

Use of Alternative Materials to Produce Lithium Silicate for CO₂ Capture

Luciene da Silva Castro*, Alexandre Carvalho Bertoli**, Ana Paula de Carvalho Teixeira**, Rochel Montero Lago**, Vanessa de Freitas Cunha Lins*

*Programa de Pós-Graduação em Engenharia Química, Universidade Federal de Minas Gerais, Avenida Presidente Antônio Carlos, 6627, Escola de Engenharia, Campus Pampulha, Belo Horizonte, Minas Gerais – Brazil.

**Departamento de Química do Instituto de Ciências Exatas, Universidade Federal de Minas Gerais, Avenida Presidente Antônio Carlos, 6627, Campus Pampulha, Belo Horizonte, Minas Gerais – Brasil.

e-mail: lucienecastro@hotmail.com; bertolialexandre@yahoo.com.br; anapct@ufmg.br; rochel@ufmg.br; vlins@deq.ufmg.br.

ABSTRACT

The development of efficient and low-cost materials for carbon dioxide capture is currently an important technological challenge. Among several materials used for CO₂ capture, lithium silicate has been proved an important alternative for several applications. The present work reviews the use of different available and low-cost mineral and waste silica sources to prepare lithium silicate. The main natural silica sources mapped were halloysite, vermiculite, kaolinite, sepiolite, wollastonite, serpentinite, diatomite, and quartz whereas the main potential wastes were slag, fly ash and rice husk ash. Based on several aspects compared for the different sources, e.g., silica content, the preparation methods, and the CO₂ capture efficiencies and a critical analysis suggested that depending on the location, cost, and availability different silica sources may stand out as a more promising source.

Keywords: CO₂ capture; lithium silicate; mineral; silica; waste.

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I. INTRODUCTION

Fossil fuel utilization and anthropogenic activities are mainly responsible for dioxide carbon emissions (CO₂) in the atmosphere, one of the main responsible gases for climate change [1]. In this scenario, technologies involving Carbon Capture and Storage (CCS) and Carbon Capture and Utilization (CCU) [2]–[4] have received much attention in the last years. In the CCS technology, the captured CO₂ is transferred to a suitable local for long-term storage and in the case of CCU, the captured CO₂ is transformed into a product [5]. Capture technologies are usually divided into three groups: post-combustion, pre-combustion, and oxyfuel [3]. In pre-combustion technology, CO₂ is separated from other gases in an intermediate step [6], [7]. In the oxyfuel process, the fuel is burnt in almost pure oxygen resulting in high temperature and CO₂ high concentration (about 80%). In post-combustion, the capture is carried out after combustion with air, obtained carbon dioxide, normally in low concentration. Among these

techniques, post-combustion is the most used [7]–[9]. The most common post-combustion separation processes are absorption, membrane separation, adsorption, cryogenic separation, and chemical looping combustion (CLC) [10]. There are different kinds of materials that can be used for CO₂ capture, for example zeolites, carbon, MOFs (metal-organic frameworks), COFs (covalent organic frameworks), and silicates [11]. One of the most interesting material for CO₂ capture is the lithium silicate [12].

II. LITHIUM SILICATE (Li₄SiO₄) FOR CO₂ CAPTURE

Lithium silicate is an important material for CO₂ capture due to its high CO₂ capture capacity (theoretical value: 367 mg CO₂/g Li₄SiO₄), excellent cyclic (until 50 cycles [13]) and thermal stability [14] [15] with relatively low regeneration temperature (> 700 °C) as compared with CaO. It also shows the possibility of using available and low-cost raw materials for its production [16]–[20].

Its CO₂ capture capacity depends on the silica source used and on the synthesis method [21].

As the literature uses different terminologies for the CO₂ removal by lithium silicate, e.g. "CO₂ absorption" [22]–[24], "CO₂ adsorption" [25], and carbonatation [26], hereon, the generic term "CO₂ capture" will be applied.

Several silica sources can be used for lithium silicate synthesis, which can be classified as synthetic, natural mineral, and waste [16]. The synthetic silica is the most used starting material for lithium silicate production [20], [27]–[36] and it is commercially available in different forms [37]. However, synthetic silica usually has relatively high costs and a more complex production process [38]. The use of silica from waste or natural mineral sources is an alternative to replace synthetic silica for large-scale lithium silicate production due to the availability and low cost of these precursors [13]. The present work reviews the use of alternative sources of silica to produce lithium silicate to carbon dioxide capture. This review will be divided into a brief report on the different types of existing lithium silicate, followed by the main alternative sources of silica (minerals and wastes) that can be used for the synthesis of lithium silicate and the use of the obtained materials for CO₂ capture. In addition, a comparison will be made of the capture capacities of materials produced from synthetic, mineral, and waste silica sources. Finally, a consideration on the use of alternative sources for lithium silicate production will be presented, considering aspects such as cost, availability, and need for pre-treatment.

III. LITHIUM SILICATES: Li₄SiO₄ AND Li₂SiO₃

Lithium orthosilicate (Li₄SiO₄) is a white solid with a monoclinic crystalline structure with lattice parameters $a = 5.303 \text{ \AA}$, $b = 6.113 \text{ \AA}$, $c = 5.154 \text{ \AA}$, and $\beta = 90.33^\circ$ [36] (Fig. 1).

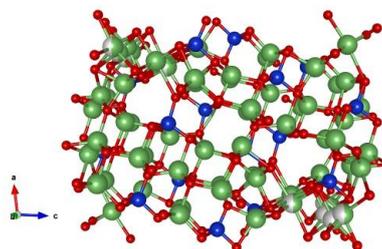


Figure 1 – Lithium silicate structure.

Lithium orthosilicate can be obtained from different synthesis methods such as solid-state reaction [14], [17], [24], [27], [32], [36], [39]–[42], sol-gel [32], [43], [44], impregnation [14], [32], [45], [46], solvo-plasma [47], spray-drying [48], and combustion [33], [49]. Lithium silicate formation always used a silica source and a lithium precursor regardless of synthesis methods. Its formation depends on different variables such as reaction temperature and time, the silica source, lithium precursor, and Li: Si molar ratio. The calcination temperature range used in the lithium silicate synthesis varied between 700 and 1000 °C, while calcination time varied between 1h [29] and 15h [50]. The Li: Si molar ratio equal to 4 favors the formation of this compound, therefore, it is the most used in the lithium silicate synthesis [32], [41], [51], [52]. Lithium silicate usually has low specific surface area, dense morphology, and low porosity [20].

The lithium metasilicate (Li₂SiO₃), an intermediate phase, is usually formed during Li₄SiO₄ synthesis [13]. This phase is present together with lithium silicate probably due to sublimation of Li₂O according to Equation 1 [53]. It can also be obtained from an incomplete reaction between lithium precursor and silica, according to Equation 2.



The lithium metasilicate has an orthorhombic crystalline structure (Fig. 2) and lattice parameters $a = 9.392 \text{ \AA}$; $b = 5.397 \text{ \AA}$; $c = 4.660 \text{ \AA}$ [54]. Li₂SiO₃ has a small CO₂ capture capacity between 300°C and 600°C [55].

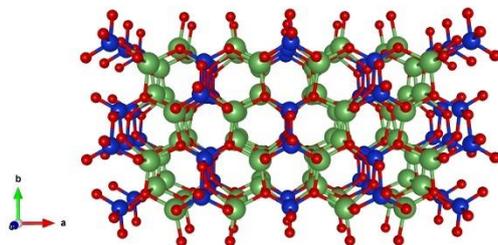


Figure 2 – Lithium metasilicate structure.

IV. ALTERNATIVE SOURCES OF SILICA FOR LITHIUM SILICATE SYNTHESIS

The main rich alternative silica sources used for lithium silicate synthesis are: rice husk ash [45], [49], [56]–[59], fly ash [46], [52], [60], [61],

wood dust ash [62], cotton stalk ash [62], slag [25], [31], [63], natural clays [64]–[66], quartz [67], [68], diatomite [69]–[71], and serpentinite [72]. Fig. 3 shows silica source belonging to the minerals class and waste and their respective SiO₂ contents.

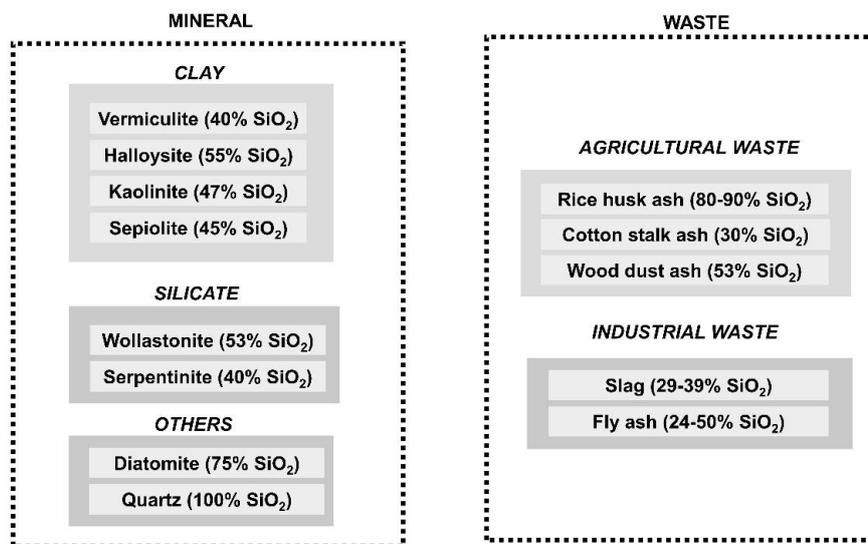


Figure 3 – Schematic drawing of the source of mineral and waste silica and their respective SiO₂ content.

The silica contents vary from 40 up to 100% for mineral sources. For waste silica sources, lower silica content is observed, from 24%, reaching up to 90%, in the case of rice husk ash.

4.1 Silica source from minerals

The silica sources derived from minerals used in the production of lithium silicate found in the literature were: vermiculite, halloysite, kaolinite, sepiolite, wollastonite, serpentinite, diatomite, and quartz.

4.1.1 Vermiculite

Vermiculite ((MgFe, Al)₃(Al, Si)₄O₁₀(OH)₂·4H₂O) is a clay mineral composed of silica (SiO₂), aluminum, iron, and magnesium [73], [74]. This low-cost and abundant mineral [75], [76] has a crystalline structure composed of a Mg ions octahedral layer between two silica tetrahedral layers [77]. The vermiculite chemical composition can vary widely from 20-40% SiO₂ and others components such as Al₂O₃, MgO, and Fe₂O₃ [78].

In the work of Zhang et al. [79], vermiculite was used as a silica source for lithium

silicate synthesis. Its composition was of 40% SiO₂, 22% MgO, 21% Al₂O₃, 7% K₂O, 5% Fe₂O₃, 3.5% CaO, 1.5% Na₂O, and 1% TiO₂ (wt.%). For lithium silicate synthesis, vermiculite was washed and dried at 120 °C and calcined at 900 °C for 2h. All materials obtained from raw vermiculite and treated vermiculite used a Li:Si molar ratio of 4 and formed Li₄SiO₄ as the main phase and MgO and Li₅AlO₄ as secondary phases [79].

4.1.2 Halloysite

Halloysite (Al₂(OH)₄Si₂O₅·2H₂O) is a clay mineral of the two-layer aluminosilicate class (a sheet formed by alumina octahedra and a sheet formed by silica tetrahedra) at a stoichiometric ratio of 1:1. It is composed of approximately 45% silica and 40% aluminum oxide and may contain some contaminants such as TiO₂, FeO, MgO, CaO, Na₂O, K₂O, SiO₂, and Al₂O₃ [80]. This mineral can be characterized as hydrated kaolinite with a lot of spacing between the layers. The morphology of this mineral can be presented as spheres, plates, or tubes[81]. Halloysite on a microscopy scale usually occurs in nanotubes form. Natural halloysite can be found in Brazil, China, France, Japan, the USA, South Korea, Turkey, and Poland [82].

Niu *et al.* [42] used halloysite nanotubes pretreated in acid as a silica source for Li₄SiO₄ synthesis by solid-state reaction method using lithium carbonate as a lithium precursor and a Li: Si molar ratio of 4:1. The material was calcined at 800°C for 4 h. The authors compared the results obtained with Li₄SiO₄ with the results obtained with SiO₂ and Li₂CO₃. Halloysite nanotube's (HNT) chemical composition was approximately 55% SiO₂, 44% Al₂O₃, and the rest were composed of ZnO, CaO, MgO e K₂O. After pretreatment in the acidic medium of the halloysite nanotubes, the silica content increased from 55% to 95%, and the concentration of Al₂O₃ decreased.

4.1.3 Kaolinite

Kaolinite, Al₂Si₂O₅(OH)₄, also known as kaolin, is a white clay mineral found in nature resulting from the granite metamorphosis [83]. This low-cost and abundant mineral [84] is formed by a layer of alumina octahedron sheet and a layer of silica tetrahedron sheet in a ratio of 1:1 [85]. Kaolin is usually composed of Al₂O₃ (ca. 40%), SiO₂ (47%), and H₂O [86].

Gao *et al.* [66] used kaolin as a silica source for lithium silicate synthesis. For it, kaolin was submitted to alkaline and acid pretreatments before calcination. The SiO₂ from kaolin was mixed with LiNO₃ with the aid of a solvent, ethanol, using a Li: Si molar ratio of 4:1, and calcined at 750 °C.

4.1.3 Sepiolite

Sepiolite is a natural fibrous clay mineral, composed of hydrated magnesium silicate (Si₁₂O₃₀Mg₈(OH)₄(H₂O)₄·8H₂O). This mineral is low-cost, non-toxic, with a relatively high surface area, and a needle-shaped material [87]–[89]. This mineral has a 2:1 structure formed by two tetrahedral silica layers and an octahedral layer formed by magnesium bonded to hydrogen and oxygen atoms [90]. The sepiolite chemical composition varies between 65-69 % SiO₂ and 19-31% MgO [91].

The sepiolite used by Zhang *et al.* [92] was composed of 45% SiO₂, 23% CaO, 16% MgO, 5.5% MnO, 2.5% Al₂O₃, and Fe₂O₃, F, and TiO₂. The raw sepiolite was subjected to an acid pretreatment, before lithium silicate synthesis using three different methods: solid-state reaction, impregnation, and impregnation-precipitation method.

4.1.4 Wollastonite

Wollastonite, composed mainly of calcium silicate (CaSiO₃), is a natural white mineral, with a circular shape similar to a needle found widely around the world. Wollastonite's chemical composition depends on its origin, but typically it is composed of 52 % SiO₂, and 48% CaO. The wollastonite showed excellent chemical resistance in several media, excellent thermal stability, and non-toxicity [93]–[95].

Wollastonite was used as a silica source for Li₄SiO₄ synthesized by Yang *et al.* [13]. Its chemical composition was 53% SiO₂, 42% CaO, and other oxides such as P₂O₅, MgO, Al₂O₃, and Fe₂O₃. The lithium precursor used was lithium hydroxide. The synthesis method was impregnation with a Li: Si molar ratio of 4.2: 1. All materials obtained were calcined at 900 °C for 4 h. In this material, the presence of the Li₂SiO₃ phase was not observed, which can be attributed to its interaction with CaO, forming the Li₂CaSiO₄ phase.

4.1.5 Serpentinite

Serpentinite with the simplified formula (Mg₃Si₂O₅(OH)₄) is a rock made up of minerals from the serpentine and magnetite groups but may contain brucite (Mg(OH)₂) in some cases, as well as secondary minerals such as talc (Mg₃Si₄O₁₀(OH)₂), magnesite (MgCO₃), and calcite (CaCO₃) [96]. The serpentine group is formed by lizardite (Mg₃Si₂O₅(OH)₄), chrysotile (Mg₃Si₂O₅(OH)₄), and antigorite ((Mg, Fe²⁺)₃Si₂O₅(OH)₄) [97]. The serpentinite has 32-38% MgO and 35-40% SiO₂ [72].

Vieira *et al.* [26] used serpentinite as a silica source for lithium silicate synthesis. The synthesis method

used was the impregnation method with different calcination temperatures of 500, 700, and 900°C for 3 h. The lithium precursor was LiOH, and the authors evaluated the effect of the addition of lithium (5, 10, and 20%) on the lithium silicate synthesis.

4.1.6 Diatomite

Diatomite (SiO_2), known as diatomaceous earth, is a biogenic sedimentary rock formed from the waste (skeleton and shell) of diatoms (algae). It is naturally available and is found in abundance in marine areas such as lakes and oceans, which usually gradually accumulates in deposits. Diatomite is a low-cost, non-toxic, and low-density material, which presents a high porosity, high surface area, and small particles enriched in amorphous silica [98]–[102]. The silica content varies according to species [103]. The typical chemical composition of diatomite is silica (80–90%), alumina (2–4%), iron oxide (0.5–2%), and impurities such as organic compounds, and other metal oxides [104]. In the work of Shan *et al.* [21], [70], [71], the diatomite chemical composition used for lithium silicate synthesis was 75% SiO_2 , 14% Al_2O_3 , 2% Fe_2O_3 , 1% CaO , and 2% K_2O .

4.1.7 Quartz

Quartz is the second most abundant mineral on planet Earth, found in igneous, metamorphic, and sedimentary rocks. This mineral is one of the most important sources of silica [105] composed of silicon and oxygen, SiO_2 . Seggiani *et al.* [67] produced pure Li_4SiO_4 and doped with potassium carbonate from amorphous silica (quartz wool and fumed silica) and crystalline quartz by using the solid-state reaction method.

Xu *et al.* [68] investigated the synthesis of Li_4SiO_4 from quartz with different particle sizes (75–180 μm , 45–75 μm , 38–45 μm e <38 μm). The authors used a Li:Si molar ratio of 4:1 for lithium silicate synthesis through the solid-state reaction method at 720°C for 6 h. The Li_4SiO_4 phase formation depends on the quartz particle size, that is, small quartz particles lead to higher Li_4SiO_4 phase formation.

Table 1 summarizes the most relevant information about the minerals used for lithium silicate synthesis, such as the chemical formula of the minerals, their composition, their availability, and where they are found. Table 2 shows the conditions for lithium silicate synthesis from these different described minerals.

Table 1 - The chemical formula of minerals, composition, where they are found, and availability.

Mineral	Chemical formula/Composition	Origin country	Global Resources/reserves	Ref.
Diatomite	SiO_2 75.26% SiO_2 , 14.33% Al_2O_3 , 2.31% Fe_2O_3 , 1.02 % CaO, 1.02-2.0% MgO, 1.56 % K_2O , and 1.17% others	It is found in the United States, Argentina, China, Denmark, France, Germany, Japan, the Republic of Korea, Mexico, New Zealand, Peru, Russia, Spain, and Turkey.	It is available 2,200 mil tons.	[21], [106]
Vermiculite	$Mg(Fe,Al)_3(Al,Si)_4O_{10}(OH)_2 \cdot 4H_2O$ 39.8% SiO_2 , 20.7% Al_2O_3 , 5.1% Fe_2O_3 , 3.4 % CaO, 21.7% MgO, 1.5% Na_2O , 6.9% K_2O , 0.9% TiO_2	It is found in many countries around the world, mainly in United States, Brazil, Bulgaria, China, India, Russia, Uganda, South Africa, and Zimbabwe.	It is available 380 mil tons.	[64], [106]
Halloysite	$Al_2(OH)_4Si_2O_5 \cdot 2H_2O$ 54.85 % SiO_2 , 43.86% Al_2O_3 , and others (ZnO, CaO, MgO, and K_2O)	It is mined from natural deposits in Brazil, China, France, Japan, the USA, South Korea, Turkey, and Poland.	-	[42], [82], [106]
Kaolin	$Al_2Si_2O_5(OH)_4$ 46.55% SiO_2 , 39.50% Al_2O_3 and 13.96% H_2O	It is mined in the United States, Brazil, China, Czechia, Germany, India, Iran, Mexico, Spain, Turkey, Ukraine, and Uzbekistan	It is available 44,000 mil tons.	[86], [106]
Wollastonite	$CaSiO_3$ 52.79% SiO_2 , 0.81% Al_2O_3 , 41.61% CaO, 1.65% MgO, 2.06% P_2O_5 and 0.73% others.	It is mined United States, Canada, China, India, Mexico, and other countries.	It is available 1,100 mil tons	[13], [106]
Sepiolite	$(Si_{12}O_{30}Mg_8(OH)_4(H_2O)_4 \cdot 8H_2O)$ 44.72% SiO_2 , 2.58% Al_2O_3 , 0.93% Fe_2O_3 , 23.26% CaO, 16.02% MgO, 5.41% MnO, 0.73% F, and 0.125% Others	It is found mainly in Spain, but it can be found in the countries such as United States, Tanzania, Kenya, China, Turkey, Japan, and England.	It is available 850 mil tons.	[92], [107], [108]
Serpentinite	$Mg_3Si_2O_5(OH)_4$ 40.00% SiO_2 , 10.00% Fe_2O_3 , 30.00% MgO, and 20% Others (Al, Ca, Ni and Mn)	It is found a deposit in Russia, the USA, Canada, Paraguay, New Zealand, Brazil, Australia, Armenia, and Italy.	It is estimated in hundreds of millions of tons global reserve.	[26], [72]
Quartz	SiO_2 100% SiO_2	Quartz deposits are found in many countries in the world as the United States and Brazil	The total reserve of quartz in the world is estimated at 82 million tons.	[109], [110]

Table 2 – Lithium silicate synthesis condition from minerals.

Silica source	Li source	Temperature (°C)	Time (h)	Molar ratio (Li:Si)	Synthesis methods	Ref.
Diatomite	Li ₂ CO ₃	700	2	4	Solid-state reaction	[21]
	Li ₂ CO ₃	700	4	4-5.6	Solid-state reaction	[71],
	LiNO ₃	600-700	4	5.2	Solid-state reaction	[70]
	Li ₂ CO ₃	700	2	5.2	Impregnation-precipitation	[70]
Vermiculite	LiNO ₃	650-850	5-7	5.2	Solid-state reaction	[79]
Halloysite	Li ₂ CO ₃	800	4	4	Solid-state reaction	[42]
Wollastonite	LiOH.H ₂ O	900	4	4.2	Impregnation	[13]
Sepiolite	LiNO ₃	750	6	4-5	Solid-state reaction	[92]
	LiNO ₃	750	6	5	Impregnation	[92]
	LiNO ₃	750	6	5	Impregnation precipitation	[92]
Serpentinite	LiOH	500-900	3	4	Impregnation	[26]
Quartz	Li ₂ CO ₃	900	4	4	Solid-state reaction	[67]
	Li ₂ CO ₃	720	6	4.1	Solid-state reaction	[68]
	Li ₂ CO ₃	700	4	4.1	Solid-state reaction	[59]
	LiOH.H ₂ O	900	4.2	4	Impregnation	[13]
Kaolin	Li ₂ CO ₃	750	4	4	Impregnation	[66]

4.2. Silica Sources from Industrial Waste

According to the literature, the silica sources derived from industrial wastes used for lithium silicate synthesis were rice husk ashes or rice straw ashes, cotton stalk ashes, wood dust ashes, slag, and fly ash.

4.2.1 Rice husk ash

Rice husk, also known as rice straw, is one of the main agricultural wastes and comes from the rice processing process, which corresponds to an average of 20% of production. In 2019, world rice production was 496 million tons [111]. Rice husk is generally used as fuel in the production of parboiled rice or as a source of energy for thermoelectric power [112], [113]. In the rice husk burning, a waste known as rice husk ash is generated, which corresponds to 22% rice husk mass [114], [115]. It is estimated that in 2019, 2.6 million tons of rice husk ash were generated.

Rice husk ash consists of 80-90% SiO₂ [116]. Other components are shown in the rice husk ash such as K₂O, Al₂O₃, CaO, MgO, Na₂O, and Fe₂O₃ [117]. Silica can be in the amorphous or crystalline form, which depends on the burning temperature [118].

Rice husk ash used for lithium silicate synthesis was mainly constituted by amorphous

silica (85-99%), but it may also contain small amounts of Al₂O₃, Fe₂O₃, CaO, MgO, and others compounds such as Na₂O, K₂O, MnO and SO₃ [56], [59], [62]. The first work that used rice husk ash as a silica source for Li₄SiO₄ synthesis was published by Wang *et al.* [56]. The rice husk was collected in rice milling located in Wuhan, China, and calcined at 650°C followed by washing with acid and distilled water. The lithium precursor was carbonate and a Li: Si molar ratio of 4.1:1 was used; the material was calcined at 800°C for 4 h. The main phase obtained was Li₄SiO₄ with morphology of polyhedral particles and agglomerates, with sizes of 60-100 μm, and surface area of 2-6 m²/g.

Wang *et al.* [59] continued their studies using rice husk ash (amorphous nano-silica) for Li₄SiO₄ synthesis, and compared it with lithium silicate from quartz and aerosil nano-silica. The rice husk's chemical composition was basically SiO₂ (83%), but it contained some impurities such as MgO, P₂O₅, SO₃, K₂O, CaO, MnO₂, Fe₂O₃, Al₂O₃, Na₂O, and Cl [56]. Wang *et al.* [62] compared cotton stalk, rice straw, and wood dust for lithium silicate production. All these biomasses were burned using an oven at 600°C for more than 1 h to obtain the biomass ash, except the rice straw ash which was pre-treated with hot distilled water to remove impurities. Based on the Si content for each biomass

ash, the SiO₂ content was estimated, being 15%, 16.5%, 21%, and 61% of SiO₂ for cotton stalk ash, wood dust ash, straw ash of rice, and pretreated rice straw ash, respectively. All these silica sources had low silica content, except the pretreated rice husk ash, compared to the other materials used as silica sources to obtain lithium silicate.

4.2.2 Fly ash

Fly ash is a fine powder waste from coal combustion [119] constituted mainly by silica, aluminum, calcium oxide, and iron oxide. This waste shows complex composition and physicochemical properties which depend on the coal type, the collector configuration, and combustion conditions [120].

Fly ash was used as silica source for lithium silicate in several works [46], [56], [60], [121], [122]. Olivares-Marín *et al.* [122] used three samples of fly ash collected at different locals of the coal-fired power plant to synthesize Li₄SiO₄ for CO₂ capture from the coal and biomass/coal mixtures burning.

Sanna, Ramli, and Maroto-Valer [61] developed a material for CO₂ capture from fly ash (FA) collected in the cyclone filter. Fly ash shows a high-content silica (ca. 50%). Zhang *et al.* [121] used fly ash collected in the coal-fired power plant as a silica source for lithium silicate synthesis. Izquierdo *et al.* [46] carried out Li₄SiO₄ synthesis from different silica sources: fly ash from oil shale power plant and pure silica.

4.2.3 Slag

Slag is mainly a byproduct generated for the iron and steel industries corresponding to 90% of the by-products generated [123]. This byproduct is classified according to the furnace where it is produced. The main types of slag are blast furnace slag, steel furnace slag, basic oxygen furnace slag (BOF), electric arc furnace slag (EAF), and shell slag [123]. Each ton of produced steel generates 130-200 kg slag which depends on the production process of the steel and the steel composition [137, 124]. The steel world production was 1.87 billion tons in 2019 [125] which results in an estimated 308 million tons of slag. Brazil is the ninth largest iron and steel producer with a production of 32 million tons in 2019, with an estimated generation of 5.3 million tons of slag [125].

The slag composition depends on processes, but the main components usually are SiO₂, CaO, Al₂O₃, Fe₂O₃, and MgO [126]. Some studies reported the use of blast furnace slag [31],

[25], and electric furnace slag [25] for Li₄SiO₄ synthesis for CO₂ capture.

Liu *et al.* [31] used three types of silica gel derived from blast furnace slag and silica gel from sodium silicate and pure silica (quartz) for Li₄SiO₄ synthesis by the solid-state reaction method. Slag composition was 32% SiO₂, 42% CaO, 14% Al₂O₃, and traces of S, Fe₂O₃, MgO, and TiO₂. The silica content of silica gel from slag was 92% SiO₂, however, the silica gel from Na₂SiO₃ showed 97% SiO₂.

Alcántar-Vázquez *et al.* [25] used blast furnace and electric arc furnace slag for Li₄SiO₄ synthesis by using the solid-state reaction method. Slag was constituted of 39-54% SiO₂, and 2-33% CaO. The specific surface area of lithium silicate from the different slags was around 1 m²/g.

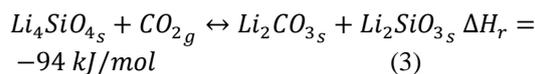
Table 3 shows the descriptions of waste used for lithium synthesis, silica content, where is found, and availability. Table 4 shows the conditions for lithium silicate synthesis from wastes.

V. LITHIUM SOURCES

The main lithium sources for lithium silicate synthesis are lithium carbonate (Li₂CO₃), anhydrous and monohydrate lithium hydroxide (LiOH e LiOH.H₂O), and lithium nitrate (LiNO₃). In addition, it was also observed to a lesser extent other lithium sources such as lithium acetate dihydrate (C₂H₃O₂Li.2H₂O), lithium oxalate (C₂Li₂O₄), lithium tartrate (C₄H₄Li₂O₆), lithium formate monohydrate (HCOOLi.H₂O), lithium citrate tribasic tetrahydrate (C₆H₅O₇Li₃.4H₂O), lithium benzoate (C₇H₅LiO₂) e lithium lactate (C₃H₅LiO₃) [16]. Another lithium source that can be used is the battery waste [127].

VI. LITHIUM SILICATE APPLICATION FOR CO₂ CAPTURE

Lithium silicate shows a high CO₂ capacity capture (theoretical value 367 mg de CO₂/g). The CO₂ capture process occurs according to Equation 3, i.e., each 1 mol of lithium silicate consumes 1 mol of CO₂ forming lithium carbonate (Li₂CO₃) and lithium metasilicate (Li₂SiO₃) [24], [50].



Lithium silicate from different minerals was used in the carbon dioxide capture. Table 5 shows the CO₂ capture conditions by lithium silicate from different minerals, however, Table 6 shows CO₂ capture conditions by lithium silicate from waste.

Table 3 – Waste characteristics, origin, location, and availability.

Waste	SiO ₂ content (%)	Origin	Origin country	Global Resources/reserves	Reference
Rice husk ash	80-90	A by-product from rice husk burning.	The main producers of rice husk are Bangladesh, Brazil, Burma, Cambodia, China, Egypt, India, Indonesia, Japan, Korea South, Nigeria, Pakistan, Philippines, Thailand, and Vietnam.	It was estimated at 2.6 million tons of rice husk ash in the world in 2019. In Brazil, it is estimated at 408 mil tons of rice husk ash in season 2019/2020.	[111], [114]–[116]
Cotton stalk ash	30	A by-product from cotton cultivation burning	The main producers of cotton are China, India, United States, Brazil, Pakistan, Uzbekistan, and Turkey.	It was estimated at 116 million tons of the cotton stalk in the world in the season of 2019/2020. In Brazil, it is estimated at 13 million tons of the cotton stalk in season 2019/2020.	[128], [129]
Wood dust ash	2-53	A by-product from the wood beneficiation process.	The main producers of sandwood are China, the United States of America, the Russian Federation, Canada, Germany, and Sweden.	It was estimated at 1.16 million of wood dust ash in the world. In Brazil, it was estimated 23 mil tons of wood dust ash in 2019.	[130], [131]
Slag	29-39	A by-product generated during the production of iron and steel	The main producers of steel are China, India, Japan, United States, Russia, South Korea, Germany, Turkey, Brazil, Iran, and Italy.	It was estimated at 308 million tons of slag in the world in 2019. It was estimated 5.3 million de tons of slag in Brazil in 2019.	[125].
Fly ash	24-50	A by-product from the thermal power plant (Waste from coal combustion)	The main producers of fly ash are India, China, United States, Germany, United Kingdom, Australia, and France.	It is generated 375 million tons of fly ash annually.	[46], [61], [121], [122], [132]

Table 4 – Lithium silicate synthesis conditions from waste.

Silica source	Li source	Temperature(°C)	Time (h)	Molar ratio (Li: Si)	Synthesis methods	Ref.
Rice husk ash	Li ₂ CO ₃	800	4	4.1	Solid-state reaction	[56]
	Li ₂ CO ₃	700	4	4.1	Solid-state reaction	[59]
Rice straw ash	Li ₂ CO ₃	800	4	4.1	Solid-state reaction	[57]
	Li ₂ CO ₃	800	4	4.1	Solid-state reaction	[62]
Cotton stalk ash	Li ₂ CO ₃	800	4	4.1	Solid-state reaction	[62]
Wood dust ash	Li ₂ CO ₃	800	4	4.1	Solid-state reaction	[62]
Fly ash	Li ₂ CO ₃	950	8	4	Solid-state reaction	[122]
	Li ₂ CO ₃	800	8		Solid-state reaction	[61]
	Li ₂ CO ₃	750	6	4	Solid-state reaction	[121]
	Li ₂ CO ₃	800	7	4.4	Solid-state reaction	[46]
	LiOH.H ₂ O	600	7	4.4	Solid-state reaction	[46]
	LiOH.H ₂ O	600	7	4.4	Precipitation	[46]
Slag	Li ₂ CO ₃	-	-	3-7	Solid-state reaction	[31]
	LiOH.H ₂ O	-	-	3-7	Solid-state reaction	[31]
	Li ₂ CO ₃	850	8	4.2	Solid-state reaction	[25]
	Li ₂ CO ₃	600-800	4-6	4	Solid-state reaction	[31]

Table 5 – CO₂ capture conditions by lithium silicate from mineral.

Silica source	CO ₂ capture conditions				CO ₂ capture Capacity(%)	Ref.
	Temperature(°C)	Time (min)	N ₂ flow	CO ₂ flow		
Diatomite	620	180	50 mL/min	50 mL/min	28	[21]
	-	-	50 mL/min	50 mL/min	30	[71]
Halloysite	700	20	50 mL/min	50 mL/min	34	[70]
	350–720	-	-	60 mL/ min	34	[42]
Wollastonite	500-600	120	-	-	18	[13]
Vermiculite	650	120	-	-	40	[96]
Sepiolite	500-700	120	-	-	33	[92]
Quartz	Room temperature-1000	-	-	50 mL/min	31	[68]

	550-680	120	-	50 mL/min	25	[59]
	500-600	120	100 mL/min	100 mL/min	27	[67]
Serpentinite	400-700	180	-	20 mL/min	25	[26]
Kaolinite	500-700	-		60 mL/min	34	[133]

VII. COMPARISON OF CO₂ CAPTURE CAPACITY BY LITHIUM SILICATE OBTAINED FROM WASTE AND MINERAL.

Fig. 4 shows the CO₂ capture capacity by lithium silicate obtained from different minerals and wastes. The CO₂ capture capacity is expressed in percentage (w/w %) which means the gain in mass of the material in relation to the initial mass of the material.

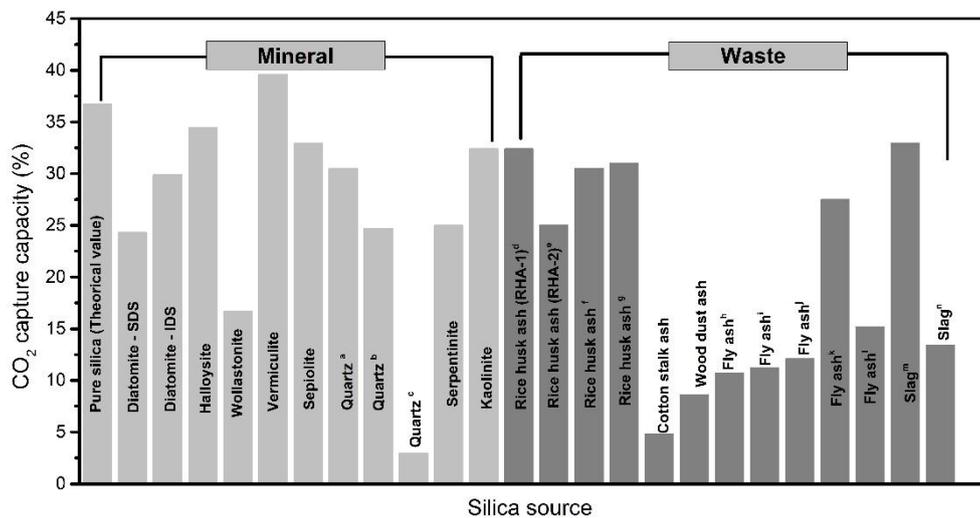


Figure 4– CO₂ capture capacity by lithium silicate obtained from mineral and waste.

The material obtained from vermiculite showed a higher CO₂ capture capacity compared to others obtained from different silica sources. It was also observed that its CO₂ capture capacity was superior of the theoretical CO₂ capture capacity of lithium silicate due to the presence of Li₂O, Li₈SiO₆, and Li₅AlO₄, which showed a higher CO₂ capture capacity than Li₄SiO₄. The obtainment of lithium silicate from halloysite, sepiolite, quartz, and kaolinite were carried out by pretreatments such as acid leaching, doping, or grinding, which caused an improvement CO₂ capture capacity. It can be seen

the discrepancy in the carbon dioxide capture capacity of the lithium silicate obtained from quartz different studies when compared with other silica sources. The result related by Xu *et al.* [68] was ten times higher than that reported by Yang *et al.* [13], which can be attributed to several factors such as synthesis conditions (temperature and calcination time), lithium precursor, and gas flow. In the lithium synthesis from waste, it was observed that lithium silicate from rice husk ash, fly ash, and slag showed different values of CO₂ capture capacity.

VIII. COMPARISON OF LITHIUM SILICATE OBTAINED FROM SYNTHETIC SILICA SOURCE

Fig. 5 shows the comparison of CO₂ capture capacity for lithium silicate obtained from main synthetic silica sources and the best silica source from waste and minerals.

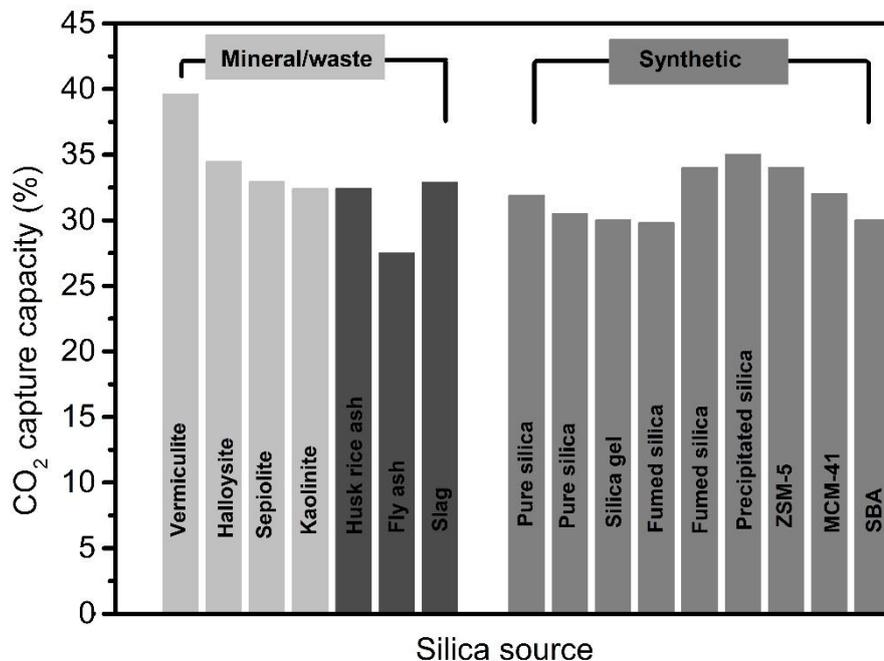


Figure 5 – Comparison of CO₂ capture capacity by lithium silicate obtained from mineral and residue with lithium silicate from synthetic silica.

It is possible to observe a certain similarity between the values of CO₂ capture capacity of these materials. Therefore, it shows that the lithium silicate obtained from waste and minerals is promising as compared to lithium silicate from synthetic silica. Lithium silicate from vermiculite showed a higher CO₂ capture capacity than lithium silicates from other minerals, wastes, and even synthetic silica.

IX. COMPARISON OF THE POSITIVE AND NEGATIVE FACTORS OF THE DIFFERENT MINERAL AND WASTE SILICA SOURCES

Table 6 shows estimates of the availability, the cost, the need of pre-treatment for the different silica sources and the CO₂ capture capacities for the lithium silicate produced. The information in Table 6 indicates that all minerals and wastes sources are potential candidates as raw materials for lithium silicate production with a good CO₂ capture capacity in general. However, the availability of these

sources in different parts of the world can vary significantly and so does their quantity and cost.

In general, the natural raw materials show a SiO₂ content between 40 and 75%, except quartz (100% SiO₂). The wastes show SiO₂ content that can vary over a wide range between 24-50% for slag and fly ash. Among the waste class, rice husk ash stands out with a content of ca. 80-90% SiO₂.

Another aspect that must be considered is the need for a pre-treatment of the raw material (silica source) to be transformed into Li₄SiO₄. For some precursors, such as halloysite, sepiolite and fly ash, an acid leaching process to remove impurities is necessary. This process introduces a degree of complexity and cost. On the other hand, minerals such as quartz and serpentinite only need a grinding process, simplifying the production of lithium silicate. For wastes such as fly ash and slag, relatively complex treatments are proposed, while rice husk ash can be used directly.

The cost of the silica mineral source can vary over a wide range, depending on the quality of the mineral and its application, being lower values

in the range of US\$10-575/ton, however, it can reach US\$ 1000 to 24250/ton. In the case of waste,

the cost of raw material tends to be much lower, in the range of US\$14-100/ton.

Table 6 – Availability, the raw material cost, pre-treatment carried out on the silica-rich raw material, and CO₂ capture capacity by the lithium silicate.

Waste/mineral	SiO ₂ content %	Availability/Quantity in the world (thousand ton)	raw material cost (US\$/ton)	Requirement of a pre-treatment of the raw material	CO ₂ capture capacity (%)
Diatomite	75	2200 ^a	10-1000 ^a	No ^o	34
Vermiculite	40	380 ^a	140-575 ^a	Washing / drying / calcination ^p	40
Halloysite	55	Not found	3000 ^h	Acid washing ^q	34
Wollastonite	53	1100 ^a	360-390 ^a	No ^r	18
Sepiolite	45	850 ^b	400-24250 ⁱ	Acid washing ^s	33
Serpentinite	40	100000 ^c	8 ^j	No ^t	25
Quartz industrial	100	82000 ^d	19-500 ^{h,l}	No ^u	31
Kaolinite	46	44000 ^a	160 ^a	No ^v	34
Rice husk ash	80-90	2600 ^e	55-100 ^m	No ^w	34
Fly ash	24-50	375000 ^f	25-75 ⁿ	Acid washing ^x	28
Slag	29-39	308000 ^g	14 ^a	Silica gel synthesis ^y	33

^a [106]; ^b [107]; ^c [72]; ^d [110]; ^e [111]; ^f [132]; ^g [125]; ^h [134]; ⁱ [135]; ^j [136]; ^k [137]; ^m [138]; ⁿ [139]; ^o [70]; ^p [64]; ^q [42]; ^r [13]; ^s [92]; ^t [26]; ^u [68]; ^v [133]; ^w [57]; ^x [121]; ^y [31].

Among the options described in Table 6, related to the minerals class, quartz stands out. It seems to be the most promising alternative, due to its high SiO₂ content, its availability, its price, and no need of pre-treatment.

As for the waste class, rice husk ash seems to be the best option, due to its increased SiO₂ content and no need of pre-treatment. However, the relatively high cost can be an impediment compared to other wastes. Therefore, fly ash becomes an alternative due to its lower cost compared to the rice husk ash and a greater availability among the wastes. Although fly ash is relatively more expensive than slag, it nevertheless requires a less complex lithium silicate synthesis process compared to slag since the slag has been turned into silica gel [31].

An important aspect to be considered is the logistics involving the place where the raw material

is available and the place where it will be transformed into lithium silicate and used for CO₂ capture. Therefore, the ideal scenario is that the raw material (silica source), the lithium silicate production industry, and the dioxide capture industry are close. In this context, the raw material, mineral or waste, must be chosen in each case, as there may be situations where these raw materials can be made available for values much lower than those reported in Table 6.

Another important aspect to be highlighted is that mineral raw materials are non-renewable natural resources, so their use must be made very judiciously. In addition, it requires the extraction of deposits that can cause environmental and social impacts, due to the exploitation of the soil and disposal in dams. On the other hand, the use of waste seems very attractive as it encourages a circular economy and sustainability. Especially rice

husk ash, which is currently a waste constantly generated in agriculture without proper disposal. This waste is a renewable source of raw material, while fly ash (coal) and slag are related to finite mineral resources.

X. CONCLUSION

Lithium silicate is an important alternative for carbon dioxide capture because of the high CO₂ capture capacity at high temperatures. Another advantage is that lithium silicate can be prepared from a wide range of sources such as synthetic, mineral, and waste sources. The use of silica sources from minerals can be an interesting alternative due to the availability and cost of many different raw materials. On the other hand, silica waste sources are interesting sustainable circular and economical alternatives compared to non-renewable minerals. It also must be considered to select the most suitable raw material for the Li₄SiO₄ the cost, logistics, and complexity of the synthesis process.

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