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Citation for published version:

Chukwuma, JS, Pullin, H & Renforth, P 2021, 'Assessing the carbon capture capacity of South Wales' legacy iron and steel slag', *Minerals Engineering*, vol. 173, 107232.
<https://doi.org/10.1016/j.mineng.2021.107232>

Digital Object Identifier (DOI):

[10.1016/j.mineng.2021.107232](https://doi.org/10.1016/j.mineng.2021.107232)

Link:

[Link to publication record in Heriot-Watt Research Portal](#)

Document Version:

Peer reviewed version

Published In:

Minerals Engineering

Publisher Rights Statement:

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Assessing the Carbon Capture Capacity of South Wales' Legacy Iron and Steel Slag

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Abstract

The industrial revolution was responsible for releasing over 1.5 trillion tonnes of carbon dioxide into the atmosphere resulting in a global temperature increase of ~1°C. It also produced billions of tonnes of alkaline materials that have the potential to react with some of that CO₂, and this carbon capture potential may be used to help prevent climate change. Here we assess the carbon capture potential of legacy iron (Fe) and steel wastes in South Wales, the United Kingdom, and show that between 0 – 77% of total carbonation potential has been realised in the samples (equating to maximum potential between 40 and 608 kg CO₂ /t of slag has been removed from the atmosphere by direct carbonation) across 10 historic works (out of 48 that operated in the region), even after >140 years. The current work suggests that there is a capture potential of up to 17 MtCO₂ by direct carbonation which represents a potential carbon sink for future emissions. Iron and steel slag contains Ca silicate minerals, often dominated by melilite group phases (gehlenite, Ca₂Al₂SiO₇ and akermanite, Ca₂MgSi₂O₇), but also includes minor phases like olivine (fayalite, (Fe,Mn)₂SiO₄ and fosterite, Mg₂SiO₄). The presence of mineral carbonate phases such as calcite, magnesite, ankerite, and kutnohorite in the material demonstrates carbonation reaction has occurred after slag formation. Given that there is still carbonation potential to be realised, and that materials produced in the future from the iron and steel industry could play an important role in meeting our climate targets, then new management strategies are needed to maximise the use of this resource.

Keywords: Carbon sequestration, carbon capture potential, direct carbonation, legacy deposits, historical iron and steel works, Ca silicate minerals.

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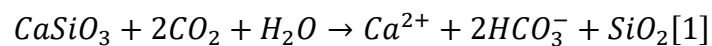
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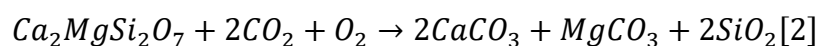
1. Introduction

38 In 2016, continuing anthropogenic emissions exceeded 40 GtCO₂ per year (1), approximately
39 half of which contributed to the build-up of carbon dioxide concentration in the atmosphere,
40 while the remainder may have been absorbed by terrestrial and ocean sinks (2). This has
41 resulted in ~1°C of warming over pre-industrial climate (3). Preventing further dangerous
42 climate change will require net-zero emissions. As it is unlikely that we will be able to
43 complexly decarbonise our economy, negative emission technologies (NETs also known as
44 ‘greenhouse gas removal’) that remove greenhouse gases from the atmosphere will be required
45 (3-4). Proposed NETs include afforestation and reforestation, soil carbon sequestration,
46 bioenergy combined with carbon capture and storage and geochemical based approaches (5).
47 The later involves reaction of calcium (Ca) or magnesium (Mg) rich silicates and oxide
48 minerals with atmospheric CO₂ to form solid or dissolved carbonate minerals in which the
49 carbon dioxide will be permanently stored (6,11). This exploits natural weathering processes
50 that already remove up to a GtCO₂ out of the atmosphere every year (7-8). Alkaline materials
51 such as blast furnace and steel slag produced during iron and steelmaking are a potential source
52 of silicate minerals (e.g., akermanite, Ca₂MgSi₂O₇ and gehlenite, Ca₂Al₂SiO₇), which when
53 dissolved in water can react with atmospheric CO₂ to form aqueous bicarbonate ions (HCO₃⁻)
54 (e.g., Eq. 1) (6,12). If these bicarbonate ions are conveyed to the ocean (e.g., in runoff and
55 rivers), they could reside in the ocean for >100,000 years (often called ‘enhanced weathering’)
56 (9). Alternatively, if sufficient Ca or Mg is dissolved from the mineral, carbonate compounds
57 can precipitate forming a permanent store of CO₂ (‘mineral carbonation’) (e.g., Eq. 2) (7-8,10).
58 While both processes will result in the sequestration of CO₂ emissions, almost twice as much
59 CO₂ is removed via enhanced weathering compared to mineral carbonation (6). However, some
60 slag can produce potentially harmful leachates (e.g., 19), in which case a self contained mineral
61 carbonation system may be more appropriate.

62



63



64 Equation (1) imply $\eta = 2$; however, due to buffering in the carbonate system, the value is
65 between 1.4 and 1.7 for typical seawater chemistry, pCO₂ and temperature (6).

66

67 Globally, approximately 500 Mt of iron and steel slag is produced every year (13). Some of
68 these by-products are reused primarily for additive purposes (e.g. aggregates in concrete, feed
69 for cement kilns, partial substitute for Portland cement, etc.), while a proportion is stockpiled.
70 In the EU, steel production emits ~182 Mt CO₂ of greenhouse gases (14), and the global steel
71 industry emits 4-5% of total emissions (15-16). Slag is a non-metallic co-product formed
72 during iron and steel production, which contains Ca, Mg, and Al silicates (22). The chemistry
73 of slag created depends on the raw materials used (21) and post-processing application.
74 Typically, iron or steel slag is catergorised into the part of steel production in which it arises
75 (e.g., ‘blast furnace’ or ‘steel slag’). Its mineralogy and physical properties are influenced by

76 how it is handled or managed (e.g., air/water cooled, grinding (23), the chemistry of the melt,
77 and subsequent management practices, which in turn influence physio-chemistry (24-25).

78 Mineral carbonation is primarily controlled by the availability of metal cations that may be
79 leached from the material and the supply of CO₂ (19). During weathering of natural minerals
80 under high temperature conditions, it is assumed that the supply of the cation is the rate-
81 limiting step in carbonation (26), which may be such for ambient weathering of artificial
82 materials, although with more rapid dissolution kinetics supply of CO₂ may also limit
83 carbonation. Therefore, slag mineralogy plays an important role in carbonation reactions
84 (15,27-29). Previous work has mostly focused on laboratory-based mineral carbonation
85 experiments under elevated CO₂ concentration or acid buffered pH levels, high temperature
86 and relatively short duration, which recorded up to 50-75% conversion over 30 minutes (11).
87 Less work has considered the ambient carbonation of slag. Although work that investigates
88 legacy deposits may provide some evidence on the long-term fate of the material (e.g., 30-32).
89 Most studies investigating legacy deposits were centred on emplaced tips (17), drainage waters
90 (19), and the surrounding environments (33). A 10-year investigation of a stainless steel slag
91 showed 10-50 gCO₂ /kg uptake varying across the 7m depth of the covered (stored in a
92 warehouse) heap (the largest CO₂ uptake recorded at the surface of the material, the location
93 not reported) (58). A >100 year old legacy iron slag heap in Pennsylvania, USA (59), while
94 trace calcite was detected using x-ray diffraction, it was not quantified. Similarly, a study on
95 ladle slags also reported calcium carbonate in X-ray diffraction, although insufficient quantities
96 were present to be detected through thermal analysis (60).

97 These have demonstrated the potential of CO₂ capture and carbonate precipitation. South
98 Wales, the United Kingdom, was a focal point for the industrial revolution, and was one of the
99 first places to exploit iron and steel manufacturing on an industrial scale. Here we examine a
100 broad geographical distribution of legacy deposits in order to assess their maximum carbon
101 capture potential at a regional level. This study supports higher resolution work that considers
102 national (20) and international (6) production/deposit estimates. Understanding the variation of
103 slag chemistry across a region allows for an assessment of the CO₂ capture potential. The
104 mineralogy and physical properties of these slag has not been previously documented. Thus,
105 this study identifies silicate minerals in legacy deposits, the extent of previous carbonation, and
106 also the remaining potential for future use.

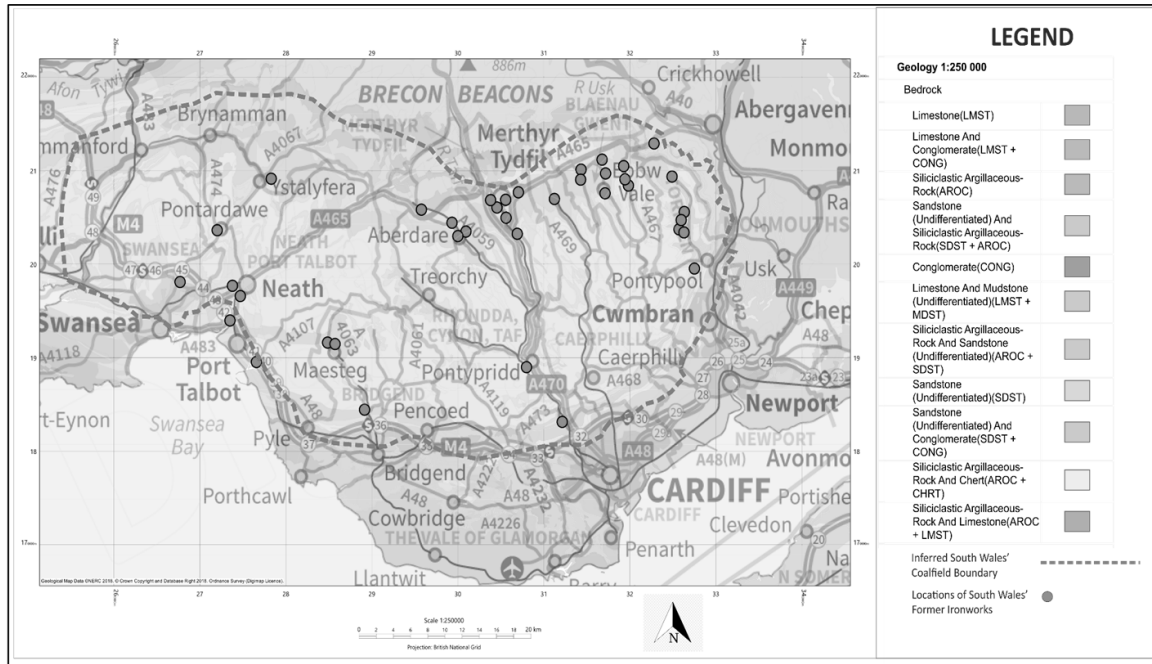
107 **2. Materials and Methods**

108 **2.2 Study Site**

109 The study sites were historical iron and steelworks that operated within the South Wales'
110 Coalfield (*Figure 1*). Iron production peaked in these areas in the Early 19th century and
111 declined in the 20th century until 1980 when most of the ironworks were closed (46-48). During
112 this time, steel production increased after the launch of basic oxygen steelmaking (BOS) in the
113 1960s (49). As was the common practice of most iron producers, slag materials were either
114 stockpiled adjacent to the ironworks (*Figure 2*) or dumped within designated landfills (17,12).
115 The historical maps and aerial photographs of the ironworks were used to select locations
116 showing little evidence of distributed slag materials.

117 The study sites overlay the Lower Carboniferous rocks, the Millstone Grit and the Coal
 118 Measures (50). The Carboniferous rocks consist of limestone and coal, which provided the raw
 119 materials for iron and steel production. Iron ore deposits are present in the lower carboniferous
 120 measures and outcrop to the north of the coalfield. However, the relatively small low-grade
 121 deposits meant that high-grade ores were imported to support the industry (51).

122
 123



124

125 **Figure 1. The map of South Wales' Coalfield in relation to its historical ironworks (Source:**
 126 **EDINA digimap, 2018).**



128

129 **Figure 2: Legacy stockpiles of slag materials at Blaenavon ironworks during active operation**
 130 **(Aerial images reproduced, courtesy Welsh Government Aerial Photographs Unit).**

132

133 **2.3 Production Data and Calculation**

134 The production data of blast furnace slag is seldom reported, and inventory assessments of
135 current stockpiles are not publicly available (6). As such, estimates of production were
136 calculated using iron and steel output, the ratio of slag to metal production (52), furnace
137 capacities (19), as well as ore grade (34). Typically, blast furnaces that utilize ore grades
138 containing 60-66% iron generates between 0.25 and 0.30 tons of slag per ton of Fe produced,
139 whereas steel is roughly around 0.13 to 0.2 tons (52). Ores with lower grades will usually
140 produce more slag (1.0 to 1.2 tons) as its chemical composition must meet certain chemical
141 requirements for optimum blast furnace operation. Therefore, the volume of raw materials
142 added to the blast furnace determines the quantity of slag generated (35). The British blast
143 furnace statistics 1790 to 1980 (46), South Wales iron industry 1750 to 1885 (47) and Early
144 history of the old South Wales ironworks 1760 to 1840 (48) were used for production
145 estimates. After the 1980s, data are from Glamorgan and Gwent Archaeological Trust Ltd,
146 Swansea (36).

147

148 **2.4 Sample Preparation and Analysis**

149 Twenty two samples were obtained using hand auger at depths <1m. Samples were prepared
150 for analysis by pulverization using a fritsch jaw crusher, siebtechnik vibrating-tema mill with a
151 tungsten carbide grinding bowl to obtain uniform particle size. Subsequently, samples were
152 oven-dried for 24 hours at 105 °C. Selected samples were investigated by X-ray diffraction
153 (XRD), X-ray fluorescence (XRF), total carbon (TC) and total inorganic carbon (TIC) analysis
154 by calcining samples at 1000°C in a muffle furnace over 8 hours.

155 **3. Results and Discussions**

156 **3.1 Production Estimates**

157 Over forty eight ironworks operated in South Wales and an estimated 330 million tonnes of
158 iron and steel was produced over 140 years, accounting for a cumulative >75 million tonnes of
159 slag. 10 ironworks were mapped for this study and estimate of the slag materials they produced
160 is presented in Table 1.

161

Regions	Ironworks	Mass of slag (Mt)	Years of operation
Ebbw Vale Group	British	0.18	53
	Sirhowy	0.09	89
Aberdare and the Cynon valley	Hirwaun	0.44	74
	Abernant	0.07	64
	Llwydcoed	0.38	67
The Neath valley	Port Talbot	56.00	87
Eastern outcrop of the Coalfield	Blaenavon	0.87	117
	Varteg	0.07	44
Northern outcrop of the Coalfield	Tredegar	0.28	95
	Cydach	0.12	53

162 **Table 1: Production output of Slag.**

163 The ironworks fall within different iron and steel production regions of South Wales, and the
164 iron producers adopted various iron-making processes during production (31). Prior to the
165 launch of the Bessemer method, iron was produced both by direct and indirect methods (31).
166 The direct method involved reduction of iron ore in a bloomery furnace via a powder
167 metallurgical process, where it reacts with fuel (e.g., wood, charcoal) to form a porous iron
168 bloom that never reaches the molten stage. During this process, SiO₂-rich slag is created, which
169 is separated as a melt. In contrast, the indirect method consists of two different steps carried out
170 in two distinct furnaces (i.e., blast furnace and fining hearth). In the blast furnace, iron ore
171 reacts with the fuel to produce molten pig iron that contains 4% carbon, together with other
172 elements such as silicon, phosphorus, and manganese. The pig iron is subsequently treated in a
173 fining hearth where it gradually becomes consolidated into a bloom, this is then hammered to
174 separate excess slag and is shaped into bars or cakes. Such difference in practice can give rise
175 to compositional variations of the end-product, thus we should expect carbon capture potential
176 to vary considerably.

177

178 **3.2 Slag Composition**

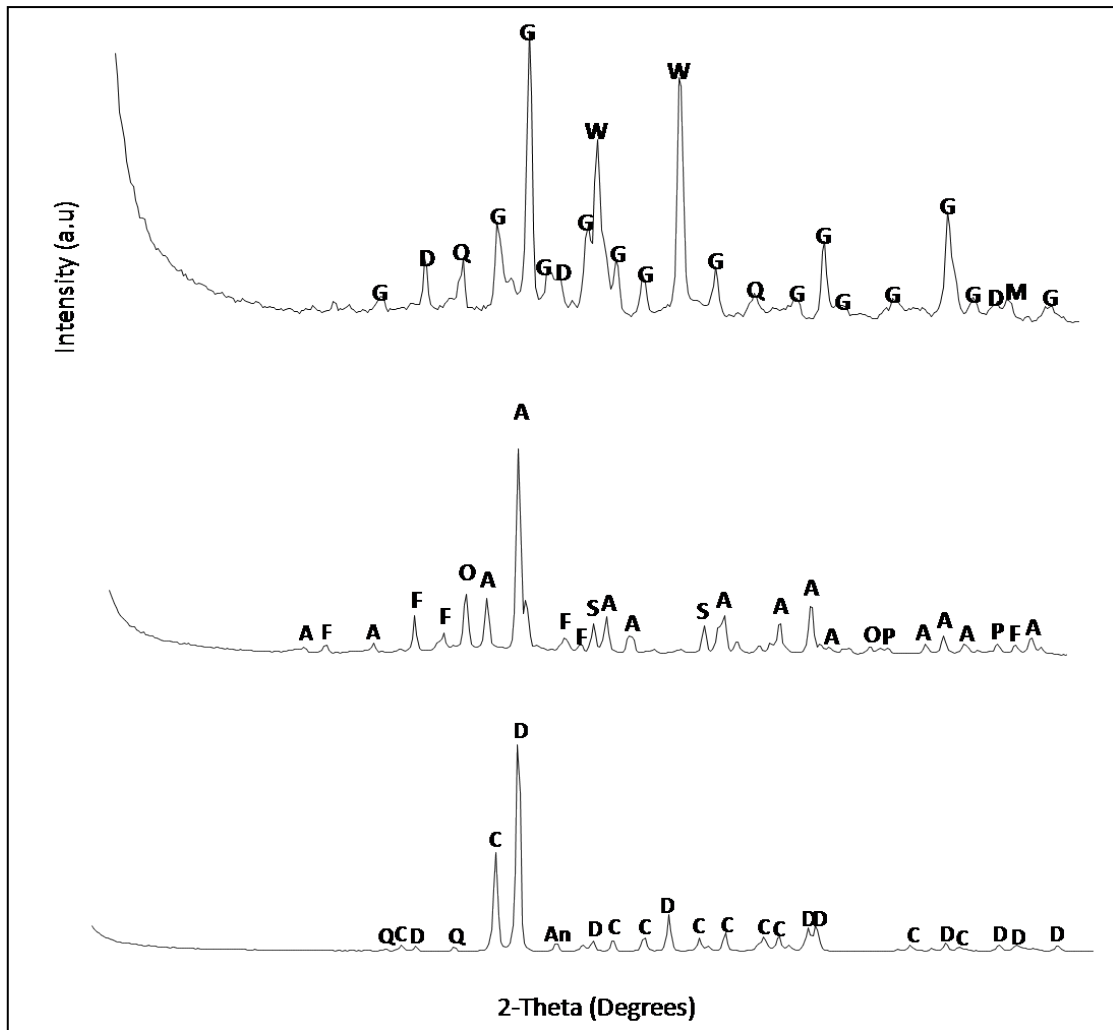
179 The slag samples consist of various primary phases including silicates, oxides, glasses, metallic
180 iron, and amorphous materials. Diverse overlapping patterns noticeable in XRD indicates
181 identifiable crystalline phases present in the legacy deposits (see Figure 3). The majority of
182 samples can be categorized as “Ca silicate-rich” with predominant melilite group minerals
183 (e.g., akermanite and gehlenite), which have been recognized as the first slag phase to
184 crystallize (37), while other minor phases such as olivine (e.g., fosterite and fayalite) is present.
185 Calcium or magnesium hydroxide or meta stable silicate phases are likely to be the most
186 reactive in slag (e.g., 61). However, relatively rapid dissolution rates have also been shown for
187 materials with a high proportion of aluminosilicates (62), such as gehlenite (56) and modelling
188 suggests high solubility in solutions that precipitate clay minerals (57). Most of the materials
189 obtained and analysed are glassy (both in appearance and composition), primarily due to high
190 SiO₂ content, which is reportedly common for legacy slags produced in Denmark, Sweden and
191 Norway (38-40). However, the Si-deficient samples from Sirhowy and Port Talbot is perhaps
192 due to insufficient temperatures or residence time in the smelting furnace which may hinder
193 complete melting of the charge (30). That way the materials are less likely to be glassy, but
194 rather resemble irregularly shaped clinker. Various sources could account for SiO₂ in the
195 samples (i.e., the ore, limestone flux, or the local geology of the area where the slag is
196 deposited).

197 Mineralogy of the samples analysed is summarized in Table 2 with the main phases given in
198 approximate order of abundance. This is useful in our understanding of the role each mineral
199 compound play in carbonation reactions as previous studies have highlighted the interaction of
200 these mineral phases with carbon dioxide under ambient conditions (53-54, 12). The calcite,
201 magnesite, ankerite, and kutnohorite identified in some samples illustrates carbonation
202 reactions have occurred after the slag was formed (41), but it is not possible to state whether
203 these reactions took place, before or after burial. The bulk chemistry of the slag is dominated
204 by CaO (6–63wt.%), SiO₂ (3–53wt.%), Al₂O₃ (9–25wt.%), and MgO (1–11 wt.%), constituting

205 between 62% and 99% mass of the samples (see Table 3); other oxides such as Na₂O (0.19–
206 2wt.%), K₂O (0.16–3wt.%), SO₃ (0.31–3wt.%), and P₂O₅ (0.27–2wt.%) are in low
207 concentrations. They are regarded as the main sequestration minerals in this study.

208 According to (31), "ores are self-fluxing, whose contribution to the slag's chemical
209 composition is minimal compared to the refractory materials found within the walls of the blast
210 furnace, made up primarily of alumina, silica, magnesia, lime and sometimes zirconia." These
211 compounds could raise concentrations of Si, Ca, Al, and Mg in the slag which is obvious in the
212 samples analysed. The high calcium content in most samples could be linked to the fluxing
213 agent (mostly limestone) used in the smelting of ores at the ironworks, rather than dolomitic
214 limestone, given the local geology of the area (42). Additionally, gehlenite is the principal
215 melilite group detected in the samples instead of akermanite, alongside other aluminosilicate
216 minerals, which explains the fairly low Mg concentrations (17). Notable variations are
217 observed in SiO₂, Al₂O₃, and MgO content, although CaO remains fairly constant in the
218 samples; these changes demonstrates varying iron-making practices across the ironworks (43).

219 The high Fe content measured in some samples is probably as a result of partial separation of
220 the Fe smelting product (or pig iron smelter feed) and the silicate slag waste at the sites (30).
221 Again this reflects the diverse and continued changing sources of ore used. The iron producers
222 of South Wales used mostly blast furnaces (fuelled by coke) to extract iron from the ore
223 between the mid-17th to late eighteenth century, that way slag is generated during ore reduction
224 to carbon (47). It is possible carbon potentially originated from the coke used, which would
225 explain the relatively high carbon content in some samples (particularly those from Abernant).
226 It appears the slag chemical composition depended on postprocessing application which
227 usually involved the manipulation of raw materials added to the bloomery/blast/puddling
228 furnaces, or fining hearth.



229

230 **Figure 3: Diffractograms of slag samples plotted for Blaenavon and Sirhowy ironworks**
 231 **with assigned mineral phases. Key: G, gehlenite; A, akermanite; D, dolomite; Q, quartz;**
 232 **C, calcite; W, wustite; F, fosterite; O, oldhamite; S, spinel; P, pyrolusite; M, magnetite.**

Sample group		British					Blaenavon		Clydach	Llwydcoed	Abernant	Varteg	Tredegar	Hirwaun	PortTalbot		
Sample ID		S1	S2	S3	S4	S5	S6	S7	S10	S11	S12	S14	S15B	S17A	S17B	S18A	S19
Name	Ideal/range composition																
Ore samples																	
Silicates																	
Quartz	SiO ₂	-	-	*	-	-	-	**	-	-	-	-	-	-	-	-	-
Akermanite	Ca ₂ MgSi ₂ O ₇	-	-	-	-	-	***	-	-	***	***	-	***	-	*	-	-
Gehlenite	Ca ₂ Al ₂ SiO ₇	-	-	-	-	-	-	-	***	-	-	*	-	-	-	-	-
Ringwoodite	(Mg,Fe) ₂ SiO ₄	***	-	-	-	-	-	-	*	-	-	-	**	-	*	-	-
Anorthite	CaAl ₂ Si ₂ O ₈	**	**	-	***	-	-	-	-	-	-	-	-	-	-	-	-
Mullite	Al ₆ Si ₂ O ₁₃	**	**	-	**	-	-	-	-	-	-	**	-	***	*	-	-
Enstatite	(Mg,Fe)SiO ₃	-	*	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Merwinite	Ca ₃ Mg(SiO ₄) ₂	-	-	-	-	-	**	-	-	-	-	-	-	-	-	-	-
Wollastonite	CaSiO ₃	-	-	-	-	-	-	-	-	-	-	-	**	-	-	-	-
Fayalite	(Fe,Mn) ₂ SiO ₄	-	-	*	-	-	-	-	-	-	-	-	-	-	-	-	-
Diopside	CaMgSi ₂ O ₆	-	-	**	-	*	-	-	-	-	-	-	-	-	***	-	-
Andradite	Ca ₃ Fe ₂ (SiO ₄) ₃	-	-	**	-	-	-	-	-	-	-	-	-	**	-	-	-
Bustamite	CaMnSi ₂ O ₆	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Larnite	Ca ₂ SiO ₄	-	-	-	-	*	-	-	-	-	-	-	-	-	-	-	-
Fosterite	Mg ₂ SiO ₄	-	-	-	-	-	-	-	-	*	**	-	-	-	-	-	-
Sillimanite	Al ₂ SiO ₅	-	-	-	-	-	-	-	-	-	*	-	-	-	-	-	-
Monticellite	CaMgSiO ₄	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Amakinite	Al ₆ Si ₂ O ₁₃	-	-	-	-	-	-	-	-	-	-	-	-	*	-	-	-
Almandine	Fe ₃ Al ₂ (SiO ₄) ₃	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Goldmanite	Ca ₃ V ₂ (SiO ₄) ₃	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	***
Carbonates																	
Dolomite	Ca(Mg,Fe)(CO ₃) ₂	-	-	-	-	-	-	***	-	-	-	-	-	-	-	-	**
Magnesite	MgCO ₃	-	-	-	-	-	***	***	-	-	-	**	-	-	-	-	-
Calcite	CaCO ₃	*	-	-	-	-	-	-	-	-	-	***	-	-	**	*	-
Ankerite	Ca(Mg,Fe(CO ₃) ₂	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Spinel	Mg(Al,Fe) ₂ O ₄	-	-	-	-	-	-	-	-	**	-	-	-	-	-	-	-

Kutnohorite	Ca(Mn,Mg)(CO ₃) ₂	-	-	-	-	-	-	-	-	-	-	*	-	-	-	-	**		
Graphite	C	-	-	-	-	-	-	-	-	-	-	-	-	-	-	***	-		
Oxides																			
Wustite	FeO	-	-	-	-	-	-	***	-	-	-	-	-	-	-	**	-	**	
Goethite	FeO(OH)	***	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
Ramsdellite/ Pyrolusite	MnO ₂	-	**	**	-	-	-	-	-	***	-	-	-	-	-	***	**	-	
Gibbsite	(Al(OH)) ₃	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
Mayenite	Ca ₁₂ Al ₁₄ O ₃₃	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
Sulfides																			
Oldhamite	(Ca,Mg,Fe)S	-	-	-	-	-	-	-	-	***	-	-	-	-	-	-	-	-	
Iron																			
Iron	Fe	***	-	-	**	*	-	-	*	-	-	-	-	-	-	-	-	-	
Cohenite	Fe ₃ C	**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
Hercynite	FeAl ₂ O ₄	-	**	-	***	-	-	-	-	-	-	-	-	-	-	-	-	***	
Hematite	Fe ₂ O ₃	-	*	-	-	*	-	-	-	-	*	-	-	-	-	-	-	-	
Magnetite	Fe ₃ O ₄	-	-	-	-	-	-	-	*	-	-	-	-	-	-	-	-	-	
Amorphous		***	***	***	***	***	***	**	***	*	***	***	*	***	***	***	***	***	
Symbols used:	- not detected	* trace phase (<6wt. % modal abundance)						** minor phase (<6-12wt. % modal abundance)						*** major phase (>12wt. % modal abundance)					
*** quantitative mineralogy suggests amorphous is a major phase in ore samples																			

Table 2: Mineral Phases Identified in the Slag samples from 9 steelworks in South Wales, UK.

Sample Group	British	Blaenavon	Clydach	Llwydcoed	Abernant	Varteg	Sirhowy	Tredegar	Hirwaun	Port Talbot
Sample ID	S1-S5	S6-S7	S9-S10	S11-S12	S14	S15B	S16B	S17A-S17B	S18A	S19
Oxides (wt. %)										
CaO	8.74	23.37	51.73	40.18	24.76	38.72	62.33	14.05	6.62	37.80
SiO ₂	29.68	16.85	28.16	36.41	32.16	34.14	3.51	52.65	39.66	5.25
MgO	2.06	2.68	3.71	3.86	1.04	6.81	11.04	-	-	-
FeO	19.61	22.72	1.01	0.91	4.17	1.21	0.24	3.18	19.96	12.10
Fe ₂ O ₃	16.94	19.63	0.88	0.79	3.60	1.05	0.21	2.76	17.24	14.69
MnO	2.56	3.23	0.40	1.37	0.06	0.85	0.00	0.48	2.98	4.28
Al ₂ O ₃	13.69	9.08	9.28	11.13	14.69	12.09	14.67	21.77	9.59	24.96
P ₂ O ₅	-	-	-	-	-	0.00	0.00	0.27	1.67	0.49
TiO	0.57	0.72	0.53	0.71	0.80	0.71	0.00	1.12	0.95	0.35
Na ₂ O	1.67	0.20	0.19	0.44	-	-	-	-	-	-
K ₂ O	1.74	0.16	1.53	1.34	1.31	1.33	-	2.86	0.92	-
SO ₃	0.95	0.55	1.05	2.18	0.98	2.63	0.31	0.31	0.40	-
C	1.79	0.81	1.53	0.68	16.43	0.46	7.69	0.55	0.00	0.08
Carbonation pot (kgCO ₂ /t)	86	210	441	345	200	364	608	107	39	294
Ewpot (kgCO ₂ /t)	159	318	674	532	310	555	912	180	66	440
Carbon stored (kgCO ₂ /t)	66	30	56	25	602	17	282	20	-	3
% Total	77	14	13	7	301	5	46	19	0	1
- Not Detected Values were normalised to 100% total										

235 **Table 3: Bulk Chemical Composition of Slag Samples determined using XRF, carbon content**
236 **through thermal analysis.**

237 Table 3 shows the level of carbonation reactions that has occurred. The amount of carbon
238 stored in each sample was used to evaluate their potential for further CO₂ sequestration. For
239 some samples, the percentage of carbon stored is almost expended given previous work on
240 similar sites (17) (e.g. the British site). The slag sample from Abernant suggests a larger carbon
241 uptake than what is possible from its chemistry, which is possibly as a result of organic carbon
242 contamination from co-disposal with coke or coal used on site.

243

244 **3.3 Assessing Carbon Capture Potential (CC_P)**

245 The maximum theoretical carbon capture potential (CC_P) for the metallurgical residues was
246 calculated using a modified version of the ‘Steinour’ equation (Equation 3 and 4) detailed in (6);
247 this uses elemental oxide composition to determine CC_P and EW_P (enhanced weathering
248 potential). For this study, we assume outputs represent maximum theoretical potential of the
249 slag materials to form carbonates and does not take into account variables such as temperature,
250 pressure, reaction rates, particle size, etc., that might affect carbonation rates. It is also presumed
251 that the carbon content in each sample is fixed from inorganic sources i.e., from
252 atmospheric/dissolved CO₂ (17,28).

253
$$CC_P = \frac{M_{CO_2}}{100} \cdot \left(\alpha \frac{CaO}{M_{CaO}} + \beta \frac{MgO}{M_{MgO}} + \gamma \frac{SO_3}{M_{SO_3}} + \delta \frac{P_2O_5}{M_{P_2O_5}} \right) \cdot 10^3 \quad [3]$$

254
$$EW_P = \frac{M_{CO_2}}{100} \cdot \left(\alpha \frac{CaO}{M_{CaO}} + \beta \frac{MgO}{M_{MgO}} + \epsilon \frac{Na_2O}{M_{Na_2O}} + \gamma \frac{K_2O}{M_{K_2O}} + \theta \frac{SO_3}{M_{SO_3}} + \delta \frac{P_2O_5}{M_{P_2O_5}} \right) \cdot 10^3 \cdot \eta \quad [4]$$

255 where CaO, MgO, SO₃, P₂O₅, Na₂O and K₂O, are the elemental concentrations of Ca, Mg, S, P, Na and K,
 256 expressed as oxides, M is the molecular mass of those oxides; coefficients α, β, ε, θ (equal to +1), γ (equal to -1),
 257 and δ (equal to -2) consider the relative contribution of each oxide; and η is molar ratio of CO₂ to divalent cation
 258 sequestered during enhanced weathering, we have used η = 1.5 in this study, which is a conservative global
 259 average. Equations (3) and (4) imply that the potential is reduced by the presence of sulphur and phosphorus within
 260 the material.

261

262 It is estimated that between 0 – 77% of total carbonation potential has been realised in the
 263 samples (equating to maximum potential between 40 and 608 kg CO₂ /t has been removed from
 264 the atmosphere by direct carbonation). This is based on volume of slag materials (i.e., 58.5 Mt)
 265 generated by ten ironworks as well as their chemical composition, typically containing 6–62%
 266 CaO, 1–11% MgO for blast furnace slag, and ~38% CaO with a depleted MgO-phase for steel
 267 slag (i.e., Port Talbot sample). The final carbon sequestration potential of what is left is
 268 represented in Table 4.

269

Ironworks	CC _P (Mt)	EW _P (Mt)
British	0.02	0.03
Sirhowy	0.05	0.08
Hirwaun	0.02	0.03
Abernant	0.01	0.02
Llwydcoed	0.13	0.2
Port Talbot	16.5	24.6
Blaenavon	0.18	0.28
Varteg	0.03	0.04
Tredegar	0.03	0.05
Clydach	0.05	0.08

270

271 **Table 4: Calculated CO₂ Capture Potentials Using a modification of the Steinhilber Equation for**
 272 **Slag Materials from the 10 Ironworks Mapped Based on Final Carbonation.**
 273

274 The current work suggests that there is a capture potential of up to 17 MtCO₂ of the slag
 275 materials by direct carbonation and perhaps 25 MtCO₂ via enhanced weathering which
 276 represents a potential carbon sink for future emissions. These potentials are dominated by
 277 resources at the Port Talbot steelworks, that remains operational. Removing billions of tonnes of
 278 CO₂ from the atmosphere every year will require methods of incentivisation (e.g., carbon tax, or
 279 removal/offset markets), which may be equivalent to scalable permanent methods such as direct
 280 air capture, that may reduce to approximately \$100/tCO₂ (44). As such, the ~17–25 Mt CO₂

281 capture potential of South Wales may constitute a \$2-3 billion resource, similar in size to the
282 carbon sequestration potential of Welsh forests (45).

283

284 **3.4 Limitations**

285 While the overall elemental composition of the slag illustrates maximum theoretical CO₂
286 capture potential, a commercial scheme for ambient carbonation has yet to be developed. The
287 results from this work represent a small CO₂ capture percentage when considering the volume
288 of silicate minerals remaining in stockpiles or buried slags. We have not covered the factors
289 that could limit carbonation reactions, however further research is required to assess the wider
290 carbon balances of reacting atmospheric CO₂ with slag materials.

291

292 **3.5 Conclusion**

293 The estimated capture potential of the samples by direct carbonation is up to 17 MtCO₂ and
294 25 MtCO₂ through enhanced weathering, but the amount to which each site has realised this is
295 variable (0 – 77%). This paper presents the range of minerals present in legacy deposits and how
296 the bulk chemistry can be used to assess carbon capture potential. The values from the legacy
297 sites is relatively small compared to that of Port Talbot which is apparent, due to large quantities
298 of waste materials produced annually and it remains operational. As some sites can reach high
299 degrees of carbonation, this suggests that passive carbonation reactions occur under ambient
300 conditions. Thus, these materials can either serve as short-lived feedstock to reduce emissions
301 close to zero over the next few decades or potentially form a resource in other waste material
302 streams.

303 We believe most of these materials are buried or stockpiled underground and that limited mass
304 transfer of CO₂ limits sequestration. However, if they are managed in a system that maximises
305 atmospheric exposure, it will enhance the process. Although, this has its accompanying
306 challenges because some slag can produce potentially harmful leachates enriched with metals.
307 From previous work, it was observed that alterations in the physical, microstructure and
308 chemical properties of slag can result in the selective release of non-carbonated products, of
309 which the generation of calcium carbonate is usually linked with volume expansion which
310 decreases porosity and promotes the retention of contaminants (28,55). Due to the presence of
311 calcium silicates, carbonates, and oxides in iron and steel slag, they become alkaline with
312 exceptional acid-neutralizing potential (22). The percentage of elements likely to pose
313 environmental concerns is within the range of typical iron and steel slag previously reported.
314 The concentration of zinc, copper, lead, cobalt, and nickel across the ironworks fall within
315 acceptable levels for both residential and industrial use, compared to arsenic, chromium, and
316 manganese showing higher values (see Supplementary file). Nonetheless, further study is
317 required to appraise the impact of accelerated carbonation on these elements in the environment.

318 The iron and steel industry can play an important role in preventing climate change by storing
319 carbon permanently either by forming mineral carbonates or dissolved bicarbonate ions that will
320 eventually be released into the ocean.

321

322 **Acknowledgements**

323 The authors thank Anthony Oldroyd and Iain McDonald (Cardiff University) for help with XRD
324 and XRF analysis, and Marco Santonastaso (School of Engineering, Cardiff University) for help
325 with Total Carbon (TC) and Total Inorganic Carbon (TIC) analysis, respectively.

326 Funding: This work is part of the UK Greenhouse Gas Removal Programme with the Natural
327 Environment Research Council (NERC), the Engineering and Physical Sciences Research
328 Council (EPSRC), Economic & Social Research Council (ESRC), and the Department for
329 Business, Energy & Industrial Strategy (BEIS), under Grant NE/P019943/1. Also, This work
330 was supported by the Chartered Institute of Wastes Management (CIWM) as bursary for an
331 MSc project.

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