

Low Cost Organic Adsorbents for Elemental Mercury Removal from Lignite Flue Gas

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1 Low Cost Organic Adsorbents for elemental mercury removal from lignite flue gas

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9 Abstract

10 Based on the knowledge of the impact of aspects tied to and utilised technology and raw materials on the
11 speciation of mercury in the flue gas, the authors have undertaken the task of examining the process of sorption
12 of mercury from flue gases using fine-grained organic materials. The main objectives of this study was to
13 recommend a Low-Cost Organic Adsorbent such as coke dust CD, Corn straw char CS-400, Brominated Corn
14 straw char CS-400-Br, Rubber Char RC-600 and Granulated Rubber Char GRC-600 to efficiently substitute
15 expensive dust-sized activated carbon. The study covered combustion of lignite from Polish field. The
16 experiment was conducted at temperatures reflecting conditions inside a flue gas purification installation. The
17 average mercury content in Polish lignite was 465 µg·kg⁻¹. The concentration of mercury in flue gases emitted
18 into the atmosphere was 17.8 µg·m⁻³. The study analysed five low-cost sorbents with the average achieved
19 efficiency of mercury removal from 18.3 to 96.1% for lignite combustion depending on the flue gas
20 temperature. Two of the tested sorbents exhibited good potential for Hg⁰ into Hg²⁺ oxidation, resulting in
21 enhanced mercury removal from the flue.

22
23 **Keywords:** *coal combustion, lignite, anthropogenic emission, mercury removal, flue gases purification, low-*
24 *cost adsorbents*

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30 *in pulverised coal-fired boiler and its flue gas cleanup with use of regression models and neural networks" (grant*
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33 **1. Introduction**

34 Ecotoxic elements, especially mercury, are particularly hazardous substances among pollutants with no
35 physiological relevance for living organisms. It is generally known that mercury exposure can inflict various health
36 issues, especially in neurological, immunological, behavioural and sensory aspect (Sloane et al. 2009, Rice et al.
37 2014). The mentioned afflictions were diagnosed in consumers of contaminated fish (Minamata Disease in Japan)
38 and crops (Iraq, Guatemala and Russia). Due to its adverse impact on health, as well as its global distribution, long
39 atmospheric lifetime and propensity for deposition in the aquatic environment and in living tissue, the US
40 Environmental Protection Agency (US EPA) (U.S. Environmental Protection Agency 2008) have classified
41 mercury and its compounds as a severe air quality threat. Coal combustion is considered as one of the most
42 significant anthropogenic sources of mercury release due to growing worldwide need for energy (U.S.
43 Environmental Protection Agency 2008, Wang et al. 2016, Xu et al. 2018a). Therefore to avoid adverse harmful
44 effects to the respiratory, nervous and immune system due to element exposition, it is crucial to decrease to
45 atmospheric mercury emission. Conducted research confirms the severe influence on the environment and warrants
46 worldwide actions for the reduction of the emission. In March 2005, the US Environmental Protection Agency
47 (EPA) announced the Mercury and Air Toxics Standards (MATS) to reduce mercury emissions from coal-fired
48 power plants (Ren et al. 2014, Yang et al. 2017). Moreover, 128 countries signed the Minamata Convention on
49 Mercury, which took effect on 16 August 2017 (Zhang et al. 2016a).

50 In Poland, solid fuels combustion, mainly coal constitutes as dominating source of mercury emission,
51 exceeding 80% of the country share. These data are backed by data published by European Environment Agency,
52 which outlines Poland contribution in 2017 to be as high as 16.9% of total mercury release of among EEA33
53 countries, which further accentuates the scale of the problem (Li et al. 2016).

54 To pertain to the issue, UE legislature requires monitoring of atmospheric mercury release with increased
55 emission standards for this element pollution from the energy sector (European Environment Agency 2020),
56 among others that is aimed to take effect in the second half of 2021.

57 Currently, under the adoption of 2010/75/UE Directive (IED – Industrial Emissions Directive) that comprise
58 the permitted industrial emissions, polish energy sector introduces Best Available Technologies (BAT) methods
59 aimed at improving ecotoxic elements capture, including mercury, often connected with other flue gas cleanup
60 systems (e.g. dust and sulfur/nitrogen oxides elimination). The concept of element removal system coupled with
61 established cleanup technologies is known as passive removal methods and require mercury monitoring for
62 successful implementation (Guidance on BAT 2015). The IE Directive enforces the use of BAT technologies

63 introduced by 2017 issue with a permissible emission limit of $1-10 \mu\text{g}\cdot\text{Nm}^{-3}$, depending on fuel used and plant size
64 (Guidance on BAT 2015, European Environment Agency 2020). At present, the vast majority of units in polish
65 powerplants does not meet the BAT-defined standards for mercury, which impose the need for technological
66 advance. As new emerging technologies such as clean coal technologies or deep desulfurization and
67 denitrogenation do not meet the European standards for mercury emission and auxiliary means of mercury removal
68 are needed in order to prevent the powerplant closure due to legislation non-compliance. Among techniques of
69 standalone mercury removal systems, injection of powdered activated carbon (PAC) to flue gasses due to the high
70 specific surface of the material is commonly utilized in the United States of America (Galbreath et al. 2000, BAT
71 2017).

72 The efficiency of mercury removal with powdered sorbent injection depends on both physical and chemical
73 properties of the material, flue gas temperature (reversely proportional, too high-temperature results in sorption
74 decrease) and flue gas constituents (presence of halogen compounds and sulfuric trioxide SO_3). Furthermore,
75 mercury speciation plays a vital role as Hg^{2+} form of mercury has good affinity for sorbents capture, whereas Hg^0
76 is practically not adsorbed. Therefore oxidation of the mercury in order to increase the share of Hg^{2+} is an
77 established practice. The oxidation can be achieved by chemical methods (chlorination or bromination) or physical
78 (UV radiation) (Favale et al. 2013). The general performance is also influenced by the intake of injected sorbent,
79 that is measured by the atomic ratio of carbon atoms in sorbent to mercury in the flue (C:Hg), level of powder-flue
80 homogenization, contact time and efficiency of dedusting that collects the sorbent (Srivastava et al. 2006, Olson
81 et al. 2009, Liu et al. 2014a).

82 The main shortcoming of activated carbon sorbent usage is the material price, which results in global research
83 (Charpentreau et al. 2007, Seneviratne et al. 2007, Zhang et al. 2016b, Zhao et al. 2017), aimed at the search for
84 new low-cost adsorbents with comparable sorptive properties that can be used as a replacement (Low-Cost
85 Adsorbents). The authors suggest that the use of waste materials with no industrial application can be the most
86 beneficial in both economic and environmental terms (Liu et al. 2014). For example, Fuente-Cuesta et al. 2012
87 have developed waste-derived sorbents from biomass and brominated flame retarded plastic for mercury removal
88 from coal-fired flue gas. Xu et al. 2018b proposed coal fly ash as low-cost material. Authors in the studies (Marczak
89 et al. 2019, Burmistrz et al. 2014) analysed six low-cost sorbents with the average achieved efficiency of mercury
90 removal from 30.6 to 92.9% for sub-bituminous coal and 22.8 to 80.3% for lignite combustion. One of the main
91 objectives of this study was to recommend a low-cost organic sorbent such as coke dust to efficiently substitute
92 expensive activated carbon for mercury removal from flue gas. Other researchers have been attempting to develop

93 alternative low-cost yet efficient adsorbents utilizing agricultural and industrial wastes (Tareq et al. 2019, Jewiarz
 94 et al. 2020, Szufa et al. 2020a). Biochars play a significant role in addressing the current demands of adsorbents
 95 for various applications (Szufa et al. 2020b, Dzikuć et al. 2020). Initial research on specially prepared chars from
 96 rubber wastes was proven beneficial for both captures of mercury (Li et al. 2015) and other ecotoxic elements
 97 capture (Chan et al 2012, Martínez et al. 2013, Selbes et al. 2015). Conducted studies suggest (Manirajah et al.
 98 2019) that activated carbon produced from rubber char in wastewater cleanup of phenolic compounds, heavy
 99 metals (Dimpe et al. 2017), disposed pharmaceuticals (Styszko et al.2017) and pesticides (Gupta et al. 2011).
 100 Taking into account the amount of mercury released fro energy sector (UN Environment 2019), the research was
 101 organized in order to determine the potential of rubber char as a sorbent for capture mercury from flue gas in
 102 different temperatures and comparison to other possible adsorbents as an alternative for expensive activated
 103 carbons.

104 The purpose of this paper was application of dust-sized sorbents for reduction of mercury emission from flue
 105 gas. The main objectives of this study was to recommend a low-cost organic asorbent such as coke dust CD, Corn
 106 straw char CS-400, Brominated Corn straw char CS-400-Br, Rubber Char RC-600 and Granulated Rubber Char
 107 GRC-600 to efficiently substitute expensive dust-sized activated carbon. The study covered combustion of lignite
 108 from Polish field. The experiment was conducted at temperatures reflecting conditions inside a flue gas purification
 109 installation.

110 2. Material and methods

111 2.1. Sorbents characterisation

112 With the purpose to compare the efficiency of sorbents, sorption studies of mercury in flue gases consisted of 5
 113 types of low-cost organic sorbents in three different forms: coke-derived char dust (CD), biomass chars (CS, CS-
 114 Br) and rubber-derived chars (RC, GRC). The commercially available activated carbon (AC) was included as
 115 industrial flue gas mercury sorbent of choice for comparative reasons. Sorbents characteristics are shown in Table
 116 1.

117 **Table 1.** Description of sorbents used in the study

No.	Sorbent	Symbol	Material origin
1	Activated carbon	AC	Commercial activated carbon, dedicated i.a. for gas-phase mercury removal. It was formed in the process of coal

			carbonisation and subsequent thermal activation of the obtained structure.
2	Coke dust	CD	By-product of large scale coke production. The dust is obtained during coke dry-cooling process, hauling and sorting.
3	Corn straw char	CS-400	Solid product (char) of corn straw torrefaction process using superheated steam at 400 °C.
4	Brominated Corn straw char	CS-400-Br	Solid product (char) of corn straw torrefaction process with the addition of bromine; blend char / Br ₂ was prepared at ratio 5:1
5	Rubber Char	RC-600	Solid product (char) of car tyre pyrolysis at 600 °C . The material was derived from industrial installations in which intact tires are subjected to high temperatures.
6	Granulated Rubber Char	GRC-600	Solid product (char) of granulated car tyre (2–4 mm particle size) pyrolysis at 600 °C.

118

119

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121 2.2. Sorbent analysis

122 The scope of sorbent analysis listed in section 2.1 includes:

123 (i) Proximate and ultimate analysis in accordance with the ISO standard (ISO 17246:2010, ISO 17247:2013).

124 (ii) Determination of chlorine content evaluated as chlorine anion content in water solution using a direct
125 reading spectrophotometer (DR/2000 HACH). A sample was combusted in AC-350 bomb calorimeter
126 (LECO) with Eschka mixture – in accordance with the ISO standard (Solid Mineral Fuels).

127 (iii) Mercury content analysis by thermal decomposition, amalgamation, and atomic absorption
128 spectrophotometry (DMA-80 Direct mercury analyser; Milestone Connect).

129 (iv) Analysis of particle size of analysed sorbents by ISO standard (ISO 728:1995).

130 (v) The porous texture of all samples was analysed using nitrogen adsorption/desorption at 77 K using
131 Autosorb[®]-1-C (Quantachrome Instruments, USA). Before measurements were made, all samples were
132 degassed at 473K for 12 h under vacuum. Interpretation of textural properties: specific surface area by

133 Brunauer-Emmett-Teller (BET), the volume of micropore calculated by Dubinin-Raduszkiewicz method,
134 the volume of mesopore calculated by Barrett, Joyner and Halenda method (BJH) and total volume of pores
135 was carried out in accordance with the recommendations of the following standards: (NIST 2006, ISO
136 15901-2: 2006, ISO 15901-3: 2007, ISO 9277: 2010).

137 (vi) Bromine content with X-ray spectrometry with wavelength dispersion in PROMUS II sequential
138 spectrometer (Rigaku) using a bespoke research procedure (ISO 5069-2: 1983).

139 Values of these parameters were determined for the air-dried basis of the sample.

140

141 2.3. Mercury adsorption system

142 Figure 1 shows a schematic of a stand for measuring mercury sorption from flue gases generated by the combustion
143 of solid fuels. The bench-scale measurement setup consists of tube furnace (3) with temperature and gas flow
144 regulation with quartz tube (4) as the combustion chamber, gas source (1) and sorbent holder (8). Utilised
145 equipment allows for controllable gas heating in specified points (9). The measurement procedure consists of air-
146 fueled coal sample (5) combustion and analysis of flue-gas mercury captured by the sorbent.

147 The experiment was conducted with predetermined temperature conditions for sample combustion and sorbent
148 temperature with similar combustion times and airflow. The 1g of the sample was positioned in a ceramic boat-
149 shaped crucible and progressively transported to the centre of combustion zone with a metal rod (2). The
150 standardised measurement time after the crucible introduction to the centre of the combustion zone determined for
151 20 minutes. More detailed description of the experimental conditions presented in the publication (Marczak et al.
152 2019). Additionally, a sorbent trap (7) is placed after a tested sorbent. The trap measures the concentration of
153 mercury emitted from the combustion gases into the atmosphere.

154

155

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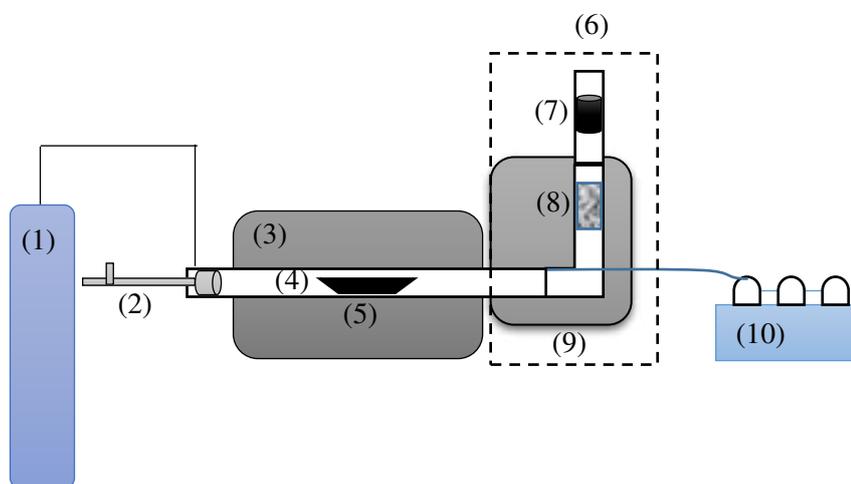
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162 **Figure 1.** (1) gas source - air, (2) metal rod, (3) tube furnace, (4) quartz tube, (5) coal sample, (6) flue mercury
163 adsorption device (7) sorbent trap, (8) sorbent holder, (9) furnace for heating of flue gases, (10) ice bath with
164 series of impingers for measurement of mercury speciation.

165 Before the test, mercury content was measured in both fuel and sorbent using the methods described in
166 section 2.2. After the procedure, mercury content was analysed in ash, sorbent and the sorbent trap.

167 For the combustion process lignite was used and underwent ultimate and proximate analysis, with the additional
168 steps of mercury and chlorine determination in accordance with the methodology described in section 2.2.

169 Sample preparation was applied in accordance with respective ISO standards (ASTM D6784-02). The
170 characteristic of combusted coal is shown in section 3.3.

171 2.3.1 Mercury speciation testing

172 The Ontario Hydro Method (OHM) (ASTM D6784-02) was used to determine mercury speciation in flue
173 gas samples. For the period of OHM setup mercury speciation ascertainment, the flue mercury adsorption device
174 (Fig. 1. point 6) was disengaged. Flue gases were flowing by a series of impingers immersed in an ice bath (Fig.1.
175 point 10). The first three impingers containing 1 N potassium chloride solution were connected to absorb oxidised
176 mercury (Hg^{2+}). The fourth impinger containing acidified hydrogen peroxide was used to absorb elemental
177 mercury, and elemental mercury was mainly captured in the fifth, sixth and seventh impingers which contained
178 the solutions of acidified potassium permanganate. Also, an eighth impinger containing silica gel was provided to
179 ensure that the flue gas was thoroughly dried before it left the impinger train.

180

181 2.4 Methodology of mercury adsorption

182 To check the reliability of the tests performed, the balance of mercury in the laboratory installation was
183 calculated for each experiment in accordance with the following model:

$$184 \quad m_c \cdot C_0 - m_{ash} \cdot C_{ash} = m_{sorb} \cdot (C''_{Hg} - C'_{Hg}) + m_{trap} \cdot (C''_{trap} - C'_{trap}) \quad (1)$$

185 where: m_c – mass of combusted coal [kg],

186 C_0 – mercury content in coal [$\mu g \cdot kg^{-1}$],

187 m_{ash} – mass of ash from coal combustion [kg],

188 C_{ash} – mercury content in the ash remaining after combustion of coal [kg],

189 m_{sorb} – weight of tested sorbent [kg],

190 C'_{Hg}, C''_{Hg} – mercury concentrations in sorbent before and after sorption [$\mu g \cdot kg^{-1}$],

191 m_{trap} – mass of sorbent trap [kg],

192 $C'_{\text{trap}}, C''_{\text{trap}}$ – mercury concentrations in the trap before and after the experiment [$\mu\text{g}\cdot\text{kg}^{-1}$].
 193 On the basis of obtained data and measurements flue gas mercury concentration before and after adsorption the
 194 Hg adsorption capacity (q , $\mu\text{g}\cdot\text{g}^{-1}$) of the sorbents samples were calculated by:

$$195 \quad q = \frac{Q_{fg}}{m_{\text{sorb}}} \int_0^t (C''_{fg} - C'_{fg}) dt \quad (2)$$

196 where: Q_{fg} – gas flow rate ($\text{m}^3\cdot\text{min}^{-1}$),

197 C''_{fg}, C'_{fg} – inlet and outlet Hg concentrations in flue gases ($\mu\text{g}\cdot\text{Nm}^{-3}$) at combustion time t (min).

198 Additionally, mercury removal efficiency of tested sorbents (MR, %) was calculated by:

$$199 \quad MR = \frac{c''_{Hg} - c'_{Hg}}{c_0 - c_{\text{ash}}} \cdot 100\% \quad (3)$$

200 3. Results and discussion

201 3.1. Sorbent characteristics

202 AC contained $6.1 \mu\text{g Hg}\cdot\text{kg}^{-1}$. Likewise, CD and CS-400, contained relatively small quantities of mercury:
 203 8.9 and $2.7 \mu\text{g}\cdot\text{kg}^{-1}$, respectively (Table 2). CD and AC are materials obtained in the carbonisation process.
 204 Therefore they also contain minimal quantities of volatiles: approximately 3.2 wt% for CD and approximately
 205 15% for AC. RC-600 and GRC-600 have high mercury content, at 158 and $73 \mu\text{g}\cdot\text{kg}^{-1}$ and ash (19.8 and 19.6%).
 206 Commercial activated carbon (AC) and rubber waste chars (RC-600, GRC-600) had from 3.5 to 18 more higher
 207 sulphur content than other examined sorbents. Sorbents had bromine content in the range of 50 -730 ppm.

208 **Table 2.** Properties of analysed sorbents

Sorbent	Proximate analysis (wt %)			Ultimate analysis (wt %)			(wt %)		($\mu\text{g}\cdot\text{kg}^{-1}$)
	M _{ad}	V _{ad}	A _{ad}	C _{ad}	H _{ad}	S _{ad}	Br _{ad}	Cl _{ad}	Hg _{ad}
AC	9.2	15.09	26.2	59.5	1.45	2.11	0.017	0.021	6.1
CD	0.4	3.19	9.8	85.0	0.16	0.59	0.014	0.023	8.9
CS-400	2.6	23.7	22.1	61.7	3.20	0.15	0.005	0.037	2.7
CS-400-Br	-	-	-	-	-	-	0.033	-	2.9
RC-600	1.3	3.2	19.8	75.4	0.91	2.55	0.068	0.063	158.1
GRC-600	2.3	3.2	19.6	76.3	0.95	2.66	0.073	0.075	73.0

209
 210 Coke and rubber waste chars are macroporous materials with a moderately developed mesoporous and poor
 211 microporous structure (Table 3). Specific surface (S_{BET}) of CD, RC-600 and GRC-600 amounts to several dozen
 212 $\text{m}^2\cdot\text{g}^{-1}$, but in AC this parameter is $670.5 \text{m}^2\cdot\text{g}^{-1}$. The specific surface area of Corn Staw Char was $4.8 \text{m}^2\cdot\text{g}^{-1}$.

213 However, mesoporous structure is essential parameter for mercury adsorption, as due to particle size of mercury
 214 mesopores are considered to be dominant areas of its deposition. RC-600 and GRC-600 sorbents had the highest
 215 mesoporous surface among the analysed samples: $0.38 \text{ cm}^3 \cdot \text{g}^{-1}$ and $0.17 \text{ cm}^3 \cdot \text{g}^{-1}$, respectively.

216

217 **Table 3.** Parameters of porous structure for analysed sorbents, based on nitrogen vapour sorption/desorption
 218 isotherms measured at 77 K.

Sample	BET surface area ($\text{m}^2 \cdot \text{g}^{-1}$)	Micropore surface area ($\text{cm}^3 \cdot \text{g}^{-1}$)	Mesopore volume ($\text{cm}^3 \cdot \text{g}^{-1}$)	Total pore ($\text{cm}^3 \cdot \text{g}^{-1}$)
AC	670.5	0.307	0.055	—
CD	24.3	0.006	0.009	0.018
CS - 400	4.8	0.001	0.010	0.023
CS – 400 - Br	4.6	0.001	0.008	0.018
RC-600	70.3	0.025	0.380	0.396
GRC-600	74.7	0.023	0.171	0.239

219

220 3.2. Properties of lignite

221 Lignite contained an average of $465 \mu\text{g} \cdot \text{kg}^{-1}$ of mercury (Table 4). The average content of halogens
 222 (chlorine and bromine), which are a supporting factor in the oxidation of mercury from Hg^0 to Hg^{2+} in analysed
 223 lignite was equal to 30 ppm Cl and 3.9 ppm Br. The average sulphur content for lignite was 1.8 wt%. Lignite
 224 contained 23.7 wt% ash, 41.20 wt% volatiles and 12.9 wt% moisture.

225 **Table 4.** Characteristic of coal used in the experiment

Coal	Proximate analysis (wt %)			Ultimate analysis (wt %)			(wt %)			(ppm)	($\mu\text{g} \cdot \text{kg}^{-1}$)
	M_{ad}	V_{ad}	A_{ad}	C_{ad}	H_{ad}	S_{ad}	Ca_{ad}	Br_{ad}	Cl_{ad}	Hg_{ad}	
lignite	12.9	41.20	23.7	43.5	4.90	1.80	2.42	3.9	30	465.0	

226

227 3.3 Determination mercury speciation in flue gases: bench-scale

228 In view of the way the experiment is conducted, ash was a solid residue after coal combustion, and nearly
 229 all the mercury contained in the coal passed to the flue gas in gaseous forms (Hg^0 and Hg^{2+}). Only 2% remained
 230 in the ashes in the form Hg_p . The share of Hg^0 in the analysed flow, reaching 70% is relatively high. Such $\text{Hg}^0:\text{Hg}^{2+}$
 231 ratio (5:2) is determined by the chemical composition of the fuel. Lignite is characterised by low content of chlorine

232 and bromine (Table 4), which lead to oxidation of Hg^0 to Hg^{2+} , but the effect can be partially stopped by the
233 relatively high presence of calcium (2.42 wt%), which is capable of chemical deactivation of chlorine due to their
234 chemical affinity (Zhang et al. 2008). Resulting fuel composition determines the share of Hg^{2+} to be no more
235 significant than 28%. Due to behavioural differences between both forms, Hg^0 is generally more difficult to remove
236 from flue gas by adsorption process. Therefore the determining factor in the selection of sorbent and process
237 condition for mercury removal should be the chemical composition of the sorbents as well as sorption temperature.

238

239

240

241 **3.4 Hg adsorption performance during combustion of lignite**

242 Table 5 shows the Hg adsorption capacity (q) of tested sorbents and the effectiveness of mercury removal
243 from flue gas (MR) for all tested process temperatures. At the lowest temperature - 95 °C, commercial activated
244 carbon, currently used in active flue gas mercury removal methods was the most efficient, removing the mercury
245 almost entirely. CD also shows high mercury sorption efficiency – at 93.8%. RC-600 and GRC-600 sorbents
246 ensured mercury removal during combustion of lignite at 81.5 and 65.7%, respectively. Observations have shown
247 Corn staw char to be the worst sorptive material during coal combustion, decreasing the concentration of mercury
248 in flue gas by only 32.4%. CS-400-Br was more efficient in mercury removal than CS-400.

249 In the combustion process of lignite with a mercury content of $465 \mu\text{g}\cdot\text{kg}^{-1}$, raw flue gas concentration of
250 mercury amounted to $17.8 \mu\text{g}\cdot\text{m}^{-3}$. Most of the mercury from the combustion of lignite was removed by AC and
251 CD. After completing the process of coal combustion, Hg adsorption capacity was 102.6 and $100.2 \mu\text{g}\cdot\text{kg}^{-1}$,
252 respectively for AC and CD. The concentration of mercury in flue gas from lignite was reduced to a value of 0.7
253 and $1.1 \mu\text{g}\cdot\text{g}^{-1}$. Application of Corn Staw Chars resulted in incomplete mercury removal. Hg adsorption capacity
254 for CS-400 and CS-400-Br was 34.3 and $53.4 \mu\text{g}\cdot\text{kg}^{-1}$, which enabled a reduction of mercury concentration in the
255 flue gas to a value of 12.1 and $8.9 \mu\text{g}\cdot\text{m}^{-3}$. Low sorptive capabilities of CS-400 can be attributed to its specific
256 surface of $4.8 \text{m}^2\cdot\text{g}^{-1}$. More value of q was obtained for Rubber Chars, mercury content determined in the flue did
257 not exceed $87.0 \mu\text{g}\cdot\text{kg}^{-1}$ for RC-600 and $70.2 \mu\text{g}\cdot\text{kg}^{-1}$ and GRC-600. The result was a reduction in the concentration
258 of mercury in flue gas, respectively, to 3.3 and $6.1 \mu\text{g}\cdot\text{m}^{-3}$.

259

260 Effect of flue gas temperature on mercury sorption ability

261 The all tested sorbents were chosen to explore the effect of adsorption temperature on Hg removal
 262 performance. As shown in Table 5, the average mercury removal efficiency for AC, CD and CS-400 it decreased
 263 with increasing temperature, was the highest at 95 °C and the lowest at 185 °C. The change in adsorption
 264 temperature had a different effect on the RC-600, GRC-600 and CS-400-Br sorbents. The average mercury
 265 removal efficiency increased as the adsorption temperature increased, the lowest was at 95 °C and the highest at
 266 185 °C.

267 Average mercury concentration in flue gases during lignite combustion was $17.8 \mu\text{g}\cdot\text{m}^{-3}$. Average mercury
 268 concentration in flue gases after adsorption process in the temperature range (95 – 185 °C) was respectively for
 269 AC, CD, CS-400: $1.1 - 5.8 \mu\text{g}\cdot\text{m}^{-3}$, $0.7 - 3.8 \mu\text{g}\cdot\text{m}^{-3}$, $12.1 - 14.6 \mu\text{g}\cdot\text{m}^{-3}$. CS-400 showed poor Hg removal
 270 performance at all tested temperatures. On the other hand, brominated CS-400-Br showed an increase of Hg
 271 removal efficiency with increasing temperature, similarly to RC-600 and GRC-600.

272

273 **Table 5.** Effectiveness of sorbents for mercury sorption

Sorbent	Temperature	q	MR
	°C	$\mu\text{g}\cdot\text{g}^{-1}$	%
AC	95	102,6	96.1
AC	125	94,2	88.2
AC	155	91,8	86.0
AC	185	84,0	78.7
CD	95	100.2	93.8
CD	125	95.4	89.3
CD	155	73.2	68.5
CD	185	72,0	67.4
CS-400	95	34.3	32.1
CS-400	125	29.4	27.5
CS-400	155	24.2	22.7
CS-400	185	19.5	18.3
CS-400-Br	95	53.4	50.1
CS-400-Br	125	55.2	51.7
CS-400-Br	155	58.8	55.1

CS-400-Br	185	61,2	57,3
RC-600	95	87,0	81,5
RC-600	125	93,6	87,6
RC-600	155	99,0	92,7
RC-600	185	101,7	95,2
GRC-600	95	70,2	65,7
GRC-600	125	84,0	78,7
GRC-600	155	88,8	83,1
GRC-600	185	97,2	91,0

274 The increase of Hg removal efficiency was mainly because the reactants could obtain more kinetic energy
275 at high temperature (Yang et al. 2019). However, the deciding factor for Hg removal efficiency can be the share
276 of Hg⁰ in the flue. For AC, CD and CS-400 the Hg removal efficiency decreased with further increasing the
277 adsorption temperature and for CS-400-Br, RC-600, GRC-600 - was increased.

278 It was speculated that high adsorption temperature cause Hg desorption from the sorbent surface. However, the
279 experiment showed that sorption temperature increase followed with a higher share of Hg⁰ in the flow. The higher
280 hg removal of Cs-400-Br, RC-600, GRC-600 with the increase in temperature can be appointed to higher Br and
281 Cl intrinsic content.

282

283 3.4.1 Effect of bromine content in the sorbent on Hg⁰ oxidation

284 Flue gases from lignite combustion are determined by the high share of unoxidised Hg⁰ mercury. It is a
285 volatile constituent of the flue with its ratio versus Hg²⁺ increasing with sorption temperature. With increase in the
286 Hg⁰ content, the average Hg removal efficiency for AC, CD and CS followed the decreasing order: 95°C > 125°C
287 > 155°C > 185°C.

288 AC, CD and CS-400 sorbent was determined by low bromine content which resulted in lesser Hg²⁺ and therefore
289 higher Hg⁰ amounts. The surface of the sorbent allows for Hg²⁺ adsorption, which can explain the pattern of higher
290 sorptive properties of AC, CD and CS-400 in higher sorption temperatures as the Hg⁰ content increases.

291

292 On the other hand, RC-600, GRC-600 and CS-400-Br were characterised by 2-14 times higher bromine content.
293 Sorbent-present Br oxidised Hg⁰ in the flue to Hg²⁺ form in higher sorption temperatures, which obtained higher
294 Hg²⁺ concentrations confirms, along with higher removal capabilities (Yang et al. 2019). The average Hg removal

295 for RC-600, GRC-600 and CS-400-Br followed the ascending order: $95^{\circ}\text{C} < 125^{\circ}\text{C} < 155^{\circ}\text{C} < 185^{\circ}\text{C}$. The
296 proposed Hg^0 oxidation and Hg^{2+} adsorption mechanism on RC-600, GRC-600 and CS-400-Br surfaces were
297 presented in Figure 2. Hg^0 was first oxidation at the activated site on the sorbent, and then Hg^{2+} sorption was
298 followed.

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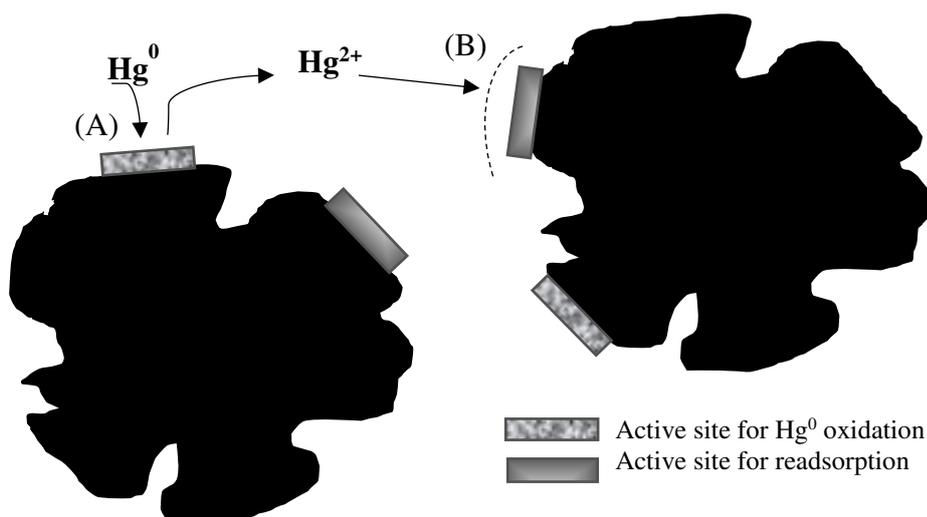
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Suggested Hg removal mechanisms of Wa-2 and Sa-4:

(A) Hg^0 oxidation at the activated site.

(B) Readsorption of the resultant oxidized mercury at the site of RC-600/ GRC-600 available for adsorption.

312 **Figure 2.** Effect of bromine content in the sorbent on Hg^0 oxidation

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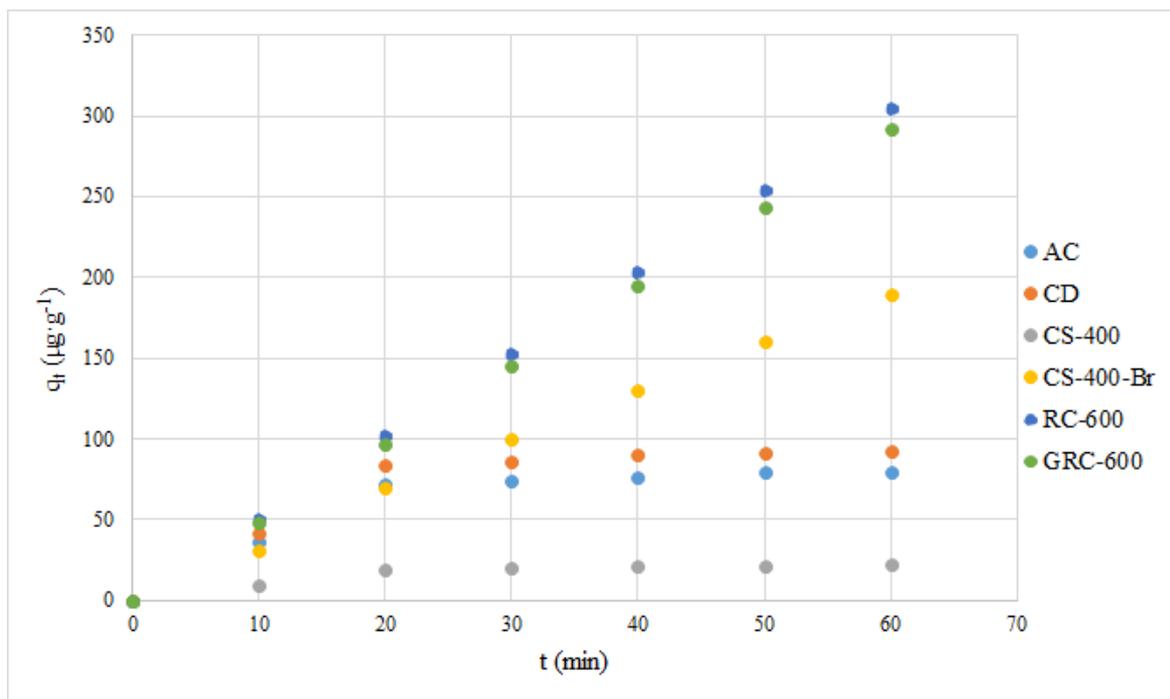
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To further assess the application prospect of such rubber waste-derived sorbents, the comparison the accumulative Hg adsorption capacity of RC-600 and GRC-600 with other sorbents was conducted under similar experimental conditions (Fig. 3). It demonstrated that with the elevation of sorption time (10 – 60 min, every 10 min.), the accumulative properties for bromine-present sorbents increase. No time influence was observed for bromine-free (AC, CD, CS-400) sorbents.



318

319 **Figure 3.** Accumulative Hg adsorption capacity for the tested sorbents

320 **4. Conclusions**

321 The results of studies of mercury adsorption from lignite flue gas using six organic sorbents (AC, CD, CS-400, CS-400-Br, RC-600, GRC-600) allow for the formulation of the following conclusions. The highest mercury removal from flue gas at 95 °C from lignite was achieved with AC (96.1%) and CD (93.8%), followed by RC-600 (81.5%) and GRC-600 (65.7%). The CS-400-Br exhibited better Hg removal performance compared to virgin biochar CS-400. The doping of sorbents with bromine resulted in a higher share of Hg⁰ oxidation to Hg²⁺ species. Low mercury removal efficiency by CS-400 was caused by its low mesopore volume (0.01 cm³·g⁻¹). The Hg removal efficiency for AC, CD and biochars (CS-400) decreased with the increase of temperature, caused by a higher amount of sorption inactive Hg⁰ mercury in the flue.

329 On the other hand, CS-400-Br, RC-600 and GRC-600 enhanced their mercury adsorption capacity with an increase in the temperature. Those sorbents had a higher content of bromine (between 2-14 times) than other sorbents. Therefore it can be confirmed that bromine had positive effects on Hg⁰ removal due to its influence on better Hg⁰ oxidation as well as adsorption on the free sites of the surface. In this study, a novel sorbent was created with one-step pyrolysis of tyre waste towards for Hg⁰ removal from lignite flue gas. This method could combine municipal solid waste disposal and mercury sorbent preparation in one process. The Hg adsorption capacity of RC-600 and GRC-600 was close to those of commercial activated carbons.

336

337 **Ethics approval and consent to participate**

338 Not applicable.

339 **Consent for publication**

340 Not applicable.

341 **Availability of data and materials**

342 The datasets used and/or analysed during the current study are available from the corresponding author on
343 reasonable request.

344 **Competing interests**

345 The authors declare that they have no competing interests.

346 **Authors' contributions**

347 SB performed mercury sorption on sorbents (on a laboratory scale). BT prepared two sorbents for further tests. SS
348 prepared two sorbents for further research and performed their analyzes. KK did did the proximate and ultimate
349 analysis of coal and sorbents. PB participated in the preparation of the research concept and final version of the
350 manuscript. MMG analyzed and interpreted the patient an data presented in the manuscript and was a major
351 contributor in writing the manuscript. All authors read and approved the final manuscript.

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Figures

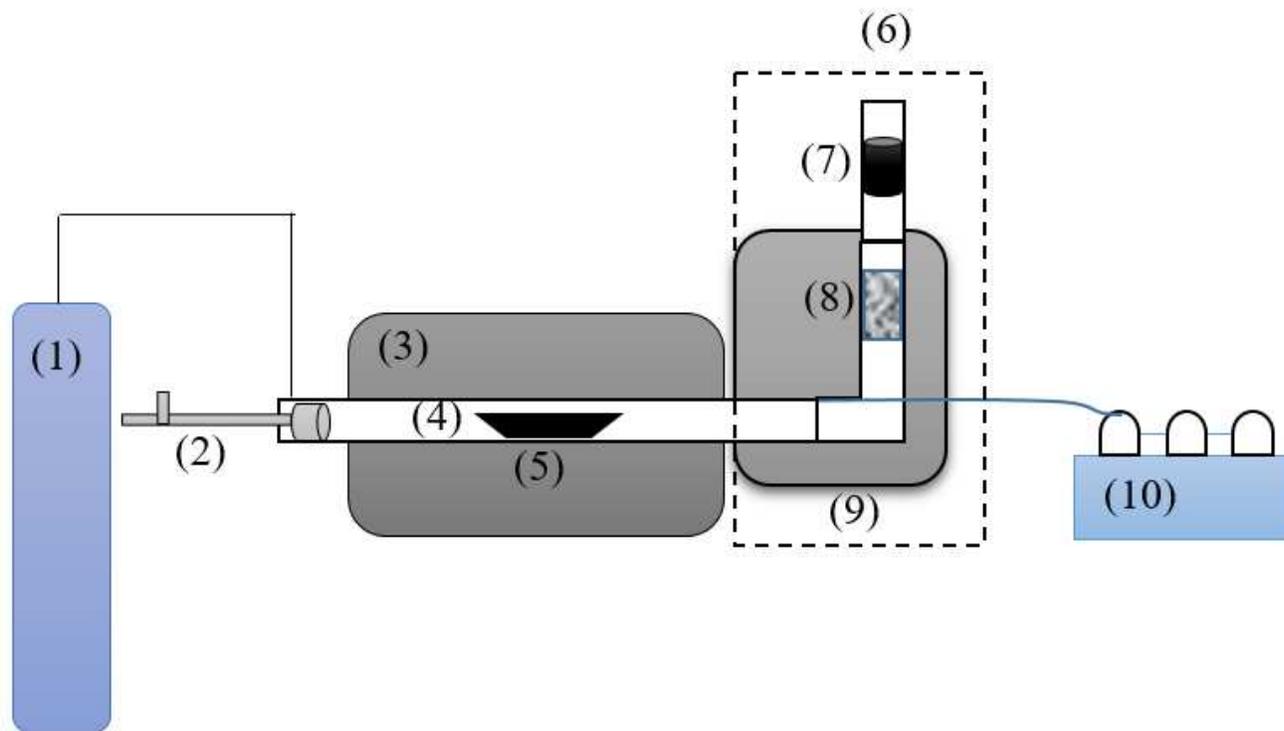
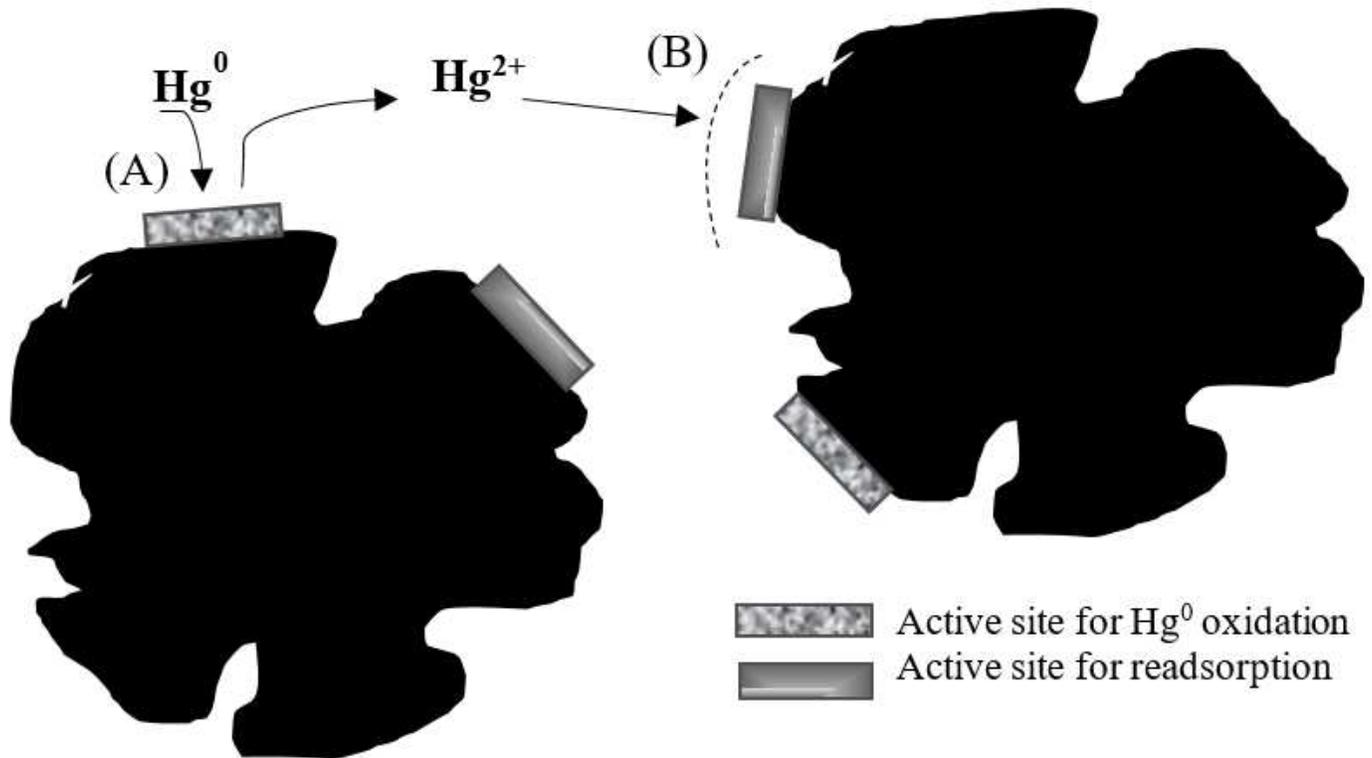


Figure 1

(1) gas source - air, (2) metal rod, (3) tube furnace, (4) quartz tube, (5) coal sample, (6) flue mercury adsorption device (7) sorbent trap, (8) sorbent holder, (9) furnace for heating of flue gases, (10) ice bath with series of impingers for measurement of mercury speciation.



Suggested Hg removal mechanisms of Wa-2 and Sa-4:

(A) Hg^0 oxidation at the activated site.

(B) Readsorption of the resultant oxidized mercury at the site of RC-600/ GRC-600 available for adsorption.

Figure 2

Effect of bromine content in the sorbent on Hg^0 oxidation

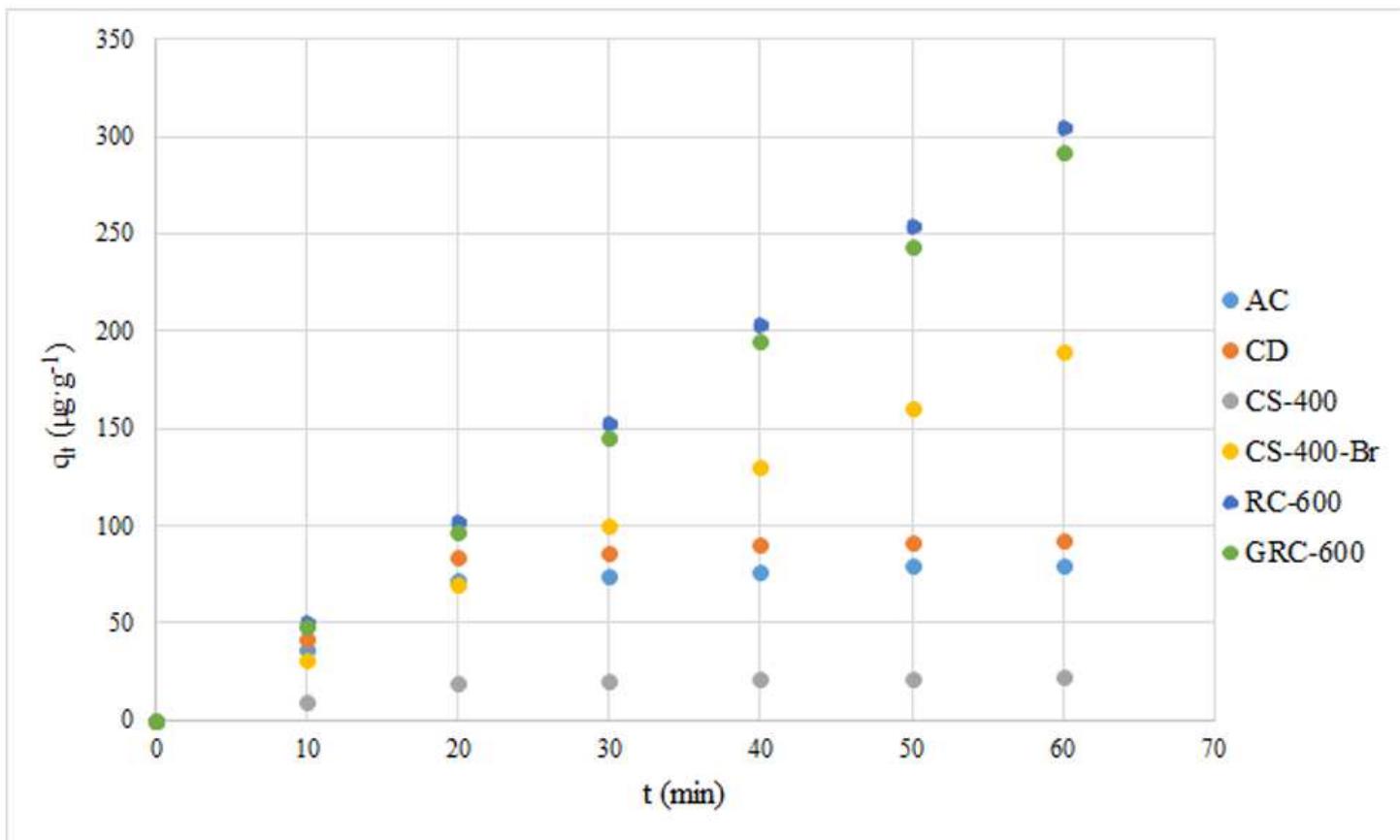


Figure 3

Accumulative Hg adsorption capacity for the tested sorbents

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