

1 A Thermal Imaging Methodology To Study
2 Evaporation Kinetics in Mine Tailings

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9 coefficient, water transport mechanisms, evaporative fluxes, fugitive dust emissions, water
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11 residues.

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19 ABSTRACT

20 Predicting why, how, and when mine tailings disposal sites become prone to dust scattering events
21 is often hampered by our limited understanding of the factors that affect the drying rates from their
22 surface layers. As a case study, thermal imaging is demonstrated here to be a valuable tool to
23 study the evaporation mechanisms and rates from bauxite residues as a function of their thickness
24 and physicochemical properties, as well as environmental conditions. These investigations reveal
25 their that late stage drying rates are limited by gas phase diffusion through the interstitial air within
26 their internal microporosity. The smallness of the effective diffusion coefficient indicates that
27 water adsorption on bauxite residues surfaces is the dominant phenomenon responsible for their
28 slow water vapour transport kinetics, a phenomenon that ultimately controls their late stage drying
29 rates, that is when dust scattering is most likely to occur. As such, application of this thermal
30 imaging methodology in the field may also contribute to improve the accuracy of risk assessment
31 protocols, support intervention and mitigation strategies, underpin optimization efforts for mining
32 residues management, and improve forecasting of fugitive dust emissions from mine tailings by
33 enabling more accurate predictions of the evolution in their surface drying state.

34

35 INTRODUCTION

36 Fugitive dust emissions emanating from mining residues can be a significant environmental
37 concern facing tailing storage facilities, challenging their prevention and mitigation efforts that, as
38 a result, become increasingly costly and labor intensive (Allan, 1995; Power, Gräfe, & Klauber,
39 2011). Unfortunately, forecasting and risk assessment efforts are hampered by our limited

40 understanding of the impact of meteorological conditions, as well as of mine tailings
41 microstructural and physicochemical properties, on their drying mechanisms and rates.

42 To tackle this problem, a bauxite residues storage area (BRSA) was selected as a case study
43 allowing meteorological records to be scrutinized in order to highlight the environmental
44 conditions that led to, and triggered, past fugitive dust emission events (Maurais et al., 2018). This
45 enabled the investigation of drying kinetics to be steered towards the most critical and relevant
46 environmental parameters for laboratory studies. Numerous meteorological parameters are
47 continuously monitored by a weather station nearby the selected BRSA namely, wind speed and
48 direction, precipitations, as well as atmospheric temperature and relative humidity (RH). This
49 latter was shown to be the most aggravating environmental parameter leading to dust particles
50 scattering events (Maurais et al., 2018). Therefore, thermal imaging techniques were brought to
51 bear on this issue by evaluating whether they could help quantify the evaporation kinetics of
52 bauxite residues. They are demonstrated herein to be convenient and effective tools for
53 investigation into evaporation rates that govern the drying process and to provide invaluable
54 insight into the underlying water transport mechanisms (Fuchs & Tanner, 1967; Pfister,
55 McDonnell, Hissler, & Hoffmann, 2010; Price, 1980).

56 Previously reported methods for measuring water contents, drying rates and evaporative fluxes
57 from porous media include frequency/time-domain reflectometry (Topp & Davis, 1985; Wang,
58 Xia, Wang, & Lu, 2012; Yu & Drnevich, 2004), dielectric permittivity measurements (Wang et
59 al., 2012), ground penetrating radar dielectric constant measurements (Van Dam, 2014), electrical
60 conductivity or resistivity (Brevik, Fenton, & Lazari, 2006; Sheets & Hendrickx, 1995), visual
61 inspection of the evolution of the drying front within the sample (Shokri, Lehmann, & Or, 2009;
62 Shokri & Or, 2011), heat-pulse probes (Bristow, 1998; Bristow, Campbell, & Calissendorff, 1993;

63 Hopmans, Šimunek, & Bristow, 2002), to name only but a few. Unfortunately, these methods
64 either lack the sensitivity or selectivity towards the surface layer, and/or cannot be easily deployed
65 for continuous in-situ monitoring and field applications (Amano & Salvucci, 1989). They are thus
66 unsuitable for the specific context of evaluating surface drying states and rates as part of risk
67 assessment protocols for dust emissions from BRSA. Thermal imaging techniques possess those
68 desirable attributes and may thus prove to be equally effective at probing drying kinetics under
69 controlled conditions in the laboratory, as well as providing a potentially powerful management
70 tool to characterize the evaporation rates from bauxite tailing surfaces at BRSA (Price, 1980).

71 Drying kinetics from porous materials are very complex, displaying contributions from a number
72 of different water transport mechanisms resulting in distinctive regimes and stages that all depend
73 sensitively on their water content, their microstructural and physicochemical properties, as well as
74 on environmental conditions (McKenna Neuman, Boulton, & Sanderson, 2009). Indeed, when the
75 water content is elevated, as in the early stages of drying, capillary flow is expected to dominate
76 water transport to the surface (a regime thereafter referred to as stage I) allowing large evaporation
77 fluxes from wet residues to be sustained (Neriah, Assouline, Shavit, & Weisbrod, 2014; Or,
78 Lehmann, Shahraeeni, & Shokri, 2006; Shahraeeni & Or, 2010; Teng, Zhang, Zhang, Zhao, &
79 Sheng, 2019; Thiery, Rodts, Weitz, & Coussot, 2017). Eventually, the rupture of the wetting layer
80 at the percolation threshold causes hydraulic discontinuity within the porous media (Moldrup,
81 Olesen, Komatsu, Schjønning, & Rolston, 2001). This shifts the limiting water transport
82 mechanism from capillary flow to gas phase diffusion thereby yielding a significant slowing down
83 of the evaporation rates (a regime thereafter referred to as stage II) (Griend & Owe, 1994; Or et
84 al., 2006; Saravanapavan & Salvucci, 2000). The water content at which the transition from stage
85 I to stage II occurs has been correlated to the microstructural features of the porous media (Shokri

86 & Or, 2011). Therefore, it is expected that the evaporation kinetics from bauxite tailings should
87 not only depend strongly on their water content, but also on their chemical composition, as well
88 as on their morphological and physicochemical properties, attributes that govern which water
89 transport mechanism should be most effective under specific meteorological conditions (i.e.,
90 temperature and relative humidity) (Saravanapavan & Salvucci, 2000). Given the role of capillary
91 forces in maintaining the cohesion of dust particles with tailing sites surfaces, forecasting of
92 fugitive dust emissions from BRSA depend most critically on a detailed quantitative understanding
93 of bauxite tailings evaporation kinetics as well as an the ability to predict their surface drying state
94 (Shao, 2001).

95 In this work, we describe how thermal imaging techniques enable evaporation rates from bauxite
96 residues to be quantified. This methodology should help describe, probe, understand, and predict
97 bauxite residues evaporation kinetics, as well as their dependence on environmental parameters
98 representative of those found at the BRSA (Neriah et al., 2014). Collectively, these investigations
99 should improve our ability to predict the drying state of bauxite residues surfaces, forecast and
100 prevent the occurrence of fugitive dust emissions, as well as devise efficient risk assessment and
101 monitoring tools to assist in mining residues management and environmental mitigation efforts.

102

103 MATERIALS AND METHODS

104 A methodology based on thermal imaging techniques was devised allowing for the
105 measurements of drying kinetics from porous media. The surface-sensitivity and selectivity of
106 optical methods (i.e., tens to a few hundreds of microns) enable measurements of surface
107 temperatures from mine tailings under controlled ambient temperature and relative humidity
108 conditions in the laboratory. Furthermore, the portability and ease of use of the commercial

109 infrared camera used in this work (FLIR® Model A320) are attractive features for field
110 applications. These may thus enable thermal imaging techniques to be brought to bear on real-time
111 monitoring of the evaporation process from the surface of mine tailings storage facilities.

112 Under given environmental conditions (i.e., ambient T and RH), water vapour concentration and
113 temperature gradients can establish rapidly in the superficial layers of evaporating porous media,
114 evolving continuously thereafter throughout the drying process. These gradients provide a driving
115 force for the long-range diffusive/convective transport of heat and water vapour, processes that are
116 required to sustain water evaporation from the samples surface, and that explain the strong
117 dependency of the drying kinetics on environmental parameters (Neriah et al., 2014). In addition,
118 it is thus also expected that the drying kinetics should depend strongly on sample thickness, as
119 well as on their microstructure and physico-chemical properties, in addition to their water content
120 (Grifoll, Gastó, & Cohen, 2005). Under quasi-steady state conditions (Monteith, 1981), equation
121 1 indicates that the surface temperatures observed for evaporating bauxite residues can be
122 understood by comparing the latent heat flux of evaporation from the samples surface (calculated
123 from the evaporation flux, J_{evap} , and the vaporization enthalpy, ΔH_{evap}) to the sensible heat flux
124 sustained by their thermal conductivity, K , through the gradient in their temperature depth-profile,
125 ∇T (Kosky, Balmer, Keat, & Wise, 2013):

$$126 \quad K \cdot \nabla T = J_{evap} \cdot \Delta H_{evap} \quad [1]$$

127 Therefore, surface temperature measurements can straightforwardly and directly inform on the
128 evaporative fluxes emanating from drying mine tailings. The magnitude of evaporative cooling at
129 the sample surface is reported using thermal imaging measurements of the difference in surface
130 temperatures (ΔT , with a precision of ± 30 mK) between drying bauxite residues samples and those
131 from reference samples. These latter are bauxite residues that have been let to reach equilibrium

132 with the environmental conditions, at the selected ambient temperature and relative humidity
133 (Monteith, 1981).

134 Evaporation kinetics from porous media also depend strongly on their water content along with
135 their physicochemical and morphological properties (Or et al., 2006). Indeed, at water contents
136 above a critical threshold value, capillary flow supplies the evaporating surface with liquid water
137 and is the dominant water transport mechanism that controls drying rates (Neriah et al., 2014).
138 This critical threshold volumetric water content, θ_{thr} , is defined by the minimal water content for
139 which long-range transport by solute diffusion in the liquid phase is possible (Moldrup et al., 2001;
140 Shokri & Or, 2011; Thiery et al., 2017). Below θ_{thr} , the hydraulic continuity is disrupted, and the
141 evaporation kinetics should thus become increasingly limited by water vapour diffusion from the
142 vaporisation front to the sample's surface (Moldrup et al., 2001). Therefore, θ_{thr} informs us on the
143 dominant transport mechanism likely to limit the evaporation of water and can be evaluated using
144 a non-linear empirical relationship that relates θ_{thr} with the specific surface area (SSA) of the
145 porous medium (Moldrup et al., 2001):

$$146 \quad \theta_{thr} = 0.039 \cdot SSA_{vol}^{0.52} \quad [2]$$

147 Equation 2 indicates that materials with large SSAs, such as bauxite residue (44-87 cm²/g) (Li,
148 1998, 2001; Li & Rutherford, 1996), yield in an elevated value of θ_{thr} expressed as V_w/V_t , where
149 V_w is the volume of water and V_t the total volume of wet bauxite residues. Using the SSAs reported
150 in the literature (Li, 1998, 2001; Li & Rutherford, 1996) along with the macroscopic densities
151 measured for the bauxite residues used in the present study, $\rho_{bulk} = (1.2 \pm 0.1) \text{ g/cm}^3$, θ_{thr} is found
152 to vary between 31 % and 44 %, with an average value of $(38 \pm 4) \% (V_w/V_t)$.

153 The bauxite residue samples, having reached different drying stages, were collected from various
154 locations at the selected BRSA. They were rinsed, dried and prepared with an initial solid fraction

155 of 70 % (m_b/m_t , where m_b is the mass of dry bauxite residues and m_t the total mass of wet bauxite
156 residues), an initial drying state representative of the late stages of drying where fugitive dust
157 emissions are most likely to occur. Furthermore, as it corresponds to an initial volumetric water
158 content of 37 %, a contribution from capillary flow driven evaporation (i.e., stage I) is therefore
159 likely to be observed in the early stages of the drying process for the experimental conditions
160 explored in this work. Subsequently, gas phase water diffusion and adsorption on bauxite residues
161 surfaces are expected to become the dominant mechanisms that should limit water transport from
162 the vaporisation plane to the evaporating samples' surface (i.e., stage II).

163 In order to provide insight into the conditions under which the diffusive and/or capillary
164 processes limit water transport to the surface and thus, the evaporation rate, sample holders with
165 various depths, $d = 2, 4, 6,$ and 8 mm, were used. These thicknesses encompass the critical
166 superficial layers that are prone to dust scattering from BRSA surfaces. Surface temperature
167 measurements were performed using compacted samples of wet bauxite residues until completion
168 of water evaporation (i.e., when drying bauxite residues surface temperatures reach those of the
169 reference sample, within the thermal imaging temperature measurements accuracy of ± 30 mK).
170 Wet bauxite residues and reference samples were introduced in a controlled environmental
171 chamber (Humidity test chamber; VWR® Model 9005L), maintained at a fixed temperature and
172 ambient pressure (i.e., 310 K and 1 atm), and exposed to controlled relative humidity (RH)
173 conditions of 35 %, 40 %, 50 %, 60 % and 70 %. The thermal camera was placed in the
174 experimental chamber and was connected to a data processing software, allowing continuous and
175 simultaneous measurements of the reference and wet bauxite residues surface temperatures to be
176 performed for each sample thickness, as can be gleaned from the representative snapshots of a
177 typical experimental time series reported in Figure 1.

178 Finally, water adsorption isotherms were performed using initially dry bauxite residue
179 samples that have been let to equilibrate with the environmental conditions (i.e., $T = 310\text{ K}$, $P = 1$
180 atm, at the selected RHs). Once thermodynamic equilibrium was reached between the bauxite
181 residue sample and the selected environmental conditions, the gain in mass of the sample due to
182 the adsorbed water mass (m_w) was determined using a high precision analytical balance enabling
183 their equilibrium water content to be measured.
184

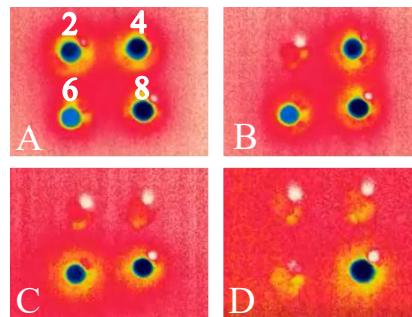


Figure 1. Snapshots from a time series showing thermal images of evaporating wet bauxite residue for sample thickness, $d = 2\text{ mm}$ (top left), 4 mm (top right), 6 mm (bottom left) and 8 mm (bottom right) : as prepared (**A**), and upon complete drying of the 2 mm (**B**), 4 mm (**C**) and 6 mm (**D**) thick sample.

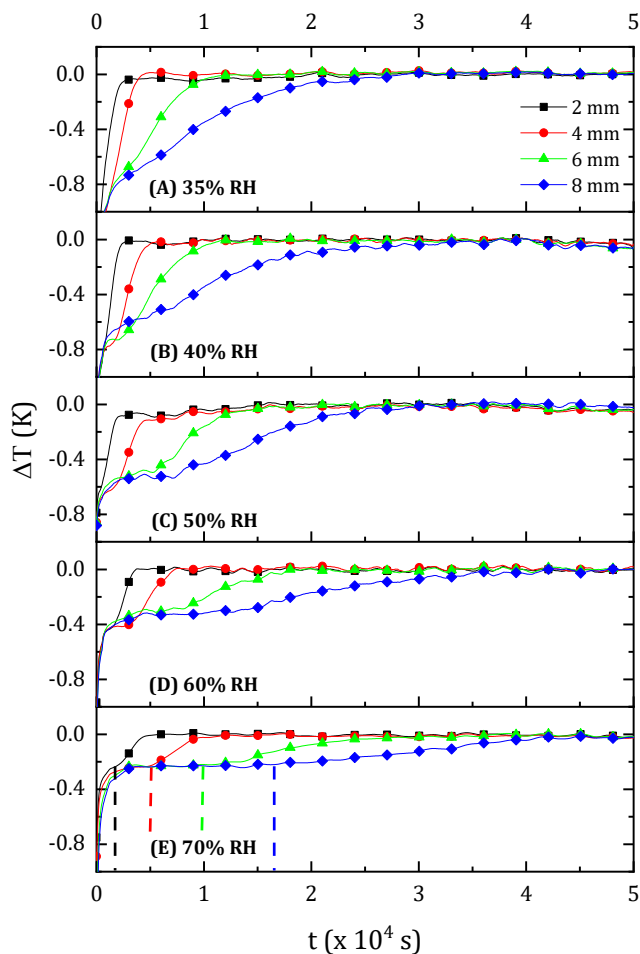


Figure 2. Evolution of the temperature difference (ΔT) between the sample and the reference surfaces during drying of bauxite residues prepared with an initial solid fraction of 70% (m_b/m_t) at $T = 310$ K. Sample thicknesses of 2 mm (black squares), 4 mm (red circles), 6 mm (green triangles) and 8 mm (blue diamonds) were monitored at a relative humidity of 35% (A), 40% (B), 50% (C), 60% (D) and 70% (E). The vertical lines in E indicate the onset of the transition from stage I to stage II evaporation regimes.

186 The evolution of the temperature difference between the bauxite residues and reference surfaces
187 (ΔT) as a function of time for sample thicknesses, $d = 2, 4, 6,$ and 8 mm (black squares, red circles,
188 green triangle, and blue diamonds, respectively), at a selected relative humidity ($RH = 35 \%, 40$
189 $\%, 50 \%, 60 \%,$ and 70% , reported in Figure 2A, 2B, 2C, 2D, and 2E, respectively), reveals the
190 complex nature of bauxite residues drying kinetics. As expected, the evaporation rates depend
191 strongly, and in a non-linear way on d , indicating that long-range transport through the bauxite
192 residues porous microstructure must play an essential role in the drying process. Lower initial ΔT
193 values are observed for each RH in Figure 2, reported as $\Delta T_{\text{free, evap.}}$ increase from ~ -800 mK at 35
194 $\% RH$ to $\Delta T \sim -250$ mK at $70 \% RH$ appear to be conspicuously independent of d . This reveals
195 that the initial drying rates remain constant during stage I evaporation, and that they recede as the
196 ambient RH increases. This is most likely due to free water evaporation from wet bauxite residues
197 in their early stages of drying, a process that is sustained by rapid capillary flow from the bulk to
198 the surface (Or et al., 2006). As such, these initial ΔT value are reported as $\Delta T_{\text{free, evap.}}$ Figure 2
199 reveals that the duration of this capillary-driven free water evaporation regime increases with
200 sample thickness and that it decreases with increasing RH .

201 Stage I evaporation is followed by an evaporation regime where surface temperatures slowly
202 rise until the samples eventually reach thermal equilibrium with their environment. Indeed, as
203 water content in evaporating bauxite residues decreases, it compromises the hydraulic continuity
204 between the vaporisation plane, buried in the bulk of the sample, and their surface. Therefore,
205 during stage II evaporation, the surface temperatures gradually rises as the evaporation rates
206 become increasingly limited by the much slower diffusion of water vapour towards the sample
207 surface until ΔT tends asymptotically to zero (i.e., near-equilibrium conditions). As a result, the
208 dominant mechanism and rate-limiting process that sustains the evaporation flux from the samples

209 surface shifts from capillary flow (stage I) to diffusive water vapour transport (stage II). In Figure
 210 2E, vertical dashed lines indicate the onset of the transition from stage I to stage II evaporation

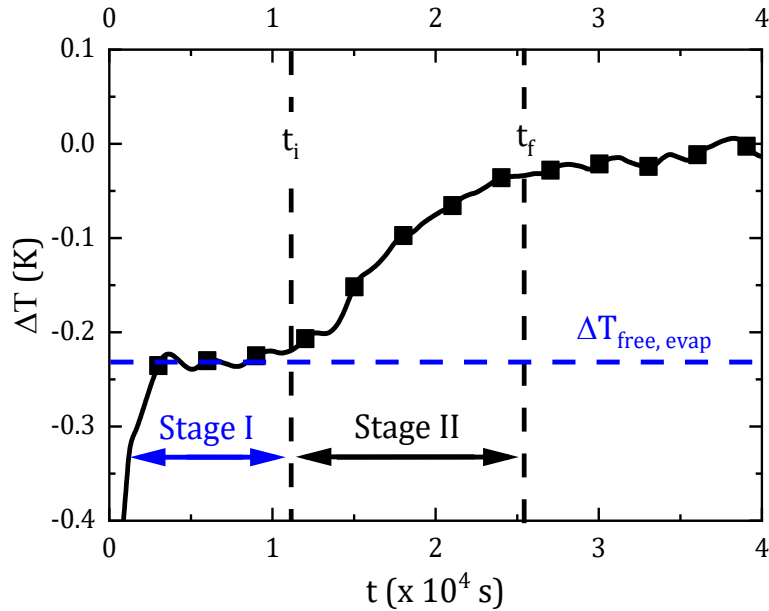


Figure 3. Evolution of the temperature difference (ΔT) between the sample and the reference surfaces during drying of a 6 mm thick bauxite residues sample with an initial solid fraction of 70% (m_b/m_t), at RH = 70% and T = 310 K. Stage I is characterised by a constant $\Delta T = \Delta T_{\text{free, evap}}$ that is independent of d suggesting that free water evaporation from wet bauxite residue is sustained by rapid capillary flow. From the onset of stage II (at $t = t_i$), the surface temperature rises as evaporation becomes increasingly limited by slow gas phase diffusion until it reaches thermodynamic equilibrium (at $t = t_f$) with its environment at specific T and RH.

211 regimes for each sample thickness at 70% RH.

212 As drying kinetics slow down significantly with increasing RH, the distinct evaporation regimes
 213 can be most clearly observed, described and analysed under these more elevated RH conditions.

214 Experimental data obtained from the 6 mm thick sample at RH = 70 % (Figure 2E, green trace) is
215 thus reproduced in Figure 3 in order to illustrate the most salient features used in the subsequent
216 discussions and analysis. The time at which surface temperatures increase to $\Delta T = \Delta T_{\text{free, evap.}} + 30$
217 mK is used to signal the onset of the smooth transition between the stage I and stage II evaporation
218 regimes and is indicated by the dashed vertical line labelled t_i , in Figure 3. Similarly, the diffusion-
219 limited stage II evaporation regime can be considered complete when $\Delta T = -30$ mK, as indicated
220 by the dotted vertical lines labelled t_f , in Figure 3. Using these somewhat arbitrary definitions, the
221 duration of the diffusion-limited stage II evaporation regime in the thermograms of Figure 2 is
222 observed to increase rapidly with sample thickness, and to decrease with RH.

223 At the end of stage II evaporation, bauxite residues samples reach a water content that should be
224 nearly equilibrated with the ambient RH at the temperature of 310 K. In order to determine the
225 water content within bauxite residues under equilibrium conditions, adsorption isotherm
226 measurements were performed at each relative humidity by studying water uptake by initially dry
227 bauxite residues samples (Figure 4). The adsorption isotherm indicates large water uptake by
228 bauxite residues under these equilibrium conditions as expected from their large SSA while an
229 important contribution must arise from their large microporosity.

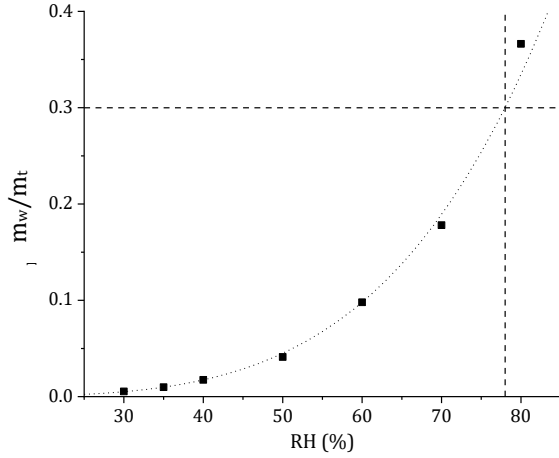


Figure 4. Adsorption isotherm of initially dry bauxite residues acquired at different RH at $T = 310$ K. The dotted line is a guide to the eye. The dashed lines highlight the fact that the equilibrium water content reaches 30 % (m_w/m_t) at $RH = 78$ %.

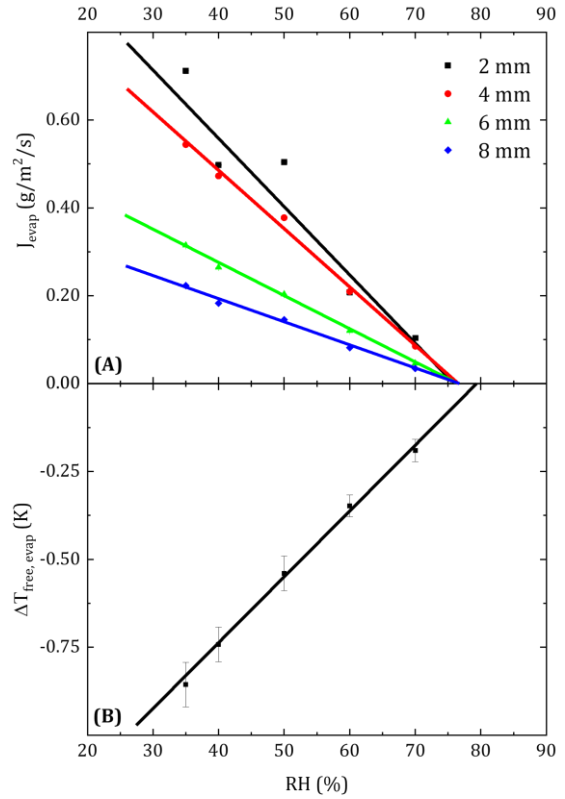


Figure 5. (A) Temperature difference in the initial stages of evaporation ($\Delta T_{\text{free, evap.}}$) as a function of relative humidity (RH). (B) Dependence of the average evaporation fluxes (J_{evap}) for each sample thickness [$d = 2$ mm (black squares), 4mm (red circles), 6mm (green triangles), 8mm (blue diamonds)] as a function of relative humidity.

230 From the initial and equilibrium water contents, the loss of water experienced during the
 231 evaporation process could be calculated. Using the total mass of water evaporated from the
 232 samples, their total evaporation time (i.e., t_f), and the sample cross section (48 ± 2 mm²), the

233 average evaporation fluxes, J_{evap} , from the bauxite residue samples could be calculated at the
234 specific RH and d of each experiment (Figure 5A). Inspection of the dependence of J_{evap} on RH
235 for each sample thickness (i.e., $d = 2, 4, 6,$ and 8 mm labeled by black squares, red circles, green
236 triangle, and blue diamonds, respectively) provides important phenomenological information. J_{evap}
237 decreases linearly with increasing RH and intercepts the x-axis at $\text{RH} = (76 \pm 4) \%$ and that,
238 irrespective of the sample thickness. This indicates that the surface boundary conditions imposed
239 by the ambient RH levels control the evaporative flux and thus, the evaporation time. In addition,
240 the temperature difference due to free water evaporation in drying stage I ($\Delta T_{\text{free, evap.}}$), reported in
241 Figure 5B, follows a linear relationship with RH, in accord with expectations from equation 1, and
242 approaches zero when $\text{RH} = (79 \pm 4) \%$. The x-intercept values for Figure 5A and 5B are consistent
243 with the adsorption isotherm of Figure 4, where the dashed lines indicate that an equilibrium water
244 content of 30% (m_w/m_t) is reached at $\text{RH} = 78 \%$. Interestingly data from Figure 5A also reveals
245 that J_{evap} increases with decreasing d which can be explained by the fact that a larger fraction of
246 the water content lies at, or near, the surface of the thinner samples where free water evaporation
247 occurs, resulting in higher average drying rates. This dependence indicates that J_{evap} is not only
248 controlled by the boundary conditions, which depend on RH, but also by long-range vapour phase
249 diffusion which inhibits water transport from the vaporisation plane to the samples' surface,
250 through the depleted evaporated interfacial zone.

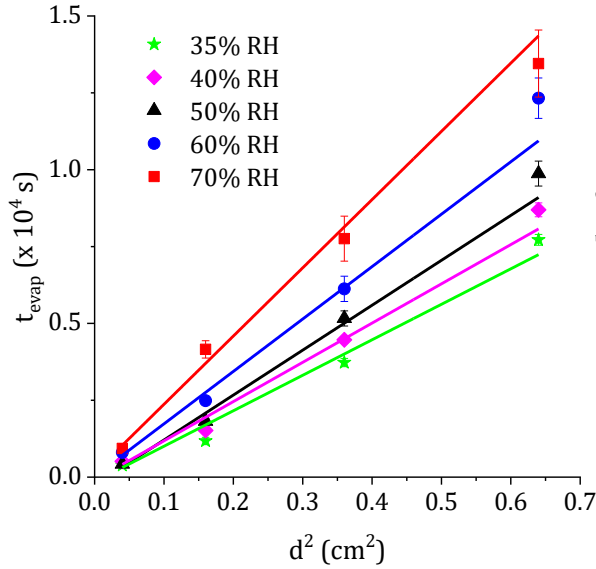


Figure 6. Stage II evaporation half-time (t_{evap}) as a function of the sample thickness squared (d^2) for RH = 35 % (green star), 40 % (magenta diamond), 50 % (black triangles), 60 % (blue circles), 70 % (red squares).

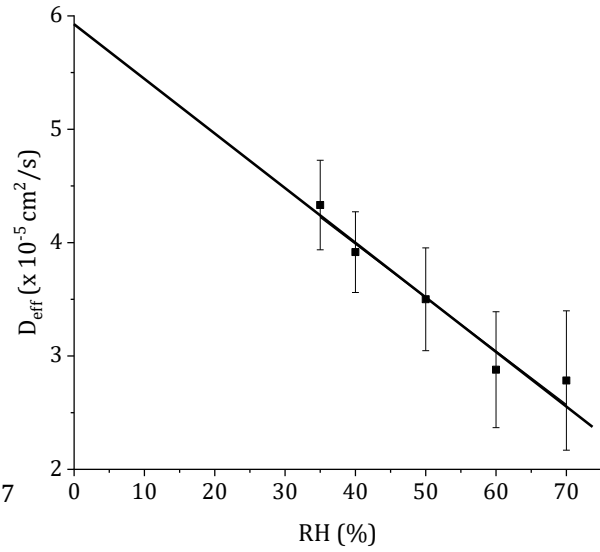


Figure 7. Dependence of the effective diffusion coefficient (D_{eff}) on the relative humidity (RH) at $T = 310$ K.

251 As stage II of the drying process is interpreted as being diffusion-limited, it is expected to follow
 252 Fick's law and to display an effective diffusion coefficient (D_{eff}). Diffusive transport would imply
 253 a linear dependence between diffusion time and the mean squared displacement of diffusing
 254 molecules whose slope could reveal the effective diffusion coefficient (D_{eff}), following $t_{evap} =$
 255 $\frac{d^2}{2D_{eff}}$, assuming 1D diffusive transport within the bauxite residues samples. Equating the mean
 256 squared displacement of diffusing molecules to the sample thickness [i.e., $\langle z^2 \rangle \sim d^2$], and using
 257 the stage II evaporation half-time (t_{evap} , defined here as $\frac{t_f - t_i}{2}$, using t_f and t_i as defined and
 258 displayed in Figure 3), Figure 6 shows that indeed, a linear relationship between t_{evap} and d^2 is
 259 obeyed. In addition to corroborating the diffusive nature of the rate limiting process for water

260 evaporation in late stage drying, an effective diffusion coefficient for water in bauxite residues can
261 be obtained using linear fits to the experimental data in Figure 6.

262 The effective diffusion coefficients obtained at each RH are reported in Figure 7 and,
263 interestingly, they decrease rapidly with increasing RH. Furthermore, these effective diffusion
264 coefficients are four orders of magnitude smaller than those commonly measured for gas phase
265 diffusion (Jabro, 2009). This must therefore reflect the fact that water diffusion is hindered by the
266 microstructure and water adsorption propensity of the medium, properties that diminish the
267 effective diffusion coefficient through the interstitial air within the bauxite residues interconnected
268 porous microstructure. Indeed, when diffusive transport occurs in porous media, D_{eff} can be
269 described using the molecular diffusion coefficient in air (D_{air}), the porosity of the sample ($\varphi =$
270 0.64 ± 0.06) and the tortuosity of its pore structure ($\tau = 1.38 \pm 0.07$) (Ho & Webb, 2006).
271 Contributions arising from adsorption of water on pore surfaces are described using parameters
272 that account for the water/bauxite residues interactions, namely an equilibrium constant for
273 adsorption (K_{ads}), for the bauxite residues mass-to-pore volume ratio ($r_{sp} = 0.7 \pm 0.1 \text{ g/cm}^3$) and
274 their specific surface area [$SSA = (5 \pm 1) \times 10^5 \text{ cm}^2/\text{g}$] (Bartels-Rausch et al., 2013; Fabre, Barnola,
275 Arnaud, & Chappellaz, 2000; Schwander et al., 1993; Seok et al., 2009):

$$276 \quad D_{eff} = D_{air} \cdot \left(\frac{\varphi}{\tau}\right) \cdot \left(\frac{1}{1 + r_{sp} \times K_{ads} \times SSA}\right) \quad [3]$$

277 Equation 3 also provides a framework to understand the dependence of D_{eff} with RH. As the
278 vaporisation plane leaves the surface of the bauxite residues, the overlying depleted zone, which
279 remains in a quasi-steady state with the environmental conditions, acts as a diffusion barrier. When
280 the RH increases, the water content in the evaporated interfacial layer, which is in a dynamic
281 equilibrium with ambient air temperature and relative humidity, also increases. Consequently, the

282 air-filled porosity decreases, and the tortuosity of the pore structure increases, yielding a decrease
283 in D_{eff} with increasing RH as observed in Figure 7.

284 Given that the morphological parameters, ϕ and τ , can only account for an utmost factor of
285 $\frac{\phi}{\tau} \sim 0.5$ reduction in D_{eff} compared to D_{air} , water adsorption on bauxite residue surfaces appears
286 to be crucial in order to explain the smallness of D_{eff} . While SSA, r_{sp} , ϕ , and τ values are well
287 established for dry bauxite residues (i.e., 0 % m_w/m_t), the equilibrium constant for adsorption,
288 K_{ads} , for water for this material is unfortunately still unknown. A linear extrapolation of D_{eff}
289 with RH enables the diffusion coefficient of water in dry bauxite residues to be estimated as D_{eff}
290 $= (5.9 \pm 0.3) \times 10^{-5}$ cm²/s at RH = 0 %. Using Equation 3, along with the known morphological
291 parameters of dry bauxite residues, a value for $K_{ads} = (5 \pm 1) \times 10^{-3}$ cm is obtained. The magnitude
292 of K_{ads} for bauxite tailings is observed to be greater than those reported for loam soils and alumina
293 active F-200 for example, that is $(6 \pm 1) \times 10^{-4}$ cm (Gary, Kohl, & Taylor, 1964) and $(6.6 \pm 0.9) \times$
294 10^{-5} cm (Serbezov, 2003), respectively. Therefore, water vapour strong propensity towards
295 adsorption onto bauxite residues pore surfaces, along with their large SSA, are suggested to be the
296 dominant contributors to the large decrease of D_{eff} compared to D_{air} , and the slow drying kinetics
297 displayed in stage II evaporation regime.

298

299 CONCLUSIONS

300 Thermal imaging was demonstrated to be an efficient and innovative tool for monitoring
301 surface evaporation from bauxite residues and establishing the underlying drying mechanisms. A
302 thorough analysis of the experimental data enabled a deeper understanding of the evaporation
303 kinetics and effective diffusion coefficients to be quantified. Water adsorption on bauxite residues

304 is demonstrated to be the dominant phenomenon in the rate-limiting mechanism in the later
305 evaporation stage and to be responsible for the smallness of the effective diffusion coefficient. The
306 evaporated interfacial layers therefore act as a diffusion barrier to water transport from the
307 vaporization plane in the deeper layers of the bauxite residues to their surface, inhibiting
308 considerably their drying rates.

309 A limitation of the thermal imaging approach is that it can only provide average evaporation
310 fluxes. Indeed, as soon as the vaporization plane leaves the sample surface, contributions from the
311 thermal gradient through the bauxite residues temperature depth profile arise, preventing
312 instantaneous drying rates to be established and finer details in the evolution of the dominant water
313 transport mechanisms to be elucidated. Concomitant gravimetric and thermal imaging
314 measurements should provide us with further valuable mechanistic insight.

315 This methodology could also be expanded to environmental conditions relevant to field
316 applications, namely the temperature dependence of the drying kinetics, especially those
317 encountered under Winter conditions where capillary transport will be strongly suppressed
318 (Feddes, Kabat, Van Bakel, Bronswijk, & Halbertsma, 1988). Environmental mitigation efforts
319 could benefit greatly from improved understanding of the drying rates under such Winter condition
320 where acute environmental consequences result from dust particles scattering from BRSA over
321 snow-covered neighbouring urban communities (Aoki et al., 2006; Warren & Wiscombe, 1980).
322 Collectively, these investigations should improve our ability to forecast and prevent the occurrence
323 of fugitive dust emissions as well as to devise efficient risk assessment and monitoring tools to
324 assist in mining residue management efforts.

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