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1 **Coupling statistical indentation and microscopy to evaluate micromechanical**
2 **properties of materials: application to viscoelastic behavior of irradiated mortars**

3 Benoit Hilloulin^{a*}, Maxime Robira^{a,b,c}, Ahmed Loukili^a

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

7 ^b *SUBATECH, Unité Mixte de Recherche 6457, École des Mines de Nantes, CNRS/IN2P3,*
8 *Université de Nantes, BP 20722, 44307 Nantes cedex 3, France – e-mail:*
9 *robira@subatech.in2p3.fr*

10 ^c *Tractebel Engineering – Le Delage, 5 rue du 19 Mars 1962, 92622 Gennevilliers CEDEX,*
11 *France*

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14 * Corresponding author: Tel.: + 33 (0)2 40 37 16 61

15
16
17 **Highlights**

18 - Data mining principles applied to materials can lead to substantial improvements in
19 the properties identification process.

20 - Coupling indentation and optical analysis reduces identified phase properties'
21 variability.

22 - γ -irradiated mortar samples exhibit greater creep modulus and hardness and lower
23 creep characteristic time than references.

24

25 **Abstract**

26 In this work, an original method coupling statistical indentation and 3D microscope
27 image analysis for heterogenous materials characterization is developed. Statistical
28 microindentation test results performed on γ -irradiated and pristine mortar specimens
29 are presented and analyzed using a clustering data mining technique. The outputs are
30 compared with the phase identification from 3D image analysis to effectively reduce
31 uncertainties in the material properties of one of the phases (cement paste). With respect
32 to the effects of irradiations on cementitious materials, a significant increase of cement
33 paste creep modulus and hardness, and a significant decrease of creep characteristic
34 time, are highlighted after an exposition of 257 kGy at 8.5 Gy /min. Young's modulus
35 of the cement paste is not significantly affected. These results confirm macroscopic
36 concrete creep observations presented in previous studies fitted with dose-dependent
37 logarithmic laws.

38
39
40 **Keywords:** Indentation, Cement Paste (D), Mechanical properties (C), Creep (C),
41 Radioactive Waste (E), Image analysis (B).

42
43 **1. Introduction**

44 The macroscopic mechanical properties of concrete mainly depend on micro-
45 mechanical properties of its binding phase (i.e., the cement paste) and more particularly
46 on the calcium silicate hydrate (CSH) gel, which exhibits significant local variations.
47 For some years, microindentation and nanoindentation have been widely investigated to
48 characterize elasto-plastic and creep properties of cementitious materials [1-4].

49 Indentation elastic parameters can then be input in homogenization schemes to
50 determine concrete elastic properties [5-7].

51 According to Oliver and Pharr theory [8], the initial elastic unloading part of indentation
52 curves can be analyzed to determine the indentation modulus and the indentation
53 hardness of homogenous materials. Because cement paste is highly heterogenous, even
54 at very small length scales, statistical nanoindentation performed at loads leading to
55 penetration depths of some hundreds of nanometers has been developed. Assuming
56 several phases may be indented at the same time, statistical indentation's main objective
57 is to collect enough data points to apply a deconvolution algorithm giving the individual
58 phase properties [9, 10]. However, two main critical aspects were identified regarding
59 the application of statistical indentation technique to cementitious materials [11]: the
60 size of the interaction volume may be larger than the size of the single phases at the risk
61 of creating spurious peaks in the probability density function (PDF) [12, 13] as well as
62 micromechanical values depending on the applied load [14], and the deconvolution
63 analysis based on Gaussian Mixture itself may converge to local minima [15].

64 Therefore, coupling indentation results to other techniques identifying the effective
65 nature of the indents is of great interest at different scales: using atomic force
66 microscope [16] or SEM [6, 17, 18]. Coupling nanoindentation and SEM-EDS to filter
67 data points, Chen et al. highlighted the presence of ultra-high density CSH/Ca(OH)₂
68 nanocomposites in low water-to-cement ratio cement paste by correlating
69 micromechanical properties, e.g. indentation hardness or indentation modulus, to the
70 portlandite volume fraction measured in volumes with approximately the same size as
71 the one investigated through nanoindentation. Localization of indents by imaging
72 techniques can also be used to differentiate the properties of several inclusions [19, 20]
73 and eventually map a restricted area depending on the measured mechanical properties

74 [19, 21]. From the microscale to the macroscale, 3D image analysis of concrete or
75 mortar surface appears to offer a promising field of research for purposes of generating
76 geometric or topological data and supplementing other experimental techniques or
77 providing input for numerical models [22-24].

78 Besides these developments concerning statistical indentation and imaging, it has been
79 found out that the long-term creep properties of concrete specimens are related to the
80 creep properties measured during minute-long microindentation or nanoindentation
81 experiments. Both creep behaviors can be described using logarithmic time-dependent
82 functions with two main variables: creep modulus and creep characteristic time [7, 25,
83 26]. Creep modulus of cement paste is linearly correlated with indentation hardness
84 which means that the lower the hardness, the greater the creep strains, though the slope
85 of the regression depends on the material: creep modulus of pure CSH is greater than
86 the one of cement paste and it decreases with Ca/Si ratio [27]. Like macroscopic creep,
87 indentation creep depends on the relative humidity [28].

88 In the context of nuclear waste disposals and, more generally, in the scope of nuclear
89 safety structures characterization, the assessment of mechanical properties of irradiated
90 concrete is of great interest to speculate over long term behavior of concrete under
91 irradiations. Concrete properties evolutions with radiations have recently been
92 summarized [29]. An extensive literature review ensures that irradiations lead to a
93 decrease of the macroscopic strength of concrete under several types of radiations (α , γ
94 and neutrons). The main mechanism behind concrete degradation under α -radiation is
95 radiation-induced volumetric expansion (RIVE) of siliceous aggregates [30, 31] at doses
96 greater than a reference dose of around 1×10^{20} n/cm² [32]. But under pure γ -radiation
97 (exposition condition of structural concrete element of disposals [33]), degradation
98 mechanisms are not understood yet and there is still a debate whether a reference level

99 of 2×10^5 kGy introduced some decades ago is relevant or not as some recent studies
100 showed degradations after the exposure to lower doses [34, 35]. Water radiolysis
101 triggered by γ -radiation is supposed to be the main phenomenon responsible of possible
102 degradations located in the cement paste [36, 37] and phase alterations occurs only at
103 very high doses of some dozens or even hundreds of MGy [38]. Concrete creep under
104 low dose irradiation (< 1 Gy / h [33]) is one of the major preoccupations concerning
105 long-term behavior of waste disposal infrastructures. To our knowledge, only one study
106 reported the smaller extent of creep of γ -irradiated concrete under compression, though
107 measurements were carried out over a relatively short period of one year without any
108 repetition [39].

109 The growing use of various data mining techniques in civil engineering and materials
110 science applications is changing the way scientists and engineers are facing issues and
111 creating promising paths of investigation at the same time. Larger datasets can be
112 obtained and the challenge is to find useful and innovative information out of them [40].
113 Data mining techniques are being developed to identify materials properties [41] and
114 even leading to the creation of novel materials. In civil engineering [42], data mining
115 has mainly been used for large scale transportation problems for some years [43].

116 A main objective of the present work is to demonstrate the potential of data mining
117 techniques for material properties identification in civil engineering at a microscale and
118 mesoscale, at the crossroads of materials science and civil engineering. For this purpose,
119 two complementary methods, namely statistical microindentation and microscopy, were
120 performed on mortar specimens and combined to detect hidden data trends. Used
121 together with adequate data mining techniques, microindentation data analysis and
122 optical microscopy image analysis are shown to reduce the uncertainties associated with
123 cement paste mechanical properties identification. An application to the determination

124 of mechanical properties of γ -irradiated mortars is then proposed. The first micro
125 mechanical dataset of this type is reported and compared with the only, because tedious,
126 measurement performed on concrete some decades ago [39]. Hundreds of measurement
127 points obtained from 3 irradiated and 3 pristine control specimens are compared to
128 highlight hidden trends due to radiation exposure.

129

130 **2. Materials and methods**

131 **2.1 Specimens preparation and irradiation conditions**

132 Mortar was prepared with CEM I 52.5 and 0/4 mm calcareous sand (to avoid the
133 activation of alkali-silica reaction by irradiation) with the proportions detailed in
134 Table 1. Any use of organic additives like superplasticizer or demolding oil was
135 avoided to not induce a possible premature degradation by the irradiations. This mortar
136 formulation was determined to be as representative as possible of a high-performance
137 concrete used in nuclear waste storage facility galleries.

138 Six mortar prisms with dimensions of 4 x 4 x 16 cm³ were cast in polypropylene molds
139 to avoid any presence of metallic compounds from the molds. After 1 day of curing
140 under sealed conditions in an air-conditioned room at a temperature of 20°C and
141 90% RH, the specimens were demolded. The specimens were further cured in lime-
142 saturated water until the age of 28 days. Mortar prisms were then dried during 14 days
143 in an oven at 45°C (a constant mass was measured at 10 days).

144 Half of the mortar prisms (MD-257kGy-I1 to MD-257kGy-I3) were then introduced in
145 an irradiator at ARRONAX (¹³⁷Cs source, 661 keV, 123.4 TBq) at the age of 42 days.
146 Specimens were exposed as close to the source as possible to guarantee a spatially
147 homogenous dosage during 3 weeks. The total γ dose received by the specimens was
148 calculated to be around 257 kGy based on a map of the γ fluxes in the irradiator realized

149 by Fricke dosimetry measurement. The other half of the specimens (MD-257kGy-S1 to
150 MD-257kGy-S3), e.g. the control specimens, were kept in an air-conditioned room at
151 relative humidity of around 65% close to the ones measured in the irradiator at the end
152 of the irradiation period.

153 **Table 1.** Mortar compositions

Cement (kg/m ³)	Calcareous Sand 0/4 (kg/m ³)	Water (kg/m ³)	W/C	Paste volume (%)
566	1344	270	0.43	45

154
155 Several mechanical and chemo-physical tests were performed on these mortars and on
156 other series as well. The results will be presented in future communications but, in the
157 sake of clarity, some results may be evocated in regards to micromechanical results
158 presented herein. No evident carbonation, pore size evolution or hydrated phases
159 transformations were measured. Thus micromechanical properties evolutions may not
160 be attributed to calcite formation in contrast with [44].

161

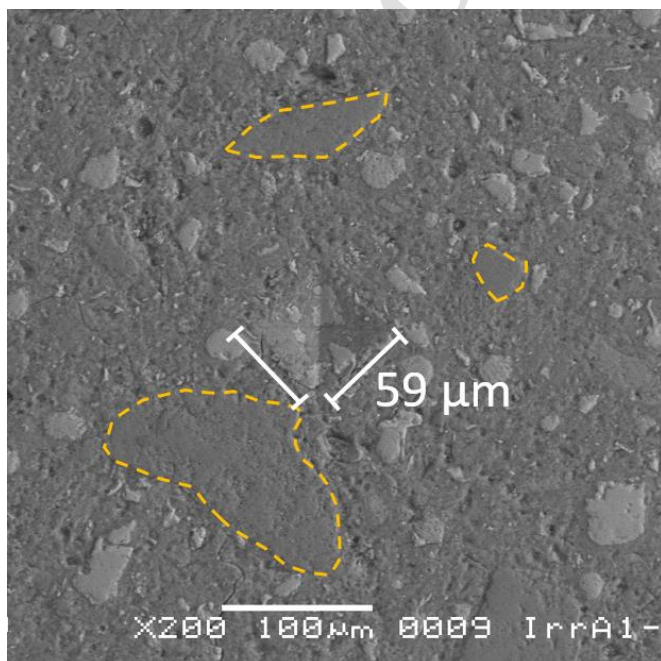
162 **2.2 Indentation setup and theory**

163 Microindentation tests were performed during the week following the irradiation period
164 with the main objective of comparing the properties of irradiated and control specimens.
165 Thus, a volume of 2 x 2 x 1.5 cm³ was sawn from the middle part of a half of the
166 4 x 4 x 16 cm³ broken by three-point bending test. This location was selected to avoid
167 damage due to the three-point bending test. The 2 x 2 cm² section was polished with Si-
168 C paper with decreasing particle size (500, 1200, 2000, 4000) using ethanol as polishing
169 liquid. Polishing times were selected around some minutes per paper to limit the risk of
170 aggregate cracking. These times are much shorter than the ones used for

171 nanoindentation on cement pastes [45] but were sufficient to obtain a surface roughness
172 R_q of around 0.5 - 1 μm which is acceptable for micro-indentation with penetration
173 depth of some microns.

174 A typical cement paste area with a typical indent is presented in Fig. 1. As it can be
175 observed, the scale of the indent is larger than the characteristic scale of the
176 microstructure of the cement paste (residual anhydrous clinker size): thus, the
177 performed microindentation tests provided the mechanical properties of the cement
178 paste or the sand, or the interface and not of the individual phases of the cement paste.

179 Microindentation was performed using a Vickers indenter probe over a grid of
180 20 x 20 points, evenly spaced by 500 μm to investigate a representative surface of
181 1 x 1 cm^2 of the mortar sample. For each indent, the load was increased linearly over
182 time in 5 s up to 2000 mN, kept constant during the 60 s holding phase, and decreased
183 linearly over time back to zero in 5 s. The very short loading time was selected to limit
184 creep during this period and do not apparently damaged the sample.



185 **Fig. 1.** SEM observation of a typical indent located in the cement paste with a width of
186 59 μm (small sand fines located near the indent are circled by orange dashed lines).

187

188 Various properties were calculated from the load – penetration curves. First, the
189 indentation hardness depending on the maximum load and the projected contact area:

$$H_{IT} = \frac{P_{max}}{A_p(h_c)} \quad (1)$$

190 For Vickers' indenter the projected contact area can be estimated by:

$$A_p(h_c) = 24.50h_c^2 \quad (2)$$

191 Where h_c can be calculated based on maximum indentation depth h_{max} assuming:

$$h_c = h_{max} - \varepsilon(h_{max} - h_r) \quad (3)$$

192 with $\varepsilon = 0.75$ and h_r is the final ideal penetration height can be determined. Martens
193 hardness was also computed according to:

$$H_M = \frac{P_{max}}{A_s(h)} \quad (4)$$

194 Where $A_s(h)$ is the contact area, depending on the indentation depth h , defined for a
195 Vickers indenter by:

$$A_s(h) = \frac{4 \sin(\alpha/2)}{\cos^2(\alpha/2)} h^2 \approx 26.43h^2 \quad (5)$$

196 With $\alpha=136^\circ$ the top angle of the Vickers pyramid.

197 The Young's modulus E_{IT} of the indented material is given by:

$$M = \frac{E_{IT}}{1 - \nu_s^2} = \frac{1}{\frac{1}{E_r} - \frac{1}{M_{ind}}} \quad (6)$$

198 Where M denotes the indentation modulus, $M_{ind} = 1140 \text{ GPa}$ the modulus of the
199 indenter, ν_s the Poisson ratio of the material (considered equal to 0.2, a possible

200 variation of some percent of EIT may be induced for other values of Poisson ratio) and
 201 E_r is the reduced modulus which is determined assuming:

$$E_r = \frac{1}{2} \left(\frac{dP}{dh} \sqrt{\frac{\pi}{A}} \right) \text{ at } h = h_{max} \quad (7)$$

202 A first creep parameter C_{IT} , e.g. the normalized indentation creep parameter, was
 203 calculated by the machine. It is defined as:

$$C_{IT}(\%) = \frac{h_{max} - h_1}{h_1} \times 100 \quad (8)$$

204 With h_1 the indentation depth at the beginning of the creep stage.

205 Energetic parameters n_{IT} and W_t were also recorded. n_{IT} is the proportion of the elastic
 206 response relatively to the total energy W_t : $n_{IT} = W_{elas} / W_t$.

207 More meaningful creep parameters C and τ can be extracted from the indentation curves
 208 (see Fig 2a)) calculating the creep function $L(t)$ [21]:

$$L(t) - L(0) = L(t) - \frac{1}{M} = \frac{2a_u \Delta h(t)}{P_{max}} \quad (9)$$

209 Where $\Delta h(t)$ denotes the indentation depth increase during the constant load phase, a_u
 210 denotes the is the radius of the equivalent projected contact area between the indenter
 211 probe and the indented surface at the onset of unloading

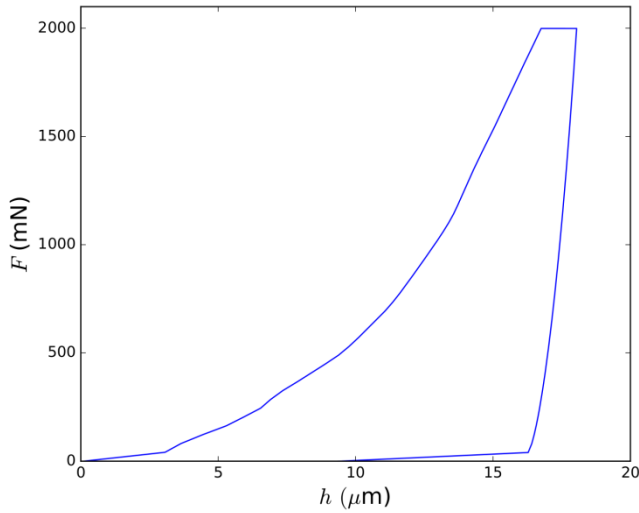
212 Assuming a logarithmic fit of the creep function, the creep parameters are defined
 213 according to [25]:

$$L(t) - \frac{1}{M} = \frac{\ln(t/\tau + 1)}{C} \quad (10)$$

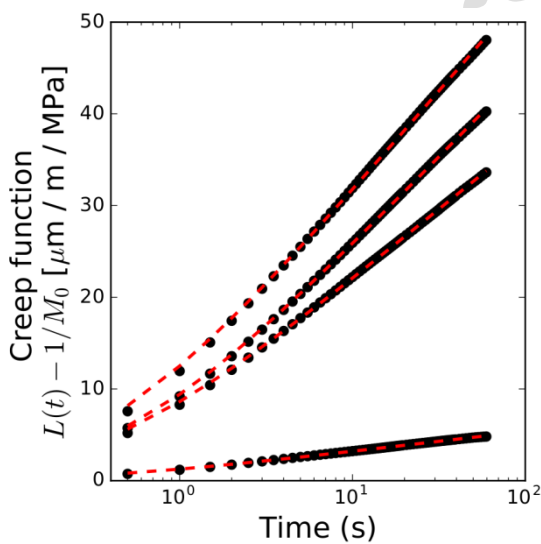
214 It was verified that this law can adequately reproduce the creep behavior of both cement
 215 paste and sand as illustrated in Fig. 2 b). In order to avoid drying during creep
 216 experiments, indentation was realized at a relative humidity close to the one imposed
 217 during the irradiation period (around 65% RH). Moreover, autogenous shrinkage of the

218 sample can be neglected at the age of 72 d. Then, the creep function is expected to
219 characterize the basic creep behavior of the specimens.
220

a)



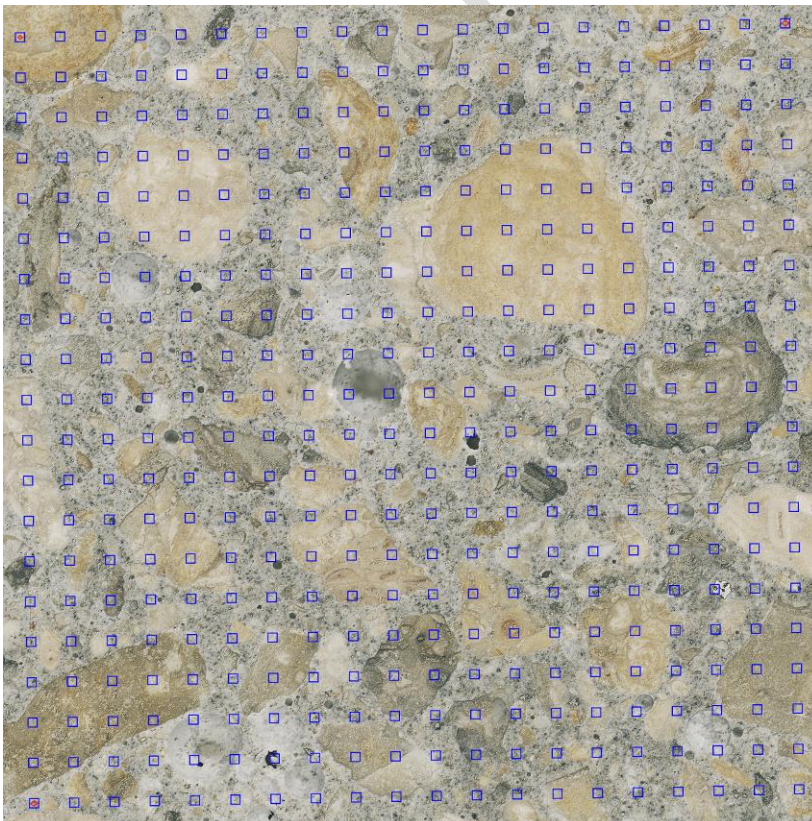
b)



221 **Fig. 2.** Microindentation outputs: a) raw load – penetration curve, b) fitting the creep
222 function by a logarithmic law (3 curves correspond to cement paste indents and 1 to a
223 sand indent)
224

225 **2.3 3D microscope measurements and analysis**

226 After indentation, a map of the indented zone was realized using a Hirox RH-2000 3D
227 microscope by merging around 200 3D reconstructed images evenly spaced along the
228 indented area. The selected magnification (x 140) led to a final horizontal resolution of
229 the 2D projected image of 1.5 μm / pix which is adequate to correctly locate the indents
230 and assess their nature. Because some indents may be hardly located, especially the
231 ones created in voids, a basic routine was implemented in Python to locate all the
232 indents based on the location of 3 of them by drawing blue squares on the original
233 image. As illustrated in Fig. 3, the 20 x 20 matrix of indents covers a sufficiently large
234 area representative of the mortar volume and the reconstructed image correctly
235 represent the matrix without image distortion due to multiple image merging (it was
236 verified that the indents were correctly located by the blue squares). The nature of the
237 indents was then manually identified because color-based filters were found to be
238 relatively imprecise in the case of such mortar images.



239

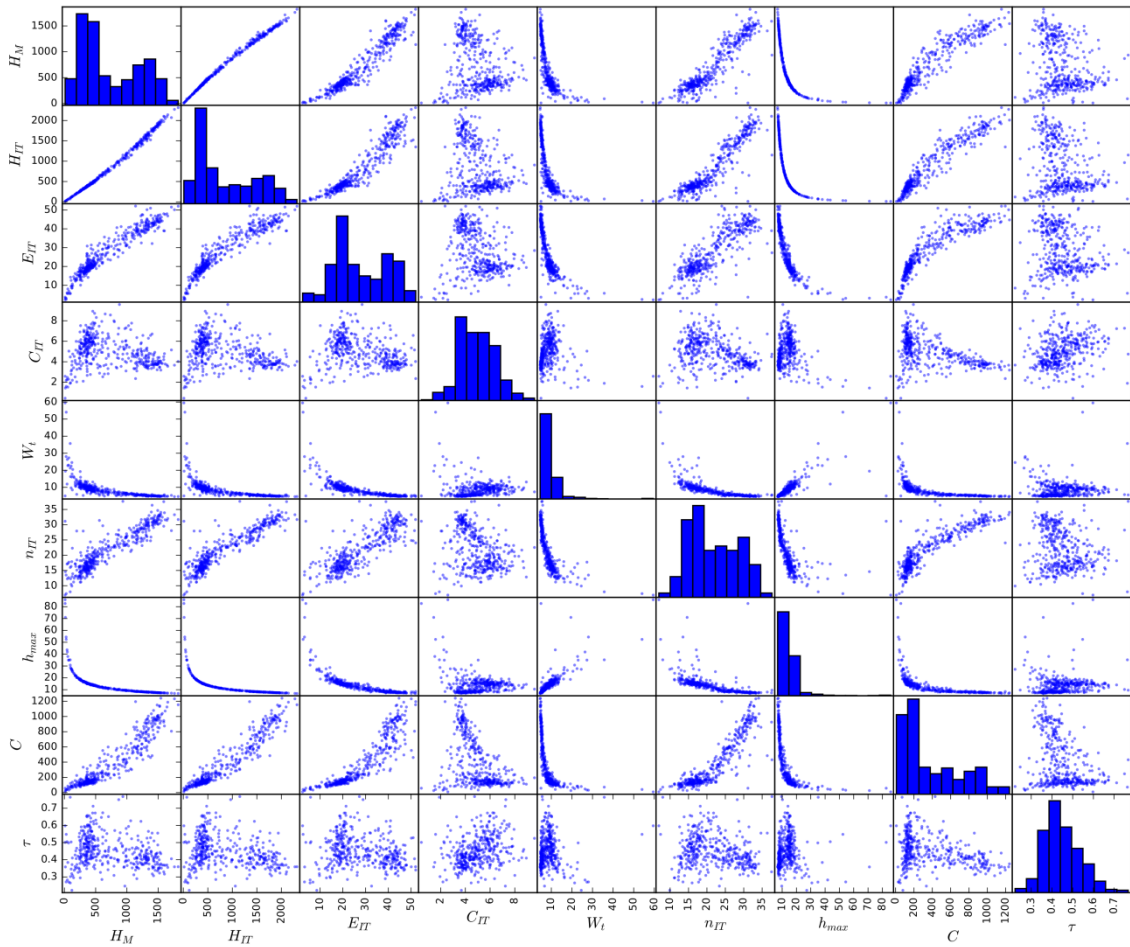
240 **Fig. 3.** 2D projected global view of the 20 x 20 matrix of indents over a 1 x 1 cm²
241 surface from 3D microscope acquisition (indents are indicated by blue squares)

242

243 **3. Results and discussion**

244 **3.1 Statistical indentation data deconvolution and its variability**

245 First, indentation outputs from each specimen were visualized by mean of a scatter plot
246 as illustrated in Fig. 4. Several basic observations directly arise from the analysis of the
247 scatter plot. Some data points are isolated from the others. These isolated points are
248 mainly indents with a maximum penetration depth h_{\max} greater than 20 μm . In order to
249 properly run data deconvolution, these points were filtered out on all samples. As we
250 will see in the next paragraph, they correspond to indents close to voids or local defects
251 and therefore do not correctly represent cement paste properties. Due to sand
252 heterogeneities, some data points exhibit indentation hardness greater than 2500 MPa.
253 Such points were also filtered out of the analysis. Already reported correlations can be
254 highlighted in the scatter plot: correlation between hardness and Young's modulus,
255 linear-shaped correlation between hardness and creep modulus. Looking at the PDFs
256 located along the diagonal of the scatter plots, one may also observe that two main
257 groups, presumably the cement paste and the sand, can be identified (especially on H_{IT}
258 and E_{IT} PDFs). However, these two groups considerably overlap each over, probably
259 due to a non-negligible proportion of the indents at the interface between cement paste
260 and sand grains.



261

262 **Fig. 4.** Unfiltered scatter plot from sample MD-257kGy-S3 (H_M and H_{IT} are expressed
 263 in MPa, E_{IT} and C in GPa, C_{IT} and n_{IT} in %, W_t in μJ , h_{max} in μm and τ in s).

264 Then, data points repartition was visualized using a Euclidian distance-based
 265 dendrogram generated from the 9 output variables as illustrated in Fig 5 a). This way,
 266 the similarities between the data points can be highlighted without subjectivity
 267 (subjectivity may come from the arbitrary choice of the number of clusters prior to
 268 deconvolution, the output variables used to generate the clusters, e.g. typically 2 [9] or 3
 269 [14] only). The data points mainly belong to two dissimilar groups joining at a high
 270 around 60 and with their subgroups differentiated at highs around 20.

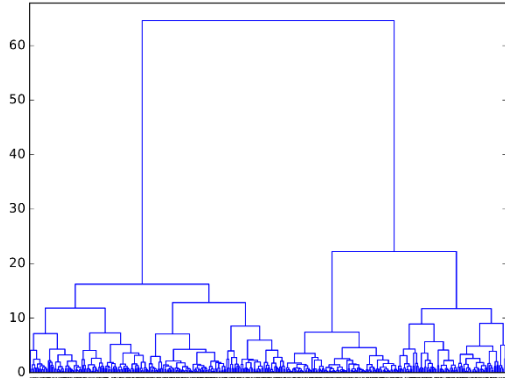
271 Cutting the dendrogram at a high of 30 leads to a 2-groups hierarchical clustering
 272 represented on a C - H_{IT} 2D plane (Fig. 5 b)) with one group probably mainly composed
 273 of cement paste and one group probably mainly composed of sand. Cutting the

274 dendrogram at a high of 20 leads to 3 groups (Fig. 5 c)). Naturally, because of
275 hierarchical clustering principles, only one of the two groups has been divided into two
276 subgroups, and for most of the mortar samples it is the group with the highest hardness
277 (presumably sand indents). Then, it means that, at the selected indentation load, sand
278 grains are more heterogeneous than the cement paste. It also leads to another
279 observation: 2-groups (and 3-groups) hierarchical clustering clearly overestimates
280 cement paste proportion, if one basically assumes that the group with the smallest mean
281 hardness corresponds to the cement paste. Indeed, reading the dendrogram, the
282 horizontal proportion of the first group corresponding to the lowest hardness is greater
283 than the one of the second group corresponding to the highest hardness. As the
284 horizontal axis is composed of all the single data points, it would mean that the number
285 of indents in the cement paste is greater than the number of indents in the sand. That is a
286 biased conclusion because the paste volume fraction is only of 45% (and at this
287 indentation load, an important number of indents are located at the interface between
288 cement paste and sand grains and they would lead to intermediate properties). This
289 erroneous prediction of the phase volume fractions has already been reported elsewhere
290 and the assignation of mean phases properties based on numerical data deconvolution is
291 questionable [15]. This basic observation motivates the use of optical techniques
292 described hereafter to filter experimental data based on indents nature.

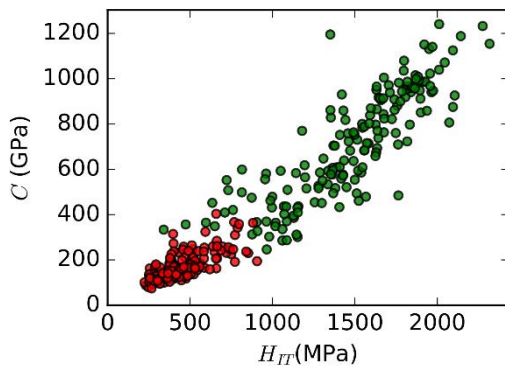
293 Reporting the nature of the indents predicted by deconvolution over the microscopic
294 image as illustrated in Fig. 6, a relatively good correspondence is noticeable: red
295 squares corresponding to the first deconvoluted group are mainly located in the paste
296 and blue ones, corresponding to the second group are mainly located in sand grains,
297 while black squares, corresponding to initially filtered values are all near voids.
298 However, some indents (2 of them were circled in orange as examples) numerically

299 identified as cement paste are located on sand grains and vice versa which emphasizes
300 the limits of numerical clustering.

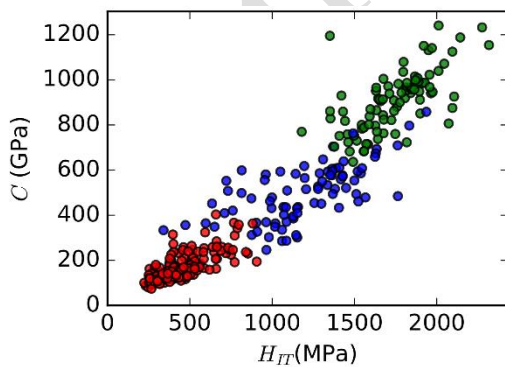
a)



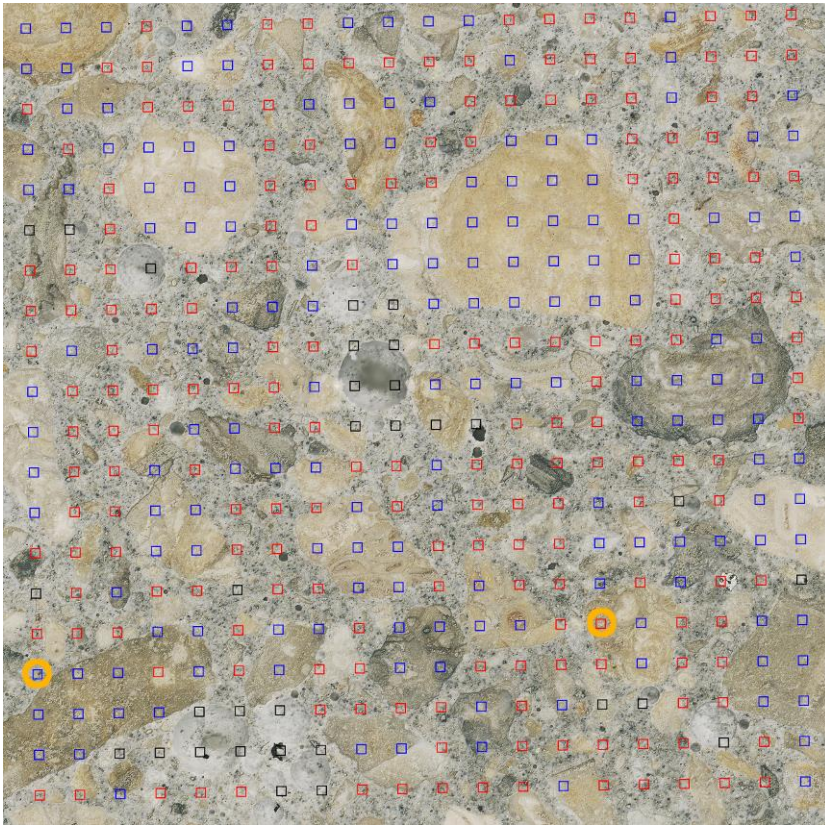
b)



c)



301 **Fig. 5.** Hierarchical clustering strategy applied to specimen MD-257kGy-S3: a)
302 Euclidian distance-based dendrogram (with the 400 individual data points on the x-
303 axis), b) and c), data points repartition along C- H_{IT} plane using a 2-groups separation
304 (b) and a 3-groups separation (c).



306

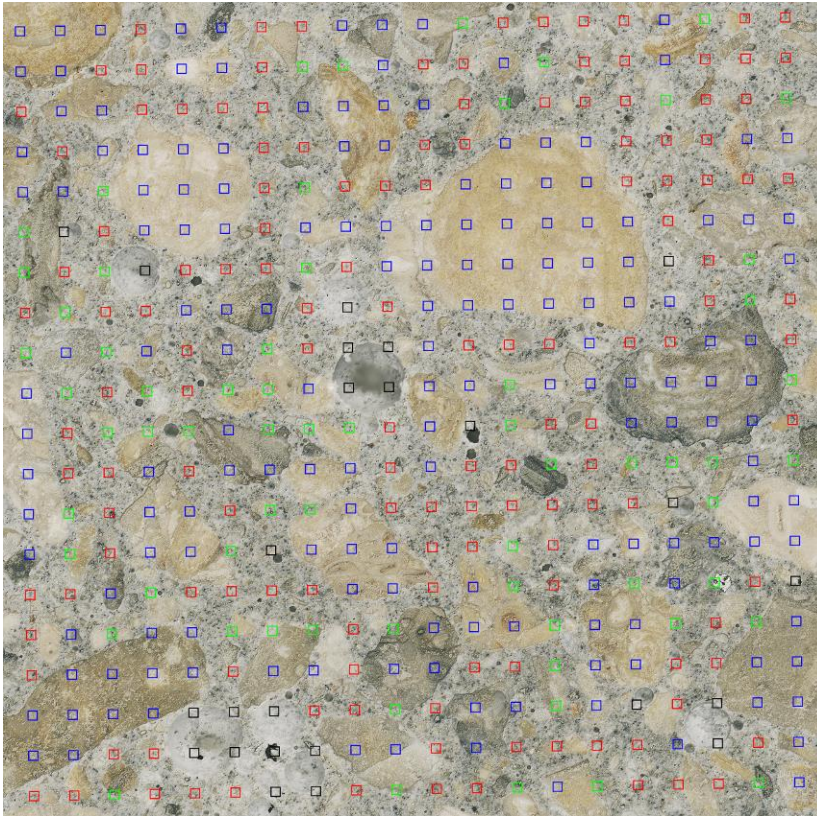
307 **Fig. 6.** Representation on the 3D microscopic image of specimen MD-257kGy-S3 of the
 308 nature of the indents predicted by data deconvolution (red squares correspond to group
 309 1, e.g. 'paste', blue squares to group 2, e.g. 'sand, and black squares to filtered values
 310 before the analysis). Orange circles indicate two indents attributed to the wrong group
 311 as an example of the limits of numerical clustering. Indentation area extends over
 312 $1 \times 1\text{cm}^2$.

313

314 3.2 3D microscope results

315 Indents were visually classified into 4 groups: voids, paste, sand and interface between
 316 paste and sand so that the latter 3 could be compared to the 3 groups obtained by
 317 hierarchical clustering. As illustrated in Fig. 7, there is an important proportion of
 318 indents located at the cement paste - sand grains interface marked by green squares.
 319 This is directly correlated to the indent size relatively to the size of the phase and the

320 volume fraction of sand. Comparing Fig. 6 with Fig. 7, one may notes that the other
321 indents are on majority of the same nature as the indents attributed to one group using
322 the hierarchical clustering method.

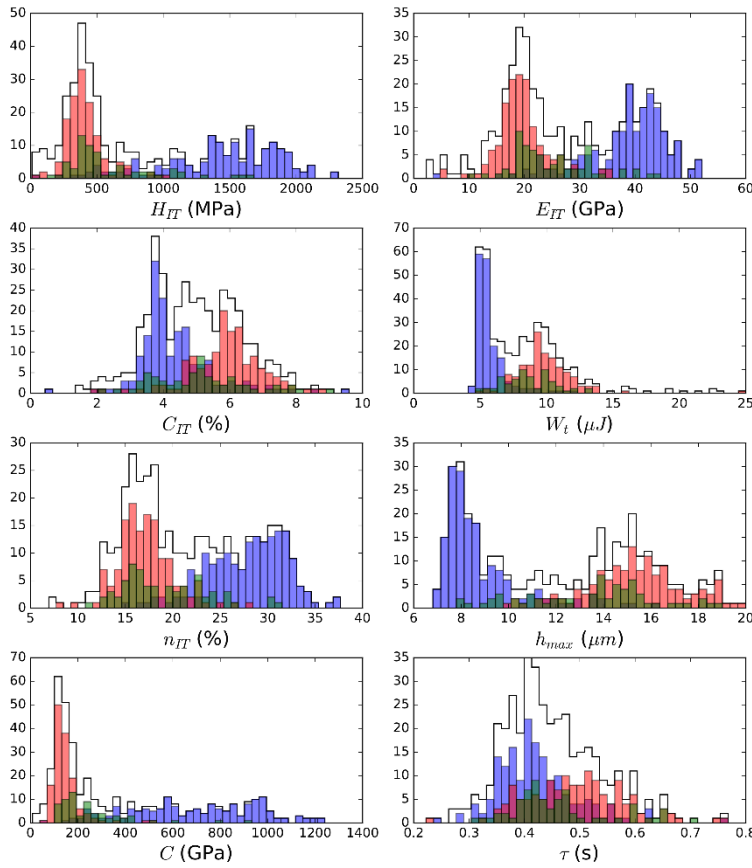


323
324 **Fig. 7.** Indents location and nature of specimen MD-257kGy-S3 (paste in red, cement
325 paste - sand grain interface in green, pure sand in blue, void in black) according the 3D
326 microscope image analysis.

327
328 From the repartition of the indented outputs variables per groups using 3D microscope
329 (Fig. 8), it can be observed that cement paste and sand indentation variables
330 distributions closely match normal distributions. On the other side, properties associated
331 with indents located at interfaces between cement paste and sand grains (green
332 histograms) are much more variables and do not follow any specific distribution. This is
333 due to the fact that indents on interfaces could either exhibit the mean value when two
334 phases properties are probed (for example there are a lot of indents on interfaces with

335 Young's modulus between 25 and 35 GPa, because this property is representative of a
336 large volume), either be very similar to cement paste or sand properties because the
337 indenter probe slide to one side during loading (most of the time, to the cement paste
338 side because of its lower altitude). For this reason, a lot of indents on interfaces have an
339 indentation hardness around 500 MPa, e.g. around the mean value of indents attributed
340 to the cement paste as this property is representative of a smaller volume.

341 Identifying phases properties based on imaging is therefore possible for predominant
342 phases like cement paste or sand. Distributions are well defined but some indents
343 properties are possibly associated with the wrong group. For example, in Fig. 8, one can
344 see that some indents with high hardness and creep modulus (around 1 GPa and
345 400 GPa resp.) were identified as cement paste while some indents with Young's
346 modulus lower than 15 GPa were identified as sand grains. More than half of these
347 values are associated with either an odd indent shape on sand or the presence of very
348 small sand grains close to an indent attributed to the cement paste (most of these indents
349 are probably rightly on cement paste but the indentation variables may be influence by
350 very close sand grains even under the indent). Thus, although the optical identification
351 of the indents should be attractive and sufficient in some cases, the combination of a
352 numerical deconvolution technique and of an imaging technique may help reduce the
353 uncertainties concerning some minor odd indents outputs.



354

355 **Fig. 8.** Histograms of distributions of the indentation output variables of specimen MD-
 356 257kGy-S3. Total distribution is represented by the continuous black line, distributions
 357 of the phases identified using the 3D microscope are colored in red (cement paste), blue
 358 (sand grains) and green (cement paste – sand grain interface).

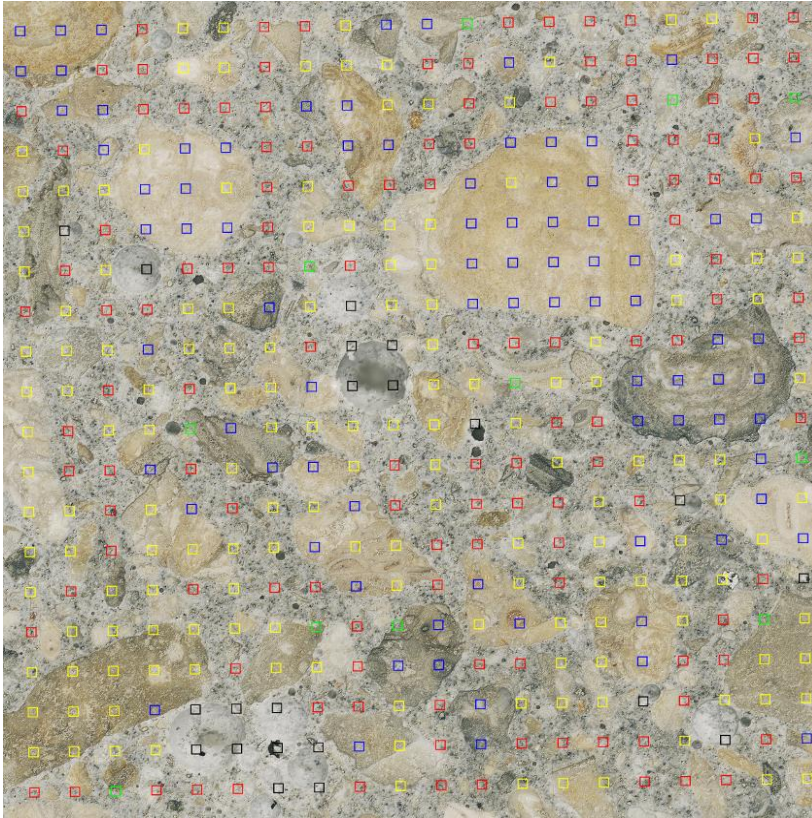
359

360 **3.3 Combination of 3D microscope and indentation results**

361 3D microscope and indentation data deconvolution phase assignments were combined
 362 to reduce the variability of the identified phase properties. Comparing the indent natures
 363 from the two first order techniques (3D microscope 3 groups identification vs 3 groups
 364 hierarchical clustering), the indents with corresponding natures were considered as
 365 representative of a given phase while the others were considered as non-reliable
 366 information regarding the final application of this study, e.g. cement paste
 367 characterization. This coupling may address the two main preoccupations of the first

368 order analysis: concerning numerical clustering this method can effectively provide a
369 supplementary information about the nature of the indents and, concerning optical
370 clustering, the method can considerably decrease the risk of false identification (either
371 due to the operator or to the image precision) by selecting clusters of indents with close
372 micromechanical properties.

373 From a visual inspection of the spatial repartition of the nature of the indents in one of
374 the worst cases (Fig. 9), it should be observed that there is a huge proportion of
375 noncorresponding indent natures (yellow squares). Indeed, when comparing 3 groups
376 from 3D microscope identification and 3 groups from data deconvolution, around 35%
377 of indents nature do not correspond between the two analysis, while this proportion is
378 usually around 25% for the comparison between 3 groups from 3D microscope and 2
379 groups from data deconvolution. A majority of noncorresponding indents are located in
380 sand grains or at their periphery and this is the reason explaining why there is less error
381 using 2 groups data from deconvolution data: data points numerically identified as
382 belonging to the intermediate group were mostly visually identified as sand grains and
383 not sand grain – cement paste interface. Only a very few indents were identified as
384 interfaces by combining the two methods which means that groups identified by
385 clustering algorithms with intermediate properties are not necessarily different phases
386 (here sand grain – cement paste interface). Moreover, the variability of sand grain
387 properties could also be visualized as for some sand grains almost all indents are
388 corresponding ones, while for others, probably with smaller hardness, indents are
389 mostly noncorresponding ones (identified as sand for one analysis, interface for the
390 other).

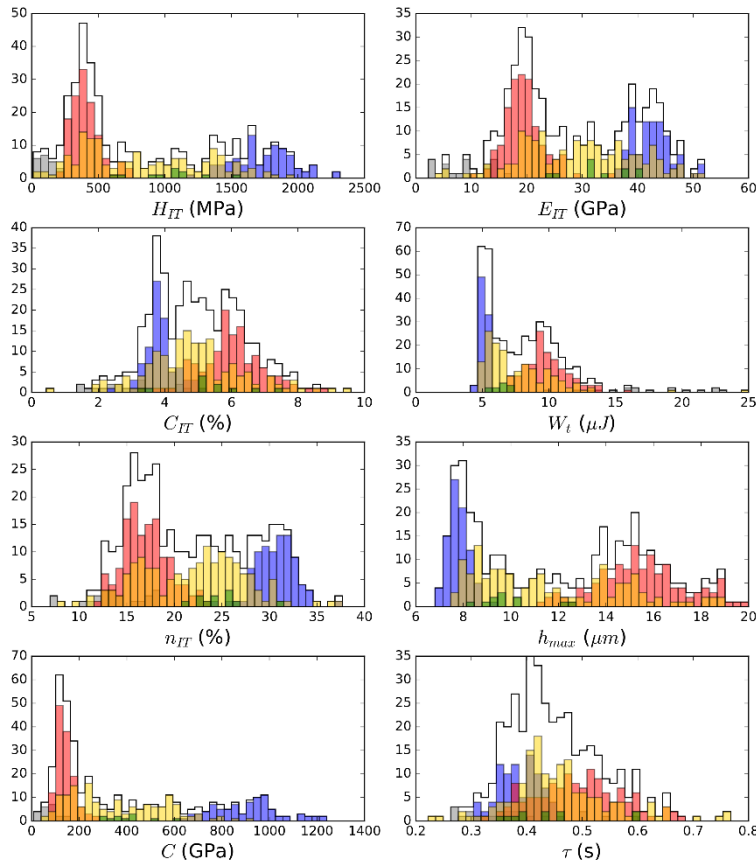


391

392 **Fig. 9.** Indents location and nature of specimen MD-257kGy-S3 comparing 3D
393 microscope image analysis and 3 groups hierarchical data deconvolution (paste in red,
394 cement paste - sand grain interface in green, pure sand in blue, void in black and indents
395 with noncorresponding natures between the two analyses in yellow).

396

397 A detailed analysis of the microindentation output variables through histograms (Fig.
398 10) reveals that noncorresponding indents are mainly the ones with intermediate
399 properties as predicted by the visual observation. Interestingly, the initial goal of this
400 comparison method seems to have been achieved: cement paste and sand distributions
401 are tightened and do not overlap for Young's modulus, hardness and creep modulus.
402 Moreover, some of the extreme values are filtered out. Thus, the comparison of
403 numerical data deconvolution and optical data clustering efficiently reduces the
404 uncertainties concerning phase identification because of the complementarity between
405 the two methods.



407

408 **Fig. 10.** Histograms of distributions of the indentation output variables of specimen
 409 MD-257kGy-S3 after the combined use of numerical and clustering and image analysis.
 410 Total distribution is represented by the continuous black line, distributions of the phases
 411 are colored in red (cement paste), blue (sand grains), green (sand grain – cement pastes
 412 interface). The distribution of noncorresponding indents is represented in yellow.

413

414 **3.4 Creep properties of γ -irradiated mortar specimens**

415 Using the method described in the previous paragraphs, cement paste properties of
 416 irradiated and control mortar specimens were compared. Tables 2 to 4 summarize the
 417 indentation outputs associated with hierarchical clustering, 3D-microscope image
 418 analysis and the method coupling numerical indentation data clustering and 3D-
 419 microscope image analysis. As explained, the main interest of the technique is to reduce

420 the variability of the indentation outputs attributed to one phase, here the cement paste.
421 This objective is achieved for most of the outputs as the variances of the outputs
422 variables distributions in Table 4 are smaller than the corresponding ones on Table 2
423 and 3 both on individual specimens and on the two series merging data from the 3
424 specimens per series. Moreover, mean values of hardness, Young's modulus, creep
425 parameter are smaller on Table 4 because some data points with intermediate properties
426 were filtered out comparing numerical clustering and optical clustering.

427 One may finally observe that it is rather difficult to draw some conclusions on the
428 evolution of the output variables due to irradiations from these data as the standard error
429 (square-root of the variance) is close or greater than the difference between the two
430 series. Thus, indentation outputs were grouped together whether they belong to
431 irradiated specimens or not. The merged data is represented on Fig. 11. Analyzing non-
432 diagonal graphics, one may observe that data points are rather well concentrated and
433 dispersion limited. On diagonal probability distributions (PDF), a clear shift of C due to
434 irradiations can be observed towards greater values while τ decreases. Indentation
435 hardness also seems to be affected, though statistical analysis is necessary to conclude
436 concerning a possible influence of irradiations.

437

438 **Table 2.** Indentation outputs for cement paste indents identified using hierarchical
 439 clustering (means and variances in brackets).

	HM (MPa)	HIT (MPa)	EIT (GPa)	CIT (%)	Wt (μ J)	nIT (%)	hmax (μ m)	C (GPa)	τ (s)
MD-257kGy-I1	377.4 (5424)	402.7 (6192)	19.5 (5.2)	5.16 (1.22)	10.2 (2.0)	16.7 (4.2)	15.40 (2.30)	173 (1111)	0.384 (0.005)
MD-257kGy-I2	432.0 (13035)	466.6 (16610)	21.0 (15.5)	5.20 (1.51)	9.3 (2.8)	18.1 (8.1)	14.54 (3.30)	197 (3534)	0.416 (0.007)
MD-257kGy-I3	438.8 (16321)	478.8 (22917)	21.3 (28.2)	5.04 (1.02)	9.2 (2.8)	19.0 (14.9)	14.46 (3.92)	210 (8623)	0.447 (0.005)
MD-257kGy-S1	396.9 (9088)	415.4 (10205)	20.2 (8.4)	6.00 (1.29)	10.1 (2.4)	16.7 (6.2)	15.26 (3.13)	149 (1006)	0.465 (0.005)
MD-257kGy-S2	369.3 (6869)	395.2 (8737)	19.9 (12.5)	4.99 (1.89)	9.7 (3.0)	17.5 (10.5)	15.58 (2.79)	182 (4653)	0.426 (0.006)
MD-257kGy-S3	416.1 (16467)	441.0 (20063)	20.8 (18.1)	5.77 (1.43)	9.7 (3.2)	17.3 (7.4)	15.01 (4.34)	165 (3481)	0.481 (0.007)
MD-257kGy-I	421.8 (13019)	456.2 (17314)	20.7 (18.2)	5.13 (1.27)	9.5 (2.7)	18.1 (10.4)	14.71 (3.43)	196 (4998)	0.420 (0.006)
MD-257kGy-S	396.2 (12102)	420.2 (14600)	20.4 (14.4)	5.55 (1.73)	9.8 (3.0)	17.2 (8.3)	15.25 (3.62)	168 (3539)	0.459 (0.007)

440

441 **Table 3.** Indentation outputs for cement paste indents identified using 3D-microscope
 442 image analysis (means and variances in brackets).

	HM (MPa)	HIT (MPa)	EIT (GPa)	CIT (%)	Wt (μ J)	nIT (%)	hmax (μ m)	C (GPa)	τ (s)
MD-257kGy-I1	440.8 (19742)	476.8 (27060)	21.2 (12.9)	5.24 (1.29)	9.4 (2.9)	18.0 (11.9)	14.50 (3.92)	199 (4667)	0.400 (0.005)
MD-257kGy-I2	434.1 (17383)	468.9 (23191)	20.6 (16.4)	5.33 (1.20)	9.4 (3.0)	17.9 (8.3)	14.60 (3.82)	192 (4943)	0.425 (0.007)
MD-257kGy-I3	431.9 (15769)	464.6 (21090)	20.4 (20.5)	5.63 (1.15)	9.3 (2.2)	18.6 (12.4)	14.64 (3.50)	177 (4922)	0.478 (0.007)
MD-257kGy-S1	480.2 (22752)	509.0 (32818)	22.4 (15.9)	6.39 (0.97)	9.1 (2.3)	18.1 (10.6)	14.01 (3.09)	166 (5071)	0.558 (0.013)
MD-257kGy-S2	412.5 (28845)	443.9 (42071)	20.3 (18.6)	5.44 (1.62)	9.6 (2.8)	17.6 (9.7)	15.17 (4.73)	180 (8569)	0.435 (0.005)
MD-257kGy-S3	402.8 (16326)	425.0 (20613)	20.0 (18.6)	6.00 (0.96)	9.7 (3.2)	17.3 (8.1)	15.29 (4.33)	151 (3661)	0.495 (0.007)
MD-257kGy-I	435.7 (17592)	470.2 (23734)	20.8 (16.5)	5.40 (1.24)	9.4 (2.7)	18.2 (10.9)	14.58 (3.74)	190 (4898)	0.434 (0.007)
MD-257kGy-S	432.2 (23588)	459.5 (32752)	20.9 (18.8)	5.95 (1.32)	9.5 (2.8)	17.7 (9.6)	14.82 (4.36)	165 (5814)	0.497 (0.011)

443

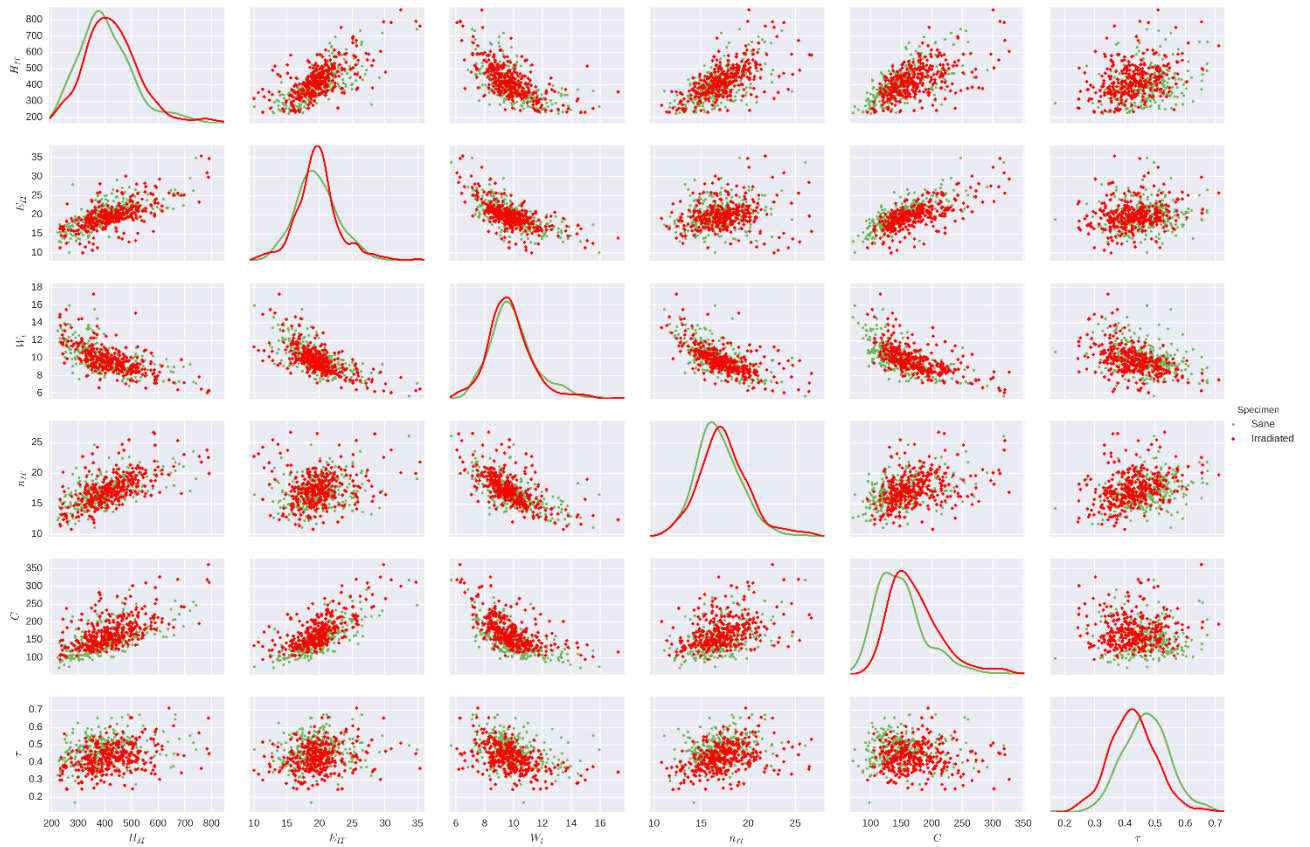
444 **Table 4.** Indentation outputs for cement paste indents identified using the proposed
 445 method coupling numerical clustering and image analysis (means and variances in
 446 brackets).

	HM (MPa)	HIT (MPa)	EIT (GPa)	CIT (%)	Wt (μ J)	nIT (%)	hmax (μ m)	C (GPa)	τ (s)
MD-257kGy-I1	376.0 (5267)	400.6 (5997)	19.4 (4.8)	5.25 (1.21)	10.2 (2.1)	16.6 (4.6)	15.44 (2.26)	168 (919)	0.391 (0.004)
MD-257kGy-I2	416.9 (11407)	448.1 (14084)	20.2 (12.8)	5.33 (1.22)	9.6 (2.8)	17.6 (6.6)	14.79 (3.20)	183 (2896)	0.429 (0.007)
MD-257kGy-I3	397.8 (8279)	428.1 (11073)	19.7 (19.3)	5.35 (0.91)	9.6 (2.1)	18.1 (12.2)	15.09 (2.79)	174 (4012)	0.454 (0.004)
MD-257kGy-S1	411.2 (8037)	428.4 (9145)	20.4 (7.9)	6.33 (0.77)	10.2 (2.4)	16.4 (4.8)	15.00 (2.59)	144 (773)	0.476 (0.004)
MD-257kGy-S2	372.3 (9381)	395.0 (11590)	19.6 (13.0)	5.40 (1.63)	9.9 (2.5)	16.9 (5.5)	15.63 (2.92)	162 (2310)	0.439 (0.005)
MD-257kGy-S3	387.5 (10432)	406.7 (12111)	19.5 (12.7)	6.03 (0.88)	9.9 (2.7)	17.0 (5.8)	15.48 (3.54)	142 (1247)	0.493 (0.006)
MD-257kGy-I	399.7 (8965)	428.8 (11226)	19.8 (12.7)	5.32 (1.12)	9.7 (2.5)	17.5 (8.1)	15.06 (2.87)	176 (2723)	0.427 (0.006)
MD-257kGy-S	386.0 (9739)	406.1 (11455)	19.7 (12.0)	5.85 (1.26)	9.9 (2.6)	16.9 (5.5)	15.45 (3.17)	150 (1639)	0.470 (0.006)

447

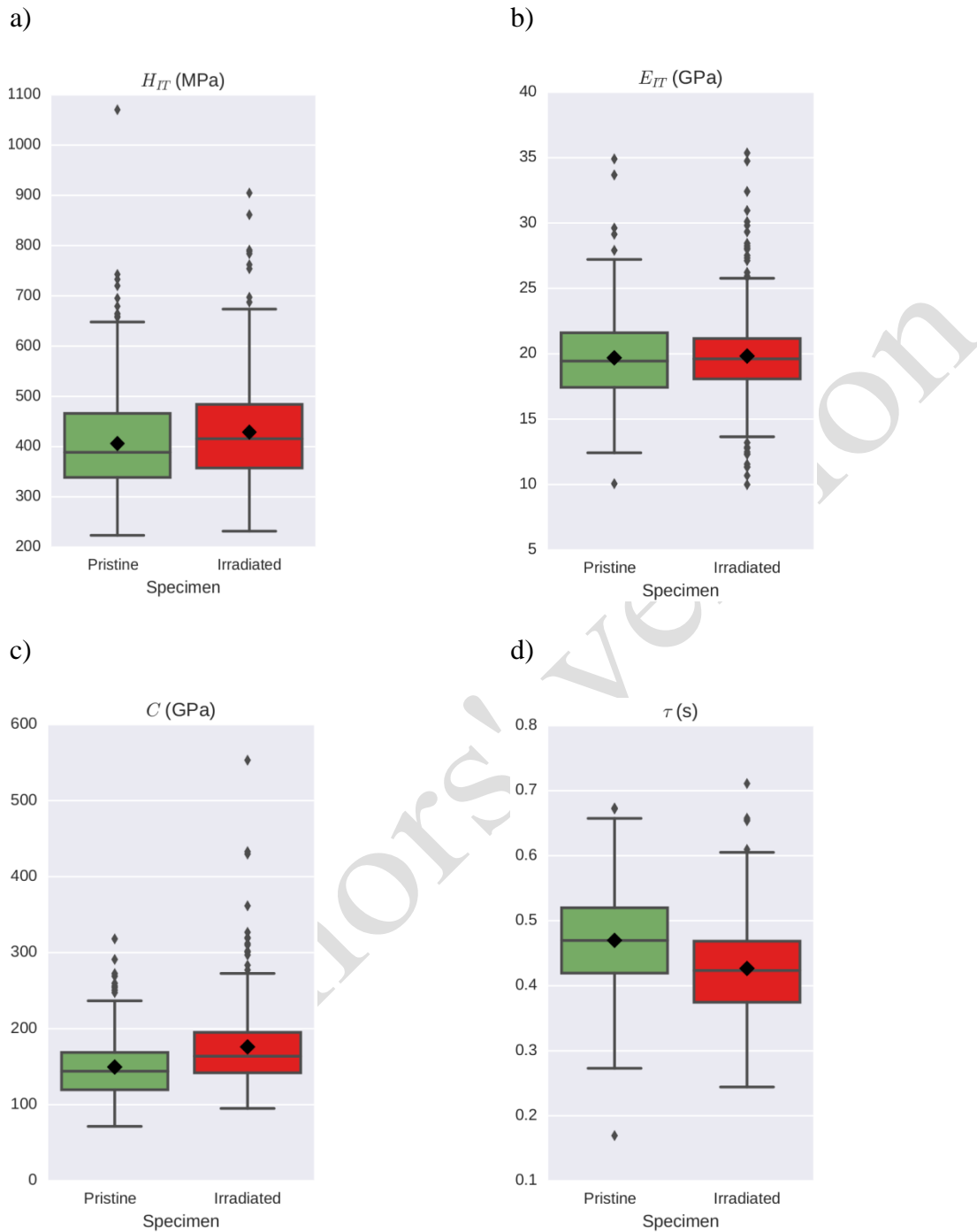
448 Statistical analysis was performed using Gnumeric software. Because normality of these
 449 new distributions could not be guaranteed using normality algorithms and variances
 450 could differ slightly, irradiated and control specimens data points were compared using
 451 the Welch's t-test (mean comparison assuming normal distributed variables but
 452 sufficiently robust to deal with non-normal samples if there are enough data points) and
 453 Wilcoxon-Mann-Whitney test (median comparison without normality assumption) as a
 454 verification both at a confidence level of 5%. There is no significant difference
 455 concerning Young's moduli in agreement with the tables. After a total irradiated dose of
 456 257 kGy, there is a significant increase of the creep modulus C of about 17% and a
 457 significant decrease of indentation characteristic time τ of about 9% comparing data
 458 points obtained from the method herein presented. Box plots summarizing the results
 459 are presented in Fig. 12. These proportions are similar to the ones computed from

460 hierarchical or 3D-microscope analysis outputs but may not be statistically different in
 461 the latter cases, especially when data points are identified using 3D-microscope only.
 462 Creep modulus increase is correlated with a slight indentation hardness increase of
 463 about 6% which is significant for data points obtained using the coupled method.
 464 Consequently, h_{\max} slightly decreases while n_{IT} slightly increases with irradiations. The
 465 indentation energy W_t remains unchanged proving that only slight changes are
 466 occurring.
 467



468 **Fig. 11.** Scatter plot of indentation data points attributed to the cement paste by
 469 coupling data deconvolution and image analysis for irradiated and control specimens
 470 (H_{IT} is expressed in MPa, E_{IT} and C in GPa, n_{IT} in %, W_t in μ J and τ in s).
 471

472



474 **Fig. 12.** Box plots of indentation data points attributed to the cement paste by coupling
 475 data deconvolution and image analysis for irradiated and pristine specimens: a) H_{IT} , b)
 476 E_{IT} , c) C and d) τ , Center horizontal line of the box represents the median value while
 477 thick diamond stands for the mean value.

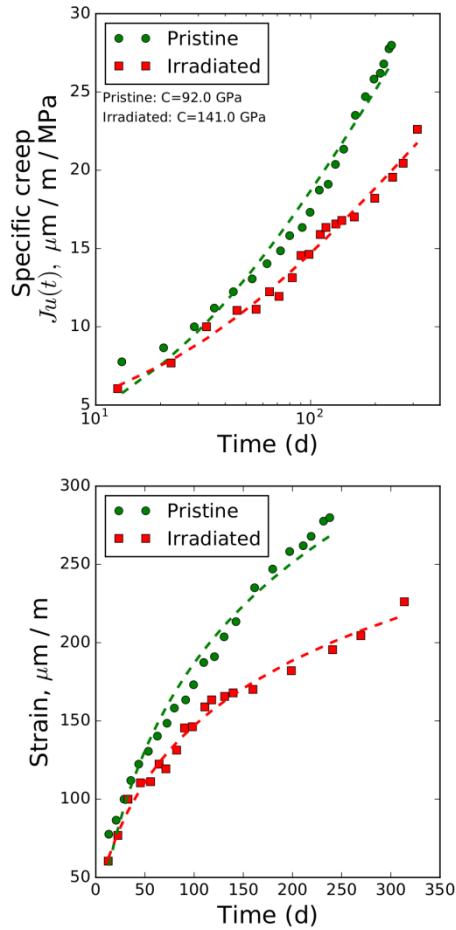
479 From our knowledge, these creep results are the first microindentation results
 480 concerning irradiated concrete specimens. Therefore, it could be interesting to make a
 481 comparison with the only macroscopic creep results under γ -irradiation reported a long
 482 time ago [39] and illustrated as markers in Fig. 13. The experimental macroscopic
 483 specific creep function was first fitted using a classic 3-parameters logarithmic law:

$$J_u(t) - \frac{1}{E_0} = \frac{\ln(t/\tau + 1)}{C} \quad (11)$$

484 This first fit led to an increase of the creep modulus of 74% (132 GPa for irradiated
 485 sample and 76 GPa for control sample) and a decrease of τ of around 49 % (47 d
 486 vs 24d). Though the raw increases are dramatic and somehow affected by the lack of
 487 experimental values compared to the number of parameters, this trend qualitatively
 488 agrees with the microindentation results. In order to obtain more realistic creep modulus
 489 variations, it has been decided to fit the experimental data using a two parameters law
 490 written as:

$$J_u(t) - \frac{1}{E_0} = \frac{\ln(t/cste + 1)}{C} \quad (11)$$

491 Using a characteristic time *cste* of 25 d (same order of magnitude as the characteristic
 492 time using a 3-parameters fit. but still a relatively small value regarding the dose
 493 (around 70 kGy) at this time), this type of law correctly describe the creep behavior for
 494 times greater than τ . As illustrated in Fig. 13, the fit is acceptable for times greater than
 495 approximately 40 d. When applied on the regression curves proposed by McDowall,
 496 this fit leads to a more reasonable creep modulus increase of 53% after a total dose
 497 820kGy in approximately 300 d (141 GPa for the irradiated specimen and 92 GPa for
 498 the control specimen). This increase is around three times greater than the increase
 499 measured by indentation after a total dose of 257 kGy.



500

501 **Fig. 13.** Comparison between experimental creep results from [39] (markers) and a 2-
 502 parameters logarithmic fit (dashed line).

503

504 Therefore, in order to compare creep modulus' increase at similar total doses, we
 505 performed 2-parameters fitting up to increasing doses with the assumption that
 506 irradiation gradually affects cementitious materials. For this purpose, we approximated
 507 the initial fit of the evolution of strain proposed by McDowall in order to feed the
 508 algorithm with continuous data. Then logarithmic 2-parameters fitting was realized
 509 between dose segments of growing sizes beginning by a first segment of [0 kGy,
 510 20 kGy] and gradually increasing the size of the segment by 0.5 kGy. The general trend
 511 of the optimization problem for the ratio of the creep modulus evolution assessment,
 512 depending on time t and dose d , is summarized by eq. 12 where J' and J are the specific

513 creep function of the irradiated and control specimens. The resulting evolution of the
 514 ratio between the creep moduli of irradiated concrete specimen and control specimen
 515 over the total dose is illustrated in Fig. 14.

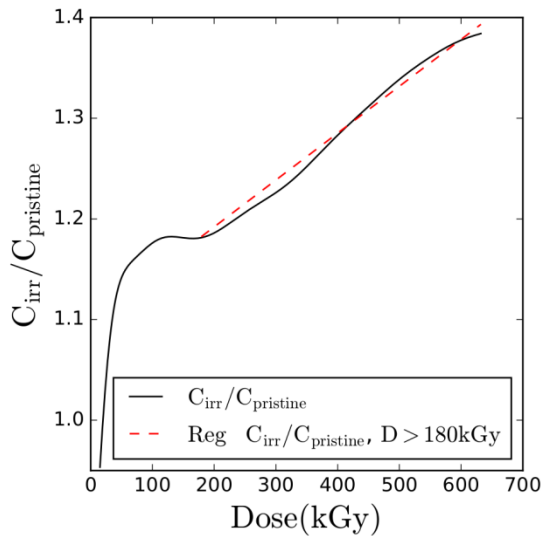
516

$$\frac{C_{Irr}(d)}{C_{sane}} = \frac{\min_{c,\tau=25d} \int_0^d J'_{exp}(t, d) - J'_{num}(t, d)}{\min_{c,\tau=25d} \int_0^d J'_{exp}(t) - J'_{num}(t)} \quad (12)$$

517

518 Because the effect of radiations is supposedly small at low doses, creep moduli ratio is
 519 close to 1 for small segments and. because the effect of radiation increases with time the
 520 ratio gradually increases. It is difficult to say whether the quick increase of the ratio
 521 before 50-60 kGy is representative of the concrete behavior or if the fit reliability is
 522 affected by the logarithmic function selected (irradiation times are close to the creep
 523 characteristic time at low dosage). However, one may observe the ratio exhibits a quasi-
 524 linear increase after 180 kGy (doses at which time $\geq 3 \times$ creep characteristic time). A
 525 linear regression with a correlation coefficient of 0.994 is proposed after 180 kGy. From
 526 this regression, it can be stated that the increase rate is of $5 \times 10^{-2} \% / \text{kGy}$, which leads,
 527 considering an initial creep modulus of around 100 GPa, to a rate of 50 GPa/MGy. For
 528 a dose of 257 kGy, the increase of the creep modulus from this method is around 20%
 529 which is in good agreement with the increase calculated from the microindentation
 530 experiment.

531



532

533 **Fig. 14.** Evolution of the ration between creep moduli calculated for irradiated and
 534 control concrete specimens from [39] relatively to the final dose considered for a 2-
 535 parameters fitting logarithmic law.

536

537 **4. Conclusion and perspectives**

538 In this study, the potential of coupling indentation and optical microscopy for
 539 heterogenous materials characterization has been demonstrated. A novel method has
 540 been introduced to reduce the variability of the micromechanical properties associated
 541 to an indented phase. This method relies on the comparison of the nature of indents
 542 numerically predicted by statistical indentation data deconvolution and by 3D-optical
 543 microscope image analysis. This method robustly counteracts the uncertainty of
 544 numerical clustering methods by introducing optical information. In our study, the data
 545 points cluster of interest generated by the method associated with cement paste likely
 546 exhibits a normal distribution and a systematic reduced variance compared to each of
 547 the technique alone. This herein described method could be applied to a wide range of
 548 materials and studies.

549 The proposed method was successfully applied to the assessment of micro-mechanical
 550 properties of γ -irradiated mortar specimens. The method led to the identification of the

551 cement paste elasto-plastic parameters affected by irradiation at relatively low
552 cumulated dose of 257 kGy. It has been found out that creep modulus C significantly
553 increases by around 17% and indentation characteristic time γ significantly decreases by
554 around 9% after irradiation. The creep modulus increase is correlated with a slight
555 increase of the indentation hardness H_{IT} and of the proportion of the elastic response
556 relatively to the total energy n_{IT} . On the other hand, Young's modulus of irradiated
557 specimens does not significantly differ from the one of control specimens.
558 These micro-mechanical observations agree with the only macroscopic uniaxial
559 compressive creep measurements under γ -irradiation published to our knowledge [39].
560 Using a logarithmic dose-dependant fit of the specific creep function proposed in this
561 study, it can be calculated that the creep modulus increases and the creep characteristic
562 time decreases leading to less creep of the irradiated samples. Then a reverse analysis
563 was proposed to quantify the evolution of the irradiated vs pristine creep moduli ratio
564 with the time increasing γ -dose. Although supplementary similar experiments should be
565 carried out at both microscale and macroscale with various dose rates, the creep
566 modulus appears to linearly increase with the γ -dose for doses between 180 and
567 700 kGy.

568

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575

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