

## Carbon Capture Performance Enhancement of Solid State Synthesized $\text{Li}_4\text{SiO}_4$ Powders by Using Different Kind Steel Slags as $\text{SiO}_2$ Source

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### Abstract

The potential of lithium orthosilicate ( $\text{Li}_4\text{SiO}_4$ ) as a material to capture carbon dioxide ( $\text{CO}_2$ ) is being explored due to its excellent capture capability and thermal stability. Currently, a significant portion of the world's energy needs are met through the burning of fossil fuels, which leads to an increase in atmospheric  $\text{CO}_2$  and contributes to global warming. While fossil fuels will remain the primary source of energy for several more years, it is crucial to develop cost-effective and environmentally friendly technology for large-scale  $\text{CO}_2$  capture and storage before its release into the atmosphere in order to mitigate the associated greenhouse effect. This study focuses on optimizing the synthesis of lithium orthosilicate using a solid-state method to enhance  $\text{CO}_2$  capture. The research involves assessing the key characteristics of the synthesized material that improve  $\text{CO}_2$  capture efficiency. The starting materials used for comparison include lithium carbonate ( $\text{Li}_2\text{CO}_3$ ), BF (blast furnace), BOF (blast oxygen furnace) and EAF (electric arc furnace) slags, and pure reagents of silicon dioxide ( $\text{SiO}_2$ ). The effect of process conditions, such as synthesis temperatures ranging from 850 to 950°C and varying holding times of 4, 6, 8, and 10 hours in a muffle furnace, were studied and compared. X-ray diffraction (XRD) analysis was performed to characterize the samples and slags.  $\text{CO}_2$  capture performance of the samples were evaluated in thermobalance test under 92 vol%  $\text{CO}_2$  ( $\text{N}_2$  balance) gas concentration at 600°C. As to conclude, carbon dioxide capturing sorbent material was produced by utilizing the slags generated as a by-product in steel production. Thus, considering the principles of circular economy, a material defined as waste was used as raw material for the production of another product used for lowering atmospheric  $\text{CO}_2$  levels.

### Keywords

Solid state synthesis; Slag utilization; Carbon Capture; Global Warming

### 1. Introduction

Global warming, a component of climate change, pertains to the sustained elevation of global temperatures over an extended period. It is a concerning phenomenon driven by the increased concentration of greenhouse gases in the Earth's atmosphere, primarily resulting from human activities such as the combustion of fossil fuels in industrial processes, deforestation, and agricultural practices [1]. These activities contribute to the release of significant amounts of carbon dioxide into the atmosphere.

When carbon dioxide ( $\text{CO}_2$ ) and other air pollutants and greenhouse gases accumulate in the atmosphere, they have the ability to trap heat and solar radiation that would otherwise escape into space. This trapped heat creates a greenhouse effect, leading to a rise in global temperatures and a disruption of the Earth's climate patterns. The consequences of global warming include rising sea levels, more frequent and severe weather events, and adverse effects on ecosystems and biodiversity.

To address the urgent need to mitigate the impact of carbon dioxide emissions, researchers have been actively exploring

various approaches to reduce its concentration in the atmosphere. Several technologies have emerged as potential solutions, including absorption, adsorption, membrane gas separation, and gas hydrate technologies. These methods aim to directly capture carbon dioxide from the air or industrial sources, thereby curbing its contribution to the greenhouse effect.

In the pursuit of more effective carbon capture and storage methods, novel materials are being investigated for their ability to capture and store carbon dioxide. One such material is lithium orthosilicate ( $\text{Li}_4\text{SiO}_4$ ), which shows promise due to its high capture capacity for carbon dioxide and its thermal stability. By optimizing the synthesis of lithium orthosilicate and understanding its key characteristics, researchers aim to enhance its efficiency in capturing carbon dioxide, paving the way for a more sustainable and environmentally friendly approach to combating global warming [2].

The exploration of lithium orthosilicate ( $\text{Li}_4\text{SiO}_4$ ) and its potential applications has extended beyond its use in lithium-ion batteries and  $\text{CO}_2$  doping. This ceramic material has garnered significant attention as a robust sorbent for  $\text{CO}_2$  capture due to its immediate absorption and desorption capabilities. Its unique properties enable it to quickly and efficiently capture carbon dioxide from various sources, including industrial emissions and the atmosphere.

The research and development efforts surrounding lithium orthosilicate aim to optimize its performance as a  $\text{CO}_2$  sorbent. Researchers are investigating methods to enhance its absorption capacity, improve its stability during cyclic  $\text{CO}_2$  capture and release processes, and explore its compatibility with different operating conditions.

The utilization of lithium orthosilicate for  $\text{CO}_2$  capture aligns with the global pursuit of sustainable and environmentally friendly technologies. By capturing and storing carbon dioxide, this ceramic material has the potential to mitigate the impacts of greenhouse gas emissions, contributing to the worldwide efforts to combat global warming and climate change.

Moreover, the versatile nature of lithium orthosilicate opens up possibilities for its application in other areas related to carbon capture and utilization. Researchers are exploring its potential in catalytic processes, carbon dioxide conversion into value-added products, and even as a potential component in advanced energy storage systems.

In summary, the ongoing exploration of lithium orthosilicate as a robust  $\text{CO}_2$  sorbent showcases its significance in addressing the challenges of global warming. Through further advancements and optimization, this ceramic material holds promise for enabling more efficient and sustainable carbon capture and utilization strategies, ultimately driving us towards a greener and more environmentally conscious future [3, 4].

### 1.1. Slag and Slag Types

Slag is a glass-like by-product that remains after separating a target metal from the raw ore. Slag is usually composed of metal oxides and silicon dioxide. Slags can include metal sulphides and elemental metals. Significant volumes of waste materials, such as slags, are generated during the iron and steel production process.

### 1.2. Blast Furnace (BF) Slag

Iron ore, iron scrap, and fluxes, such as limestone and/or dolomite, are introduced into a blast furnace together with coke, serving as fuel, to facilitate the production of iron. Within the furnace, coke undergoes combustion, resulting in the formation of carbon monoxide, which acts upon the iron ore, transforming it into a molten state. This molten iron possesses the capability to be moulded into various iron-based products, although its primary application lies in its utilization as a raw material in the manufacturing of steel. As a by-product of the blast furnace process, slag emerges, representing a non-metallic substance predominantly composed of silicates, aluminosilicates, and calcium-alumina-silicates.

In a study, silica extracted from blast furnace slag by using an acid leaching process was used to prepare  $\text{Li}_4\text{SiO}_4$  using the solid-state reaction method with  $\text{Li}_2\text{CO}_3$  at  $873^\circ\text{C}$ . At  $600\text{--}650^\circ\text{C}$ , the adsorption potential in pure  $\text{CO}_2$  was  $100.8\text{ mg CO}_2/\text{g}$  sorbent, and it rose to 98% conversion at  $700^\circ\text{C}$ . This was linked to the slag's small particle size and the presence of metal impurities like potassium and calcium [5].

### 1.3. Basic Oxygen Furnace (BOF) Slag

The slag from a basic oxygen furnace (BOF) has high alkalinity, a high angularity, a hard surface, and relatively good mechanical properties [6]. For every ton of crude steel produced in a BOF, approximately  $100\text{--}150\text{ kg}$  of slag is extracted as waste, depending on the hot metal condition and steel-making activity. Slag from the BOF process contains approximately half of  $\text{CaO}$  and is used to flux the oxides of silicon (Si), phosphorus (P), sulphur (S), and manganese (Mn) formed during the liquid steel processing process. These oxides react with the dissolved calcium oxide ( $\text{CaO}$ ) to form the BOF slag. Many steel plants only partly reprocess this solid waste, and a large amount is either discarded or used for ground filling also for cement production for construction [7].

BOF slag is considered to have highly heterogeneous surfaces. The heterogeneity of the BOF slag surface is caused by two factors: geometrical and chemical. Geometrical heterogeneity (porosity) is caused by variations in the size and shape of pores, as well as pits, and vacancies meanwhile the chemical heterogeneity is correlated with different functional groups on the surface as well as different surface contaminants. BOF slag's distinctive adsorption properties are influenced by both chemical and geometrical heterogeneities [8].

#### 1.4. Electric Arc Furnace (EAF) Slag

Electric arc furnace (EAF) slag, a by-product of steelmaking recovered during the oxidizing process, can be used as aggregate in hydraulic concrete. Several studies have been conducted on the characteristics of EAF slag in relation to its use in the building industry, specifically on its material properties, possible expansivity, and chemical reactivity. [9, 10]

EAF slag is a non-metallic by-product composed primarily of silicates and oxides produced during the processing of molten steel. Depending on the ferrous oxide material, raw EAF slag may appear as grey or black lumps. This slag typically has a coarse surface texture. In addition, higher iron content in EAF slag may result in a higher density. [9, 10, 11].

It is well known that EAF slag from various regions and factories may have varying appearances and physical properties, depending on the composition of steel scrap used as feed materials, the form of the furnace, steel grades, and processing processes. Regardless of the variations in chemical compositions, EAF slag usually has Mohs hardness values in the range of 6–7 [11, 12].

Characteristics of BOF slag and EAF slag for CO<sub>2</sub> capture have been studied earlier. In this aforementioned study, CO<sub>2</sub>-capture characteristics were compared. It has been found that in general, the higher the temperature, the better the calcium (Ca) conversion of slag. The Ca and MgO content of slag is an important factor in determining CO<sub>2</sub> sorption potential at various temperatures and CO<sub>2</sub> concentrations. Ca use of slag increased with rising temperature in the 400–500°C range, likely due to reaction kinetics and reactive gas diffusion also the exothermic character of the crystallization of CaCO<sub>3</sub> as the higher temperatures may impede also a more complete carbonates generation. Though for a 50% and 75% CO<sub>2</sub> concentration for BOF slag and 75% concentration for EAF slag, Ca usage at 550°C was found to be lower than at 500°C. One possible explanation is that the carbonation reaction is driven not only by reaction kinetics but also by reactive gas diffusion. Steel slag is a porous material thus CO<sub>2</sub> will diffuse to the slag's surface and then into the slag's pores to react with CaO. Due to this, the thick product layer of calcium carbonate (CaCO<sub>3</sub>) will cover the surface of CaO, preventing further interaction of CO<sub>2</sub> molecules with CaO. Also, due to the different molar concentrations of CaCO<sub>3</sub> and CaO, the high-volume substance CaCO<sub>3</sub> would fill the pores within the slag and reduce the available CaO surface area, slowing the reaction even more. In general, for the two types of steel slag, namely BOF slag and EAF slag, the Ca usage will be at its lowest at 50% CO<sub>2</sub> at 450 and 550°C. Higher values may occur when the CO<sub>2</sub> concentration is smaller, about 10% or higher, which is more than 75%. As a conclusion, in terms of both CO<sub>2</sub>

reactivity and Ca usage, EAF slag outperformed BOF slag [13].

## 2. Materials and Methods

### 2.1. Materials

Three different slags were used, these slags are named; BF (blast furnace), BOF (basic oxygen furnace) and, EAF (electric arc furnace) slags. These three different slags were obtained from different iron and steel Turkish industries. The chemical compositions of above-mentioned slags are given in the Table 1 below.

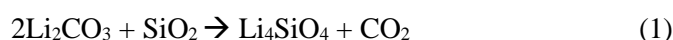
**Table 1.** Chemical compositions of slags (wt.%).

Slag	FeO	SiO <sub>2</sub>	CaO	Al <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>	MnO
BF	0.36	37.20	39.55	13.14	5.89	1.56	0.42
BOF	18.43	12.25	48.04	2.12	3.72	0.50	2.80
EAF	23.86	15.06	37.07	6.63	6.40	0.63	4.63

### 2.2. Method

#### 2.2.1. Solid state synthesis of Li<sub>4</sub>SiO<sub>4</sub> powders

Li<sub>4</sub>SiO<sub>4</sub> can be synthesized by the reaction of a homogeneous mixture of lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>) and silica (SiO<sub>2</sub>) in a 2:1 molar ratio. Li<sub>4</sub>SiO<sub>4</sub> produced by chemical reaction at solid state is as shown (1). This reaction is the overall reaction for the solid-state synthesis of Li<sub>4</sub>SiO<sub>4</sub>. Firstly, Li<sub>2</sub>SiO<sub>3</sub> forms and this formed Li<sub>2</sub>SiO<sub>3</sub> reacts with Li<sub>2</sub>CO<sub>3</sub> to result in obtaining Li<sub>4</sub>SiO<sub>4</sub>.



In this work, lithium orthosilicate adsorbents have been prepared based on 10 grams of Li<sub>2</sub>CO<sub>3</sub> using three separate slags and pure reagent of SiO<sub>2</sub> as silica sources, and the following step was the preparation of the mixture with a molar ratio of 2.1:1 to avoid evaporation of lithium carbonate at high temperatures. Each sample has been mixed for 30 minutes in a turbula mixer to ensure a homogeneous mixture. The mixtures have been put into alumina boat crucibles for calcination and the calcination has been done in muffle furnace at a temperature range of 750-850-950°C and a time duration (holding time) of 4-6-8-10 hours for varying samples. After cooling down under normal conditions, the samples were grinded with an agate mortar in order to shape them into a powder form for XRD analysis.

In review of Table 2, a comprehensive set of 36 experiments was carried out, encompassing various experimental conditions shown in Table 2. Specifically, among these 36 experiments, 12 were specifically focused on investigating the properties of BF slag, while the remaining 24 experiments were dedicated to the evaluation of other slag types and pure SiO<sub>2</sub> reagent.

To investigate and compare the outcomes, a series of experiments were conducted involving BF (blast furnace) slag, BOF (basic oxygen furnace) slag, and EAF (electric arc

furnace) slag. These experiments were conducted under varying temperatures ranging from 850°C to 950°C, and holding times of 4, 6, 8, and 10 hours. To establish a baseline for comparison, pure SiO<sub>2</sub> reagents were also employed in the study.

**Table 2.** Experimental parameters.

Experiment No. #	Temperature / Time	SiO <sub>2</sub> Source
1	950°C / 8h	BF
2	950°C / 4h	BF
3	950°C / 6h	BF
4	950°C / 6h	BOF
5	950°C / 10h	BF
6	950°C / 10h	BOF
7	850°C / 10h	BF
8	850°C / 10h	BOF
9	850°C / 8h	BF
10	850°C / 8h	BOF
11	950°C / 10h	SiO <sub>2</sub>
12	950°C / 8h	SiO <sub>2</sub>
13	850°C / 10h	SiO <sub>2</sub>
14	850°C / 8h	SiO <sub>2</sub>
15	950°C / 6h	SiO <sub>2</sub>
16	950°C / 4h	SiO <sub>2</sub>
17	850°C / 6h	SiO <sub>2</sub>
18	850°C / 4h	SiO <sub>2</sub>
19	950°C / 4h	BOF
20	950°C / 8h	BOF
21	950°C / 4h	EAF
22	950°C / 6h	EAF
23	950°C / 8h	EAF
24	950°C / 10h	EAF
25	850°C / 4h	EAF
26	850°C / 6h	EAF
27	850°C / 8h	EAF
28	850°C / 10h	EAF
29	850°C / 4h	BF
30	850°C / 6h	BF
31	850°C / 6h	BOF
32	850°C / 4h	BOF
33	750°C / 10h	BF
34	750°C / 8h	BF
35	750°C / 6h	BF
36	750°C / 4h	BF

Furthermore, additional experiments were carried out specifically focusing on BF slag at a temperature of 750°C. These experiments included holding times of 4, 6, 8, and 10 hours to further assess the behaviour and properties of BF slag under these specific conditions.

### 2.2.2. CO<sub>2</sub> uptake tests

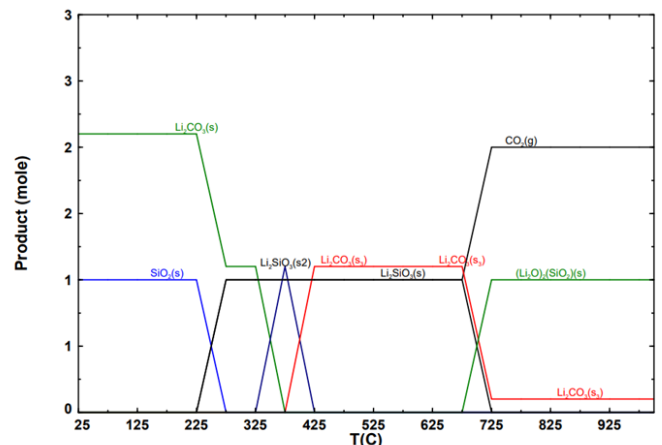
The CO<sub>2</sub> sorption performance of synthesized powders was evaluated using a TGA equipment (Setaram Setsys Evolution). First, these samples were treated at 100°C in N<sub>2</sub> atmosphere to eliminate possible previous carbonation. Subsequently, they were heated up to 600°C with an increase rate of 20°C/min.

Afterwards, they were again subjected to a 10 minutes long process under N<sub>2</sub> gas. Then, tests were carried out for 2 hours under a flow rate of 65 ml/min with a gas atmosphere of 92 vol% CO<sub>2</sub> (balance N<sub>2</sub>).

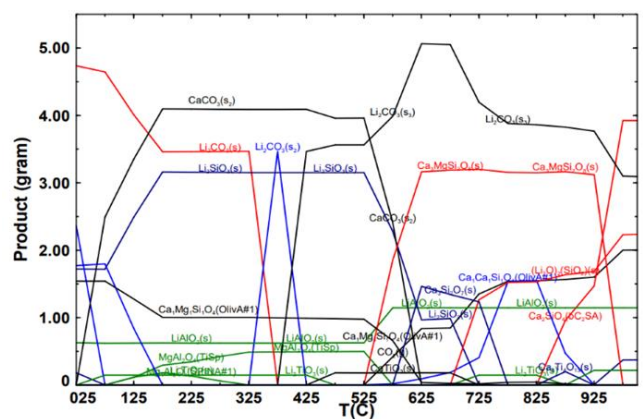
## 3. Results and Discussion

### 3.1. Results of thermodynamical investigations

The thermodynamical modelling for the production of the sorbents was performed using the thermochemical simulation software FactSage 8.1. From Figure 1, it can be said that at 225°C, lithium carbonate and silicon dioxide start to react, their moles are decreasing and lithium metasilicate (Li<sub>2</sub>SiO<sub>3</sub>) moles are starting to increase which means that Equation (1) takes place and both lithium metasilicate and carbon dioxide starts to form. Then, this carbon dioxide, reacts with the excess lithium and forms lithium carbonate again in between 325°C and 425°C according to simulation. Afterwards, the remaining lithium carbonate reacts with lithium metasilicate and starts to produce lithium orthosilicate and carbon dioxide. This explains the molar increase of carbon dioxide and the formation of lithium orthosilicate at 675°C. The products are lithium orthosilicate, carbon dioxide and excess lithium carbonate due to 2.1:1 molar ratio. Lithium orthosilicate is shown as (Li<sub>2</sub>O)<sub>2</sub>(SiO<sub>2</sub>) in the figure.

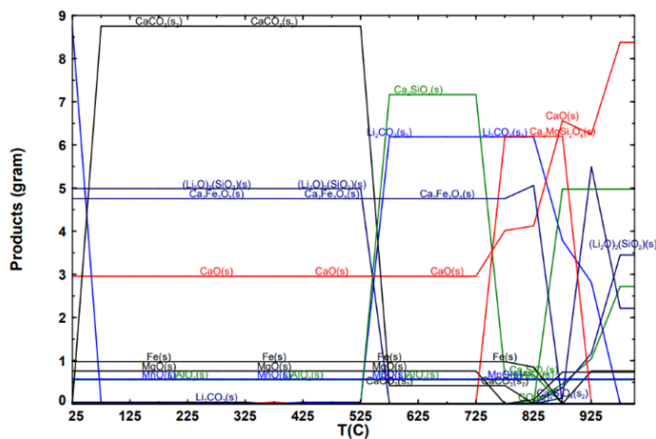


**Figure 1.** Modelling of the production for Li<sub>4</sub>SiO<sub>4</sub> using SiO<sub>2</sub>.



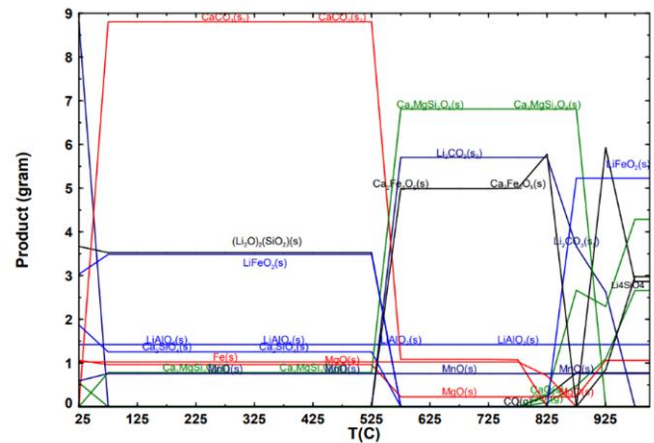
**Figure 2.** Modelling of the production for Li<sub>4</sub>SiO<sub>4</sub> using BF slag.

FactSage makes calculations and simulates the reactions based on closed systems. Slags contain lots of residues and this makes a system very complicated. It can be seen in Figure 2 that the same reaction took place in between 325°C and 425°C, where carbon dioxide, reacts with the excess lithium and forms lithium carbonate, according to simulation. Formation of lithium orthosilicate can also be seen, starting at 675°C. Between 425°C and 475°C, calcium carbonate and lithium titanite reacts to form a small amount of calcium titanite. Then, this calcium titanite starts to react with lithium carbonate at around 675°C to form lithium titanite and thus, calcium titanite converts to lithium titanite. In addition, at 525°C, reaction of lithium carbonate and lithium metasilicate can be seen, after this reaction, carbon dioxide starts to form as it is visible in the figure. It can be seen that the same reaction took place in between 325°C and 425°C, where carbon dioxide, reacts with the excess lithium and forms lithium carbonate, according to simulation.



**Figure 3.** Modelling of the production for  $\text{Li}_4\text{SiO}_4$  using BOF slag.

According to this simulation, lithium carbonate immediately converts to calcium carbonate without any action and this calcium carbonate starts to form dicalcium silicate and lithium carbonate by reacting with lithium orthosilicate at 525°C. Afterwards, at 725°C, dicalcium silicate reacts with magnesium oxide to form merwinite ( $\text{Ca}_3\text{MgSi}_2\text{O}_8$ ). This simulation has discrepancies and therefore does not reflect accurate information.



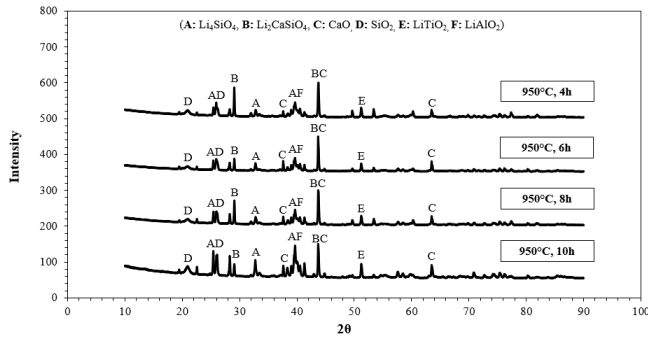
**Figure 4.** Modelling of the production for  $\text{Li}_4\text{SiO}_4$  using EAF slag.

In this simulation, calcium carbonate is formed without any interference right at the start from lithium carbonate. At 525°C, calcium carbonate reacts with lithium orthosilicate and lithium ferrite to form merwinite, lithium carbonate, and  $\text{Ca}_2\text{Fe}_3\text{O}_5$ . At approximately 850°C,  $\text{Ca}_2\text{Fe}_3\text{O}_5$  starts to convert to lithium ferrite. At the end, resulting product has lithium ferrite.

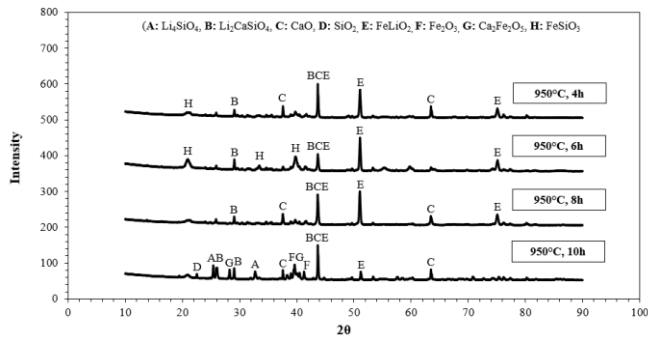
Ultimately, it should be noted that FactSage simulates calculations as finite equilibrium. This means that the consistency of these simulations is only consistent if all kinetic conditions are met. In this respect, there might be some inconsistencies.

### 3.2. Results of solid-state synthesis experiments

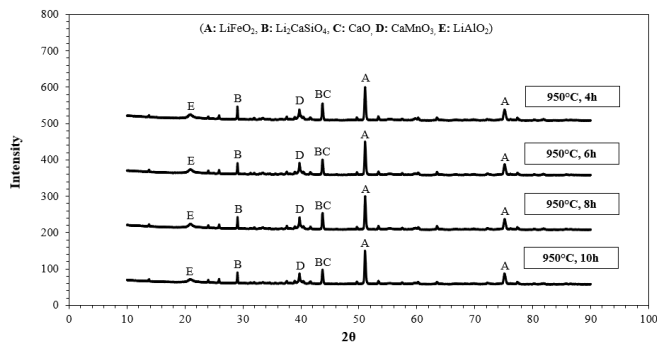
Results show that the samples made from BOF with the increasing temperatures decrease the formation of lithium silicates and these samples tend to contain a form of lithium iron oxide. According to the XRD results, with the increase of temperature and decrease of holding time, the samples tend to have more lithium silicate and calcium doped lithium silicates, which improves the  $\text{CO}_2$  capture according to other studies made on this topic [3, 14]. The involvement of calcium phases and small quantities of Mg, Fe, and Al in slag-derived lithium silicates may increase both the absorption of  $\text{CO}_2$  and the kinetic behaviour and the inclusion of calcium phases in the slag-derived materials explain the variations in  $\text{CO}_2$  adsorption capacities between pure  $\text{Li}_4\text{SiO}_4$  and slag-derived lithium silicates [15].



**Figure 5.** Comparative XRD results for BF slag derived product for 950°C and different holding times.



**Figure 6.** Comparative XRD patterns for BOF slag derived products for 950°C and different holding times.



**Figure 7.** Comparative XRD results for EAF slag derived product for 950°C and different holding times.

The involvement of calcium phases and small quantities of Mg, Fe, and Al in slag-derived lithium silicates may increase both the absorption of CO<sub>2</sub> and the kinetic behaviour and the inclusion of calcium phases in the slag-derived materials explain the variations in CO<sub>2</sub> adsorption capacities between pure Li<sub>4</sub>SiO<sub>4</sub> and slag-derived lithium silicates [5].

XRD results show that BF slag-derived samples have much higher lithium orthosilicate (Li<sub>4</sub>SiO<sub>4</sub>) and dilithium calcium silicate (Li<sub>2</sub>CaSiO<sub>4</sub>) when compared with other type of slag-derived samples. They also include much less untransformed slag and it can be seen that the conversion rates to lithium orthosilicate (Li<sub>4</sub>SiO<sub>4</sub>) and dilithium calcium silicate (Li<sub>2</sub>CaSiO<sub>4</sub>) are satisfying. BF slag-derived samples that are synthesized under 750°C have unconverted lithium carbonate

in their compositions. LiTiO<sub>2</sub> in small quantities exist in BF slag-derived samples synthesized at 850°C. BOF slag-derived samples that were made at 950°C have a lot of residual materials as it can be seen from XRD peaks. In addition, BOF slag-derived and EAF slag-derived samples have high content of ferrous oxide in their composition. Ferrous oxide hinders the formation of lithium orthosilicate and promotes the formation of lithium ferrite. Because lithium orthosilicate is the main element for capturing large volumes of CO<sub>2</sub> in this study, a low content, or even a complete lack of lithium orthosilicate, limits the performance accordingly. Unlike BOF slag-derived samples, tricalcium silicate content can be seen in EAF slag-derived samples at 850°C. CO<sub>2</sub> reacts with tricalcium silicate; however, it does not have a good uptake performance. This will lead to higher CO<sub>2</sub> uptake and this can be proved by CO<sub>2</sub> uptake tests, which is mentioned in CO<sub>2</sub> uptake results.

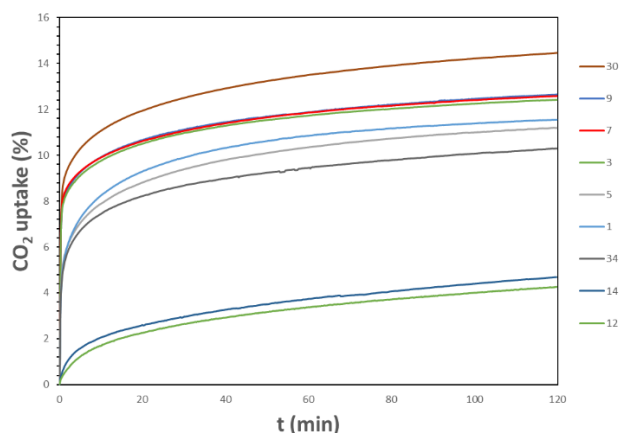
Specific surface area determination of the samples was carried out by N<sub>2</sub> physisorption at 77°K by Micromeritics ASAP2020 with performing the Brunauer, Emmett, and Teller (BET) method according to the ISO 9277 Standard. The results for the BET method are given in Table 3.

Exp. No.	Surface Area (m <sup>2</sup> /g)
7	1.242
9	1.309
30	1.557
5	0.941
1	0.801
3	0.999
34	0.784
14	0.771
12	0.742

Specific surface areas calculated with the BET method are 1.557 m<sup>2</sup>/g, 1.309 m<sup>2</sup>/g, 1.242 m<sup>2</sup>/g, 0.999 m<sup>2</sup>/g, 0.941 m<sup>2</sup>/g, 0.801 m<sup>2</sup>/g, 0.784 m<sup>2</sup>/g, 0.771 m<sup>2</sup>/g and, 0.742 m<sup>2</sup>/g for samples, respectively.

### 3.3. Results of CO<sub>2</sub> uptake experiments

According to Figure 8, out of these nine samples, the best performance was shown by Sample 30. It reached a value of 14.46% CO<sub>2</sub> uptake at the end of the test. Sample 7, 9 and, 3 almost have the same performance by CO<sub>2</sub> uptake values of 12.58%, 12.65% and, 12.41% respectively.



**Figure 8.** CO<sub>2</sub> uptake values of the samples.

Theoretically, samples that were produced with pure SiO<sub>2</sub> source have 33.78% of CO<sub>2</sub> uptake. However, they performed very poorly compared to samples produced from BF slag as SiO<sub>2</sub> source.

Maximum theoretical CO<sub>2</sub> uptake calculations are made for these samples for 92% CO<sub>2</sub> containing atmosphere and compared to experimental results which can be found below (Table 4).

**Table 4.** Theoretical and experimental CO<sub>2</sub> uptake values.

Exp. No	Temperature (°C) / Hours	SiO <sub>2</sub> Source	CO <sub>2</sub> Experimental Uptake (wt%)	CO <sub>2</sub> Theoretical Uptake (wt%)
7	850 / 10	BF Slag	12.58	39.26
9	850 / 8	BF Slag	12.65	29.77
30	850 / 6	BF Slag	14.46	40.12
5	950 / 10	BF Slag	11.55	24.47
1	950 / 8	BF Slag	11.19	27.13
3	950 / 6	BF Slag	12.41	35.34
34	750 / 8	BF Slag	10.29	26.03
14	850 / 8	SiO <sub>2</sub>	4.69	33.78
12	950 / 8	SiO <sub>2</sub>	4.26	33.78

Carbon capture efficiencies of the samples; 32.04%, 42.49%, 36.04%, 64.24%, 51.47%, 46.96%, 39.53%, 13.88%, and 12.61% respectively. Sample 1, which were synthesized at 950°C from BF slag for 8 hours has showed best efficiency by means of comparing with maximum theoretical uptake values while sample 12, which were synthesized at 950°C from pure SiO<sub>2</sub> for 8 hours has showed the worst performance. This confirms that, slag-derived samples are showing greater performance than pure lithium orthosilicate samples made with pure SiO<sub>2</sub> source. The low carbon capture efficiencies of pure SiO<sub>2</sub> samples are likely attributable to the long treatment periods used during synthesis, which has resulted in increased crystal diameter [16]. These results are also in good agreement with the BET results.

#### 4. Conclusion

In this work, lithium orthosilicate adsorbents were effectively synthesized employing three different steel slags which are BF, BOF and EAF as silica sources in addition to pure SiO<sub>2</sub> for

comparison. The produced materials were also characterized and tested for carbon dioxide (CO<sub>2</sub>) uptake. All BF slag-derived samples showed better conversion rates and thus a better CO<sub>2</sub> uptake percentage. It can be said with certainty that lithium orthosilicate (Li<sub>4</sub>SiO<sub>4</sub>) captures more CO<sub>2</sub> than dilithium calcium silicate (Li<sub>2</sub>CaSiO<sub>4</sub>). These findings indicate that slag-derived silicates are attractive materials and good candidates for application in high-temperature CO<sub>2</sub> capture processes. Samples produced from BF slag as SiO<sub>2</sub> source at 950°C have the best performance, followed by again BF slag-derived samples which were produced at 850°C and 750°C. At 850°C and 4 hours of holding time, BF slag-derived sample contains 57.9% lithium orthosilicate, 7.7% dilithium calcium silicate, and 21.3% calcium oxide. At 850°C and 6 hours of holding time, BF slag-derived sample contains 70.5% lithium orthosilicate, 6.2% dilithium calcium silicate, and 21.4% calcium oxide. At 850°C and 8 hours of holding time, BF slag-derived sample contains 61.3% lithium orthosilicate, 8.2% dilithium calcium silicate, and 11% calcium oxide. At 850°C and 10 hours of holding time, BF slag-derived sample contains 69.5% lithium orthosilicate, 6.1% dilithium calcium silicate, and 20.7% calcium oxide. As it can be seen, samples produced from BF slag at 850°C have the best conversion rates compared to other samples. This can be explained when the compositions of the slags are reviewed. BF slag has a higher SiO<sub>2</sub> percentage and also less FeO (ferrous oxide) percentage. This high SiO<sub>2</sub> percentage is an important variable since it converted to lithium orthosilicate (Li<sub>4</sub>SiO<sub>4</sub>) during solid-state synthesis which has a high yield for CO<sub>2</sub> uptake performance. It has been shown that samples synthesized from BOF slag and EAF slag in this study had ineffective and unfavorable conversion rates when compared to samples created with BF slag, which is also supported by Rietveld studies made by HighScore Plus.

Furthermore, BOF slag-derived and EAF slag-derived samples have showed poor conversion rates. This is due to the high content of ferrous oxide of these slags. Ferrous oxide hinders the formation of lithium orthosilicate and promotes the formation of lithium ferrite. Since lithium orthosilicate is the key material to capture high amounts of CO<sub>2</sub>, low content, or in some cases lack of lithium orthosilicate, hampers potential performance. Unlike BOF slag-derived samples, in EAF slag-derived samples, at 850°C tricalcium silicate formation can be seen. CO<sub>2</sub> reacts with tricalcium silicate; however, it does not have a good uptake performance.

Low carbon capture efficiencies of samples made with pure SiO<sub>2</sub> is probably due to the long treatment times during synthesis and as a result crystal diameter have increased [16].

High temperature during solid-state synthesis may lead to sintering during the synthesis of lithium orthosilicate. This might result in low efficiencies in carbon capture due to synthesized lithium orthosilicates' cores not reacting with carbon dioxide [16].

In addition, in samples synthesized at 750°C, unconverted lithium carbonate ( $\text{Li}_2\text{CO}_3$ ) was found in zabuyelite form according to XRD analysis made with HighScore Plus whilst using the same parameters (heating rate and holding time) as other samples. This shows that at lower synthesis temperatures, lithium carbonate content does not have enough time to react fully and also that it needs to be at higher temperatures. Furthermore, the percentage of lithium carbonate in these samples increased with less holding time which means that it is inversely proportional, the higher the holding time, the less content of lithium carbonate. In the carbon dioxide uptake experiments, the blast furnace-derived samples outperformed the two samples made from pure  $\text{SiO}_2$  which converted to 100% lithium orthosilicate, by approximately three times.

On the other hand, other  $\text{SiO}_2$  containing waste sources such as industrial wastes, glass wastes, and fly ash or natural resources of  $\text{SiO}_2$  like rice hulls might be used in pilot experiments in order to increase waste utilization and feasibility. High porosities might help because it will lead to more reactions with carbon dioxide. Addition of potassium can also be considered since it has improved sorption capabilities in some studies. Sol-gel synthesis approach with LiOH might be recommended for a better capture performance according to the data from other studies. Sol-gel synthesis may also increase the surface area and thus would help with capture performance even more. Palletisation might be done, considering that the obtained powders are too fine. Instead of using wastes containing high content of  $\text{SiO}_2$ , lithium containing wastes might be used to lower costs.

### Authors' Contributions

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### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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