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1 ϵ -greedy automated indentation of cementitious materials 2 for phase mechanical properties determination

3
4 B. Hilloulin¹, M. Lagrange², M. Duvillard³, G. Garioud³

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6 ¹ *Institut de Recherche en Génie Civil et Mécanique (GeM), UMR-CNRS 6183, Ecole Centrale de Nantes, 1*
7 *rue de la Noë, 44321 Nantes, France – e-mail: benoit.hilloulin@ec-nantes.fr*

8 ² *Laboratoire des Sciences du Numérique de Nantes – CNRS, Ecole Centrale de Nantes, 1 rue de la Noë,*
9 *44321 Nantes, France – email : mathieu.lagrange@cnrs.fr*

10 ³ *Ecole Centrale de Nantes, 1 rue de la Noë, 44321 Nantes, France*

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 ϵ -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 • ϵ -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; ϵ -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, ϵ -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 ϵ -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 ϵ -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 / m³)

| 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 $4 \times 4 \times 16 \text{ cm}^3$ 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

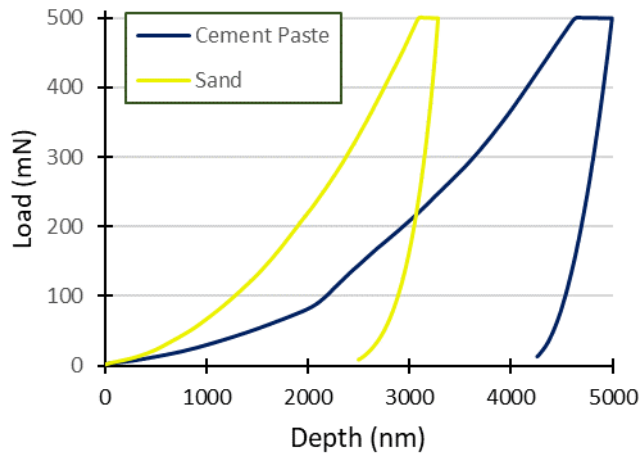
131 2.1.2 Grid indentation tests

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 40×40 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

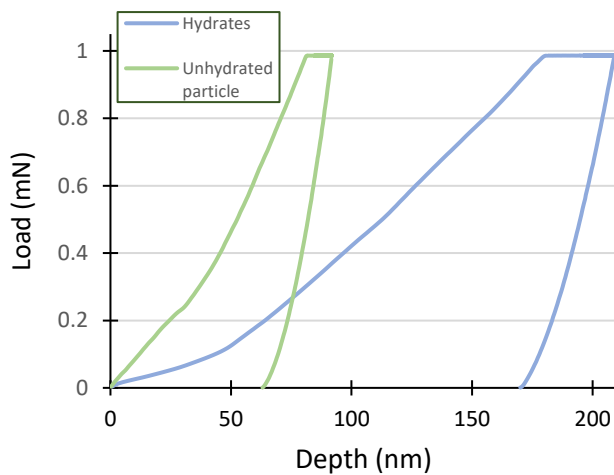
141 indentation curves of the two major phases in the mortar sample and the cement paste samples
142 are represented in Fig. 1.

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.

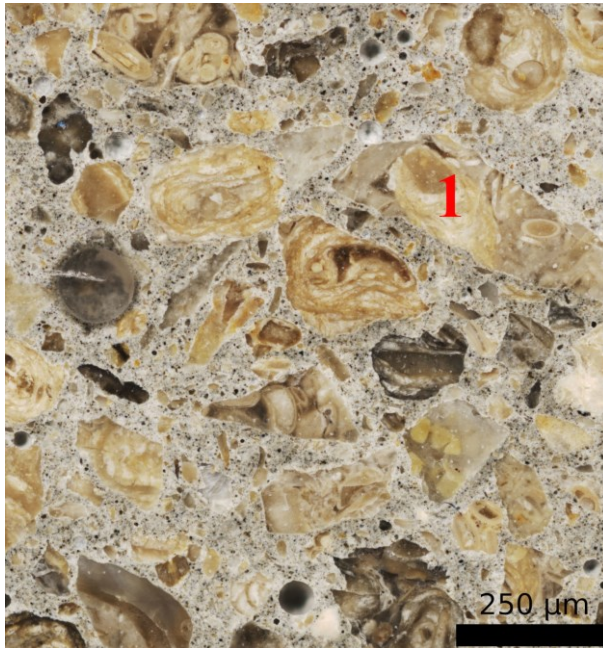
164 As illustrated in Fig. 2 (b), a SEM image of the cement paste sample was obtained using an
165 acceleration voltage of 15 kV and a working distance of 7 mm and a low vacuum pressure of
166 60 Pa to limit the risk of cracking. A relatively large light gray unhydrated particles was found
167 in the 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.

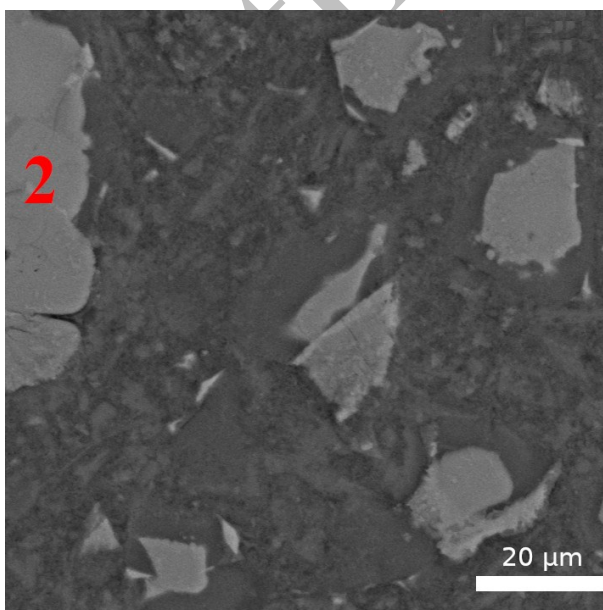
171

172

a)



b)



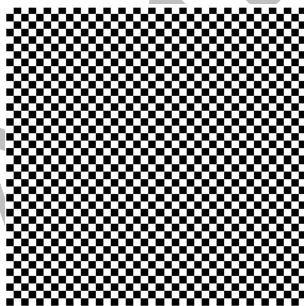
173 **Fig. 2.** Microscopic image of the samples: a) 2D optical microscopic image of mortar sample
174 with one sand particle labeled 1 (1cm × 1cm zone indented using a 40 × 40 matrix of 500mN
175 indents), b) SEM image of cement paste sample with one unhydrated particle labeled 2
176 (80μm × 80μm zone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

178 2.2 Numerical methods

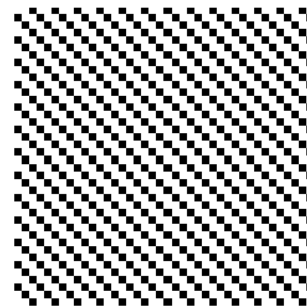
179 2.2.1 Kriging

180 Initial values, either indent nature based on the microscopic images or indentation curves, were
181 acquired initially before running the ϵ -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 x 40), the initial number
187 of indents after the initial Kriging stage ranged from 80 ($K=20$) to 800 ($K=2$).

a) $K=2$

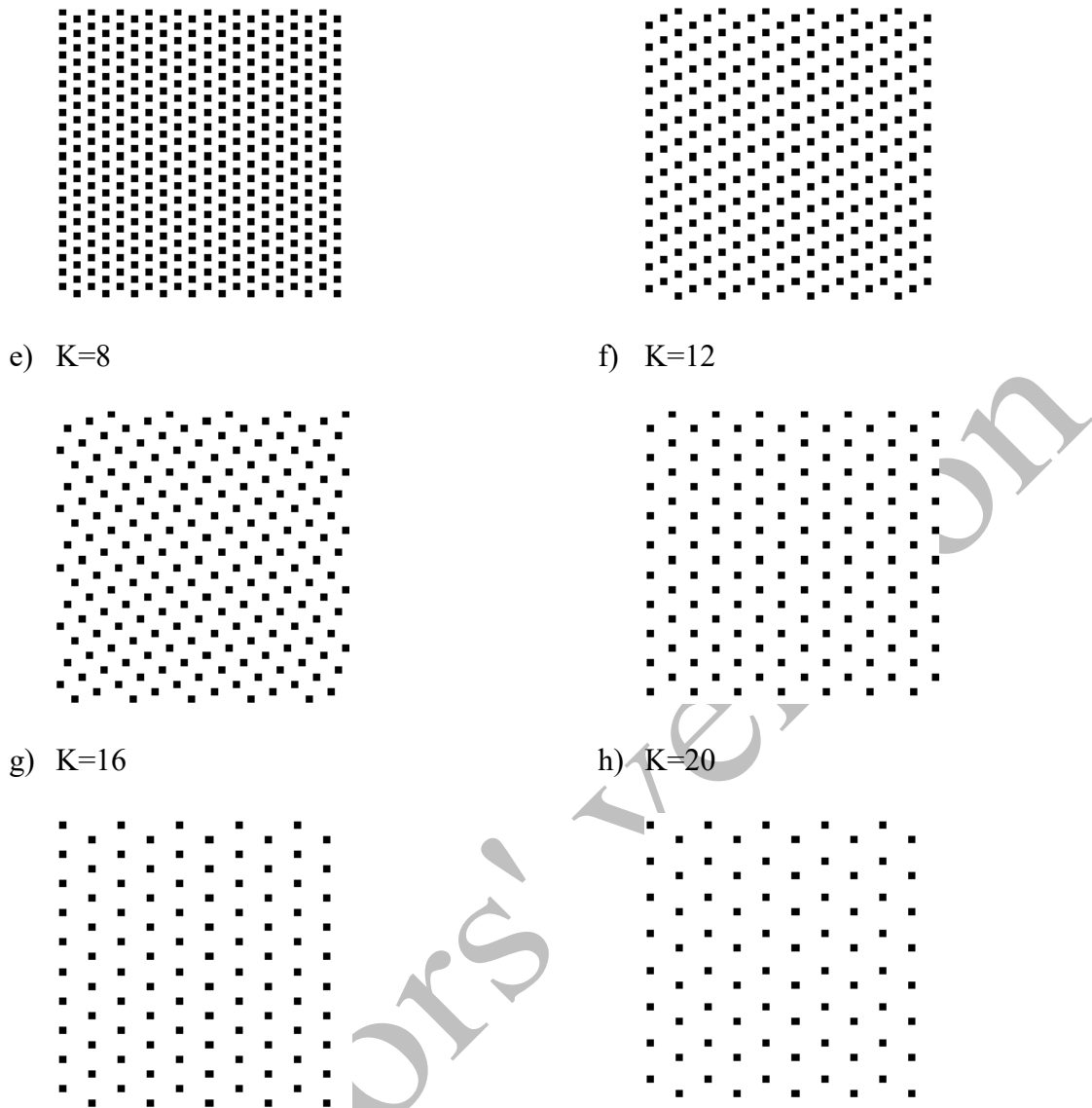


b) $K=3$



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 α 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 α 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:

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

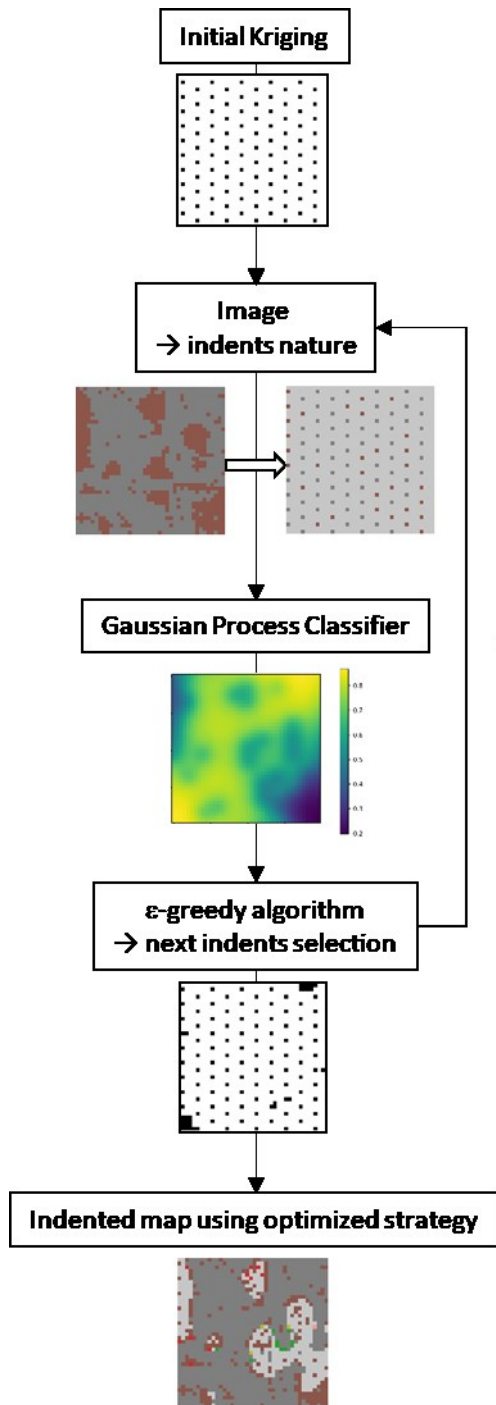
236 This kernel is parameterized by the σ_0 parameter. The dot product kernel is commonly combined
 237 with exponentiation. For this reason, Matern + Dot Product and Matern + Dot Product² have
 238 been studied herein. For all tested configurations, kernel parameters were optimized during the
 239 training procedure.

240

241 2.2.3 ϵ -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 ϵ -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 – ϵ -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 ϵ -greedy strategy. The influence of these parameters will be discussed in section
259 3.1.



Version

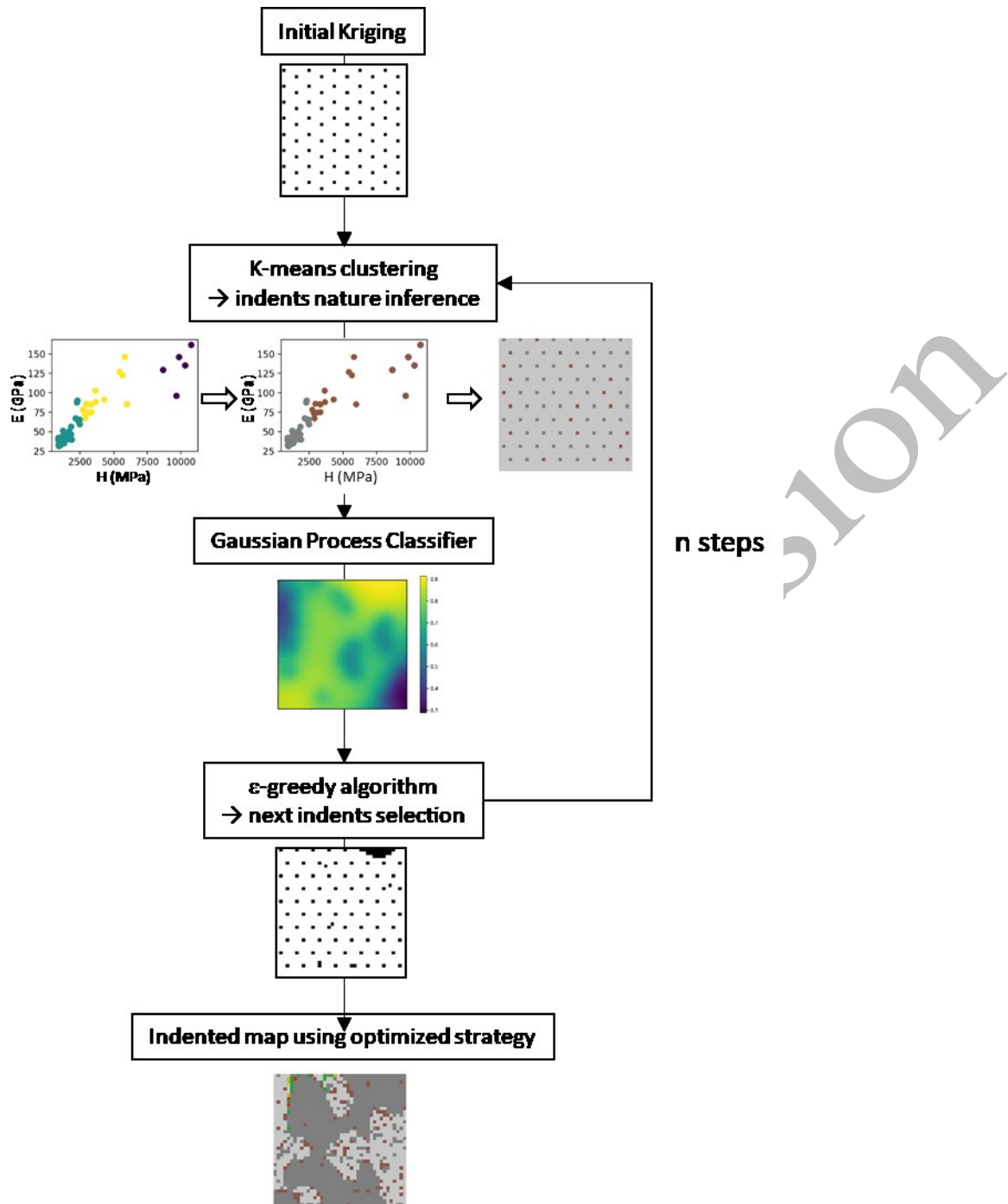
260

261 **Fig. 4.** Flowchart of the ϵ -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 ϵ -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 – ϵ -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 ϵ -greedy algorithm for indents location selection using indentation

287 curves.

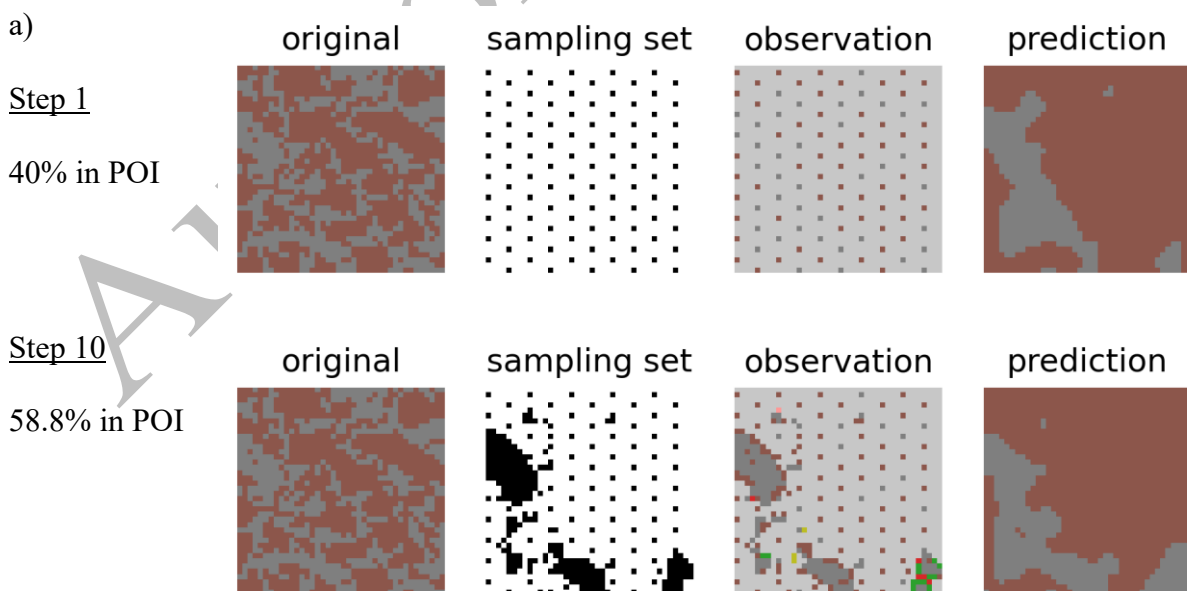
288

289 **3. Results and discussion**

290 *3.1 Algorithm using geometrical information only*

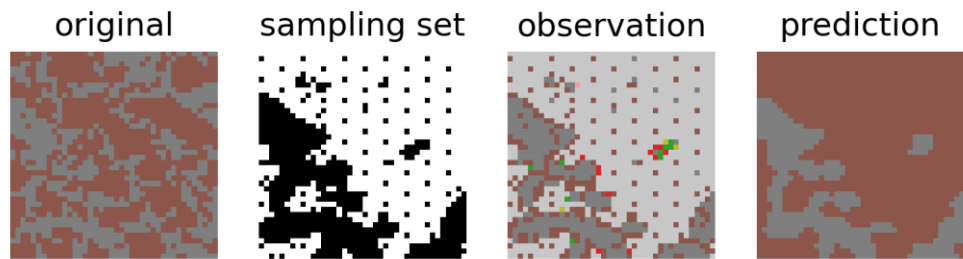
291 3.1.1 Convergence of the algorithm

292 The efficiency of the ϵ -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.



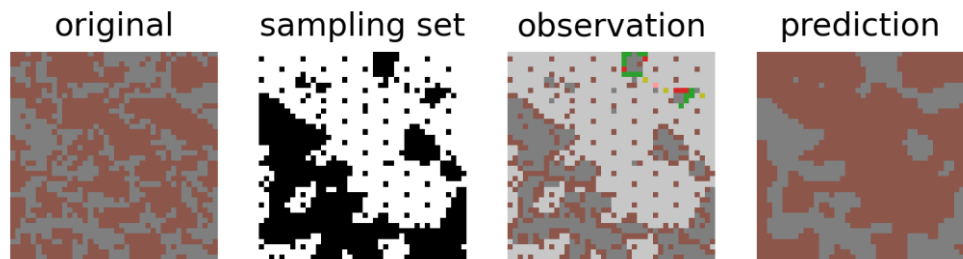
Step 20

56.1% in POI



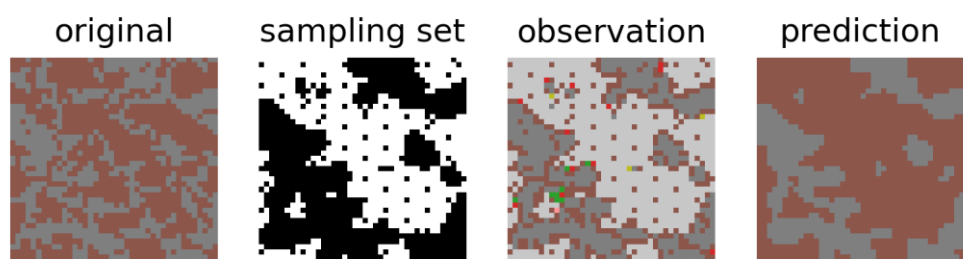
Step 30

56.2% in POI



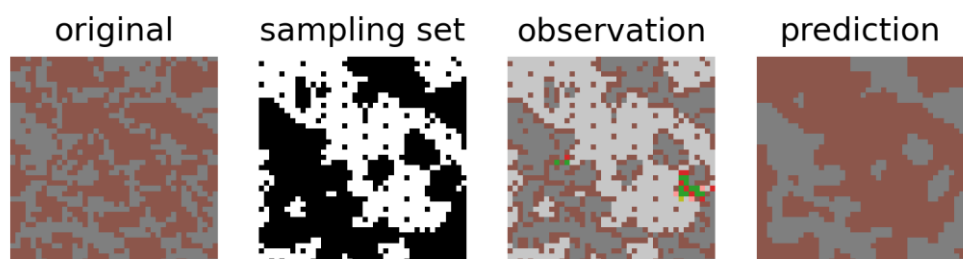
Step 40

56.0% in POI



Step 50

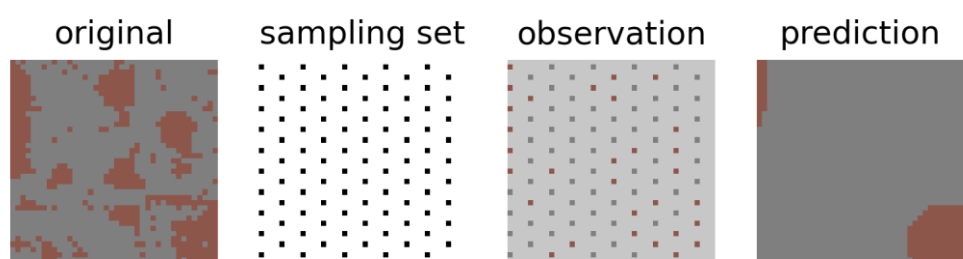
55.6% in POI



b)

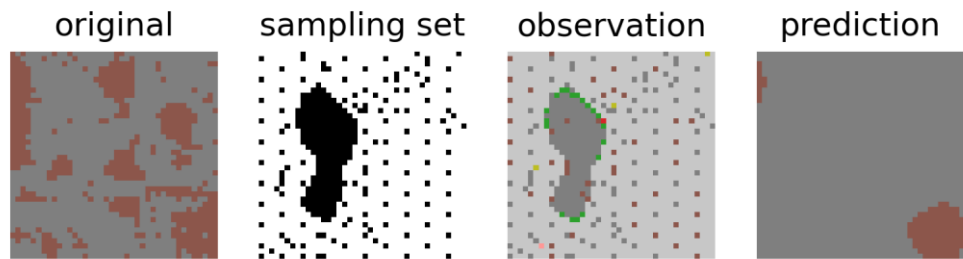
Step 1

69.0% in POI



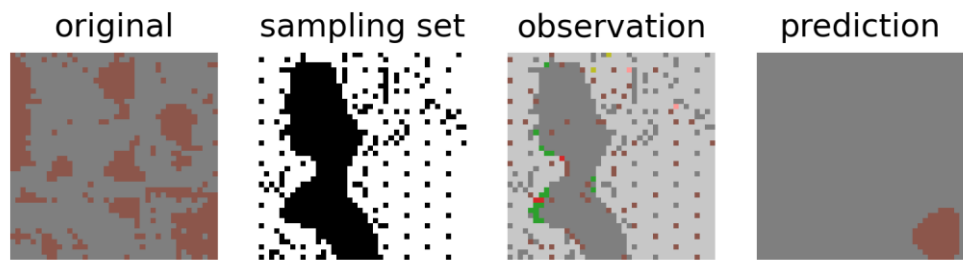
Step 10

84.9% in POI



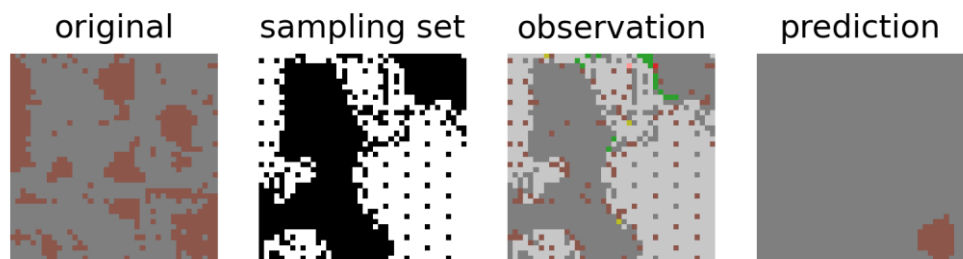
Step 20

87.8% in POI



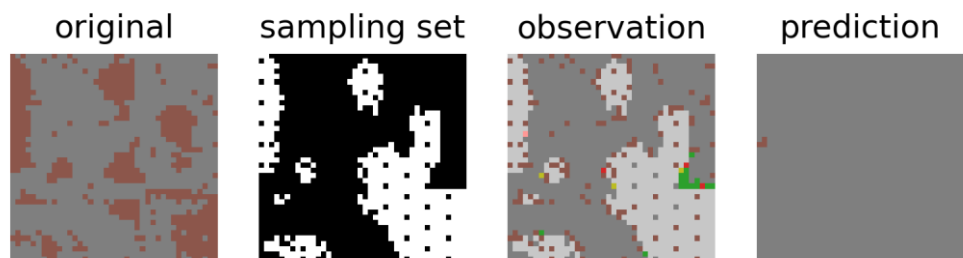
Step 30

86.5% in POI



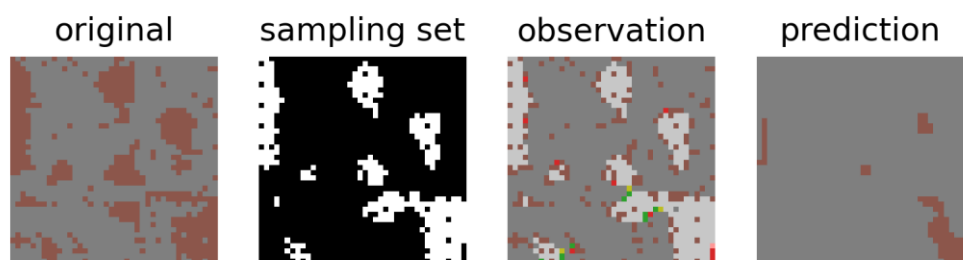
Step 40

86.5% in POI



Step 50

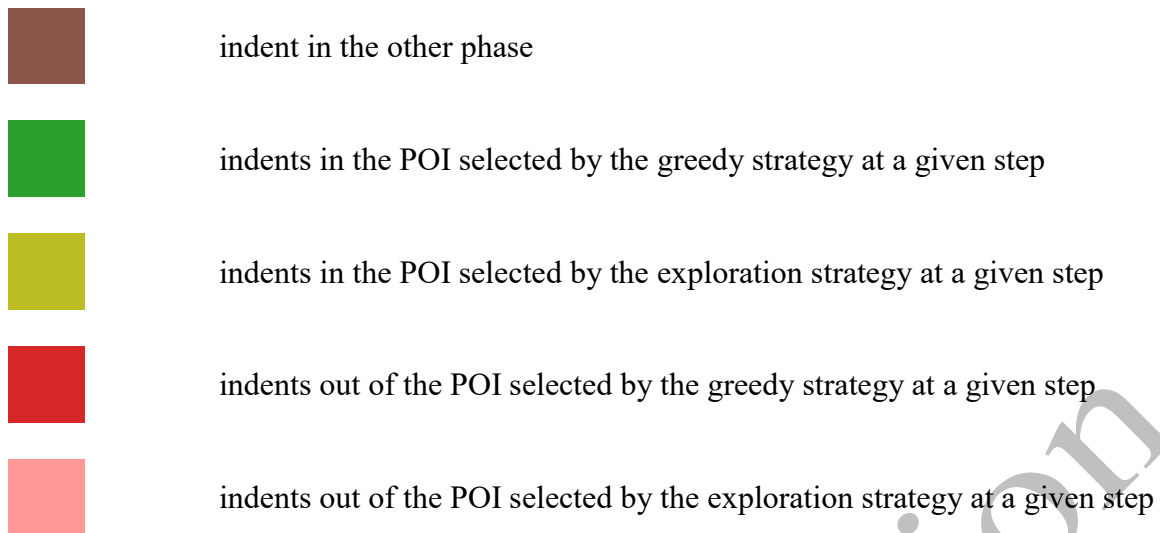
84.6% in POI



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

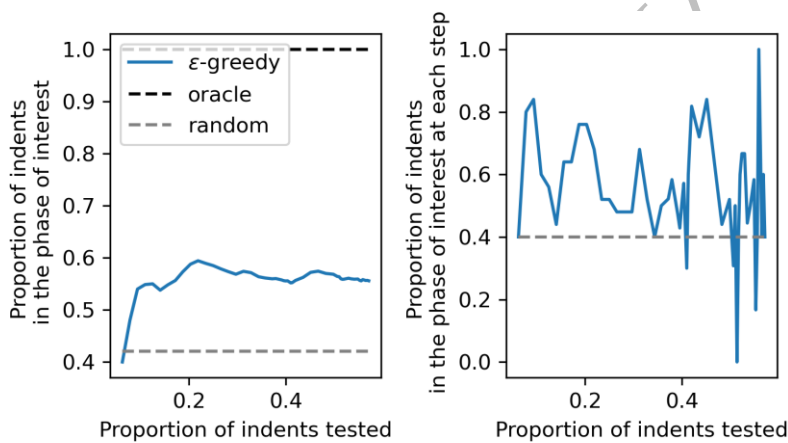
321 probabilities of finding paste indents were found in some of these specific zones, leading to the
322 testing of close areas according to the greedy strategy. Therefore, new areas containing paste
323 were found using the exploratory strategy. Several paste zones could be found on the sample
324 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.

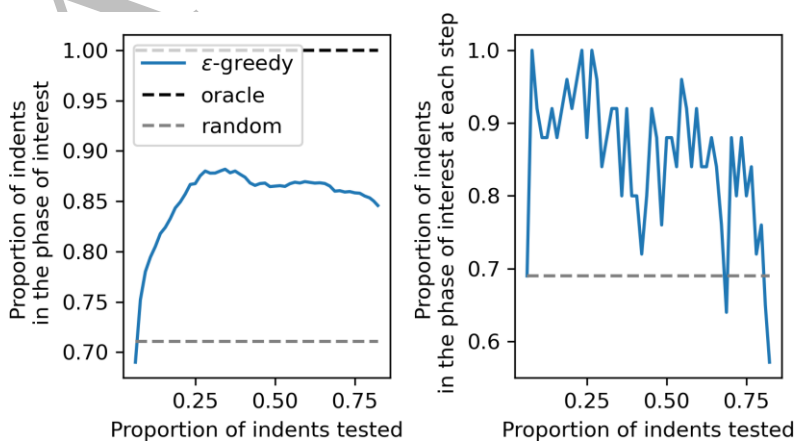
343 The algorithm's efficiency has been evaluated by measuring the proportion of indents in the
344 phase of interest relative to the ratio of indents tested in the area. As illustrated in Fig. 7, in both
345 the mortar and cement paste cases, the proportion of indents located in the phase of interest

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)

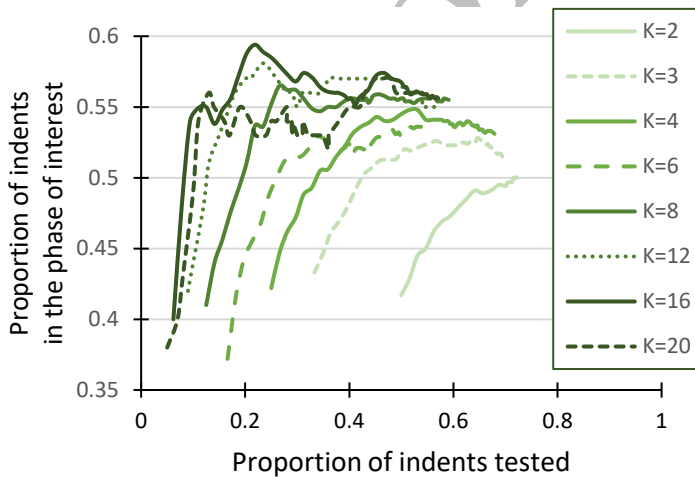


356 **Fig. 7.** Typical evolution of indents in the phase of interest over time: a) mortar, b) cement paste
357

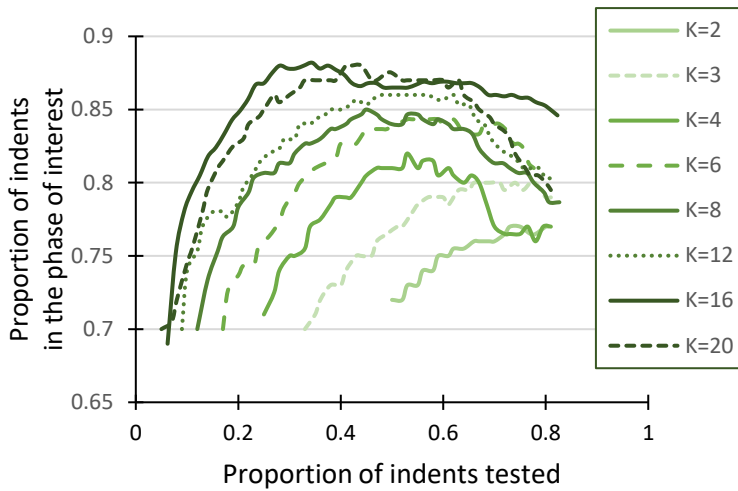
358 3.1.2 Influence of initial Kriging parameters.

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 ϵ -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

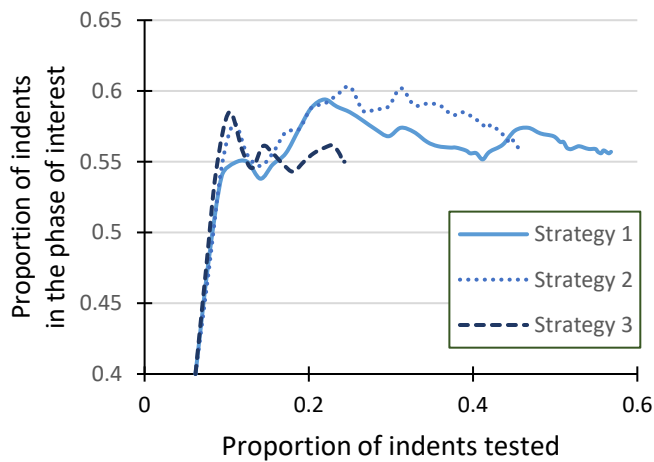
388 The exploration strategy is an essential factor affecting ϵ -greedy algorithms' performance. For
389 this reason, three strategies have been evaluated with the ratio between exploration and
390 exploitation being constant equal to 20% (5 and 20 indents resp.):

- 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).

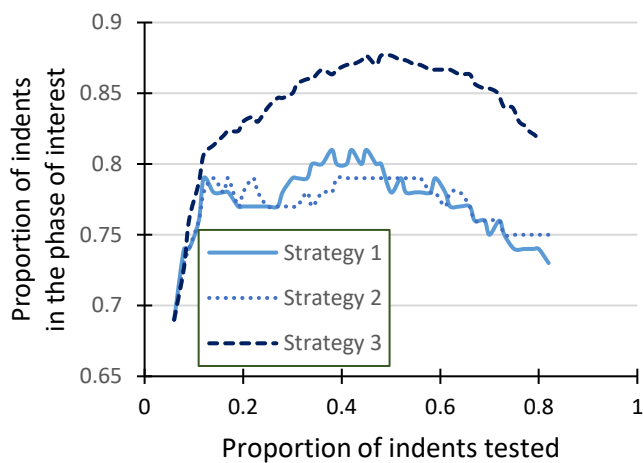
401 Strategy n°3 was considered an intermediate option between strategies 1 and 2, specifying
402 probabilities of finding the phase of interest though keeping some randomness aspect. Recent
403 works suggest promising exploration strategies optimization during the learning process, which
404 would be 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

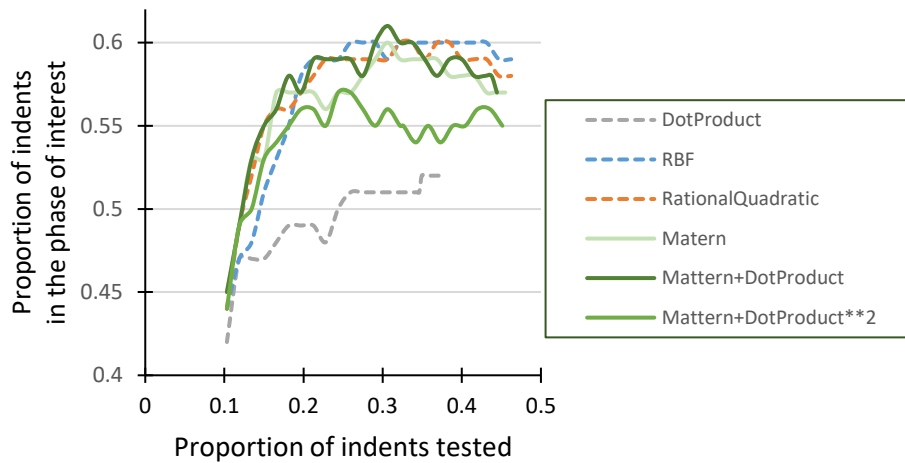
418 The influence of the kernel used with the GP Classifier has been assessed. As illustrated in Fig.

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

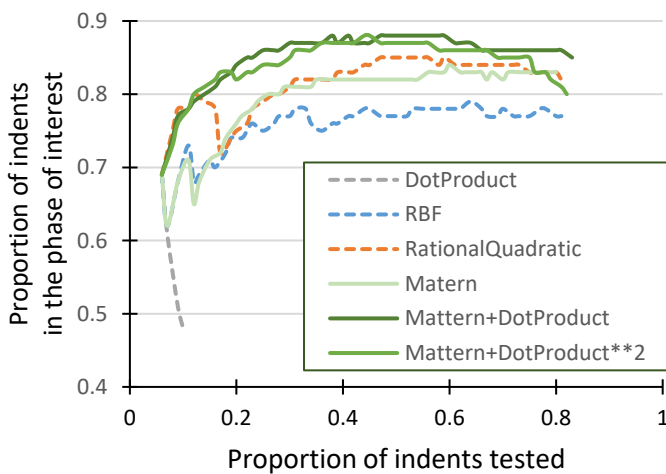
420 satisfactorily convergence in the cement paste case and led to a smaller maximum proportion
 421 of indents in the phase of interest and a slower increase of this proportion in the mortar case.
 422 Some kernels did show promising results in both cases, e.g., Matern + DotProduct and, with
 423 slightly more variable results, Matern + DotProduct² and RationalQuadratic. For this reason,
 424 most of the simulations reported in this manuscript were performed using Matern + DotProduct
 425 or Matern + DotProduct² 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.

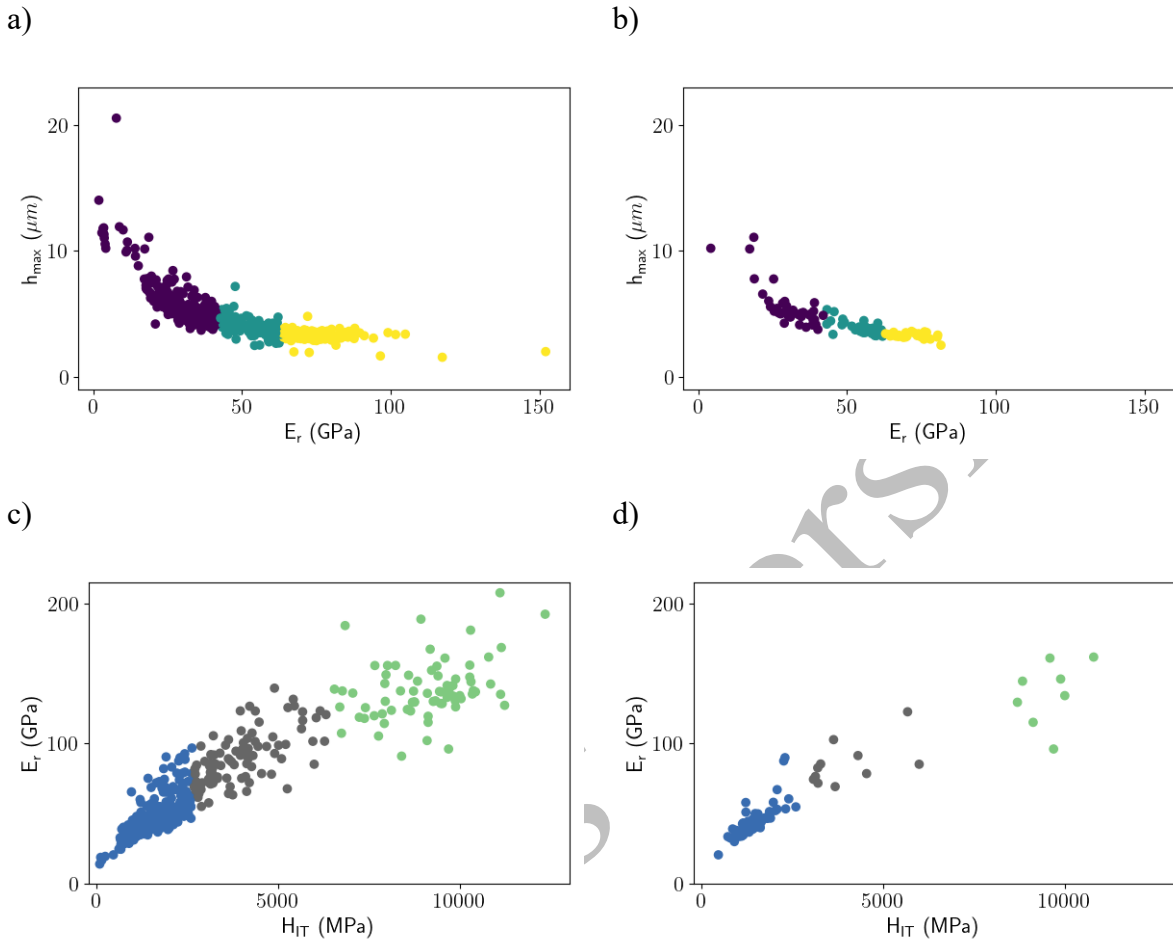
429

430 *3.2 ϵ -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 ϵ -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

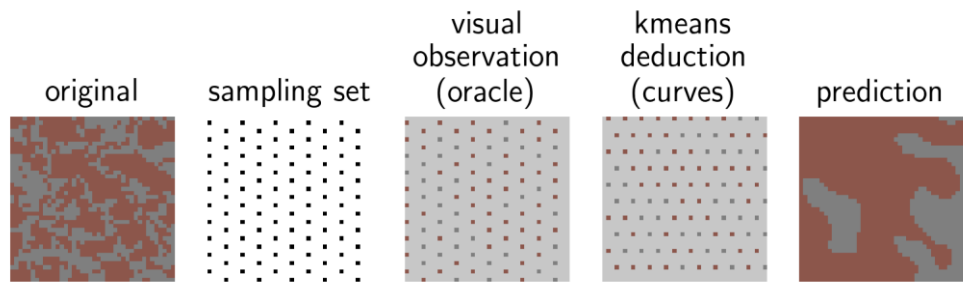
457 The online algorithm, using indentation curves only, showed excellent convergence results with
458 the aforementioned optimized parameters. After unsupervised clustering using the indentation
459 curves as detailed in the previous paragraph, the GP classifier has been used to infer the indents'
460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

a)

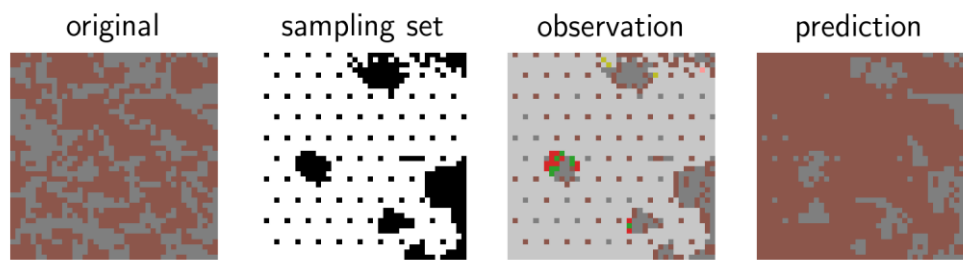
Step 1

41.0% in POI



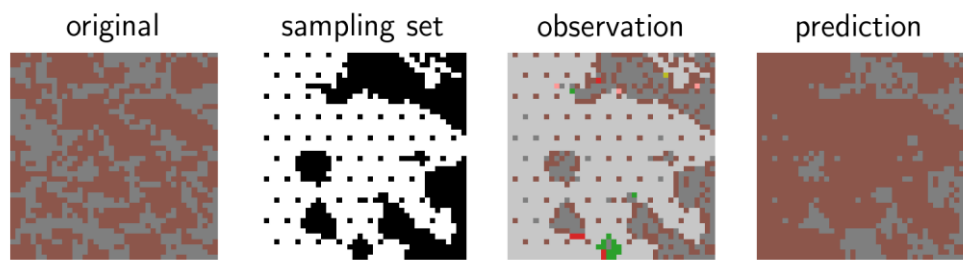
Step 10

51.7% in POI



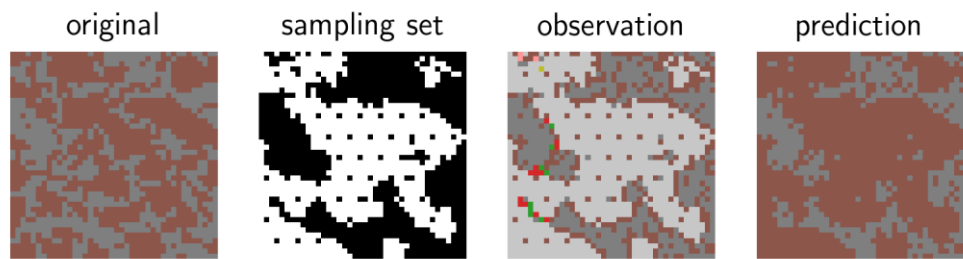
Step 20

54.4% in POI



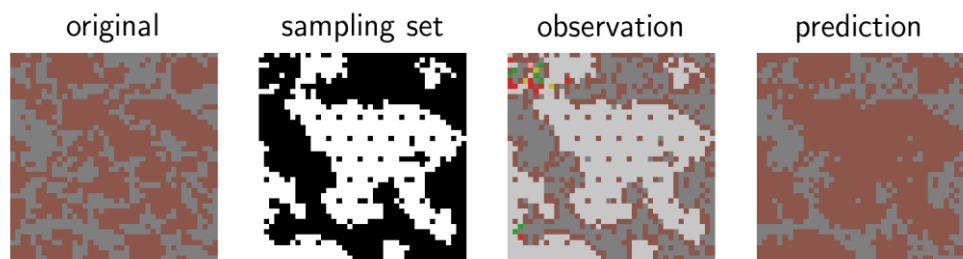
Step 30

56.0% in POI



Step 35

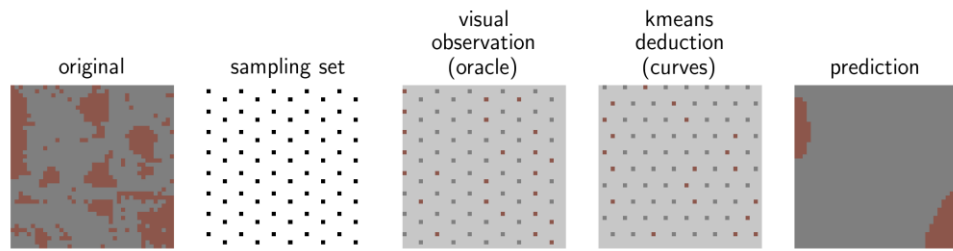
55.8% in POI



b)

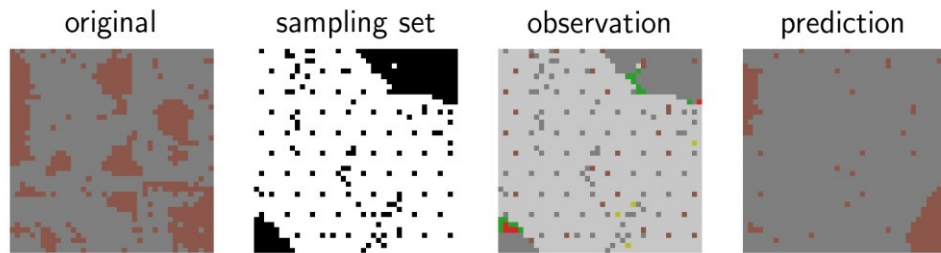
Step 1

75.0% in POI



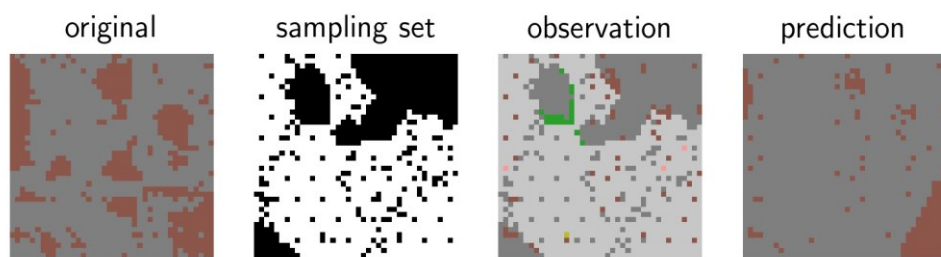
Step 10

88.5% in POI



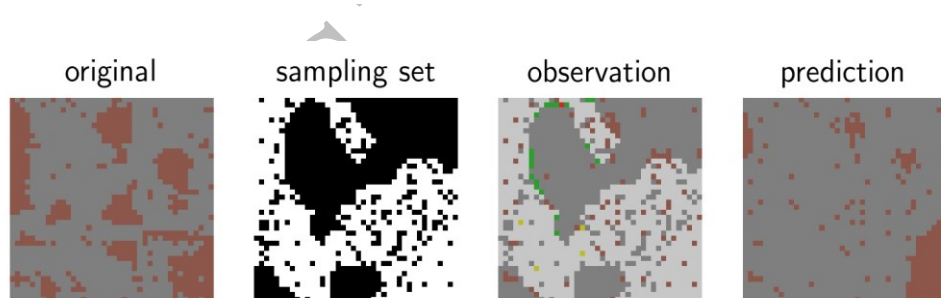
Step 20

87.9% in POI



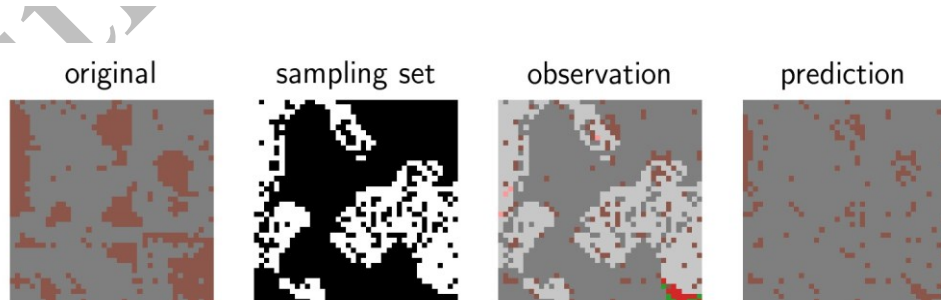
Step 30

89.0% in POI



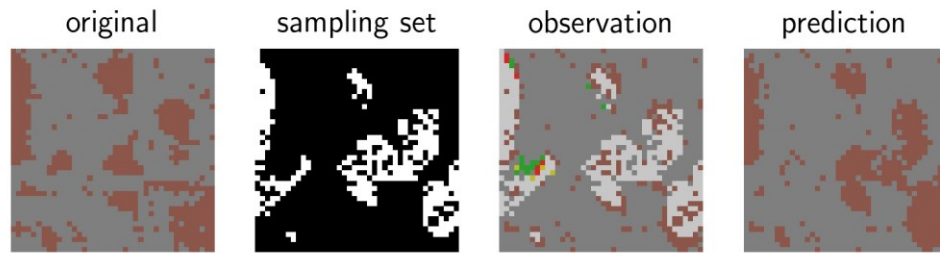
Step 40

87.4% in POI

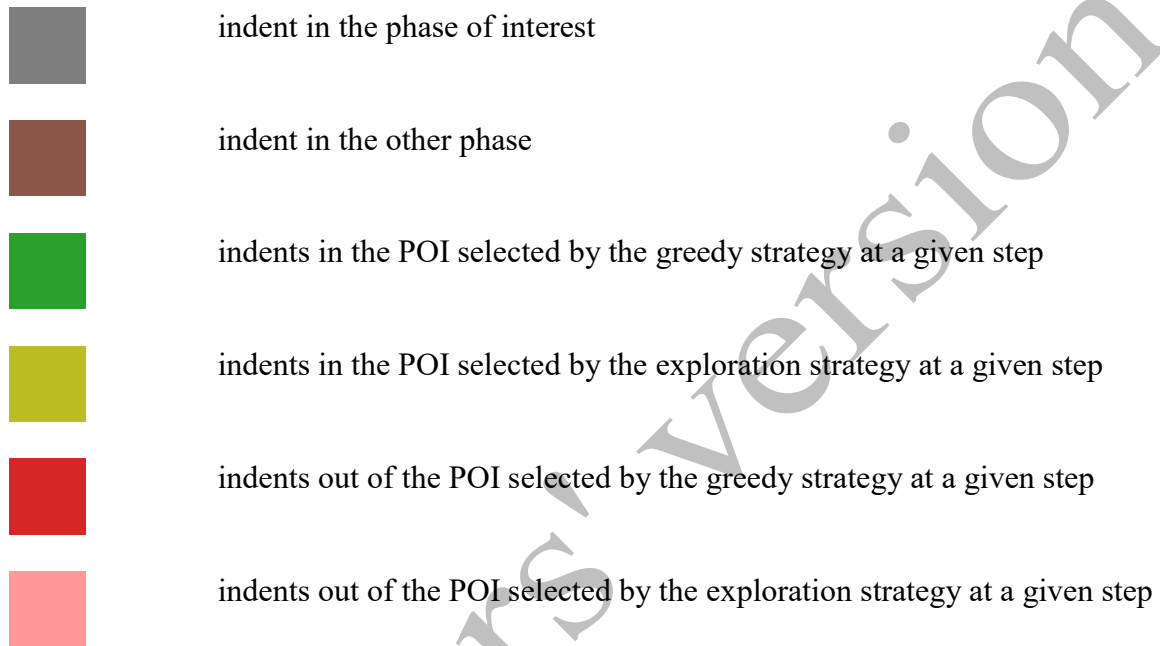


Step 50

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 ϵ -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.

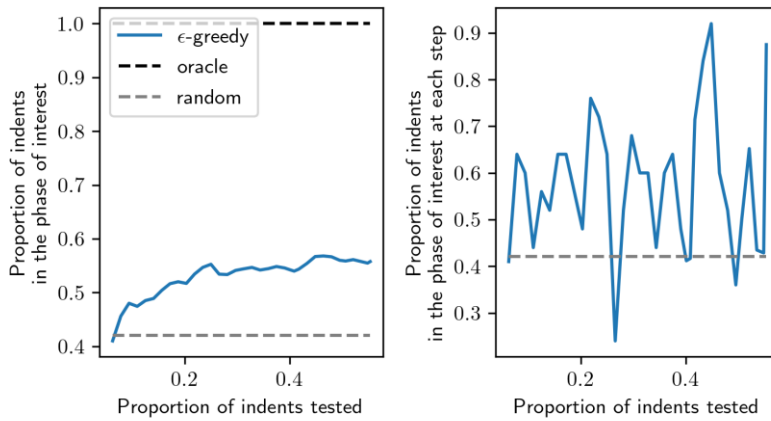
476 In the case of the cement paste specimen, as illustrated in Fig. 12 (b), the number of indents
477 performed in the phase of interest quickly increased. Similar to the case where the microscopic
478 nature of the indents was checked during the algorithm, the algorithm using the indentation
479 curves 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.

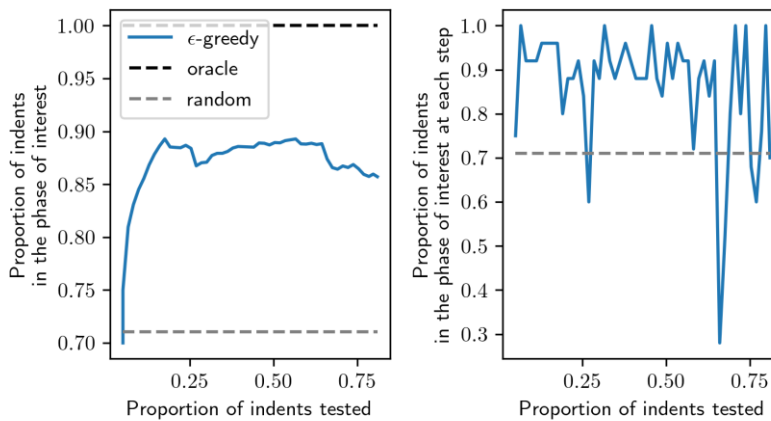
493 Interestingly, as illustrated in Fig. 13, the proportion of indents in the phase of interest, was
494 almost always higher than the random proportion of the phase of interest in the indented area.

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 ϵ -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 ϵ -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, ϵ -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 ϵ -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., 1/16th 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.

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