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# 1 $\epsilon$ -greedy automated indentation of cementitious materials 2 for phase mechanical properties determination

3  
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5  
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11

## 12 **ABSTRACT**

13 Microindentation and nanoindentation tests are very useful to assess the micromechanical  
14 properties of materials. However, statistical indentation tests require large matrices of indents  
15 for heterogeneous materials, which can be time-consuming. Given that specific properties of  
16 only one phase of interest can be looked for, a novel procedure is proposed to locate indents  
17 automatically. The procedure uses an  $\epsilon$ -greedy strategy to determine the next indents to  
18 perform. The strategy is informed by the nature of predicted indents using unsupervised  
19 clustering of the indentation curves and Gaussian Process Classification. The influence of  
20 several parameters has been assessed, and the best combination performance has been  
21 quantified in the case of the microindentation of a cementitious mortar sample (500 mN) and  
22 the nanoindentation of a cement paste (1 mN). The proportion of indents in the phase of interest  
23 increased by around 20% in both cases, paving the way for faster indentation experiments.

## 24 HIGHLIGHTS

- 25 •  $\epsilon$ -greedy strategies can automatically locate the next indents to perform in a phase of  
26 interest during indentation.
- 27 • The technique is used for cementitious materials at the micro- and the nanoscale.
- 28 • The proportion of indents in the phase of interest can increase by around 20%.
- 29 • Indentation curves are interpreted by unsupervised learning and Gaussian processes.
- 30 • The algorithm's parameters can be tuned to increase the global performance.

## 31 KEYWORDS

32 Indentation; Machine Learning; Phase properties;  $\epsilon$ -greedy strategy; Material Characterization;  
33 Cementitious materials

## 34 1. Introduction

35 Indentation is one of the primary techniques used to assess the mechanical properties of  
36 materials at various scales and can typically help design new materials or assess materials'  
37 durability against degradation. Microindentation and nanoindentation have been extensively  
38 developed during the last decades to measure the elastic [1,2] and viscoelastic properties of  
39 material phases [3–5] or hardening properties [6]. Load ranges of around some millinewtons,  
40 resp. nanonewtons, employed in microindentation, resp. nanoindentation, typically induce  
41 penetration depth of some hundreds of nanometers or micrometers, resp. tens of nanometers,  
42 providing information about the local mechanical properties. Hardness, indentation modulus  
43 [7], and eventually parameters influencing time-dependent properties such as the creep modulus  
44 can be derived based on the indentation measurements [8–11].

45 The grid indentation technique, consisting of performing a regular grid of hundreds or  
46 thousands of indents over of representative area of heterogeneous material, has been employed

47 to supplement single indentation results [12,13]. Grid indentation measurements usually take  
48 some tens of hours under precisely-controlled experimental conditions. Using deconvolutions  
49 techniques on the outputs such as least square optimization [14], unsupervised clustering such  
50 as k-means [15], hierarchical clustering [16] or Gaussian mixture models [17], the relative  
51 proportions of the various phases and their main mechanical properties can be inferred. The  
52 phase properties can then help monitor the material evolution during manufacturing or any  
53 eventual degradation. The elastic or viscoelastic properties of the phases also help assess the  
54 global material properties using analytical or numerical homogenization techniques [18–21] or  
55 the full-field measurement approach [22].

56 Importantly, some phases in heterogeneous materials might need specific attention, as they  
57 constitute the most sensitive phase subjected to mechanical properties evolutions and  
58 alterations. For example, in cementitious materials such as concrete, the major phases at a pluri-  
59 micrometric scale are aggregate, sand and cement paste. Cement paste evolution attracts most  
60 of the researchers' attention as it might develop more or less stiffness and strength during  
61 hydration, depending on the initial mix or because of the various degradations it might face due  
62 to adverse environmental conditions such as leaching [18,23], shrinkage [24], sulfate attack  
63 [25,26], chloride ingress [27] or irradiations [28]. At the nanoscale, cement paste comprises  
64 several hydrated phases, the major ones being calcium silicate hydrates (C-S-H) gel and  
65 portlandite crystals. Depending on the nature of the phenomenon inducing mechanical  
66 properties changes, one of the two phases is generally altered first, e.g., portlandite concerning  
67 leaching for example, or, for instance, C-S-H during the initial hydration depending on the mix  
68 composition and the relative proportion of supplementary cementitious materials used to lower  
69 the environmental impact of concrete [29]. For this reason, more precise information about a  
70 phase of interest can be required. Additional tests have been coupled with indentation, such as  
71 imaging [16,30,31] or chemical analysis [32], both at a microscale [33,34] or a macroscale [35].

72 Although adding experimental work, these tests provide interesting information that can help  
73 refine the deconvolution and allow the identification of a phase of interest more precisely.  
74 Besides, Bayesian approaches, based on indentation curves alone, have recently been developed  
75 to assess more precisely the mechanical properties of various phases during nanoindentation  
76 [36].

77 To circumvent the drawbacks of grid indentation to assess the properties of one particular phase  
78 of interest in a heterogeneous material, the possibility of locating indents in a phase of interest  
79 during the indentation process, i.e., online, is investigated herein. Innovative online algorithms  
80 and, more particularly, exploration-exploitation strategies are being developed for some years  
81 for automating the speeding of specific tasks. Map exploration problems can be tackled without  
82 particular initial knowledge, for example, to guide robots exploration effectively [37,38]. Using  
83 artificial intelligence techniques and increasing machine knowledge during exploration [39],  
84 objectives, such as finding specific objects in a map or exploring this map, can be fulfilled in  
85 several environments such as households [40] or industrial sites [41].

86 In this study, an exploration-exploitation strategy, namely,  $\epsilon$ -greedy, has been developed to  
87 locate the highest number of indents in a phase of interest during the microindentation or  
88 nanoindentation process of typical heterogeneous materials, e.g., mortar and cement paste.  
89 According to the strategy, after a rather loose initial grid indentation test, some specific indents  
90 were gradually selected among the remaining potential indent locations. To determine those  
91 locations, the proposed algorithm is based on an  $\epsilon$ -greedy strategy informed by a Gaussian  
92 process classification to determine the nature of unknown indents, and unsupervised clustering  
93 to separate the performed indents into groups related to the phases in the material. The proposed  
94 method is satisfactorily compared to grid indentation performance, e.g., the random selection  
95 of the indents.

96 The article is structured as follows: the first section describes the two materials used in the  
97 study, e.g., cementitious mortar and cement paste, and the numerical methods are then  
98 explained. Two versions of the algorithm are detailed to decouple the relative influence of the  
99 input parameters: one offline algorithm that can regularly check the indents nature based on  
100 microscopic information, and one online algorithm only informed by indentation curves. The  
101 results are presented in the following section. First, the convergence of the algorithm, the main  
102 parameters of the GPC and the  $\epsilon$ -greedy strategy are discussed using the ‘offline’ algorithm.  
103 Secondly, the efficiency of the online algorithm that considers only the unsupervised clustering  
104 of the indentation curves is then evaluated. Conclusions are finally drawn concerning the  
105 interest of these methods, and future research directions are proposed.

## 106 2. Materials and methods

### 107 2.1 Experimental methods

#### 108 2.1.1 Preparation of cement paste and mortar samples

109 Mortar specimens and cement paste specimens were prepared using CEM I 52.5 cement and  
110 0/4 mm calcareous sand. The mortar formulation was determined to be as representative as  
111 possible of high-performance concrete. Both formulations are reported in Table 1.

112  
113 **Table 1.** Mortar and cement paste compositions (kg / m<sup>3</sup>)

Specimen	Cement	0/4 Sand	Water	w/c ratio
Mortar	566	1344	270	0.43
Cement paste	1338	0	575	0.43

114

115 One mortar and one cement paste prisms with dimensions  $4 \times 4 \times 16$  cm<sup>3</sup> were cast in  
116 polypropylene molds to avoid any presence of metallic compounds from the molds. After one

117 day of curing under sealed conditions in an air-conditioned room at a temperature of 20°C and  
118 90% RH, the specimens were unmolded and further cured in lime-saturated water until the age  
119 of 28 days.

120 After 28 days, a 1.5 cm-thick  $4 \times 4 \text{ cm}^2$  slice was cut in the center of the specimen using a  
121 precision saw. The samples were cut again to obtain a  $1.5 \times 2.0 \times 2.0 \text{ cm}^3$  cubic specimen.  
122 These two specimens were embedded in resin and automatically polished before the indentation  
123 measurements. Mortar specimen and cement paste samples were polished with Si-C paper with  
124 decreasing particle size (500, 1200, 2000, 4000 grit) using alcohol-based polishing liquid to  
125 avoid any reaction with unhydrated cement particles. Polishing times were selected from some  
126 seconds (500 paper) to around some minutes per paper (4000 paper) to limit the risk of  
127 aggregate cracking. Finally, the samples were polished using  $1 \mu\text{m}$  diamond paste for 15  
128 minutes. A root mean square of the surface roughness ( $R_q$ ) of around 200 nm, resp. 80 nm, was  
129 measured for the mortar, resp. cement paste sample.

130

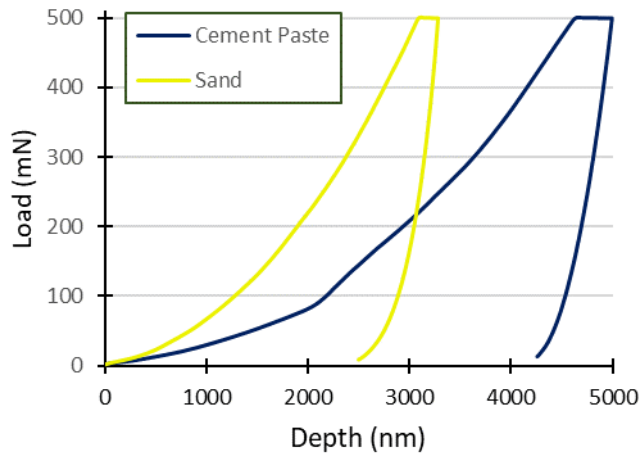
### 131 2.1.2 Grid indentation tests

132 To investigate representative surfaces of the mortar sample and the cement paste sample resp.,  
133 microindentation and nanoindentation tests were performed using a Berkovitch indenter  
134 (Bruker TS 77) probe over a grid of  $40 \times 40$  points (1600 indents), evenly spaced by  $500 \mu\text{m}$ ,  
135  $2 \mu\text{m}$  resp. For each indent, the load was increased linearly over time in 5 s up to 500 mN, resp.  
136 1 mN, kept constant during the 100 s holding phase, and decreased linearly over time in 5 s.  
137 The short loading time was selected to limit creep during this period and did not damage the  
138 sample (as checked under a microscope of an SEM after indentation). In total, the test lasted  
139 around four days to perform the entire grid, which motivates the development of a faster yet  
140 reliable method to assess the mechanical behavior of heterogeneous materials. Typical

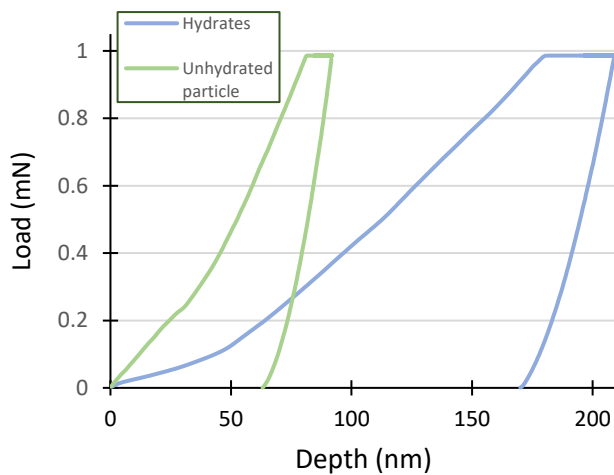
141 indentation curves of the two major phases in the mortar sample and the cement paste samples  
142 are represented in Fig. 1.

143

a)



b)



144 **Fig. 1.** Typical indentation curves: a) cement paste and sand phases in the mortar specimen, b)

145 hydrate and unhydrated particle in the cement paste..

146

147 Reduced modulus  $E_r$  and indentation hardness  $H_{IT}$  were calculated according to equations (1)

148 and (2):



$$E_r = \frac{1}{2} \sqrt{\frac{\pi}{A_c}} S \quad (1)$$

149

$$H_{IT} = \frac{P_{max}}{A_c} \quad (2)$$

150 Where  $A_c$  is the projected contact area and  $S$  the slope of the unloading curve using Oliver and  
151 Pharr approach [7]. No specific filtering was performed on the indentation curves to represent  
152 a general indentation acquisition.

153

### 154 2.1.3 Acquisition of indented area image and generation of representative artificial images

155 Photographs of the indented zone were acquired after indentation to visually identify indent  
156 location or, resp., deduce indent location, in the case of the mortar and cement paste specimens  
157 resp.

158 As illustrated in Fig. 2 (a), a picture of the indented zone on the mortar sample has been obtained  
159 using a Hirox RH-2000 3D microscope by merging around 200 3D reconstructed images evenly  
160 spaced along the indented area using a  $140\times$  magnification leading to a final horizontal  
161 resolution of the 2D projected image of  $1.5 \mu\text{m} / \text{pix}$  that is adequate to locate the indents and  
162 assess their nature. The high proportion of sand particles of various sizes, creating an optimized  
163 granular skeleton typical of high-performance concrete, is worth noting.

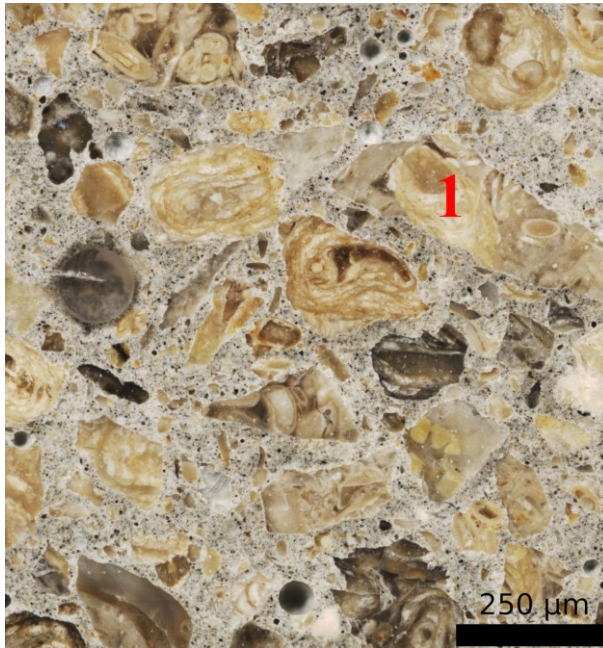
164 As illustrated in Fig. 2 (b), a SEM image of the cement paste sample was obtained using an  
165 acceleration voltage of 15 kV and a working distance of 7 mm and a low vacuum pressure of  
166 60 Pa to limit the risk of cracking. A relatively large light gray unhydrated particles was found  
167 in the left part of the indented zone, while four smaller unhydrated particles were located in the  
168 right part of the image. Most of the indents were not visible due to the relatively small load

169 compared to the roughness of the sample, except some small imprints on the unhydrated  
170 particles.

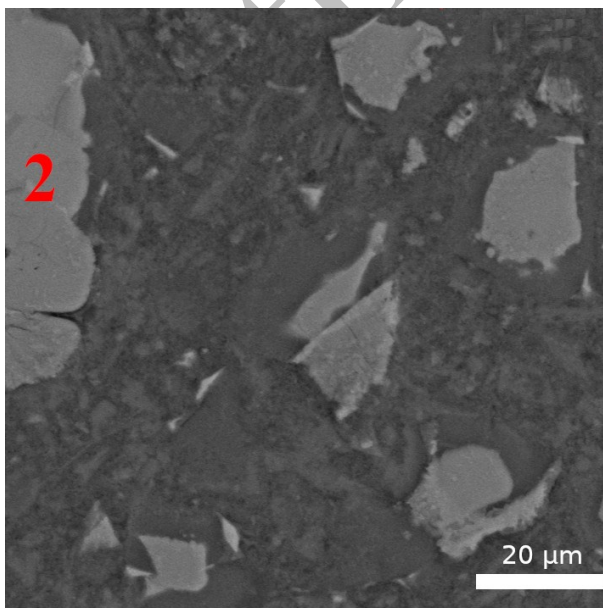
171

172

a)



b)



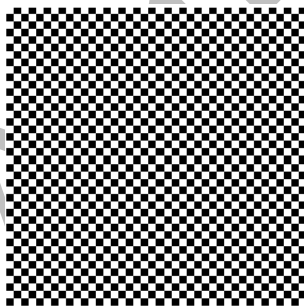
173 **Fig. 2.** Microscopic image of the samples: a) 2D optical microscopic image of mortar sample  
174 with one sand particle labeled 1 (1cm  $\times$  1cm zone indented using a 40  $\times$  40 matrix of 500mN  
175 indents), b) SEM image of cement paste sample with one unhydrated particle labeled 2  
176 (80 $\mu$ m  $\times$  80 $\mu$ m zone indented using a 40  $\times$  40 matrix of 1 mN indents (indents are not visible)).  
177

## 178 2.2 Numerical methods

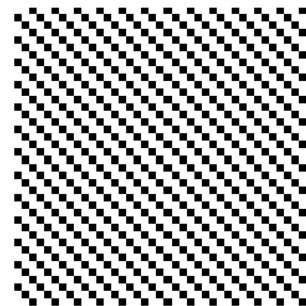
### 179 2.2.1 Kriging

180 Initial values, either indent nature based on the microscopic images or indentation curves, were  
181 acquired initially before running the  $\epsilon$ -greedy procedure in order to gain initial knowledge  
182 before inferring the unknown indent nature using Gaussian Process Classification. To this end,  
183 an initial regular Kriging stage has been performed with various Kriging parameters from K=2  
184 (half of the indents were initially artificially performed) to K=20 (1/20th of the indents were  
185 selected). The initial sampling sets for the different Kriging parameters are illustrated in Fig. 3.  
186 Considering the maximum possible number of indents being 1600 (40  $\times$  40), the initial number  
187 of indents after the initial Kriging stage ranged from 80 (K=20) to 800 (K=2).

a) K=2

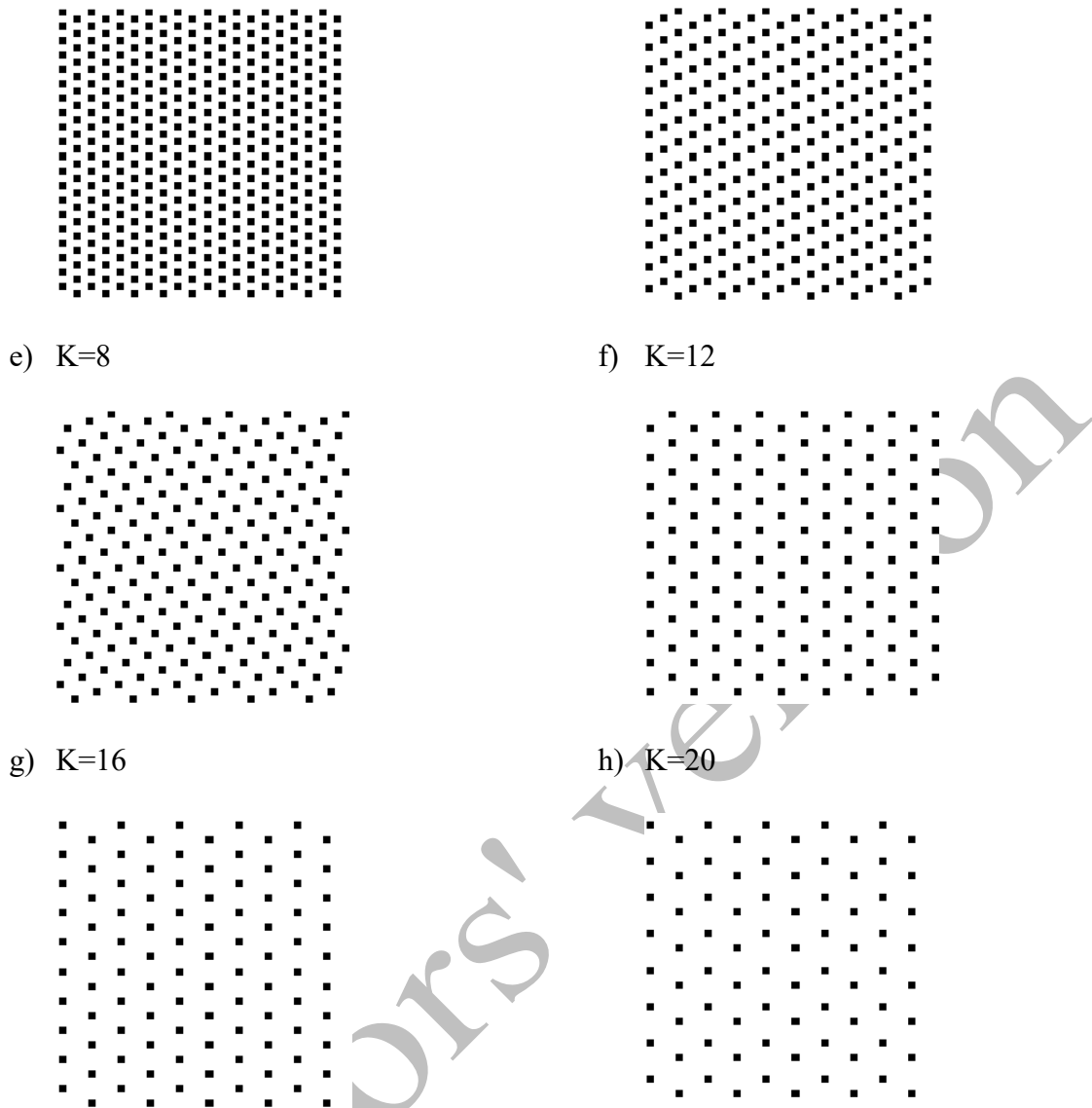


b) K=3



c) K=4

d) K=6



188 **Fig. 3.** Initial sampling sets of indents performed on the sample using various Kriging  
 189 parameters (dense to loose Kriging): a) K=2, b) K=3, c) K=4, d) K=6, e) K=8, f) K=12, g)  
 190 K=16, h) K=20.

191

### 192 2.2.2 Gaussian Process Classifier

193 After the initial Kriging stage, and regularly during the algorithm iterations, Gaussian Process  
 194 (GP) classifiers were used to infer the nature of unknown indents based on the knowledge of  
 195 the nature of the surrounding indents. GP classifiers [42] are non-parametric classifiers, relevant  
 196 to the resent problem as they require few examples to perform correctly. Given data points  $x_i$

197 from a domain  $X$  with corresponding class labels  $y_i$  in  $[-1; +1]$ , one would like to predict the  
198 class membership probability for a test point  $x$ . This is achieved using a latent function  $f$  whose  
199 value is mapped into the unit interval employing a sigmoid function  $\sigma : \mathbb{R} \rightarrow [0; 1]$ , used  
200 because of its desirable mathematical properties, such that the class membership probability  
201  $P(y = +1 | x)$  can be written as  $\sigma(f(x))$ . Under some conditions, the likelihood can be written  
202 as  $P(y | x) = \sigma(yf(x))$ .

203 A GP [42] is a stochastic process fully specified by a mean function  $m(x) = E[f(x)]$  and a  
204 positive definite covariance function  $k(x, x') = V[f(x), f(x')]$ . This means that a random  
205 variable  $f(x)$  is associated with every  $x$  in  $X$ , such that for any set of inputs  $X$ , the joint  
206 distribution  $P(f | x) = \mathcal{N}(f | m_0, K)$  is Gaussian with mean vector  $m_0$  and covariance matrix  
207  $K$ , conveniently termed kernel.

208 The factorial likelihood being non Gaussian, the posterior over the latent values is also not  
209 Gaussian. In this paper, the Laplace approximation is used for approximating the non-Gaussian  
210 posterior by a Gaussian.

211 Assuming without loss of generality  $m_0 = 0$ , one still has to define  $K$ , whose design will  
212 enforce specific properties of the metric space of  $X$ . Kernels encode the assumptions on the  
213 function being learned by defining the ‘similarity’ of two  $x_i$  combined with the assumption that  
214 similar  $x_i$  should have similar target values.

215 Among the wide range of kernels available, the most common stationary and non-stationary  
216 kernels with a small number of parameters have been tested in this paper: the Radial Basis  
217 Function (RBF) kernel with its rational quadratic extension, the Matern one and the Dot Product  
218 kernel. The combination of Matern and Dot Product kernels, and Matern and Dot Product  
219 squared have been also evaluated.

220 The RBF kernel is the most commonly used kernel and also known as the ‘squared exponential’  
221 kernel and is given by the following:

$$k(x_i, x_j) = \exp\left(-\frac{d(x_i, x_j)^2}{2l^2}\right) \quad (3)$$

222 Where  $d(\cdot, \cdot)$  is the Euclidean distance and  $l$  a length-scale parameter.

223 The matern kernel is a generalization of the RBF. It has an additional parameter that controls  
 224 the smoothness of the resulting function. When set to  $3/2$  it ensures that the functional function  
 225 is differentiable at least once, and gives:

$$k(x_i, x_j) = \left(1 + \frac{\sqrt{3}}{l} d(x_i, x_j)\right) \exp\left(-\frac{\sqrt{3}}{l} d(x_i, x_j)\right) \quad (4)$$

226

227 The RationalQuadratic kernel is given by the following:

$$k(x_i, x_j) = \left(1 + \frac{d(x_i, x_j)^2}{2\alpha l^2}\right)^{-\alpha} \quad (5)$$

228 Where  $\alpha$  is a scale mixture parameter.

229

230 This kernel can be seen as a scale mixture of RBF kernels with different characteristic length-  
 231 scales. It is parameterized by a length-scale parameter  $l$  and a scale mixture parameter  $\alpha$  that  
 232 must both be positive. Those kernels are termed stationary because they depend solely on the  
 233 radial distance between  $x_i$ .

234 The last kernel evaluated in this study was the dot product kernel. Conversely, the dot product  
 235 is a non-stationary kernel as it depends on the value of the input coordinates themselves:

$$k(x_i, x_j) = \sigma_0^2 + x_i \cdot x_j \quad (6)$$

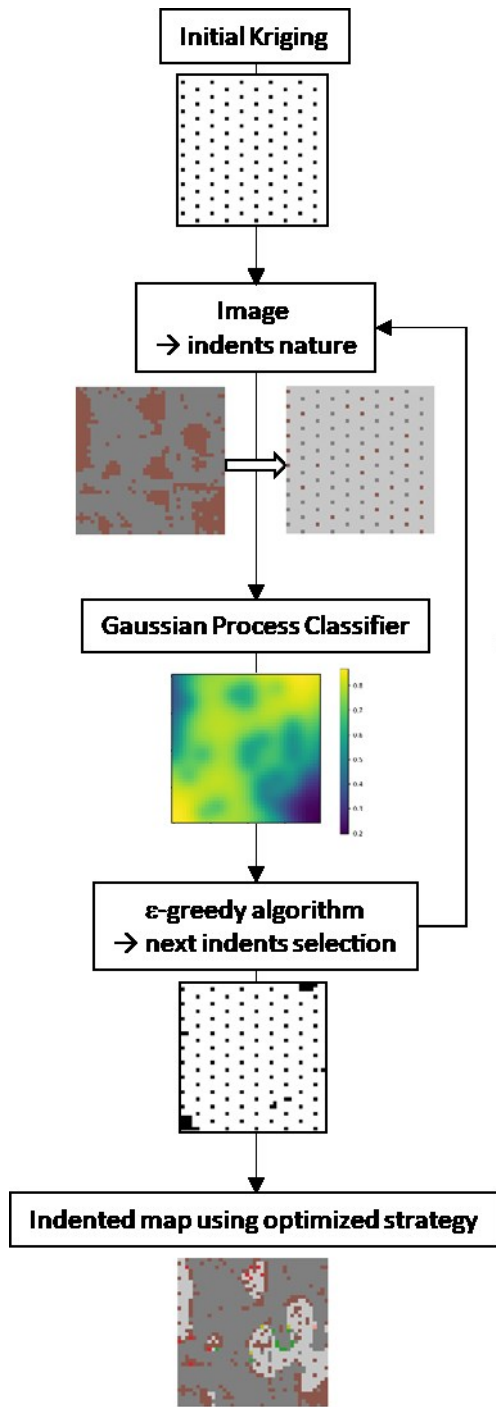
236 This kernel is parameterized by the  $\sigma_0$  parameter. The dot product kernel is commonly combined  
 237 with exponentiation. For this reason, Matern + Dot Product and Matern + Dot Product<sup>2</sup> have  
 238 been studied herein. For all tested configurations, kernel parameters were optimized during the  
 239 training procedure.

240

### 241 2.2.3 $\epsilon$ -greedy algorithm for indents location selection

#### 242 2.2.3.1 *Prediction using indents nature identified by microscopic images as priors*

243 The methodology diagram reported in Fig. 4 describes the algorithm that considers the indents  
244 nature identified by microscopic images (offline algorithm) as priors to predict the next indent  
245 to be performed. After an initial Kriging step, the nature of the selected indents, either the phase  
246 of interest or not, was obtained based on microscopic image analysis through greyscale  
247 thresholding in the case of the cement paste or manual annotation in the case of the mortar  
248 specimen (due to the low contrast between cement paste and sand particles). Using this  
249 information, a GP classifier was then used to infer the nature of the other indents in the  
250 indentation map. Based on this inference, the next indents to perform were selected following  
251 an  $\epsilon$ -greedy strategy, i.e., 80% of the indents was selected greedily where the GPC predicted  
252 the highest probability of finding the phase of interest and 20% of indents was selected  
253 according to an exploration strategy whose influence will be discussed in the results. The nature  
254 of these indents has been assessed using microscopic images and the sequence ‘indents nature  
255 verification – GPC –  $\epsilon$ -greedy strategy’ process has been repeated during tens of steps to  
256 perform as many indents as possible in the phase of interest given the indentation zone. The  
257 main goal of this algorithm was to independently study as many parameters as possible from  
258 GPC and the  $\epsilon$ -greedy strategy. The influence of these parameters will be discussed in section  
259 3.1.



Version

260

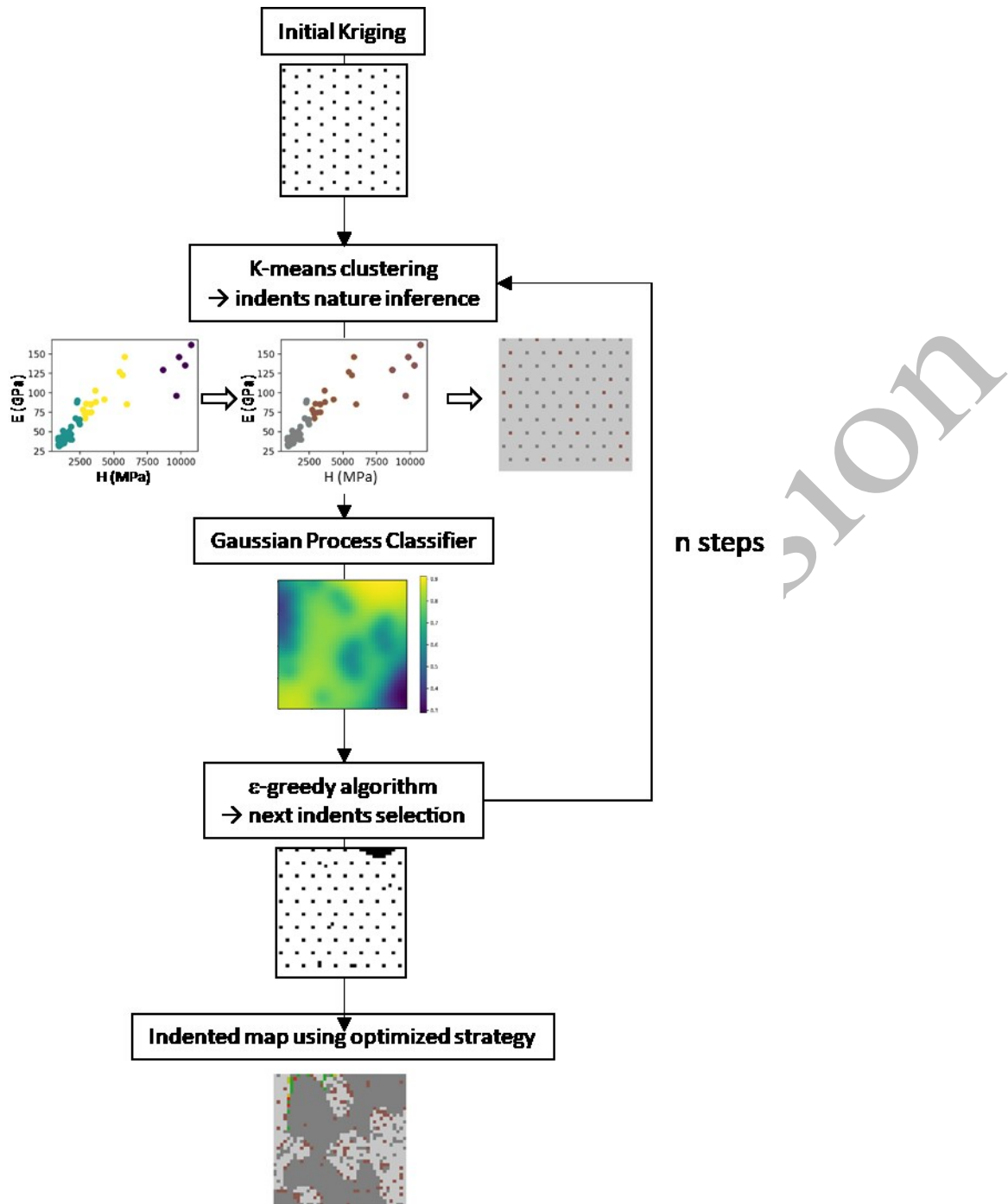
261 **Fig. 4.** Flowchart of the  $\epsilon$ -greedy algorithm for indents location selection using geometrical  
 262 information.

263 *2.2.3.2 Algorithm using indentation curves without geometrical information*

264 The methodology diagram reported in Fig. 5 shows the principle of the full-fledged algorithm  
 265 that performs online estimation of phase of interest. This algorithm could be implemented on



266 indenters to locate as many indents as possible in a phase of interest during the indentation of a  
267 heterogeneous material. After an initial rather loose Kriging step, the nature of the selected  
268 indents was inferred using unsupervised clustering (k-means) performed on the calculated  
269 micro-mechanical properties. The unsupervised algorithm inputs were  $E_r$  and  $h_{max}$ , the  
270 maximum penetration depth for the mortar specimen and  $E_r$  and  $H_{IT}$  in the case of the mortar,  
271 resp. cement paste specimen to guarantee the convergence of the overall procedure in both  
272 cases. k-means algorithm has been randomly initialized ten times, and convergence has been  
273 obtained during the first 300 runs with a relative tolerance of  $1e-4$  with regards to Frobenius  
274 norm. Three classes were estimated corresponding to indents in a) the phase of interest, b)  
275 indents in the other phase and c) indents with intermediate properties, likely located at  
276 interfaces. The latter two classes were grouped considering indents should not be similar to  
277 these indents. Based on this information, a GP classifier was then used to infer the nature of all  
278 the indents in the indentation map. From this inference, the next indents to perform were  
279 selected following the same  $\epsilon$ -greedy strategy as described in the previous section, i.e., 80% of  
280 the indents were selected greedily where the GPC predicted the highest probability of finding  
281 the phase of interest and 20% of indents were selected according to an exploration strategy. The  
282 nature of these indents has then been assessed using unsupervised clustering, and the overall  
283 'indents nature verification – GPC –  $\epsilon$ -greedy strategy' process has been repeated during tens  
284 of steps to perform as many indents as possible in the phase of interest in the indentation zone.



285

286 **Fig. 5.** Flowchart of the  $\epsilon$ -greedy algorithm for indents location selection using indentation

287 curves.

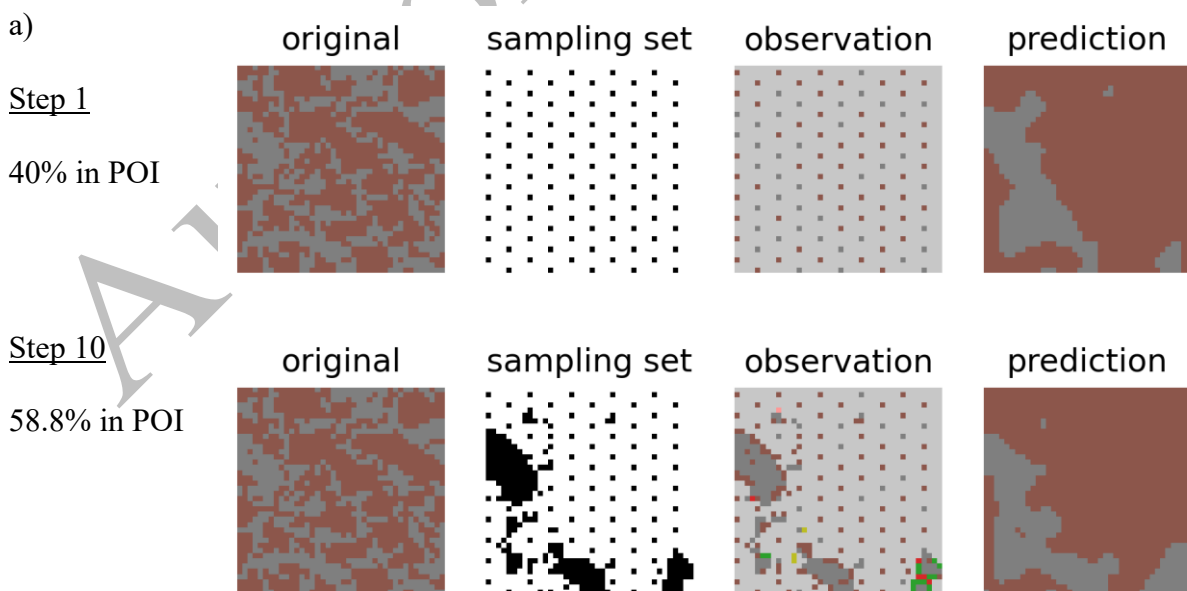
288

289 **3. Results and discussion**

290 *3.1 Algorithm using geometrical information only*

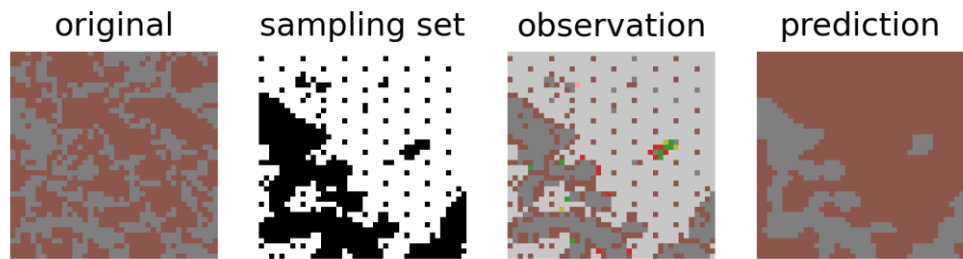
291 3.1.1 Convergence of the algorithm

292 The efficiency of the  $\epsilon$ -greedy algorithm using indent nature given by a microscopic image has  
293 been validated for the microindentation test of the mortar sample and the nanoindentation test  
294 of the cement paste sample. As illustrated in Fig. 6, after the first initial Kriging stage that  
295 provides information about the nature of about 100 indents located on the area of interest, the  
296 proposed algorithm was able to detect zones with higher probabilities of finding indents of  
297 interest. The exploration-exploitation strategy automatically then selected the next indents to  
298 perform. The algorithm has been run during several steps, typically from 20 to 50 steps,  
299 selecting 20 indents (1.25% of the total number of indents) greedily at each step. Those indents  
300 are selected as the ones with the highest probability of locating paste or hydrates areas in the  
301 case of the mortar sample, cement paste sample, resp. Additionally, 5 indents have been selected  
302 according to the exploration strategy.



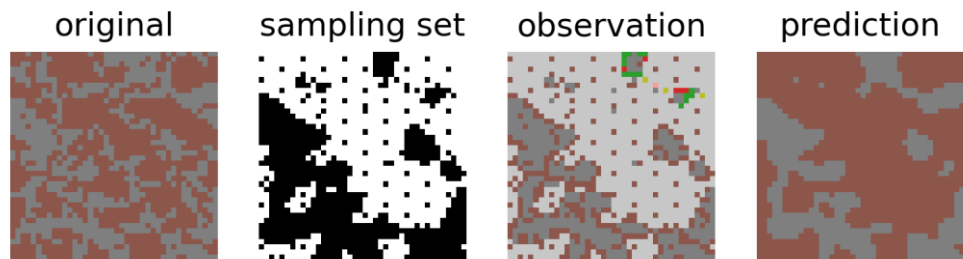
Step 20

56.1% in POI



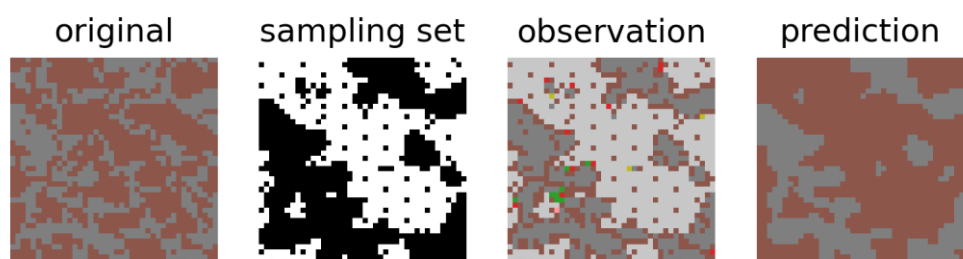
Step 30

56.2% in POI



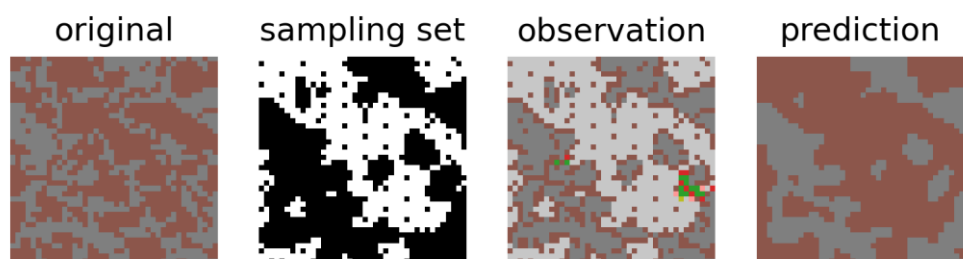
Step 40

56.0% in POI



Step 50

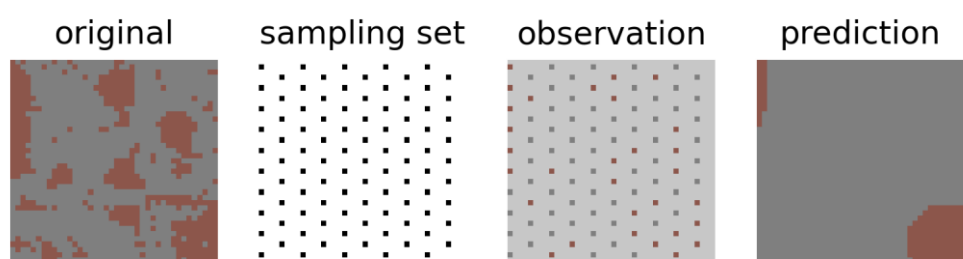
55.6% in POI



b)

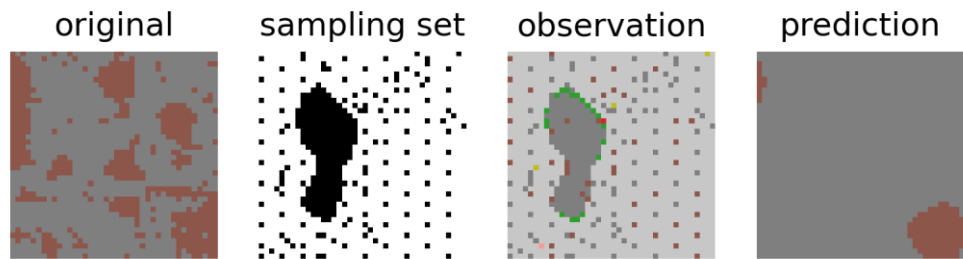
Step 1

69.0% in POI



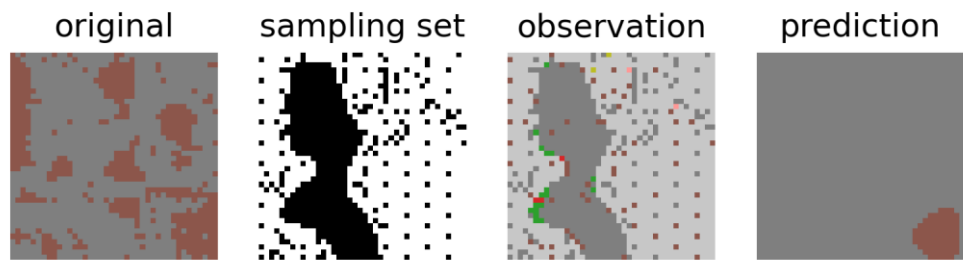
Step 10

84.9% in POI



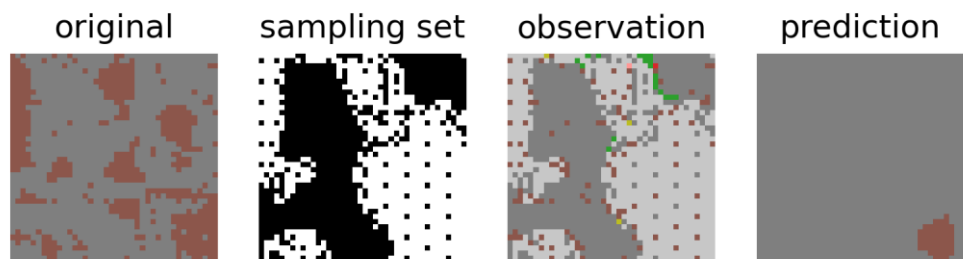
Step 20

87.8% in POI



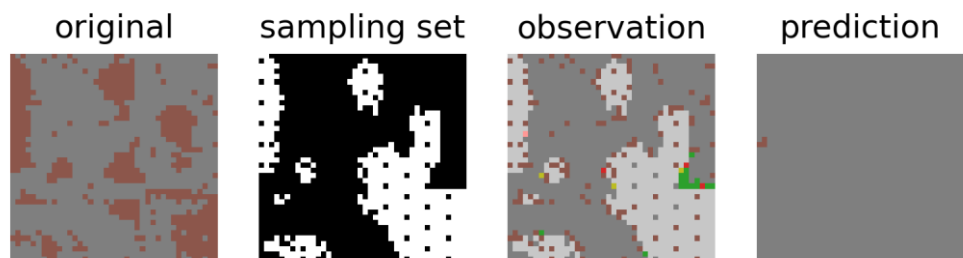
Step 30

86.5% in POI



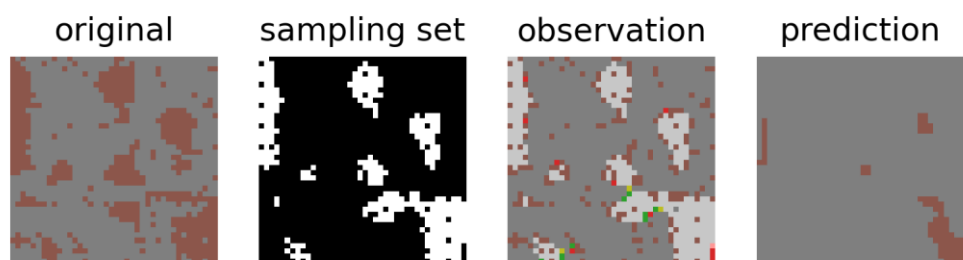
Step 40

86.5% in POI



Step 50

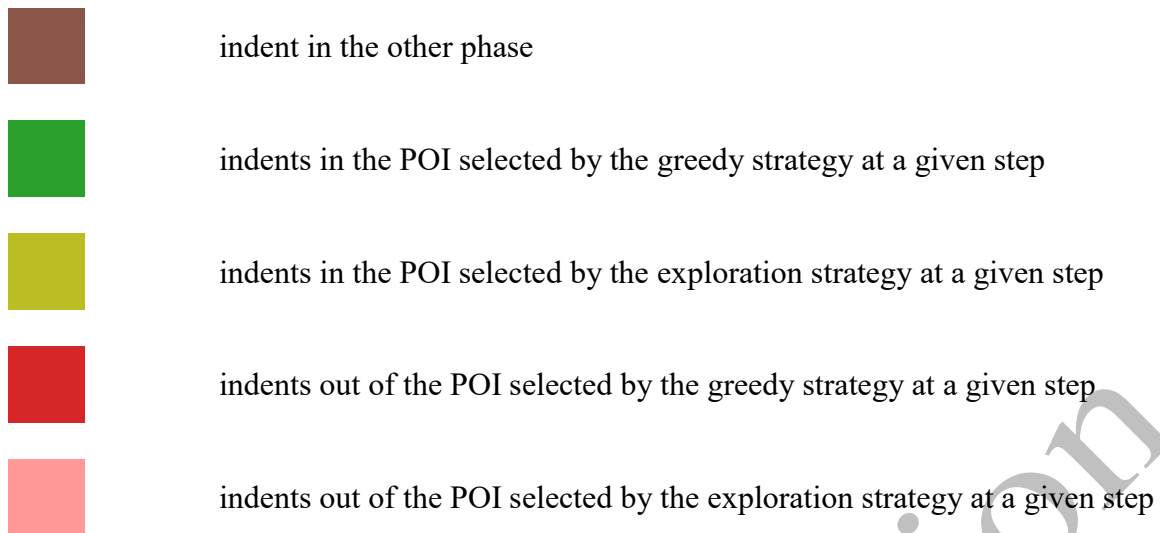
84.6% in POI



Legend:



indent in the phase of interest



303 **Fig. 6.** Maps of the selected indents over time with Kriging parameter equal to 16, GPC kernel  
 304 Matern + DotProduct: a) mortar sample, b) cement paste sample. Proportion of indents in the  
 305 phase of interest (POI) is reported at each step.

306  
 307 More specifically, in the case of the mortar sample illustrated in Fig. 6 (a), an initial Kriging  
 308 stage with a sampling parameter equal to 16 leads to 100 indents to be performed on the surface  
 309 out of the 1600 possible indents. Then, the GP classifier predicted paste indents in the left part  
 310 of the area, notwithstanding the relative lack of precision in the prediction (even if GPC training  
 311 accuracy was around 0.850 to 0.910). The relative lack of precision of the initial prediction,  
 312 which was a result of a tradeoff chosen to increase execution speed, was not found to limit the  
 313 overall capability of the model to find zones in the phase of interest. The algorithm then greedily  
 314 located the next indents in this zone. Most of these first indents corresponded to paste indents  
 315 according to the microscopic images, GPC predictions were then refined using these indents  
 316 nature and more indents were performed in the uncovered area during the first iterations of the  
 317 algorithm. Due to the appropriate exploration strategy, whose influence will be discussed in the  
 318 next section, some indents were performed in the bottom part of the area between steps 5 and  
 319 10, and in the top part of the area between 10 and 15. Because some of these exploratory indents  
 320 corresponded to paste zones, GPC predictions were revised with new information and high

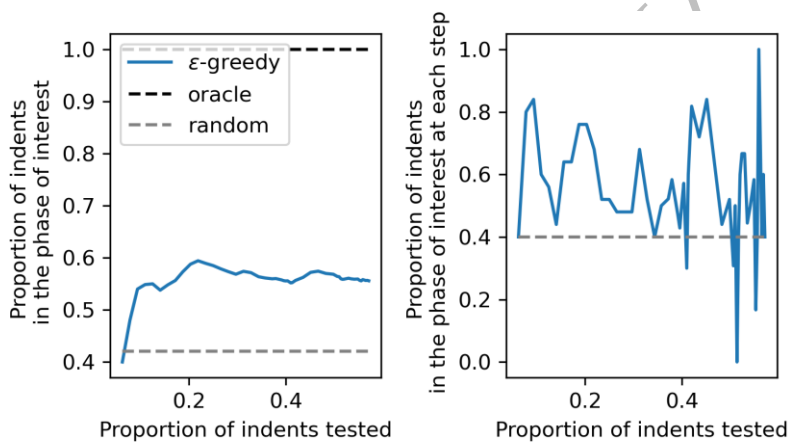
321 probabilities of finding paste indents were found in some of these specific zones, leading to the  
322 testing of close areas according to the greedy strategy. Therefore, new areas containing paste  
323 were found using the exploratory strategy. Several paste zones could be found on the sample  
324 even though the maximum 'resolution' of the indented map was relatively poor and the overall  
325 area contained a small proportion of the phase of interest, e.g., cement paste, as opposed to the  
326 other phase sand aggregates (around 40% and 60% resp.).

327 In the case of the cement paste sample, the overall area contained a higher proportion of  
328 hydrates, the phase of interest, as opposed to the other phase, the clinker particles,  
329 corresponding to regularly spaced particles (69% and 31% resp.). For this reason, the algorithm  
330 effectively found where to locate indents corresponding to the phase of interest and a relatively  
331 good initial prediction of the position of clinker particles was made by GPC after the initial  
332 Kriging stage, as illustrated in Fig. 6 (b). The locations selected by the greedy strategy to  
333 perform indents corresponded to hydrate areas located far away from the clinker phases leading  
334 to a good efficiency during the first steps. Due to the exploration strategy, several initial  
335 positions were found to generate kernels of indents in the phase of interest as in the previous  
336 case. Therefore, between step 5 and 30, three clusters containing a high proportion of the phase  
337 of interest were found, one in the top right part of the area, one in the middle left part, and one  
338 in the bottom part. Then, it was observed that the greedy exploitation strategy recommended  
339 performing indentations at the borders of these clusters leading to their extension as long as  
340 indents were performed in the phase of interest. Finally, the algorithm found most of the phase  
341 of interest and indents location was selected by the algorithm around the clinker particles, as it  
342 can be seen at steps 40 and 50.

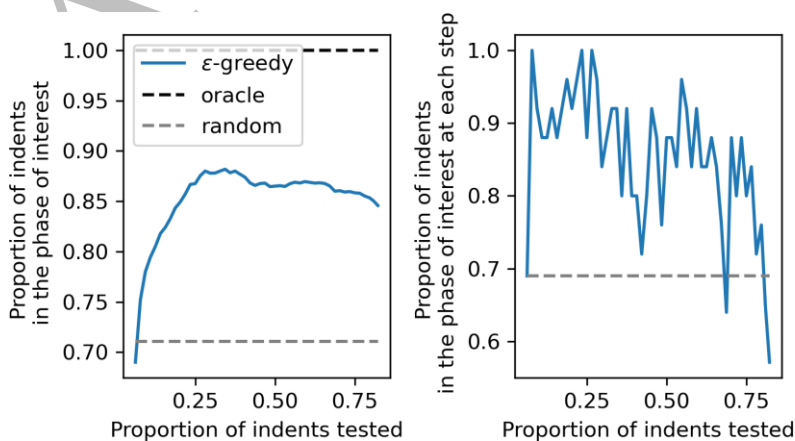
343 The algorithm's efficiency has been evaluated by measuring the proportion of indents in the  
344 phase of interest relative to the ratio of indents tested in the area. As illustrated in Fig. 7, in both  
345 the mortar and cement paste cases, the proportion of indents located in the phase of interest

346 quickly increased from the value obtained after the initial random Kriging (40% and 69% for  
 347 mortar and cement paste sample, resp.) to around 58% and 88% for the mortar and cement paste  
 348 samples, resp. At each step, the proportion of indents in the phase of interest, 58% and 87% on  
 349 average for the mortar and cement paste resp. (for proportions of tested indents smaller than  
 350 40% and 70% resp.), was higher than the random proportion of the phase of interest in the entire  
 351 area, 42% and 71% for the mortar and the cement paste, resp., as illustrated in the right-side  
 352 figures. After around 60% of the indents location in the cement paste (Fig. 7 b) were tested, a  
 353 drop in the overall efficiency associated with highly variable instantaneous precision can be  
 354 observed in the case of the cement paste (Fig. 7 b). This drop can be explained because most  
 355 indents in the face of interest have been found (and the global objective fulfilled).

a)



b)



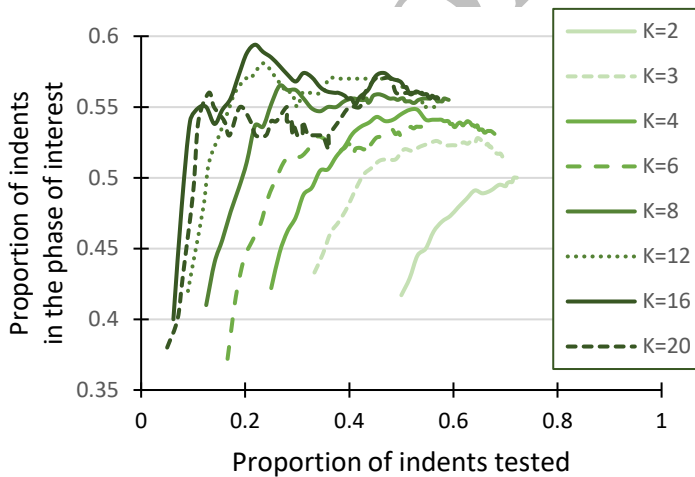


356 **Fig. 7.** Typical evolution of indents in the phase of interest over time: a) mortar, b) cement paste  
357

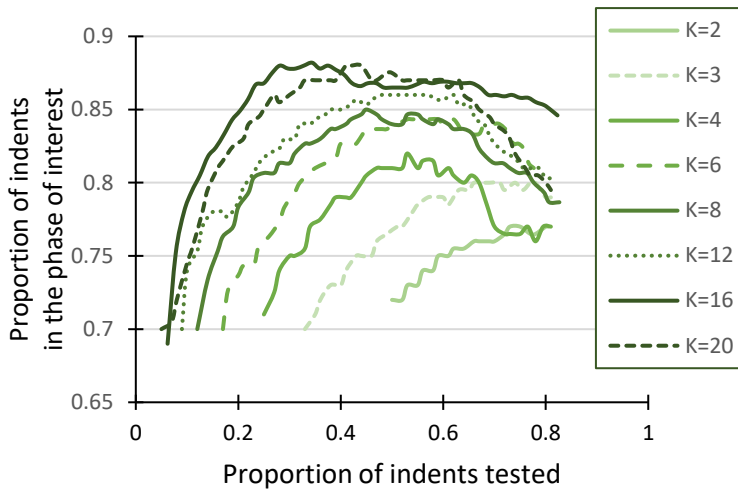
### 358 3.1.2 Influence of initial Kriging parameters.

359 The influence of the initial Kriging parameters has been assessed by running the algorithm  
360 using several Kriging parameters (K) from K=2 to K=20 in the two cases. As illustrated in Fig.  
361 8, in both cases, the maximum proportion of indents in the phase of interest globally increased  
362 with increasing K values. This observation can be explained because random initial Kriging  
363 relative importance decrease with increasing K values. For example, in the case K=2, half of  
364 the possible indents (800/1600) were performed randomly during the initial Kriging phase,  
365 explaining the relatively small room for improvement when the  $\epsilon$ -greedy algorithm is then  
366 activated. However, it is worth noting that the exploration-exploitation algorithm could select  
367 indents in the phase of interest in both the mortar and the cement paste cases after an initial  
368 Kriging stage with K=2, because of the information gathered during the Kriging stage.

a)



b)



369 **Fig. 8.** Evolution of indents in the phase of interest over time for several initial Kriging  
 370 parameters : a) mortar specimen, b) cement paste specimen.

371

372 The maximum proportion of indents in the phase of interest increased from  $K=2$  to  $K=12$  and  
 373 from  $K=2$  to  $K=16$  in the case of the mortar specimen, resp., cement paste specimen, while the  
 374 performance curves were similar for higher values of  $K$ . This highlights the increasing  
 375 algorithm's efficiency, and optimal  $K$  values of around 16 can be selected. With  $K=16$ , only  
 376 6.25% of the indents were performed during the initial random Kriging stage. Then, the  
 377 exploration-exploitation algorithm determines where to perform the next indents, and the  
 378 proportion of the indents in the phase of interest quickly grew and remained relatively stable  
 379 even though only 20 to 40% of the indents were performed (320 to 640 indents over 1600  
 380 possible indents, corresponding to a standard number of indents in statistical nanoindentation  
 381 of heterogeneous materials). In the case of the mortar (Fig. 8 a), the performance of the  
 382 algorithm with an initially loose Kriging ( $K=20$ ) is decreased compared to  $K=16$ . There is not  
 383 enough information to infer the indents' nature to start the exploration of the regions with the  
 384 highest proportions of the phase of interest, and auxiliary inputs such as the phase proportion  
 385 could help the algorithm initiation.

386

### 387 3.1.3 Exploration strategy adaptation

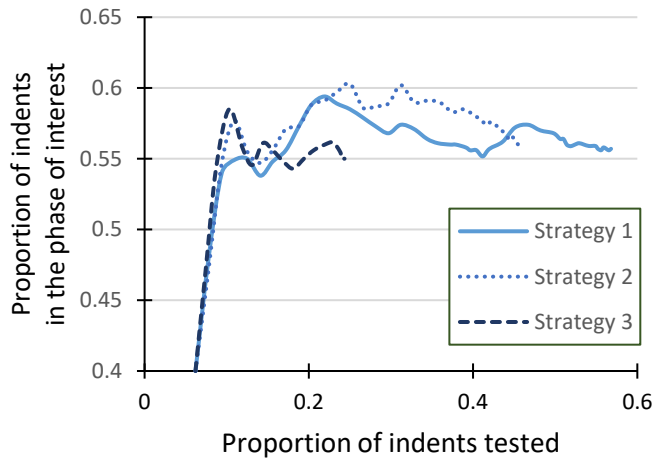
388 The exploration strategy is an essential factor affecting  $\epsilon$ -greedy algorithms' performance. For  
389 this reason, three strategies have been evaluated with the ratio between exploration and  
390 exploitation being constant equal to 20% (5 and 20 indents resp.):

- 391 i) strategy 1: most uncertain not-of-interest phase positions (sand or clinker particles)  
392 were selected to perform indents to obtain information at the boundary between the  
393 phase of interest and the other phase,
- 394 ii) strategy 2: most uncertain phase of interest positions were selected to obtain a  
395 similar kind of information but with a higher probability of selecting positions of  
396 the phase of interest and,
- 397 iii) strategy 3: particular phase of interest positions, i.e., positions where the probability  
398 of finding the phase of interest was higher than 75% or higher than 55% if no  
399 position associated with probability  $>75\%$  was found, were tested randomly  
400 (conservative exploration strategy).

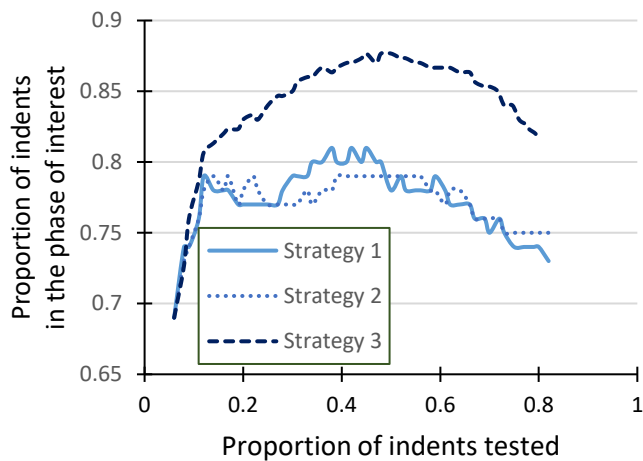
401 Strategy n°3 was considered an intermediate option between strategies 1 and 2, specifying  
402 probabilities of finding the phase of interest though keeping some randomness aspect. Recent  
403 works suggest promising exploration strategies optimization during the learning process, which  
404 would be relevant to implement in future studies [43,44]. As illustrated in Fig. 9 (a), the  
405 exploration strategy did not influence the algorithm's performance in the case of the mortar  
406 sample. This could be due to the relatively small number of positions with probabilities of  
407 finding the phase of interest higher than 75%. Therefore, the three exploration strategies  
408 selected similar indents. Conversely, in the case of the cement paste sample, exploration  
409 strategy 3 performed better than the other strategies as illustrated in Fig. 9 (b). This can be  
410 attributed to the effective selection of the locations to explore. They mostly corresponded to the  
411 phase of interest located relatively to positions tested by the greedy algorithm. Then, new

412 'seeds' can be selected on the indentation map to find new positions where the phase of interest  
413 was located, as illustrated in Fig. 6 (b).

a)



b)



414 **Fig. 9.** Evolution of indents in the phase of interest over time for several initial Kriging  
415 parameters : a) mortar sample, b) cement paste sample.

416

#### 417 3.1.4 Kernel selection

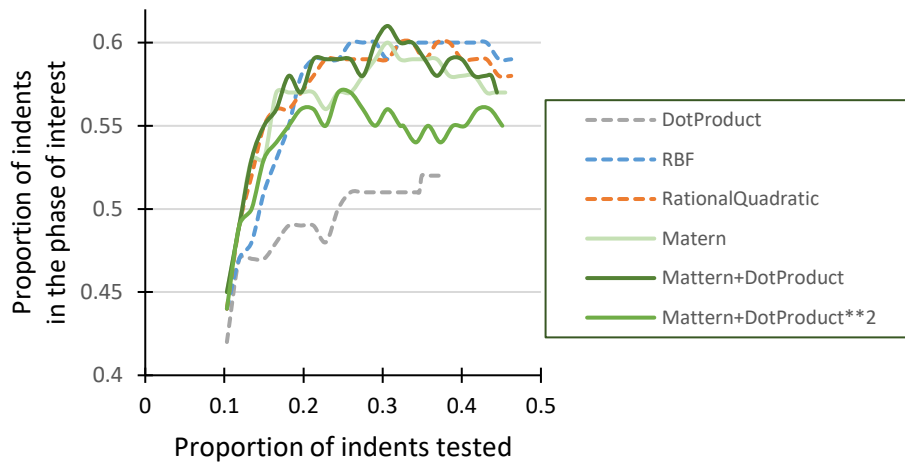
418 The influence of the kernel used with the GP Classifier has been assessed. As illustrated in Fig.

419 10, most of the tested kernels performed well except the DotProduct kernel that did not lead to

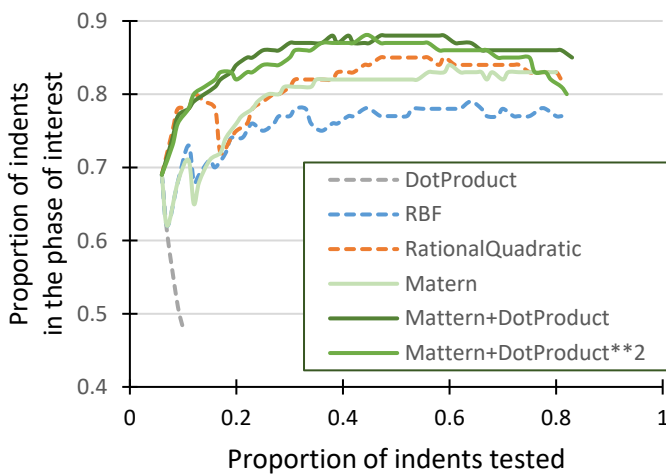
420 satisfactorily convergence in the cement paste case and led to a smaller maximum proportion  
 421 of indents in the phase of interest and a slower increase of this proportion in the mortar case.  
 422 Some kernels did show promising results in both cases, e.g., Matern + DotProduct and, with  
 423 slightly more variable results, Matern + DotProduct<sup>2</sup> and RationalQuadratic. For this reason,  
 424 most of the simulations reported in this manuscript were performed using Matern + DotProduct  
 425 or Matern + DotProduct<sup>2</sup> kernels.

426

a)



b)



427 **Fig. 10.** Evolution of indents in the phase of interest over time for several kernel natures and  
428 parameters: a) mortar sample, b) cement paste sample.

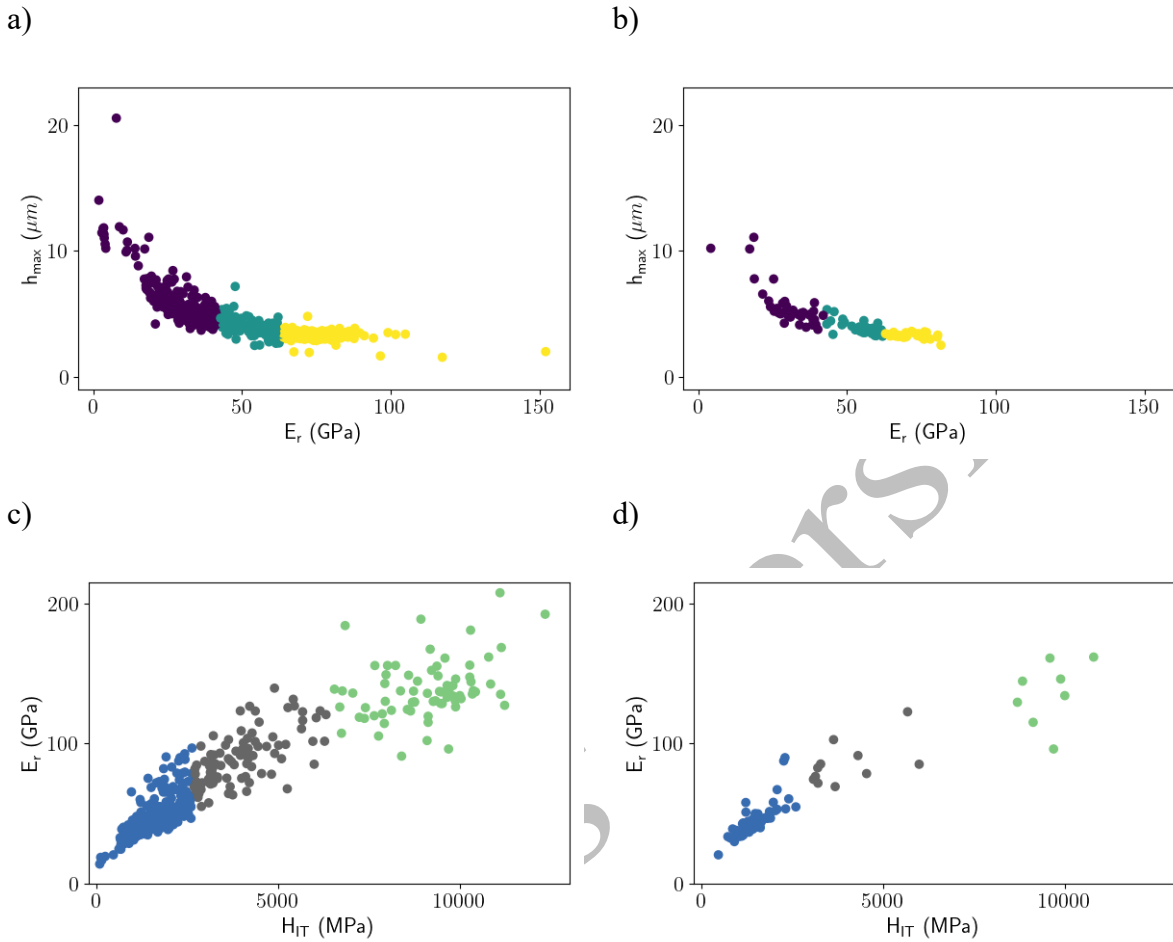
429

430 *3.2  $\epsilon$ -greedy algorithm using priors from indentation curves only*

431 *3.2.1 Deconvolution of indentation curves*

432 The effectiveness of deconvolution after the initial Kriging step has been evaluated to build an  
433 algorithm that considered indentation curves to get rid of the need of geometrical information  
434 as defined in section 2.2.3.2 to predict the prior information regarding the phase of the existing  
435 indents. As illustrated in Fig. 11, the indentation curves can be correctly clustered into three  
436 groups by an unsupervised algorithm in the case where  $K=16$  whatever the micro-mechanical  
437 properties used as inputs ( $E_r$  and  $h_{max}$ , the maximum penetration depth for the mortar specimen  
438 and  $E_r$  and  $H_{IT}$  in the case of the mortar, resp. cement paste specimen). The clusters obtained  
439 using 100 curves ( $1/16^{th}$  of the 1600 curves) illustrated in Fig. 11 (b) and (d) were very similar  
440 to the clusters obtained using half of the curves in both the mortar and the cement paste cases  
441 ( $K=2$ ). The reduced modulus cluster center value of the phase of interest was 30.4 GPa and  
442 30.0 GPa in the case of the mortar specimen with  $K=16$  and  $K=2$  resp., which corresponds to  
443 broadly reported values of cement paste properties [16] and 43.3 GPa and 43 GPa in the case  
444 of the hydrates properties in the cement paste for  $K=16$  and  $K=2$  resp. Although slightly  
445 elevated, these last two values were very close to each other and are in the order of magnitude  
446 of the reported values for the cement paste hydrates (high-density CSH with indentation  
447 modulus values close to 30 GPa and portlandite with values close to 42 - 44 GPa [45]).  
448 Therefore, clustering of initial curves obtained through Kriging can provide sufficient  
449 information, and the same algorithm trained once can be used for the entire  $\epsilon$ -greedy procedure.  
450 In case of a very loose initial Kriging or an important number of phases, the clustering algorithm

451 could be trained several times during the algorithm or consider information from similar  
452 materials.

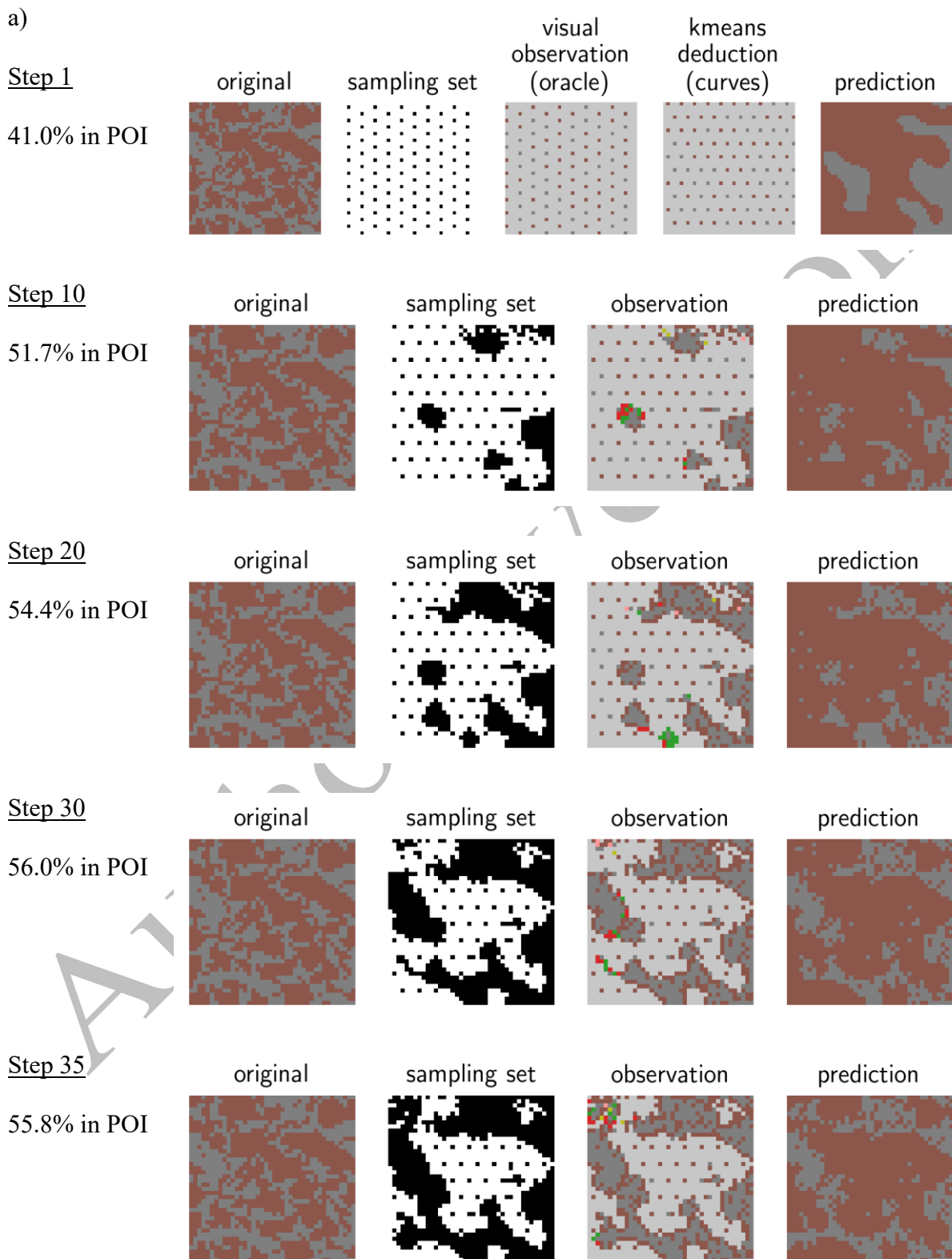


453 **Fig. 11.** Deconvolution results of the indentation curves after the initial Kriging step: a) mortar  
454 sample with  $K=2$ , b) mortar sample with  $K=16$ , c) cement paste sample with  $K=2$ , d) mortar  
455 sample with  $K=16$

### 456 3.2.2 Convergence of the algorithm

457 The online algorithm, using indentation curves only, showed excellent convergence results with  
458 the aforementioned optimized parameters. After unsupervised clustering using the indentation  
459 curves as detailed in the previous paragraph, the GP classifier has been used to infer the indents'  
460 nature based on the clustering results and the  $\epsilon$ -greedy procedure has been applied to select the

461 next indents. As illustrated in Fig. 12, the algorithm was able to find the zones with the highest  
 462 probabilities of finding indents in the phase of interest during 30 to 50 steps.

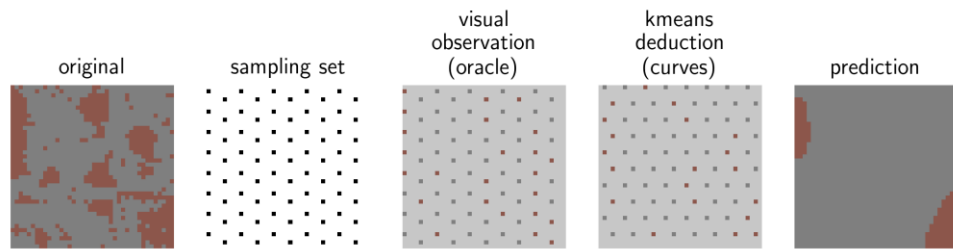




b)

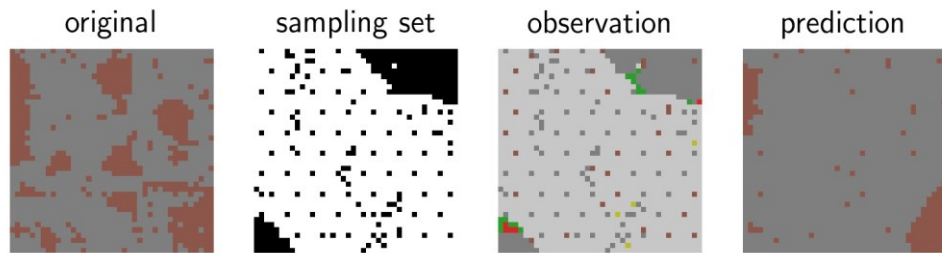
Step 1

75.0% in POI



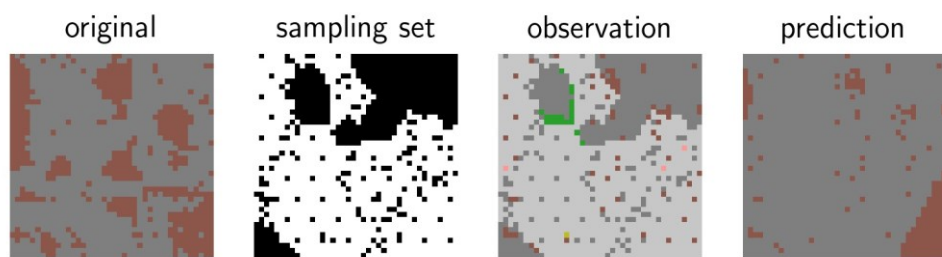
Step 10

88.5% in POI



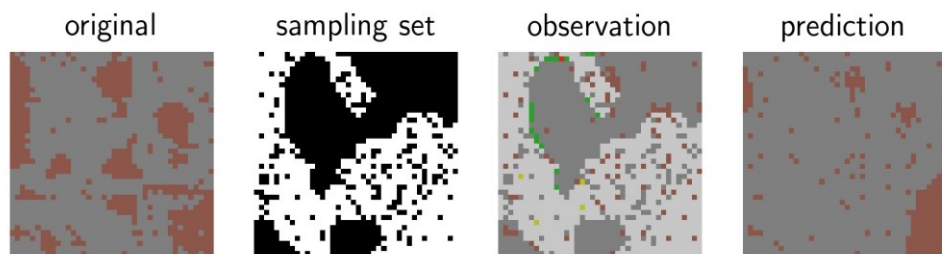
Step 20

87.9% in POI



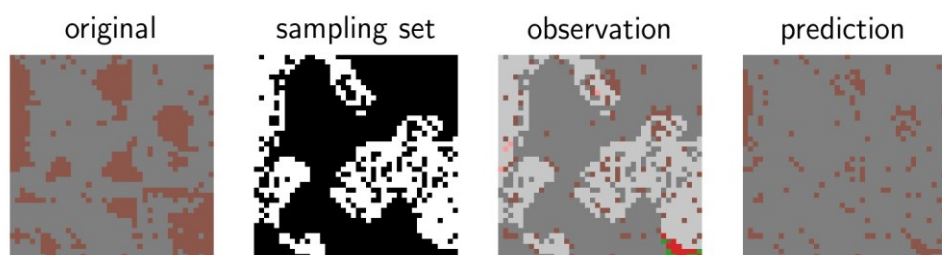
Step 30

89.0% in POI



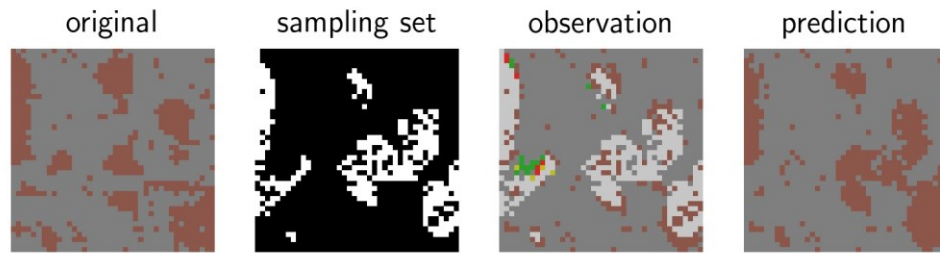
Step 40

87.4% in POI

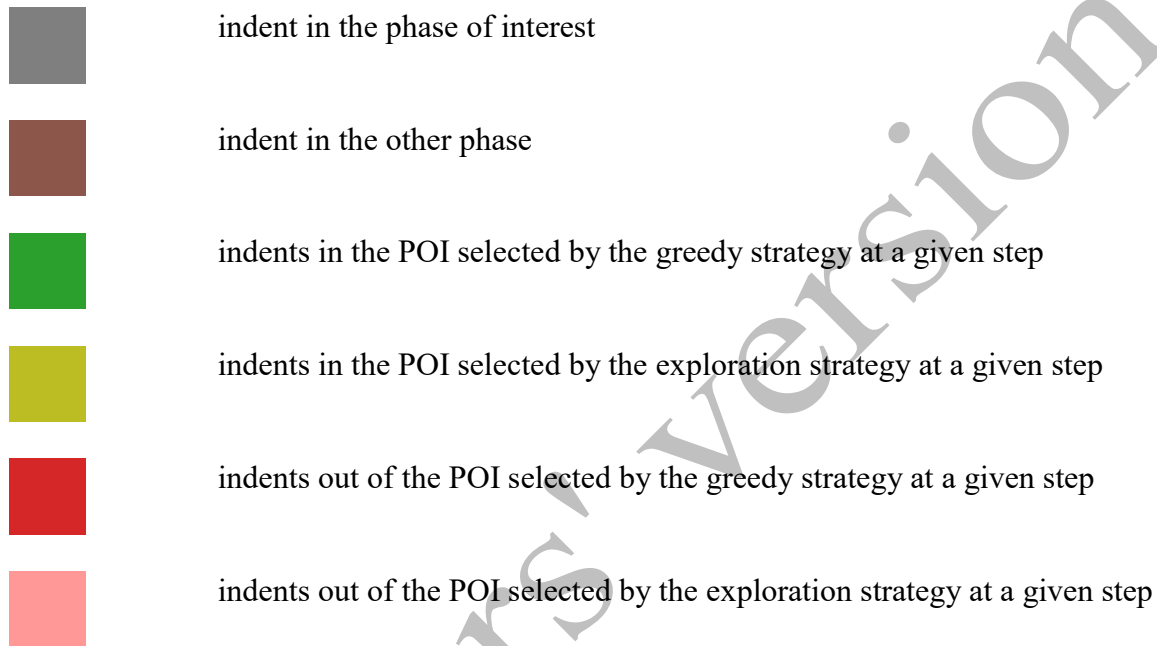


Step 50

85.7% in POI



Legend:



463 **Fig. 12.** Maps of the selected indents over time: a) mortar sample, b) cement paste sample.

464 Proportion of indents in the phase of interest (POI) is reported at each step.

465

466 In the case of the mortar specimen (Fig. 12 (a)), the initial inference of the GP classifier based  
467 on the indentation curves corresponded well with the microscopic observations obtained after

468 the indentation tests. Using the inference, the  $\epsilon$ -greedy procedure could find potential positions

469 of the phase of interest within the indented map. Several potential positions of the phase of

470 interest were selected during the first 10 iterations, then some of these zones were extended

471 performing new indents (top right part, middle left part and bottom part), and after 35 steps,

472 most of the largest zones including the phase of interest were found. Therefore, even though

473 the clustering predictions did not coincide with the nature of the indents inferred from

474 microscopic observation, the zones investigated by the algorithm corresponded well with zones  
475 with a higher proportion of the phase of interest.

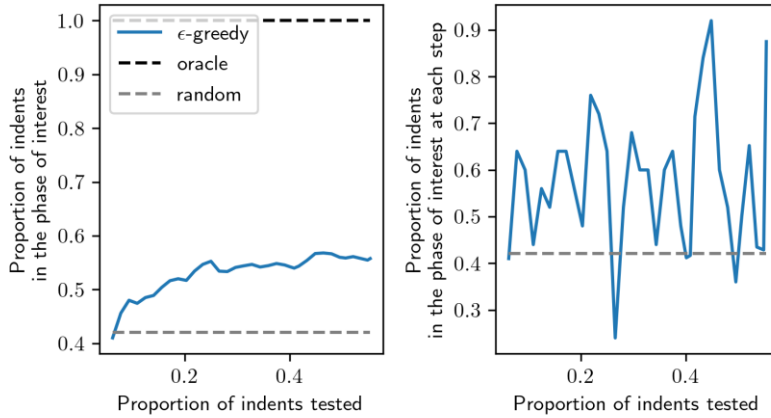
476 In the case of the cement paste specimen, as illustrated in Fig. 12 (b), the number of indents  
477 performed in the phase of interest quickly increased. Similar to the case where the microscopic  
478 nature of the indents was checked during the algorithm, the algorithm using the indentation  
479 curves only correctly advised to perform indents around the clinker particles. This performance  
480 could be explained by the distinct mechanical parameters derived from the indentation curves,  
481 which can be attributed to the different phases.

482 The algorithm's efficiency has been reported in Fig. 13. In both the mortar and cement paste  
483 cases, the proportion of indents located in the phase of interest quickly increased from the value  
484 obtained after the initial random Kriging (41% and 70% for mortar and cement paste sample,  
485 resp.) to around 58% and 88% for the mortar and cement paste samples, resp. At any step, the  
486 proportion of indents in the phase of interest, 56% and 89% on average for the mortar and  
487 cement paste resp. (for proportions of tested indents smaller than 45% and 22 to 60% resp.),  
488 was higher than the random proportion of the phase of interest in the entire area 42% and 71%  
489 for the mortar and the cement paste resp. The drop in efficiency occurring when most indents  
490 in the phase of interest have been found was confirmed by the decrease in the proportion of  
491 indents in the phase of interest. In the cement paste case, almost all the points in the phase of  
492 interest can be found (1055 / 1137) though only 75% of the positions have been tested.

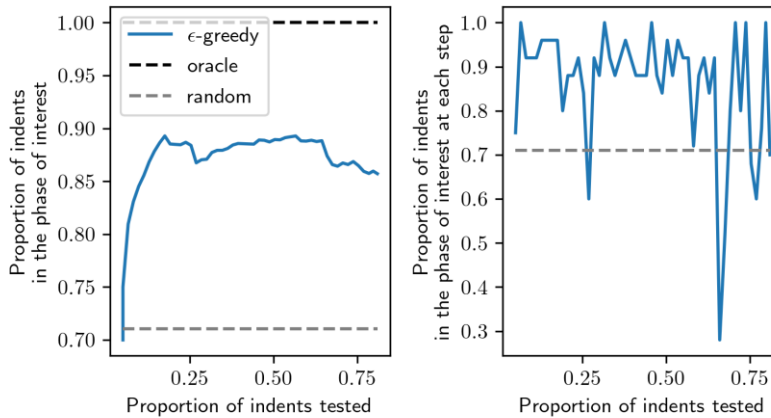
493 Interestingly, as illustrated in Fig. 13, the proportion of indents in the phase of interest, was  
494 almost always higher than the random proportion of the phase of interest in the indented area.

495 This observation highlights the performance of the proposed algorithm.

a)



b)



496 **Fig. 13.** Evolution of indents in the phase of interest over time using the online algorithm: a)  
497 mortar sample, b) cement paste sample.

#### 498 **4. Conclusions**

499 The main objective of the present work was to demonstrate the interest of  $\epsilon$ -greedy strategies  
500 to automatically select the location of indents to perform during a microindentation or  
501 nanoindentation process. Two strategies have been tested: i) an offline algorithm using  
502 geometrical information only to infer the next indent location using microscopically identified  
503 indents nature and ii) an online algorithm based only on available indentation curves to infer  
504 indents nature using unsupervised clustering to feed the  $\epsilon$ -greedy algorithm that selects the next

505 indents to perform. Both algorithms have been tested on cementitious materials, namely, a  
506 mortar specimen composed of two major phases, e.g. cement paste and sand, indented with a  
507 500 mN load, and a cement paste specimen composed of hydrates and anhydrous clinker  
508 particles indented with a 1 mN load. The main results can be summarized as follows:

- 509 - After an initial random Kriging stage giving information about the nature of around 100  
510 indents,  $\epsilon$ -greedy strategy led to the selection of indents mostly located in the phase of  
511 interest during tens of iterations.
- 512 - The proportion of indents in the phase of interest considerably increased with the use of  
513  $\epsilon$ -greedy strategies. In the case of the mortar specimen, it increased from 40% to 60%  
514 (considering the cement paste as the phase of interest). In the case of the cement paste  
515 specimen, the proportion of hydrates within the indents rose from 70% to almost 90%.
- 516 - The best parameters associated with the algorithm were found for both samples, and  
517 both the exploration strategy and the Gaussian process (GP) classifier kernel's nature  
518 can impact the algorithm's efficiency.
- 519 - Optimal initial Kriging parameters leading to the best convergence of the algorithm  
520 equal to 16, i.e., 1/16<sup>th</sup> of the possible indents are performed initially, provided sufficient  
521 information to deconvolve the indentation curves using an unsupervised clustering  
522 algorithm.
- 523 - The algorithm's efficiency is only slightly affected by using indentation curves only  
524 compared to using geometrical information.

525 Therefore, the proposed method can be successfully applied to select indents in a phase of  
526 interest during indentation experiments. By allowing the indentation procedure to be more  
527 effective, the proposed approach may open up novel research paths regarding the fast and  
528 accurate assessment of the mechanical properties of particular phases of interest in  
529 heterogeneous materials during, for example, time-dependant evolutions or after degradation.

530 Several research paths might be investigated to increase the algorithm performance, like initial  
531 knowledge usage (phase proportions) that could help decrease the need for initial Kriging,  
532 enhance initial GPC predictions and adapt the strategy to refine the mechanical properties of the  
533 phases.

## 534 Acknowledgements

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