

1 **Recycling of spent adsorbents for oxyanions and heavy metal ions**
2 **in the production of ceramics**

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24 **Abstract**

25 Spent adsorbents for oxyanion forming elements and heavy metals are classified as hazardous
26 materials and they are typically treated by stabilization/solidification before landfilling. The
27 use of lime or cement for stabilization/solidification entails a high environmental impact and
28 landfilling costs are high. This paper shows that mixing spent adsorbents in the raw material
29 for the production of ceramic materials is a valuable alternative to stabilize oxyanion forming
30 elements and heavy metals. The produced ceramics can be used as construction material,
31 avoiding the high economic and environmental impact of stabilization/solidification followed
32 by landfilling. To study the stabilization of oxyanion forming elements and heavy metals
33 during the production process, two series of experiments were performed. In the first series of
34 experiments, the main pollutant, Mo was adsorbed onto iron-based adsorbents, which were
35 then mixed with industrial sludge (3 w/w %) and heated at 1100 °C for 30 minutes. Mo was
36 chosen, as this element is easily adsorbed onto iron-based adsorbents and it is the element that
37 is the most difficult to stabilize (i.e. the highest temperatures need to be reached before the
38 concentrations in the leachate are reduced). Leaching concentration from the 97/3
39 sludge/adsorbent mixture before heating ranged between 85-154 mg/kg; after the heating
40 process they were reduced to 0.42-1.48 mg/kg. Mo was actually stabilized, as the total Mo
41 concentration after addition was not affected by the heat treatment. In the second series of
42 experiments, the sludge was spiked with other heavy metals and oxyanion forming elements
43 (Cr, Ni, Cu, Zn, As, Cd and Pb) in concentrations 5 times higher than the initial
44 concentrations; after heat treatment the leachate concentrations were below the regulatory
45 limit values. The incorporation of spent adsorbents in ceramic materials is a valuable and
46 sustainable alternative to the existing treatment methods, saving raw materials in the ceramics
47 production process and avoiding the use of stabilizing agents. Besides, spent adsorbents added

48 to the raw material for ceramic products, may improve their aesthetic and structural
49 properties.

50 **Keywords**

51 Recycling, Spent adsorbents, Oxyanions, Heavy metals, Ceramic materials

52 **1 Introduction**

53 Oxyanion forming heavy metals like As, Cr, Mo, Sb, Se, V and W are nowadays high on the
54 environmental agenda due to their toxicity and high mobility (Cornelis et al., 2008). Their
55 removal from wastewater before discharge was already studied extensively in literature.

56 Several techniques exist to remove oxyanion forming elements (and other heavy metals) from
57 wastewater: (a) (co)precipitation, (b) adsorption, (c) ion exchange and (d) membrane
58 techniques (nanofiltration, reverse osmosis and electrodialysis). All these techniques finally
59 result in clean water and some sort of hazardous solid waste that contains the element
60 removed: a sludge in (a), a column packing in (b) and (c), and a concentrate from which the
61 water can be evaporated to give a sludge for (d) (Holm and Wilson; 2006, Mohan and Pitman,
62 2007).

63 Adsorption is a simple and cheap technique, capable of removing different oxyanions
64 simultaneously. Examples of adsorbents that can be used to remove oxyanions are activated
65 carbon, manganese/aluminum/titanium oxides, and iron oxides and hydroxides. Activated
66 carbon is a relatively expensive adsorbent, which can readily adsorb organic compounds like
67 PCBs and PAHs, pesticides, dyes and aromatic solvents. Inorganic components do not bind
68 well to activated carbon, and to obtain good removal capacities e.g. for metals and oxyanions,
69 the surface of activated carbon should be pretreated or coated, by impregnating the surface
70 with e.g. iron oxides (Vaughan and Reed, 2005; Chen et al., 2007), or by modifying the

71 surface e.g. by cationic surfactants (Choi et al., 2009). Iron oxides and hydroxides can
72 however be produced at low-cost and are widely used for the adsorption of oxyanions, as they
73 have a high affinity for oxyanions; their adsorption efficiency is highest at low pH (Adegoke
74 et al., 2012, Hua et al., 2012, Gallegos-Garcia et al., 2012). To achieve a high capacity, use
75 can be made of iron oxide nanoparticles with a high specific surface. Such nanoparticles
76 cannot be used as such in an adsorption column, the most widely used continuous adsorption
77 setup in wastewater treatment, because of high pressure requirements. For column
78 applications, nanosized materials must be coated on a larger sized carrier materials. Therefore
79 adsorbents for oxyanions, consisting of magnetite supported by a zeolite or perlite matrix
80 were developed (Verbinnen et al., 2012, 2013b, 2015; Vaclavikova et al., 2010)

81 One of the main questions that arises when discussing adsorption is whether or not the
82 adsorbent can be regenerated after usage. For the adsorption of oxyanion forming elements
83 and heavy metals on adsorbents, the regeneration of the spent adsorbent is not straightforward
84 in most cases. Due to the strong interactions (i.e. inner-sphere complex formation) of most
85 other oxyanion forming heavy metals with adsorbents, techniques to regenerate the adsorbent
86 are limited and/or expensive (Verbinnen et al., 2012). For some elements, eluting with
87 alkaline solutions can be sufficient to remove the adsorbate from the adsorbent and to reuse
88 the adsorbent after regeneration (Bujnakova et al., 2013). This again leads to a contaminated
89 waste stream. When regeneration is economically not feasible, the adsorbent needs to be
90 discharged after a single use, and this is often considered the main drawback of adsorption.

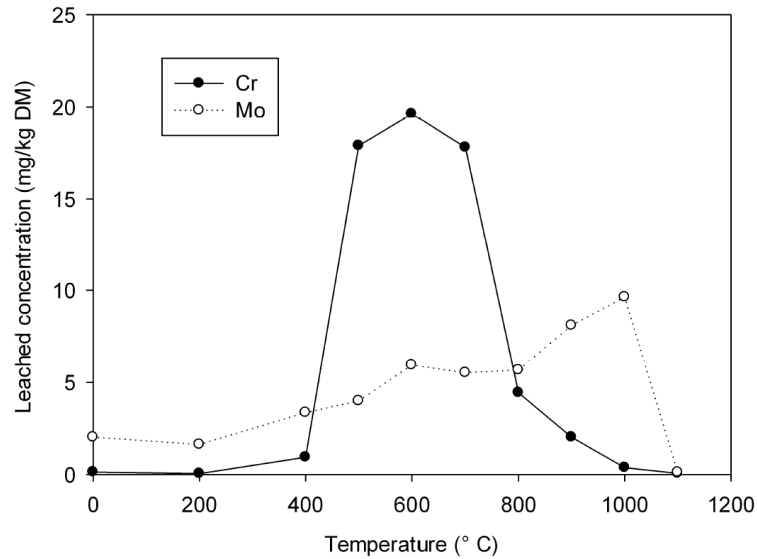
91 The main disposal strategies for spent adsorbents are either incineration or landfilling (Mohan
92 and Pittman, 2007). Incineration is mainly used for carbonaceous adsorbents, so landfilling is
93 the preferred option for most adsorbents (i.e. iron-based compounds) for oxyanions. The toxic
94 characteristics of many oxyanion forming elements require solidification/stabilization of the
95 adsorbent before landfilling. Cement and lime were proven to be effective to stabilize/solidify

96 spent (iron-based) adsorbents, but at least 25% of lime and/or cement has to be added to bring
97 leaching below the regulatory limit values (Kundu and Gupta, 2008). Although being low-
98 cost materials, the environmental impact of both lime and cement is high due to the release of
99 carbon dioxide in the production process. After stabilization/solidification, the spent
100 adsorbents need to be landfilled, entailing extra costs, i.e. landfilling taxes.

101 In this paper, an alternative strategy to dispose spent inorganic adsorbents is proposed. By
102 blending adsorbents with the raw materials for ceramics, primary resources for the ceramic
103 production can be saved, and the costs and the environmental impact of landfilling the
104 adsorbent can be avoided. Examples of such ceramic products can be (but are not limited to)
105 bricks, floor and roof tiles as well as lightweight and dense aggregates. Contaminated sludge
106 and sediments have often been used to produce ceramics (Alonso-Santurde et al., 2008;
107 Gonzalez-Corrochano et al., 2012). From earlier research on recycling of sludge from soil
108 cleaning for the production of ceramics (Verbinnen et al., 2014), it appeared that upon heating
109 inorganic sludge, the leaching concentrations of the toxic elements Ni, Cu, Zn, As, Cd and Pb
110 could be reduced (Table 1). This was also reported for other waste streams that were used to
111 produce ceramics (Chang et al., 2007; Xu et al., 2008; Quijorna et al., 2012), and is related to
112 removal of organic acids responsible for the formation of mobile organo-metallic complexes,
113 and/or a change in pH, and/or incorporation of the heavy metals in stable phases from which
114 they do not leach easily (Xu et al., 2008).

115 Contrarily, the leaching of the oxyanion forming elements Cr and Mo increases upon heating
116 (Table 1). This was also observed by other researchers producing ceramics from different
117 types of waste: Waelz slag (Quijorna et al., 2012), aggregate sludge and fly ash (Gonzalez-
118 Corrochano et al., 2012), and sewage sludge (Xu et al., 2008). At temperatures below 700°C,
119 Cr(III)- and Mo(VI)-compounds are oxidized and leach as their mobile (and for Cr most
120 toxic) hexavalent oxidation state. At temperatures above 700 °C, the mobile Cr compounds

121 are incorporated into low-melting silicates, and the leaching concentrations are reduced
 122 (Figure 1). For Mo compounds, temperatures above 1000 °C are needed to incorporate them
 123 into silicates.



124
 125 Figure 1: Leaching behavior of Cr and Mo from industrial sludge fired at temperatures between 200 and 1100 °C
 126 for 4 h

127 It is evidenced from the literature cited above that some hazardous waste streams can be
 128 stabilized by producing ceramics. Nevertheless, attempts to incorporate other heavily polluted
 129 waste streams, like spent adsorbents, in the raw material for ceramics production, in view of
 130 stabilizing the oxyanions and heavy metals present in the waste, are scarce to non-existent in
 131 literature. These materials are normally landfilled.

132

133 Table 1: Leachate concentrations of Cr, Ni, Cu, Zn, As, Mo, Cd and Pb from contaminated sludge heated at 400,
 134 600 and 800 °C for 4 h, d.l.= detection limit (0.01 mg/kg), n=2

Temperature	Cr	Ni	Cu	Zn	As	Mo	Cd	Pb
(°C)	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg

Not heated*	0.13±0.00	1.31±0.02	4.43±0.08	3.78±0.05	0.21±0.00	2.02±0.02	0.02±0.00	< d.l.
400	0.94±0.06	0.19±0.00	0.35±0.03	0.17±0.03	0.43±0.05	3.36±0.11	0.02±0.00	< d.l.
600	19.62±1.24	0.04±0.00	0.02±0.00	0.56±0.53	0.04±0.01	5.94±0.31	0.01±0.00	< d.l.
800	4.44±0.96	0.04±0.00	< d.l.	0.04±0.06	0.12±0.05	5.67±0.06	0.02±0.00	< d.l.

135 * As reported by Verbinnen et al. (2014)

136 The aim of this paper is to explore the possibility of adding spent adsorbents to contaminated
137 sludge and produce ceramic materials in order to stabilize the oxyanion forming elements and
138 heavy metals. More specifically, it is investigated whether spent zeolite or perlite-supported
139 iron-based adsorbents saturated with oxyanions can be mixed with industrial sludge (see
140 section 2.2) and subsequently fired at high temperature to obtain aggregates in which the
141 oxyanions are stabilized. In a first series of tests, Mo was loaded onto iron-based adsorbents,
142 mixed with the raw material for ceramics (the industrial sludge), and subsequently fired at
143 1100 °C to produce ceramics, from which the leaching concentrations were measured. Mo
144 was chosen, as this element is easily adsorbed onto iron-based adsorbents (Verbinnen et al.,
145 2012) and it is the element that is the most difficult to stabilize (i.e. the highest temperatures
146 need to be reached before the concentrations in the leachate are reduced). To study the
147 stabilization after heating of other oxyanion forming elements and cation forming heavy
148 metals (Cr, Ni, Cu, Zn, As, Cd and Pb), the industrial sludge was spiked with these elements,
149 ceramics were produced and the leachate concentrations were measured.

150 **2 Materials and methods**

151 *2.1 Adsorption experiments*

152 Adsorption experiments were performed with zeolite- and perlite-supported magnetite,
153 prepared as described by Verbinnen et al. (2012, 2013b, 2015). The adsorbents were saturated
154 with Mo by contacting them with 50 mg/l Mo(VI) solutions. The ideal adsorption conditions

155 (1.5 g/l adsorbent, 24 h contact time at 160 rpm, pH 2.9) used for the adsorption experiments
156 were selected based on the knowledge gained in previous research (Verbinnen et al., 2012,
157 2013b, 2015). The adsorbents were recuperated from the solution by filtering them over a
158 paper filter. The adsorption capacity for Mo was measured to be 8.4 mg/g for zeolite-
159 supported magnetite and 13.6 mg/g for perlite-supported magnetite.

160 *2.2 Ceramics production*

161 The dried and ground (< 0.1 mm) zeolite- or perlite-supported magnetite, both loaded with
162 Mo, was mixed with dried industrial sludge contaminated with mainly inorganic
163 contaminants, in a sludge/adsorbent ratio of 97/3. The 97/3 ratio was chosen, based on the
164 adsorption capacities of the loaded adsorbents, to obtain a tenfold increase in Mo
165 concentration in the final product. The contaminated sludge was obtained after washing of
166 soils contaminated with heavy metals; it was characterized thoroughly by e.g. its elemental
167 and mineralogical composition (Tables S1 and S2 in the Supplementary information). Water
168 was added to the sludge-adsorbent mixture to obtain a water content of 20 w/w % and the
169 mixture was pelletized to obtain spheres with a diameter of about 1.5 cm and an average
170 weight of 5 g. After drying at 105 °C, the spheres were introduced into a preheated muffle
171 furnace (Heraeus Thermicon P) at 1100 °C. After 30 min, the produced aggregates were
172 removed from the furnace and allowed to cool at room temperature. The temperature and
173 residence time were chosen based on the results from earlier research (Verbinnen et al.,
174 2013a, 2014), in which was shown that 30 min at 1100 °C was optimal for stabilizing
175 oxyanion forming compounds like Cr and Mo (Figure 1), and for obtaining lightweight
176 aggregates. Other toxic elements as given in Table 1, are stabilized at lower temperatures
177 (Table 1).

178 *2.3 Leaching and destruction tests*

179 After cooling, the aggregates were ground (< 4 mm) and subjected to a 24 h leaching test with
180 an L/S ratio of 10 (EN12457-2). After 24 h, the supernatant was filtered over a 0.45 µm
181 membrane filter (Millipore) and the concentrations of the relevant elements were measured
182 using ICP-MS (Thermo Xi series). Besides the heat treated mixture (a 97/3 sludge/adsorbent
183 mixture that was heated), also the untreated sludge (sludge to which no adsorbent was added,
184 and that was not heated) and the untreated mixture (a 97/3 sludge/adsorbent mixture that was
185 not heated) were subjected to the same leaching test to compare the Mo leaching.

186 The dried and ground zeolite- and perlite-supported magnetite loaded with Mo, the untreated
187 sludge and the 97/3 sludge/zeolite or perlite adsorbent mixture, before and after the heat
188 treatment, were digested to determine their content of heavy metals and/or oxyanions.
189 Therefore, 5 ml HNO₃, HClO₄ and HF was successively added to 0.1 g of the material and
190 boiled. After digestion, the excess acid was evaporated, the solutions were diluted with
191 ultrapure water and transferred to 100 ml volumetric flasks; and the metal concentrations were
192 measured with ICP-MS.

193 *2.4 Other elements*

194 To check whether other elements could also be stabilized by incorporation into ceramic
195 materials, seven toxic and regulated oxyanion forming elements or cation forming heavy
196 metals (Cr, Ni, Cu, Zn, As, Cd and Pb) were added to the contaminated sludge in
197 concentrations that were at least 5 times the concentrations initially present. Each of these
198 elements was added as one of its most water soluble compounds, to maximize the leachability
199 of the added metals from the untreated sludge. The sludge was then treated in the same way as
200 the other sludges (heating at 1100 °C for 30 min, cooling, crushing < 4mm, leaching with L/S
201 10 for 24 h, and measurement with ICP-MS).

202 **3 Results and discussion**

203 *3.1 Leaching of Mo from ceramic materials*

204 The total Mo concentrations in the untreated sludge and in the sludge mixed with adsorbent
 205 before and after heat treatment are shown in Table 2. Mixing of 3% of Mo-loaded adsorbent
 206 with sludge increases the total Mo concentration with almost a factor 10 compared to the
 207 concentration in the untreated sludge.

208 Heat treatment did not decrease the total Mo concentrations in the sludge-adsorbent mixture,
 209 indicating that Mo does not volatilize during heat treatment. The higher Mo concentrations
 210 after heat treatment are a result of the (partial) volatilization of other compounds (carbonates,
 211 residual organic compounds,...), enriching Mo in the matrix.

212 Table 2: Total and leached Mo concentrations and standard deviations (mg/kg) for sludge, and sludge mixed
 213 with 3% zeolite- and perlite supported magnetite before and after heat treatment (1100 °C, 30 min), n=2

	Total	Leached
Sludge	33 ± 11	1.97 ± 0.04
Sludge + 3% zeolite-supported magnetite, no heat treatment	248 ± 14	85 ± 5
Sludge + 3% perlite-supported magnetite, no heat treatment	281 ± 33	154 ± 1
Sludge + 3% zeolite-supported magnetite after heat treatment	251 ± 13	0.42 ± 0.26
Sludge + 3% perlite-supported magnetite after heat treatment	429 ± 19	1.48 ± 0.50

214

215 The leached Mo concentrations for untreated sludge, and the 97/3 sludge/adsorbent mixtures,
 216 before and after heat treatment are also shown in Table 2. Mo leaching from the untreated
 217 sludge is relatively low, but when the sludge is mixed with the adsorbents, leaching increases
 218 43 (mixed with zeolite-supported magnetite) to 78 times (mixed with perlite-supported

219 magnetite). This clearly indicates that the spent adsorbents should be classified as hazardous
220 materials, as the Waste Acceptance Criteria leaching limit value for acceptance of waste on a
221 landfill for hazardous materials (WAC, established by the European Council Decision
222 2003/33/EC) is set at 30 mg/kg for Mo. Even when only 3% adsorbent is mixed with sludge,
223 this limit value is already exceeded.

224 Heat treatment drastically reduces the leaching from the sludge mixed with the adsorbents to
225 only 0.5 to 1% of the initial concentration. The concentrations in the leachate are even lower
226 than for the untreated sludge, although the total concentrations are not reduced during the heat
227 treatment. This indicates that Mo is effectively stabilized in the matrix of the sludge during
228 heat treatment. Indeed, Mo is incorporated in a glassy phase at temperatures of 1100°C
229 (Verbinnen et al., 2014), preventing it from leaching. The elevated temperature is needed to
230 form aggregates with high mechanical strength and to effectively stabilize Mo and Cr, but is
231 far lower than the temperatures that are reported for similar waste streams that are stabilized
232 by full vitrification of the materials (typically 1400 – 1500 °C, Li et al., 2007; Bingham and
233 Hand, 2006; Kavouras et al., 2003). For waste streams not containing Cr and/or Mo
234 compounds, even lower temperatures suffice to reduce the leaching concentrations (Table 1).

235 *3.2 Stabilization of other toxic elements*

236 It was demonstrated that the leaching of Mo from spent adsorbents can be reduced by
237 incorporating the adsorbents in sludge and heating it to 1100 °C. Preliminary experiments also
238 showed (Table 1) that lower temperatures are sufficient to reduce the leaching concentrations
239 from sludge for other toxic heavy metals and oxyanion forming elements. To check whether a
240 similar behavior is also observed when they are added in excess concentrations to sludge (i.e.
241 as expected when spent adsorbents for these elements are added to sludge), sludge was spiked
242 with the following oxyanion forming elements and heavy metals (Cr, Ni, Cu, Zn, As, Cd and

243 Pb), heated at 1100 °C and subsequently leached. Table 3 gives the initial element
 244 concentrations in the original sludge, and the concentrations after addition of the compounds
 245 of Cr, Ni, Cu, Zn, As, Cd, and Pb, as indicated.

246 Table 3: Total concentrations (mg/kg) of the regulated elements in the sludge, and after spiking of the sludge
 247 with mobile compounds of these regulated elements

	Cr	Ni	Cu	Zn	As	Cd	Pb
Sludge*	184	95	199	1454	10	24	509
After spiking	1000	500	1000	7500	50	150	3000
Compound added	K ₂ Cr ₂ O ₇	Ni(NO ₃) ₂ ·6H ₂ O	CuSO ₄ ·5H ₂ O	ZnSO ₄ ·7H ₂ O	Na ₂ HAsO ₄ ·7H ₂ O	3CdSO ₄ ·8H ₂ O	PbNO ₃

248 *As reported by Verbinnen et al. (2014)

249 The leached concentrations after heating at 1100 °C for both the untreated sludge and the
 250 sludge spiked with the regulated elements are shown in Table 4, together with the regulatory
 251 limit value for reuse of waste in view of recycling in Flanders (Northern part of Belgium). All
 252 elements have slightly higher concentrations in the leachate for the spiked sludge than in the
 253 untreated sludge, but are all well below the regulatory limit values.

254 Table 4: Leachate concentrations (mg/kg) of the regulated elements from the sludge, and from the sludge spiked
 255 with these regulated elements, both heated at 1100 °C for 30 minutes, d.l.= detection limit (0.01 mg/kg), n=2

	Cr	Ni	Cu	Zn	As	Cd	Pb
Sludge	0.134	0.013	0.098	0.150	0.029	< d.l.	0.019

After spiking and heating	0.218±0.037	0.061±0.013	0.266±0.030	0.379±0.200	0.062±0.008	< d.l.	0.041±0.009
Limit value	0.5	0.75	0.5	2.8	0.8	0.03	1.3

256

257 *3.3 Other advantages*

258 The incorporation of the considered adsorbents as raw material in the production process of
 259 ceramics will not have a negative effect on the other (structural) product properties. In fact,
 260 iron oxides are often blended into the raw materials for two reasons. They can be used as dye;
 261 e.g. hematite (Fe₂O₃, sometimes in combination with other elements) is used to give ceramics
 262 a red color (Pereira and Bernardin, 2012). Moreover, Fe₂O₃ is often added as it can be
 263 converted to FeO in a reducing atmosphere, releasing O₂ gas (i.e. bloating), which leads to a
 264 less dense material (Yang et al., 2015; Riley, 1951). This is beneficial in applications like
 265 lightweight aggregates and concrete, thermal and sound insulation, etc., giving the material a
 266 higher market value. Besides structural and aesthetic benefits, adding the adsorbents in the
 267 production process of ceramics saves raw materials (mainly clays). Moreover no more lime
 268 and cement, which have a high environmental impact due to the release of carbon dioxide in
 269 their production process, are needed to solidify/stabilize spent adsorbent, and the cost of
 270 landfilling the solidified/stabilized materials (i.e. landfilling taxes) is avoided.

271 **4 Conclusions**

272 In this paper a sustainable alternative to landfilling is proposed by incorporating spent
 273 adsorbents as raw material in the production process of ceramics. Mixing 3 % spent adsorbent
 274 loaded with Mo with industrial sludge, and heating the mixture at 1100 °C for 30 minutes,

275 showed that Mo is stabilized in the matrix: the concentrations in the leachate were reduced to
276 0.5 to 1% of their initial values, to concentrations lower than 1.5 mg/kg.

277 To prove that other heavy metals and cation forming elements added to industrial sludge are
278 also stabilized after heating, and can thus be incorporated in industrial sludge, Cr, Ni, Cu, Zn,
279 As, Cd and Pb were added in concentrations 5 times higher than the concentrations in the
280 sludge. After heating the sludge for 30 minutes at 1100 °C, the elements appeared effectively
281 stabilized, with the leachate concentrations well below the regulatory limit values.

282 The temperature that needs to be reached to ensure that the leachate concentrations are below
283 the limit values (1100 °C), is far lower than the temperatures that are reported for full
284 vitrification of comparable waste streams (1400 – 1500 °C). For waste streams not containing
285 Cr and/or Mo compounds, even lower temperatures suffice to reduce the leaching
286 concentrations.

287 The incorporation of small amounts of spent (iron-based) adsorbents for oxyanion forming
288 elements and heavy metals is a valuable alternative for traditional S/S treatment followed by
289 landfilling, which has a has a high environmental and economic impact: carbon dioxide is
290 emitted during their production, and landfill taxes need to be paid. In addition to a sustainable
291 method for waste treatment, mixing adsorbents loaded with heavy metals to industrial sludge,
292 has also other advantages. The use of spent iron-based adsorbents is also beneficial for the
293 aesthetic (color) and structural (lightweight) properties; the obtained aggregates represent a
294 higher market value than dense materials due to their insulating capacities. At last, it must be
295 mentioned that the method for the treatment of adsorbents loaded with heavy metals described
296 in this article, might be elaborated to other residues that are obtained after removal of toxic
297 compounds from wastewater, like coagulation/precipitation sludge.

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