CO_2 avoided, chemical inputs and products. The study was structured aiming to answer the following questions:

- How much raw material and chemical inputs are needed for the removal of CO_2 emission from the power plant under assessment?
- How much energy is required to fix 1 ton of anthropogenic CO_2 emission as carbonate?
- How many tons of CO_2 could be avoided?

Figure 2 represents the LCA of a mineral sequestration process obtained by following the norms established by ISO 14040 (ISO, 2006). LCA identifies four stages being: mining (stage 1), transport (stage 2), power plant (stage 3), and mineral sequestration with chemicals recovery (stage 4),

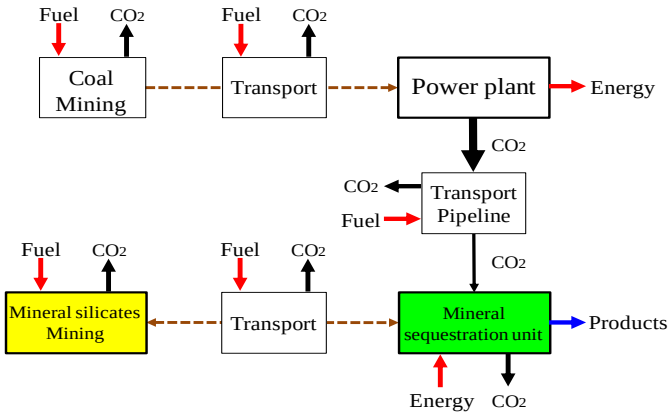

Figure 2 - LCA system for CO_2 mineral sequestration process.

The energy requirement, CO_2 emissions and products derived from each stage are represented by red, black and blue arrows, respectively.

In the stage 1 is considered the mining process of the coal and mineral silicate. Both raw materials are going to be carried out in existing mines in Brazil. The red arrow represent the energy requirement in mining stage, this energy can be provided by coal power plants or hydroelectric plants, according to the energy matrix of those cities (i.e. energy from

coal power plant for the state of Rio Grande do Sul and energy from a hydroelectric plant in the State of Goías). The black arrow represents CO_2 emission derived from use energy requirement.

Both raw materials, coal and mineral silicate, are going to be transported to the power plant and mineral sequestration unit, respectively. This transport plus the CO_2 transport are called stage 2. The transport of raw materials is realized by railway trains and the CO_2 transport is realized by pipeline from power plant until the mineral sequestration unit. In order to calculate CO_2 emissions (black rows) is assessment the quantity of fuel used (red rows), specifically in the mineral transport.

The power plant represents the stage 3, and the mineral sequestration unit represents stage 4.Once all raw materials arrive to mineral sequestration unit, the quantity of CO_2 computed will be in function of energy requirement of this process. In this work, the LCA system regards two scenarios: The first scenario considers a mineral sequestration unit by strong acid and the second considers a mineral sequestration unit by ammonium salts.

2.2 Mineral sequestration processes

The selection of the mineral sequestration process is defined by the most efficient reaction rates in laboratory research so far. The selected processes were by strong acids (Teir S. et al., 2007a; Teir et al., 2007b) and ammonium salts (Wang X. and Maroto-Valer M.M., 2011a). Both processes can be performed in atmospheric pressure, moderate temperatures, and the used chemical inputs can be recycled and reused. The aforementioned reaction involved in each mineral sequestration processes are listed in Table (2).

2.2.1 Mineral sequestration process by strong acids

Figure 3 shows the Block Flow Diagram (BDF) of the mineral sequestration process by strong acids, which uses specifically hydrochloric acid (HCl) (Teir S. et al., 2007a; Teir et al., 2007b, Teir S., 2008). For this process, four operations were considered, being Mg leaching, CO_2 mineralization, NaOH regeneration (Electrolysis) and HCl production (Fuel cells). Following the recommendations of Teir et al. (2009) and based on the works of House et al. (2007), the possibility to use energy that can be produced by a fuel cell was also assessed. Minor energy loss due to pumps, elbows, and valves were neglected.

Initially in a reactor, the mineral silicate reacts with an HCl solution 4M with 40% excess, this process corresponds to the leaching of magnesium (Mg) in the form of $MgCl_2$, according to equation (R1). After the reaction, a resulting solution with 4.7 %wt of solids is filtered, then the solids are separated obtaining 98.2% of SiO_2 and 1.8% of unreacted serpentine.

The remaining solution has a concentration of $MgCl_2$, which is necessary to mineralize CO_2 in a bubbling reactor. Moreover, a NaOH solution (50 %wt) is used in order to maintain an alkaline medium (pH 8-9) enabling carbonate precipitation in the form of hydromagnesite ($Mg_5(OH)_2(CO_3)_4.4H_2O$) as shown in the equation (R2).

After the carbonate precipitation obtained is separated by filtration, and an aqueous NaCl solution remains, which can be recycled and sent to the electrolysis process to produce H_2, Cl_2 and NaOH solution, according to equation (R3). The produced NaOH solution (22%wt NaOH) is reused in the mineralization operation (R2), and the H_2 and Cl_2 are sent to fuel cells to produce pure HCl gas, according to equation (R4). This acid can be bubbled through water to obtain an aqueous HCl solution, which is be reused in leaching to produce $MgCl_2$.

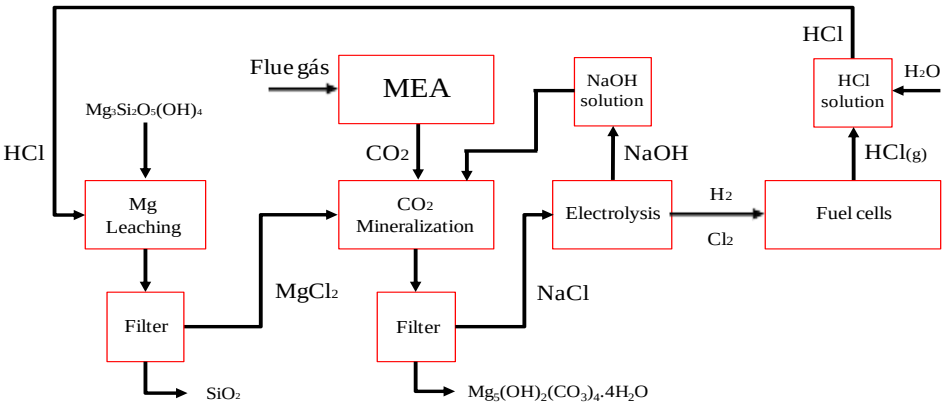

Figure 3 - BFD Mineral sequestration process by strong acids.

2.2.2 Mineral sequestration process by ammonium salts

Figure 4 shows the Block Flow Diagram (BDF) of the mineral sequestration process by ammonium salts specifically a NH_4HSO_4 solution (Wang X. and Maroto-Valer, 2011a; Wang and Maroto-Valer, 2011b). For this

process, six operations were considered being, Mg leaching, pH swing, CO_2 capture, CO_2 mineralization, reverse osmoses and thermal decomposition.

The first operation is the capture of CO_2 from flue gas with a NH_4OH solution which reacts directly with the CO_2 forming an aqueous solution of ammonium bicarbonate (NH_4HCO_3), according to equation (R5) (Resnik K.P. et al., 2004). The leaching of magnesium (Mg) from serpentine uses a NH_4HSO_4 solution 1.4 M, resulting in an aqueous MgSO4 solution (equation R6) with a 2.82 %wt of solids (87.71 %wt of SiO2 and 12.28 %wt of serpentine unreacted).

This resulting solution is filtrated and the remaining solution is treated with an aqueous NH_4OH solution (50 %wt) to convert NH_4HSO_4 excess into $(NH_4)_2SO_4$ (equation R7) in order to cause an alkaline medium (about pH 8 – 9) appropriate to carry out the carbonation reaction. The resulting solution is a mixture of $MgSO_4$, $(NH_4)_2SO_4$ and H_2O (molar ratio 0.0132: 0.0133: 0.9735).

This solution is fed into the CO_2 mineralization reactor and mixed with a NH_4HCO_3 solution from the capture CO_2. A characteristic of this reaction is that the NH_4HCO_3 solution can reduce the pH level. Thereby, to maintain an alkaline medium, an aqueous NH_4OH solution (50% wt.) is added, causing the carbonate precipitation (Nesquehonite $MgCO_3.3H_2O$), according to equation (R8). The formed carbonate is separated by filtration from the aqueous solution of 14.84 %wt. of $(NH_4)_2SO_4$. This solution is concentrated though a reverse osmoses process, after that, the concentrated solution of $(NH_4)_2SO_4$ is submitted to thermal decomposition by heating up to approximately 280°C, releasing NH_3 gas and the remaining solution is NH_4HSO_4, according to equation (R9). The water obtained in the concentration operation is sent to a clarifier and is reused again in the process.

The NH_4HSO_4 is reused in the leaching operation (equation R6) and the NH_3 gas is bubbled in water to obtain an aqueous NH_4OH solution, which is reused in the operations of CO_2 capture (equation R5), pH swing (equation R7) and CO_2 mineralization (equation R8).

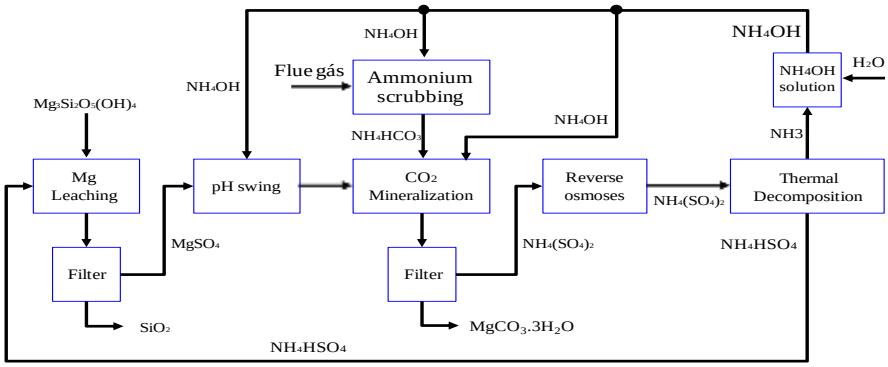

Figure 4 - BFD Mineral sequestration process by ammonium salts.

2.3 Thermodynamic analysis

The energy requirement is based on exergetic analysis (Bjorklof T. and Zevenhoven R., 2012) and the environmental analysis on LCA (Khoo H. H. et al., 2011). All calculations take into account the inputs and outputs of mass and energy streams in each stage, according to the LCA system shown in Figure 2. To determine the energy requirement, it was first made a preliminary thermodynamic analysis of all chemical reactions (Table 2), in order to obtain the operating temperatures and compare them with those found in literature. Thereby, the free enthalpy or the Gibbs energy was used, which is a thermodynamic function that predicts the spontaneity of a chemical reaction. The variation of free enthalpy ($\Delta G°$) depends exclusively on the initial and final state of the system. When the system is in constant pressure and the involved substances are in standard conditions.

Table 2 – Thermochemical conditions of the mineral sequestration process.

N°	Chemical Reaction	T	ΔH	ΔG
		(°C)	(kJ/mol)	
R1	$Mg_3Si_2O_5(OH)_{4(s)} + 6\ HCl_{(aq)} \rightarrow 3\ MgCl_{2(aq)} + 2\ SiO_{2(s)} + 5\ H_2O_{(l)}$	80	-70.70	<0
R2	$5MgCl_{2(aq)} + 10\ NaOH_{(aq)} + 4\ CO_{2(g)} \rightarrow Mg_3(OH)_2(CO_3)_4.4H_2O + 10\ NaCl_{(aq)}$	40	-234.93	<0
R3	$2\ NaCl_{(aq)} + 2\ H_2O_{(l)} \rightarrow 2\ NaOH_{(aq)} + Cl_{2(g)} + H_{2(g)}$	80	543.15	>0
R4	$Cl_{2(g)} + H_{2(g)} \rightarrow 2\ HCl_{(g)}$	200	-184.87	<0
R5	$CO_{2(g)} + NH_4OH_{(aq)} \leftrightarrow NH_4HCO_{3(aq)}$	25	-68.21	<0
R6	$Mg_3Si_2(OH)_{4(s)} + 6\ NH_4HSO_{4(aq)} \leftrightarrow 3\ MgSO_{4(aq)} + 2SiO_{2(s)} + 3\ (NH_4)_2SO_{4(aq)} + 5\ H_2O$	90	-451.28	<0
R7	$NH_4HSO_{4(aq)} + NH_4OH_{(aq)} \leftrightarrow (NH_4)_2SO_{4(aq)} + H_2O_{(l)}$	25	-77.33	<0
R8	$MgSO_{4(aq)} + NH_4HCO_{3(aq)} + NH_4OH_{(aq)} + 2\ H_2O_{(l)} \leftrightarrow MgCO_3.3H_2O_{(s)} + (NH_4)_2SO_{4(aq)}$	80	-47.96	<0
R9	$(NH_4)_2SO_{4(aq)} \leftrightarrow NH_4HSO_{4(aq)} + NH_{3(g)}$	280	165.95	<0

3 RESULTS AND DISCUSSIONS

3.1 Energy requirement of the mineral sequestration unit

Table 2 shows the energy requirement for both mineral sequestration processes. In the mineral sequestration process by strong acid, the exergy requirement is mainly due to the electrolytic process in the NaOH regeneration, reaching a value of 3381 kW/tCO_2. However, HCl production can release 993 kW/tCO_2, whether carried out in fuel cells, which can be used in the electrolysis process, thereby, the final energy requirement decreases to 2388 kW/tCO_2. The leaching of Mg and mineralization operations are exothermic, however, this quantity of energy has no practical application due to its low thermal quality (temperatures of around 80°C and 40°C, respectively).

For the mineral sequestration process by ammonium salts, the energy requirement was approximately 1196 kW/tCO_2 in the regeneration operation. Despite that the leaching of magnesium is an exothermic reaction, this release heat corresponds to a temperature of 90°C and cannot be used in the regeneration stage because it requires temperatures up to 280°C. Similarly, the exergy released in mineralization operation cannot be used due to having a low thermal quality.

Table 2 – Energy requirement in each operations for the mineral sequestration processes.

Strong acid			Ammonium salts		
Stages	Mass t/t_{CO2SEQ}	Exergy kW/ t_{CO2SEQ}	Stages	Mass t/ t_{CO2SEQ}	Exergy kW/ t_{CO2SEQ}
Leaching of Mg (R1)			CO_2 capture (R5)		
$Mg_3Si_2O_4(OH)_5$	1.75		CO_2	1.00	
HCl	1.94		NH_3	0.39	
H_2O	12.79		H_2O	0.40	
Energy requirement (80ºC)		-361	Energy requirement (25ºC)		-94
SiO_2	0.76		Leaching of Mg (R6)		
$Mg_3Si_2O_4(OH)_5$	0.01		$Mg_3Si_2O_4(OH)_5$	2.10	
Mineralization of CO_2 (R2)			NH_4HSO_4	5.23	
NaOH	1.56		H_2O	29.40	
H_2O	1.51		Energy requirement (90ºC)		-3752
CO_2	1.00		SiO_2	0.91	
Energy requirement (40ºC)		-10	Mineralization of CO_2 (R8)		
$Mg_5(OH)_2(CO_3)_4.4H_2O$	1.80		NH_3	0.39	
NaOH regeneration (R3)			H_2O	0.40	
NaOH produced	1.51		NH_4HCO_3	1.80	
H_2	0.06		Energy requirement (80ºC)		-4.09
Cl_2	1.33		$MgCO_3.3H_2O$	3.14	
Energy requirement (80ºC)		3381	Regeneration AS (R9)		
HCl production (R4)			$(NH_4)_2SO_4$	6.03	
HCl produced	1.93		H_2O	29.24	
Energy requirement (200ºC)		-993	Energy requirement (280ºC)		1196
Total energy requirement	-------	2388	NH_4HSO_4	5.21	
			NH_3	0.77	
			H_2O	29.24	
			Total energy requirements	-----	1196

AS: Ammonium Salt

3.2 Life cycle inventory (energy)

Table 9 presents a summary of the database which was used to calculate the energy requirement and CO_2 emissions on LCA system. The mining of coal has lower energy requirement compared to the serpentine. It can be noted that depending on the mineral carbonation process the required amount of mineral, and therefore increases their CO_2 emissions. For transport is considered as diesel fuel, so the energy required is the same for both the transport of coal and serpentine. The consumption of the fuel depends mainly on the mass of materials and the distance between: (1) coal mining – power plant, (2) CO_2 source – mineral sequestration unit and (3) serpentine mining – mineral sequestration unit, being approximately 4 km, 1675 km and 2374 km, respectively. The transport stage presents the lowest energy requirement. The energy requirement for the power plant stage is defined by the technology used to capture the CO_2. In the case of a mineral sequestration process by strong acid, it is often used the MEA (Monoethanolamine). For the mineral sequestration by ammonium salt, it was used an ammonia scrubber operation, which is exothermic and does not required energy input. The high energy requirement in the mineral sequestration stage is mainly due to the regeneration process. Therefore, to mineralize 1.138 t of CO_2 derived from coal power plant, the mineral sequestration process by strong acid showed higher energy requirement.

Table 3 – Summary data base for LCA system (1MWh)

Stage		Name	Value	Unit
Mining	Coal	Mass	0.672	t
		Exergy requirement	300	MJ/t
		CO_2 emission	0.021	t
	Serpentine	Mass – SA	1.992	t
		Mass – AS	2.390	t
		Exergy requirement	83	MJ/ t
		CO_2 emission – SA	0.0173	t
		CO_2 emission – AS	0.021	t
Transport	Coal	Distance	4	km
		Consumption fuel	0.008	lt/kmt
		Exergy requirement	0.036	MJ/lt
		CO_2 emission	0.000	t
	CO_2	Distance	1675	km
		Mass	1.138	t
		Consumption fuel	0.001	lt/kmt
		CO_2 emission	0.001	t/km.t
	Serpentine	Distance	2374	t
		Mass – SA	1.992	t
		Mass – AS	2.390	t
		Consumption fuel	0.008	lt/kmt
		Exergy requirement	0.036	MJ/lt
		CO_2 emission – SA	0.172	t
		CO_2 emission – AS	0.206	t
Power plant	Coal pulverized	Exergy produced	1	MWe
		CO_2 emission w/o CCS	1.138	t
		CO_2 emission w CCS	0.475	t
		Energy requirement by CCS	4000	MJ/tCO_2cap
Mineral Sequestration	Strong Acid	Exergy requirement	8532	MJ/tCO_2
		CO_2 emission	0.76	t
	Ammonium Salts	Exergy requirement	4310	MJ/tCO_2
		CO_2 emission	0.38	t
SA: Strong Acid, AS: Ammonium Salts				

3.3 Life cycle inventory (CO_2 emission)

Figure 7 shows CO_2 emissions of each stage of the LCA system per 1t of CO_2 converted in a carbonate. The mining stage presents the lowest CO_2 emission, being 0.024 and 0.025 tCO_2 for strong acid and ammonium salts, respectively. These CO_2 emissions are a function of energy requirement and the emission factor of the power source. The CO_2 emissions derived from transport stage is a function of the distance, mass of materials and fuel type used – in this case diesel oil – (emission factor = 3.06 $kgCO_2$/lt. fuel), being their consumption about 0.000801 Lt.km-1.t-1 (railway trains), thus, it can be noted that CO_2 emissions from transport are low. However, CO_2 emissions from the power plant stage are resultant from CO_2 capture technologies used (i.e. MEA is used for strong acid and a ammonia scrubber is used for ammonium salts), thereby to use MEA, produces a CO_2 emission. The CO_2 emission in mineral process by ammonium salts is due to thermal treatment on recovery stage. The final quantity of CO_2 is a result of the difference between the CO_2 emitted and carbonated (sequestered). A positive value means a emission, however, a negative indicate a quantity effective CO_2 avoided. Comparing both mineral processes, it is observed that by strong acid emit a quantity of CO_2, however, by ammonium salts present a negative value, which represent a considerably quantity of CO_2 avoided. The amount of avoided CO_2 is calculated according to Khoo H . H. et al., (2011), as it is shown below:

$$CO_{2avoided} = CO_2^{PP} - \left(CO_2^{Mining} + CO_2^{Transport} + CO_2^{PowerPlant} + CO_2^{Carbonatio}\right) \quad (1)$$

where CO_2^{PP} is the amount of CO_2 sequestrated from the coal power plant (1.138 tCO2/MWh). CO_2^{Mining}, $CO_2^{Transport}$, $CO_2^{PowerPlant}$ and $CO_2^{Carbonatio}$ are the amounts of CO_2 emission from mining, transport, carbon capture process of the power plant and mineral sequestration processes, respectively.

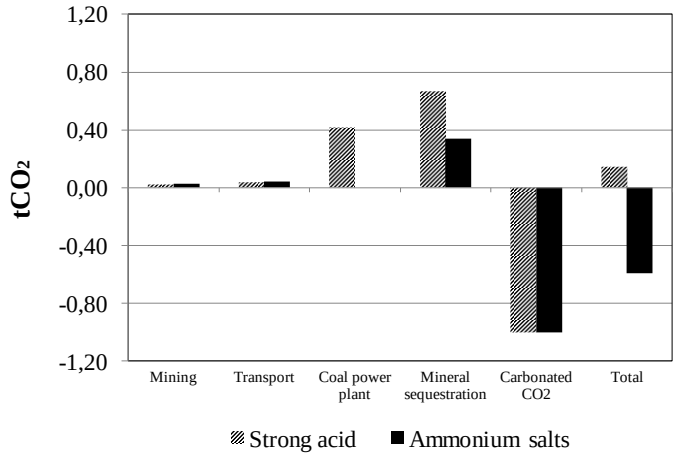

Figure 5 – CO_2 emission for each per 1 tCO_{2SEQ} for LCA system**.**

4. CONCLUSIONS

Brazil has a large amount of mineral silicate, with serpentine being the best raw material due to its higher quantities of cations such as Mg. The quantity of serpentine necessary to fix 1.138 t of CO2 (emission of Brazilian coal power plant) into carbonates is 1.99 t and 2.38 t (strong acid and ammonium salts, respectively). A mineral sequestration process by ammonium salt needs 0.39 t of serpentine more than strong acids unit, however, requires less energy than by strong acids. The mining and transport are less critical than the mineral sequestration stage, whereas the energy (mining and transport) can be increased if the energy requirement in the mineral sequestration stage is reduced (critical stage). The mineral sequestration is a critical stage due to the necessity of larger quantities of power and thermal energy, principally for the regeneration chemical inputs (i.e. 3.4 MW and 1.2 MW for strong acids (electrolysis - R3) and ammonium salts (thermal decomposition -R9), respectively). Therefore, in the mineral sequestration process by ammonium salts, it can be produced carbonates and SiO2 at around 3.14 t and 0.91 t per each 1 tCO2 fixed as carbonates, respectively. The carbonates can be used for the cement industry or soil recovery, and the SiO2 can be sold for the electronic industry. 0.54 t of CO2 avoided were obtained for 1 t of CO2 fixed as carbonate, which can be traded in the carbon market. Nowadays, the value of 1 t of CO2 avoided is approximately USD$ 25. A coal power plant in Rio Grande do sul produces around 458 MWh if the power plant works at 7740 hour/year, i.e. in 1 year, it can be obtained 1.914.256.8 t of avoided CO2 and, if it were traded, it would be obtained USD$ 47.86 million dollars. An economic assessment involving capital cost is certainty necessary, O&M cost to know the revenue value and production cost for factory of carbonates which will determine if this is economically feasible. Although the energy consumption of mineral

sequestration by ammonium salt was lower, still is not feasible for to be used for Brazilian power plants of coal pulverized.

5. ACKNOWLEDGMENT

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