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Extrusion-based additive manufacturing with cement-based materials – Pro duction steps, processes, and their underlying physics: A review

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- 15

16 Abstract

- 17 This article offers a comprehensive overview of the underlying physics relevant to an under-
- 18 standing of materials processing during the various production steps in extrusion-based 3D
- 19 Concrete Printing (3DCP). Understanding the physics governing the processes is an important
- 20 step toward the purposeful design and optimization of 3DCP systems as well as their efficient
- and robust process control. For some processes, analytical formulas based on the relevant phys-
- ics have already enabled reasonable predictions with respect to material flow behavior and
 buildability, especially in the case of relatively simple geometries.
- The existing research in the field was systematically compiled by the authors in the framework
- of the activities of the RILEM Technical Committee 276 "Digital fabrication with cementbased materials". However, further research is needed to develop reliable tools for the quanti-
- tative analysis of the entire process chain. To achieve this, experimental efforts for the charac-
- 28 terization of material properties need to go hand in hand with comprehensive numerical simu-
- 29 lation.
- 30

Keywords: Concrete technology; additive manufacturing; 3D-printing; extrusion; production;
 underlying physics

33 1. Introduction

- 34 Digitalization and automation in construction bring great potential in respect of increases in
- 35 productivity, in creating more attractive jobs, and in compensating for shortages of skilled labor
- 36 [1, 2]. Large-scale Additive Manufacturing (AM) with cement-based materials, commonly re-37 ferred to as 3D Concrete Printing (3DCP), belongs to the most promising new concrete tech-
- nologies for implementing digital data from the planning phase and ultimately to actual auto-
- mated production in factories and on construction sites [3, 4]. Not only can 3DCP facilitate and
- 40 quicken production processes considerably, it can also make it technically and economically
- 41 feasible to realize topologically optimized, geometrically complex structural elements designed
- 42 according to the principle *form follows force* and also enable the integration of various func-
- 43 tionalities. Such topological optimization allows for elegant, material-minimized, and resource-
- 44 saving structures [2, 5].
- 45 Over the last decade various 3DCP approaches have been developed, and a number of real-
- scale pilot projects have been successfully completed; see e.g. [6, 7]. According to the RILEM
- 47 process classification framework for Digital Fabrication with Concrete (DFC) technologies [8], 48 the existing enpresence of AM with semant based materials can be subdivided into different

49 groups, of which material extrusion, particle-bed binding, and material jetting are three that 50 have been demonstrated at scale. While these groups and individual approaches within the 51 groups differ considerably with respect to their respective material concepts, equipment, and 52 production steps, all of them base themselves on sound interaction between material and ma-53 chine along the processing chain. Hence, mastering material-machine interactions is a prereq-54 uisite for efficient and robust processes, and for their optimization and control. Such mastering 55 is only possible if the underlying physics of individual 3DCP processes are well understood and 56 memory follower foll

56 purposefully applied.

57 In this light and in understanding the importance of the topic with respect to successful imple-58 mentation of DFC technologies into the practice of construction, the authors dedicated consid-59 erable time and effort to the systematic analysis of the physical background of various DFC 60 technologies and their individual processing steps. Initiated as an activity of the RILEM Technical Committee 276 "Digital fabrication with cement-based materials", an important outcome 61 62 of this work is the article at hand, which focuses on the first group of AM approaches – namely, DFC technologies whose basis is material extrusion. The choice of this DFC group was straight-63 64 forward since extrusion-based 3DCP approaches dominate the field both in research and first 65 applications worldwide. Thus, at present material extrusion seems a clear favorite with respect to both the level of overall technological readiness and economic viability [4, 9]. 66

67 The goal of this contribution is to bring together in orderly fashion the relevant knowledge in physics needed to understand and to shape purposefully the relevant processes belonging to 68 69 extrusion-based 3DCP. In doing so, first, the existing approaches of this DFC group are subdi-70 vided into three categories according to the respective major concepts of material handling. Second, the main production steps common to all extrusion-based approaches are defined. Next, 71 72 the processes and underlying physics relevant to individual production steps are specified for 73 each approach category. This is all laid out in Section 2 while the subsequent sections focus on 74 the main processes, i.e., their physics, their corresponding key physical properties, and their 75 evolution over time. Given the wide spectrum of the relevant processes and their complexity, the authors have aimed at providing a comprehensive overview of crucial aspects as a basis for 76 77 a general understanding of the technology in the first place rather than trying to deliver the 78 details on every process and its underlying physics. However, numerous references to important 79 sources in the literature should help in finding additional information easily. To make the pro-80 cess and physical parameters "tangible" while providing some rough guidance with respect to 81 relevant quantitative information, the ranges of absolute values are given as first estimates for 82 the reader's consideration whenever it is possible and meaningful. Finally, attention is paid to 83 research needs, of which a great many remain.

84 2. Processing steps and their underlying physics

85 The additive manufacturing approaches of material deposition by extrusion can be subdivided 86 into three categories: i) extrusion of stiff material, similar to conventional extrusion, ii) extru-87 sion of flowable material with or without adding admixture(s) in the printhead, and iii) extrusion of material using additional energy input, e.g. vibration, which facilitates the delivery and dep-88 89 osition of stiff mixtures. The ideal case of the first category is the "infinite brick" extrusion 90 regime, where the filament and nozzle cross-sections are equal; see Figure 1a. In the second 91 category the ideal case is free flow deposition, where the material flows freely until the stress 92 induced by gravity equals the yield stress of the printable material; see [10] and Figure 1c. In 93 the context of additive manufacturing with concrete, most extrusion flows are located some-94 where between these two asymptotic deposition approaches; see Figure 1b. Note that while two 95 asymptotic cases can be mathematically modeled as described in the next sections, the prediction of more realistic final shapes for real printable, cement-based materials is not straightfor-96

97 ward due to several mechanisms and boundaries involved. These mechanisms/boundaries in-

98 clude solicitation gradient, shear-induced particle migration, non-uniform structural build-up,

99 complex nozzle geometry, and so on. In such cases adequate multi-physical, numerical tools

100 would be required to describe the extrusion/deposition process fully. Analytical formulas can,

- 101 however, still provide reasonable estimations. Finally, the third manufacturing approach, the
- 102 extrusion of material using additional energy input, has been as yet rarely applied.



103 104

Figure 1. Two asymptotic regimes of material deposition by extrusion: a) extrusion of a very stiff material (so-called infinite brick strategy) and c) extrusion of very flowable material that spreads out after deposition (adapted from [10]); regime b) shows a more realistic case of extrusion of sufficiently stiff material by means of a nozzle with a geometric reduction.

109

In general the major processing steps in these manufacturing approaches are similar since they all include: 1) transportation of build material to the printhead, 2) printhead process/extrusion by the printhead, 3) deposition of build material, accompanied by its deformation, and 4) depositions of further layers, accompanied by loading earlier deposited upper layer(s) by selfweight and process-induced forces, related deformation of build material after deposition followed by further deformation due to early-age shrinkage, early-age creep, and thermal dilation.

116 Note that a technical discussion of the initial processing step, namely "concrete mixing" is be-117 yond the scope of this publication. Thus, the first step in each presented approach is seen as "transportation". However, mixing in the printhead to disperse chemical admixtures such as 118 119 hydration-accelerating agents is addressed here as a part of the printhead process. Table 1 lists 120 three manufacturing approaches of selective material deposition by extrusion and four pro-121 cessing steps. The underlying physical processes for each processing step for a given fabrication 122 approach are listed in Table 1 as well. These underlying physical processes, such as gravita-123 tional flow, pumping, i.e., pressure-induced flow in a pipe, or extrusion or pressure-induced 124 flow through a section contraction, are presented and discussed in the following sections. How-125 ever, early-age shrinkage, early-age creep and thermal dilation of the material after deposition 126 are not covered in this article since they 1) do not represent a production step as such, b) are 127 not considered to be different to conventional concrete construction from the physical point of 128 view.

- 129
- 130

131 Table 1. Production steps, related processes and underlying physics relevant to three different

- approaches within the extrusion-based additive manufacturing with cement-based materials.
- 133

	Extrusion-based manufacturing approach					
Production step	Extrusion of stiff mate- rial	Extrusion of flowable mate- rial with or without disper- sion of admixture(s) in the printhead	Extrusion of material using additional energy input			
Transportation of build material (also primary motivation)	Pumping (over short distances) or gravita- tional flow with or without energy input	Pumping	Pumping or gravitational flow with or without energy input			
Printhead process	Extrusion using pri- mary motivation (pumping), ram extru- sion, or screw extrusion	Extrusion using primary motivation, ram extrusion, or screw extrusion; optional dispersion of admixture by high-energy mixing	Screw extrusion or gravita- tional flow supported by vi- bration			
Deformation of build ma- terial during deposition	Gravitational flow, visc	Gravitational flow and com- paction supported by vibra- tion, elastic deformations				
Behavior of build material after deposition	Deformations due to self-weight and kinetic energy of deposition; additionally: early-age shrinkage, early-age creep, thermal dilation					

134

For the sake of clarity one of the approaches is depicted in Figure 2, which illustrates the processing steps belonging to the extrusion of flowable materials without addition of admixture in the printhead.

138

Generally, the behavior of fresh cementitious materials can be considered as visco-plastic. This means that, similar to solids, they do not flow until a given critical shear stress, i.e. yield stress τ_c , is exceeded and, similar to a liquid, they flow when subjected to shear stresses greater than τ_c [11, 12]. Depending on the processing step during additive fabrication, either solid or fluid behavior is involved, e.g. during the conveying and feeding processes the material flows inside the transportation system and printhead, i.e. fluid behavior, while after deposition the material

145 has to remain static (i.e. solid behavior, viscoelastic-plastic) [10].

146 Note that cement-based materials are heterogeneous, and this can have a pronounced effect on

147 their flow behavior in specific cases. For example, in the case of pumping, shear-induced par-

ticle migration creates a lubrication layer that greatly influences the flow of the material [13].

149 As a result, neither the velocity profile nor the flow rate can be accurately predicted based on

150 viscosity and yield stress of the bulk material. The formation of a lubrication layer may occur

also during the extrusion process; see Figure 1a.

152



- 1. Transportation of build material pumping
- 2. Printhead process extrusion using primary motivation
- 3. Deformation of the build material during deposition gravitational flow, viscoelastic-plastic deformations
- 4. Behavior of build material after deposition deformations due to self-weight and kinetic energy from deposition; early-age shrinkage, early-age creep, thermal dilation
- 154 Figure 2. Processing steps in the extrusion of flowable materials.
- 155

153

156 **3.** Gravitational flow

157 During the 3D concrete printing process gravity plays a role in the transport of the material,

158 particularly between mixing and placement in a pumping or extrusion system. The gravity-

159 induced flow of material from a hopper to the pump is one of the printing steps that can be

160 studied by analogy to the discharge flows of the March cone, for example.

161 After deposition, there is "competition" between, the force of gravity and the material strength,

- 162 in rheological terms yield stress. This competition can be described by a dimensionless number,
- 163 i.e. the ratio of the gravity-induced stress ρgh over the material yield stress τ_c , thus $\rho gh / \tau_c$,

- 164 where ρ is the material density, g is the acceleration of gravity, and h is the height of the depos-
- 165 ited layer. Note that the balance between gravitationally induced stresses and the yield stress
- 166 can be studied using the slump/spread flow theory developed by Roussel and Coussot [14]; see
- 167 also Perrot *et al.* [15].
- 168 In the literature related to extrusion-based 3D printing process using cementitious materials, a
- 169 wide range of material yield stress has been reported. Depending on the dimensionless number
- 170 $\rho gh / \tau_c$ and the geometry of the nozzle's cross section, the final section of the material layer 171 can vary from rectangular to semi-ellipsoidal, cf. [10]. Figure 1a shows a high yield stress of
- 171 can vary from rectangular to semi-ellipsoidal, cf. [10]. Figure 1a shows a high yield stress of 172 over 500 Pa for a cement-based material that keeps its shape after being deposited pursuant to
- the infinite brick strategy, while Figure 1c shows a filament of fluid cement-based material that
- spreads out after deposition with a yield stress of less than 100 Pa. It is important to note here
- that for the case of printable cementitious materials with yield stress in the order of magnitude
- between hundred and thousands of Pa at the nozzle exit, the effect of the surface tension on the
- 177 final shape of the layer can be neglected [10].
- 178 In the case where the yield stress of the material is greater than the stress induced by gravity,
- the layer of cementitious material will not be deformed after deposition. This scenario applies
- 180 if the material is placed gently and relatively slowly; otherwise additional effects need to be
- 181 considered. As proposed by Roussel and Coussot [14], this ideal case can be described using a 182 purely elongational plastic model wherein the critical height of a stable layer reads $h_{stable} =$
- 182 purely congational plastic model wherein the entreal height of a stable layer reads $n_{stable} = \sqrt{3\tau_c/\rho g}$ where $\sqrt{3\tau_c}$ is the elongational yield stress of the cementitious materials conditioned
- on their following von Mises plasticity criterion [10, 14, 15, 16]. In other words, to enable
- 185 cementitious materials to keep their shape after deposition, the target yield stress value should
- be at least around 13 Pa per millimeter of layer thickness (for ρ of 2200 kg/m³). Note that the
- plasticity criterion and its time dependency are still an open research question, especially for
- 188 stiff mixtures, which can exhibit pressure-dependent behavior [17, 18].
- 189 If gravitational force overcomes the material elongational strength at the layer bottom, the ma-190 terial will deform until an equilibrium between yield stress and gravity-induced stress is 191 reached. As a result, the layer being deposited will deform and spread on the previously printed 192 layer. The limiting case of this type of behavior for a low yield-stress value tends to the case of 193 shear flow on a horizontal plate. In this shear flow regime the final height of the layer should 194 be equal to h in ρgh [14]. In many 3D printing applications the actual filament deformation 195 behavior ranges somewhere between the pure shear flow regime and the infinite brick, no-flow 196 regime; thus, the final shape of the layer will depend on its material yield stress, on the cross-197 section of the nozzle opening, and on the gap between nozzle and printed layers, and this should 198 be studied on a case-by-case basis. In this context, numerical analysis using e.g. Computation 199 Fluid Dynamics (CFD) approaches can be instrumental in understanding material behavior bet-
- 200 ter under specific boundary conditions and, prospectively, in predicting this behavior.
- The effect of the gap between nozzle and deposition area on the shape of the deposited layer can also be important because tensile forces, i.e. in under-extrusion, where the material flow rate is lower than $S_{nozzle} \cdot V$, where S_{nozzle} is the nozzle section and V the nozzle velocity, or compression forces in over-extrusion, where flow rate is higher than $S_{nozzle} \cdot V$, can be induced by the material leaving the nozzle; see Figure 3. These effects could lead to surface cracking, layer bending, or even coiling. These effects should be studied in some depth in order to allow for full control of layer geometry during printing.
- 208 Some extrusion-based, selective material deposition methods [19] purposefully establish a di-
- 209 rect interaction between the nozzle and the extruded material: The nozzle presses the exiting
- 210 material, forcing the layer to have a thickness equal to the gap between the nozzle and the layer

211 below; it is assumed that the layer below is stiff enough to sustain this additional pressure with-212 out considerable deformation. This strategy has the advantage of eliminating uncertainty in the 213 final height of the printed structures since the final printed structure height will only depend on the nozzle toolpath. While improving the overall geometry control of the printing process, the 214 215 direct interaction of the nozzle and the deposited material induces additional pressure on the 216 underlying layers at the level of the nozzle and can contribute to cracking or even to collapse of the printed element. Note that to date the tensile behavior of fresh, cement-based materials 217 has not been extensively studied. One can mention the tensile strength measurement carried out 218 219 by Mettler et al. [18] and Lo Monte et al. [20]. Nonetheless, a consistent explanation of the 220 cracks' formation and the definition of mix-design solutions to prevent it, e.g. by adding fiber 221 or polymer, are still open questions requiring extensive research.

222



a)

223 Figure 3. Challenges in extruding concrete or mortar: a) extrusion rate is too low compared to the nozzle velocity leading to discontinuous layers [21], b) extrusion rate is too fast compared 224 225 to the nozzle velocity leading to "buckled" layers [21], c) "tearing" of extruded layers due to 226 the overly high yield stress of the printed material; adapted from [22].

227 4. **Pumping and extrusion**

228 4.1 Pumping

229 Pumping is often used in transporting mixed concrete to the printhead or nozzle. It is performed 230 by pushing material through a hose with the help of positive-displacement piston pumps or progressive cavity pumps, the latter is so far the most widely implemented means in extrusion-231 232 based additive fabrication. While the pumping process as such is similar to ram extrusion, there 233 are some distinctions worth addressing: it usually covers considerably longer distances and re-234 sults in higher material discharge rates. Furthermore, ram extrusion usually implies compaction 235 and shaping of the material by the nozzle narrowing towards its opening. Concrete pumpability 236 as characterized by pressure losses depends primarily on the plastic viscosity. However, the 237 influence of yield stress increases with increasing τ_0/μ [23]. Calculations using the Bucking-238 ham-Reiner equation for laminar flow of a Bingham fluid through a pipe overestimate the 239 pumping pressure for a given flow rate by up to 5 times [13, 24, 25]. This is a consequence of 240 the formation of a lubricating layer (LL) at the inner walls of the pipeline due to shear-induced 241 particle migration (SIPM). The yield stress and plastic viscosity of the LL are generally about one fifth and one fifteenth of the corresponding values of the bulk material, respectively [24]. 242 The shear stress, shear rate, and velocity profiles of concrete during pumping are presented in 243 244 Figure 4.



$\tau_{0,LL}, \mu_{p,LL}, e_{LL}$

Figure 4. Pressure P, shear stress τ , shear rate $\dot{\gamma}$, velocity v and flowrate Q profiles of a concrete flow inside a pumping pipeline. The yield stress and plastic viscosity of concrete are $\tau_{0,B}$ and $\mu_{p,B}$, whereas the yield stress, plastic viscosity and thickness of the lubricating layer are $\tau_{0,LL}$, $\mu_{p,LL}$ and e_{LL} , adapted from [26].

251

252 Shear rate and shear stress are zero at the central axis of the pipe and are maximal at the pipe 253 wall. If the shear stresses at a given radial position are higher than the concrete bulk yield stress, 254 then shearing of the bulk takes place. As a consequence of and in dependence on the properties 255 of concrete and pumping conditions, concrete can flow in the slip flow mode, i.e. very thin slip 256 layer + unsheared plug, or in the slip-plus-shear flow mode, slip + partial shear of bulk + plug 257 [13, 27]. Kaplan et al. [27] introduced analytical models for predicting concrete pumping pres-258 sure in the cases of slip and slip-plus-shear flow modes using rheological parameters for bulk $\tau_{0,B}$, $\mu_{p,B}$ and for interface $\tau_{0,LL}$, $\mu_{p,LL}$ as well as geometrical data of the pipeline. In the case of 259 260 infinite brick extrusion with stiff material, the dynamic yield stresses and plastic viscosities are 261 a few hundred Pa, and few Pa·s, respectively.

262 Whereas in the case of free flow deposition, these values are few tens of Pa and few tens of 263 Pa·s. Although the free flow material has lower yield stress, this does not necessarily mean 264 lower pumping pressure losses in comparison to infinite brick extrusion, in which the high yield stress bulk does not deform during pumping, as proven in earlier research on pumping [23, 24]. 265 266 Assuming a flow rate of 5 m³/h in a DN125 pipeline, the pressure loss per unit length can be in 267 the range of 3.5 kPa/m for the infinite brick case and 5 kPa/m in the case of free flow deposition. This translates into pumping pressures of 70 kPa and 100 kPa for a DN125 pipeline of length 268 269 20 m, respectively. Similarly, higher pressure losses are expected in the case of free flow dep-270 osition printable concrete than infinite brick printable concrete when pumped in pipelines of 271 smaller diameter e.g. DN50.

272 The optimal rheological requirements of the material with respect to pumping are in competi-273 tion with those favorable in respect of deformation behavior after layer deposition. In general, 274 printable concretes are supposed to have high static yield stress to overcome gravitational and 275 other forces acting after material deposition. The influence of high structural buildup and thix-276 otropy rates of 3D-printable concretes, especially those used in infinite brick extrusion on 277 pumping behavior have yet to be investigated. De Schutter and Feys [13] emphasized that "short 278 interruptions during pumping lead to major difficulties in resuming pumping operations due to 279 the sometimes tremendous effect of internal structural buildup". With large-scale pumping 280 tests, they have observed that a 20-minute delay leads to an increase of pressure loss by 50% 281 for a mixture with a structural buildup rate A_{thix} of 0.3 Pa/s. Despite increasing the pumping 282 pressures to 35000 kPa, the pumping operations could not be resumed after the delay. There-283 fore, pumping very high-yield-stress concretes or, pumping chemically accelerated concrete 284 may not be an optimal solution for large-scale, extrusion-based additive fabrication.

An alternative solution is to pump low viscous and low yield stress concretes over the long distance and then activate them at the printhead, shortly before deposition. Such an approach requires the use of inline, second stage mixing techniques to ensure precise dosage and uniform homogenization of the admixture; see Section 5. Note that even in the case of free flow deposition, it is necessary to ensure that the rheological properties are sufficiently high to prevent

290 filtration under high-pressure and segregation.

The methodology in characterizing the pumping-relevant rheological properties of concrete and lubricating layer, and the prediction of pumping pressures is a considerable challenge. The in-

adequacy of empirical design charts, which consisted mostly of a slump or spread parameter

[13], has been overcome with recent, advanced test approaches and prediction models [28, 29,

- 295 30, 31, 32]. However, most of these approaches require the accurate quantification of the con-
- crete's and the LL's rheological properties, which has proven challenging for high yield-stress,
- 297 printable concretes [33].
- Numerical models can help in understanding the SIPM during pumping and characteristics of
- LL, as well as predicting pumping pressures. However, such tools have so far been simplified
- 300 by the assumption of either a single-fluid homogeneous medium [23, 24, 25, 28, 34] or discrete 301 granular elements [35]. In the case of concretes with high granular fractions, the interactions
- between aggregate and paste are crucial. Fluid-solid numerical models coupled with the mod-
- 303 elling of time- and shear-dependent variations of pumping characteristics are yet to be devel-
- 304 oped.

The pumping process can alter the rheology of concrete, for example, due to dispersion of cement particles at higher shear rates, activation of residual superplasticizer, or air-induced by pistons. However, earlier observations of pumping-induced changes in concrete yield stress are inconclusive [28, 36]. This can cause critical consequences for the continuity of the extrusion process or the stability of the resulting extruded layers, respectively, as a consequence of the increase or decrease in yield stress, thus necessitating further research to develop inline rheology monitoring tools that can be embedded, for instance, in the printhead.

312

313 4.2 Extrusion

314 In the context of additive fabrication, the extrusion step can be considered as the action of con-315 ferring the shape of its section to the deposited layer of cementitious material. This step is performed by forcing the material through a section contraction. This complex flow system has 316 317 been studied extensively, providing insights into the rheology of visco-plastic fluids [36, 37, 318 38, 39, 40]. However, when dealing with cement-based materials, especially with a view to the 319 additional plastic materials used in the "infinite brick" extrusion approach [10], i.e. to material 320 with yield stresses higher than several kPa, extrusion-induced segregation has been reported in 321 the literature due to the multiphase nature of the cementitious materials under study [33, 41, 42, 322 43]. Such behavior can be described as competition between a) the cementitious materials' 323 characteristic drainage time that is the result of the pressure drop at the extruder die and b) the 324 extrusion time [33]. If the drainage time is shorter than the extrusion time, segregation is likely 325 to occur, leading to the material's stiffening. For stiff cement-based mixtures which are close 326 to the solid randomly packed fraction, drainage can also drastically change the material's rheological behavior from plastic to frictional [29, 33, 41, 43]. Such types of behavior are likely to 327 328 induce surface defects or, more damaging, a stoppage of flow. Hence, securing a sufficient 329 extrusion flow rate helps reduce the effect of drainage.

Another solution is to add external energy in such a way as to ease the flow, reducing wall friction and locally liquefying the cementitious materials. Such energy-based approaches to extrusion are a recent field of interest that has not been widely studied yet. Among these meth-

333 ods is local vibration of the cementitious material, which was studied to reduce the apparent

- 334 yield stress of the material and to allow for a reduction of the extrusion pressure due to contrac-
- 335 tion flow [30].
- 336 The effect of vibration on an extrudate surface produced with a screw extruder can be seen in
- Figure 5, which indicates cracks which appear on the extrudate surface when no vibration is applied. When adding adapted vibration, the extrudate surface becomes smooth, and for even
- higher values of vibration energy, the extrudates are no longer able to withstand gravitational
- 340 force.

341 The strategy of using vibration not only for facilitating material transport in the large printhead 342 but also for extrusion-based material deposition was used by the HuShang Tengda company [31]. Because of the vibration of the nozzle orifice, relatively stiff, "ordinary" concrete could 343 be deposited layer-wise and compacted in the process according to the personal observation of 344 345 this article's first author. Note that for the purposeful use of such an approach a precise deter-346 mination of the area of influence of vibration is required since the action of vibration on the 347 already deposited layer could compromise the structural buildup process and lead to collapse 348 of the printed layers. Some studies on the effects and influence zones of vibration have been 349 carried out, but they are limited to the axisymmetric case of a vibration needle immersed in 350 concrete [32, 44]. Further studies on the influence of vibration on cementitious materials' ex-

trusion flow are therefore necessary, specifically in the context of additive fabrication.

352 For printable cement-based materials, which are usually highly thixotropic, understanding the 353 effect of vibration of extruded layers is a complex task, the zone of influence might be smaller 354 than is usually predicted [44]. Alternatively to vibration, it is possible to use the electric poten-355 tial difference within the steel extruder to promote the formation of a lubricating layer at the 356 extruder's inner walls [45, 46]. Local application of a magnetic field is also envisaged to change 357 the rheology of cementitious materials containing magneto-sensitive particles that can thus be uniformly oriented [2, 47]. While such approaches seem promising, substantial additional ef-358 359 forts are required to assess the effect of those external energy applications on the extrusion flow 360 of cementitious materials in the context of additive fabrication. In the first place, the determi-361 nation of the precise region of influence of external solicitation is mandatory, since such influ-362 ence could compromise the stability of the already printed layers.

363



364 365

366 367 a) No vibration

b) Frequency 7Hz Amplitude 12 mm

c) Frequency 30 Hz Amplitude 12 mm

Figure 5. Effect of external vibration on the surface of extruded mortar (same screw rotationalvelocity of 5 rpm; mix design can be found in [30].

Furthermore, the design of the die is crucial because it governs the orientation and shape of the
layer of material being deposited [6, 48]. Likewise, the velocity profile is important to control
within the cross-section of the extruder in order to ensure proper placement of the layers and
good adherence between layers [6]. Additionally, the extrusion flowrate should be controlled

- 375 and estimated based on the rheological behavior of the cementitious materials, since the veloc-
- ity of the material leaving the extruder has to be adjusted to the robotic arm to control the section

of the deposited layer; see also Figure 3. Finally, some shapes of the nozzle make it necessaryto rotate the nozzle when printing curves.

- 379 Moreover, in the case of accelerated mixes, mostly using chemical admixtures, it is important
- to use die geometries which prevent the formation of a dead zone, since the material "captured"
- there can harden during the process and eventually lead to process stoppage due to blockage.
- 382 Consequently, mastering the flow behavior within the extruder leads to better control of the
- 383 material's residence time in the die and avoid the buildup of heterogeneities, thus facilitating
- an adequate flow rate and trouble-free material deposition.

While the velocity profiles within an extruder have been studied for the simplest case of axisymmetric ram extrusion [38, 36, 41, 49], no study has dealt with the determination of the strain and stress fields within the extruder for complex die or nozzle geometries. Hence, there is need to provide numerical tools which can predict accurately the flow of cementitious materials through nozzles of any given geometry.

A last open question deals with the study of the flow of cementitious materials in a hopper screw geometry or in a progressive cavity pump extruder [21]. Predicting the cementitious flow rate from its rheological and tribological behavior and from the screw's rotational velocity is a challenge. Adequate numerical and analytical tools appropriate to the rheological properties of a given cement-based material are much required to ensure the continuous and controlled flow rate of the materials during extrusion.

396

397 5. Dispersion in the printhead

398 Many printing techniques involve a so-called acceleration of the material during or after depo-399 sition. Most of these techniques rely on the incorporation at the printhead level of either a chem-400 ical accelerator able to modify the silicate/aluminate balance and accelerate one of these hydra-401 tion reactions, or an organic flocculant able to bridge the finest particles in the system. They 402 could also rely on the mixing of the material with an alternative binder such as aluminate-based 403 substances [50]. The former strategy (dispersion) involves amounts of products below a very 404 few percent while the latter (mixing) often involves amounts above 10% by volume. All the 405 strategies above lead to a faster and enhanced phase-change of the printed material, allowing 406 for higher building rates and higher productivity. Moreover, these strategies allow for the mix-407 ing, pumping, and feeding of the robot with an extremely fluid material, which then turns into 408 a pseudo-solid once the accelerator is added. However, these strategies require the dispersion 409 of an active agent in the printhead or the mixing of a slurry, both of which give rise to several 410 difficulties in terms of printhead design. The authors have focused here on dispersion.

- 411 From a process point of view, as soon as a dispersion technology is involved as a sub-process, 412 residence (or retention) time of the material in the dispersing zone becomes a key parameter 413 [51]. Considering that the nozzle cross-section is often close to the cross-section of the layer,
- the average material velocity in the printhead is on the order of the nozzle velocity itself (typi-
- 415 cally from 1 to 10 cm/s [9]). Since most printheads' overall lengths are in the order of several
- 416 tens of centimeters, the residence time should vary between 1 and 100 s.
- 417 The accelerators used in additive manufacturing are either inorganic compounds with sizes of
- 418 less than one nanometer or organic macromolecules of sizes on the order of 100 nanometers
- 419 [52]. Although the validity of the Stokes-Einstein equation (see Eq. 1) fades when the size of
- 420 the molecules gets closer to the size of the molecules of the solvent, it is used here to estimate
- 421 the typical diffusion length from the natural diffusion coefficient of these accelerators.

$$D = \frac{kT}{6\pi r\mu} \tag{1}$$

422 where k is the Boltzmann constant, T the temperature in Kelvin, r the size of the accelerator 423 molecule and μ is the viscosity of the solvent. The typical diffusion length is of the order of 424 \sqrt{Dt} , where t is the residence time. This leads to typical diffusion lengths between a few mi-425 crometers and a few hundred micrometers. Consequently, even for the smallest printheads for 426 pastes, full dispersion of accelerators in the nozzle cross-section cannot rely on natural diffusion 427 alone, requiring additional dispersion capacity. As the viscosity of cement-based materials is 428 too high to allow for turbulent dispersion, it is in the field of convective mixing that solutions 429 do exist [53, 54].

- The idea behind convective mixing relies on the creation of a secondary flow in the nozzle so that the distribution of the accelerator molecules is allowed. By shearing the material and, hence, distributing the accelerator in sheared material layers, one can reach, after a sufficient residence time, the situation in which the typical distance between two sheared material layers is on the order of the typical diffusion length estimated above.
- 435 From a technological point of view with respect to the newest existing solutions, this translates
- 436 into either so-called static mixers or screw-mixing devices. The former are immovable, as in-
- 437 dicated by their description, and it is the overall flow of the material around the surface of the
- 438 static mixer that disperses the accelerator or other admixture, such as pigments. The latter are
- 439 simply additional mixing systems with their own controls. Both are inserted into the printhead.
- 440 The dispersion intensity of the static mixer is proportional to the flow rate in the printhead,
- 441 while at the same time it is an independently tunable parameter in the case of the screw mixer.
- 442 The above features have not been studied in detail for the specific conditions and requirements 443 of extrusion-based printing with cement-based materials as yet. Printheads and their mixing
- 444 devices are accordingly designed by trial and error. Numerical simulations [55, 56, 57] could
- 445 in the future allow for a better understanding and progress in printhead design.

446 6. Load-bearing and deformation behavior after deposition

447 6.1 Object failure during manufacturing

448

449 **6.1.1 Failure modes**

Additive manufacturing methods of cementitious materials are, by definition, set apart from "conventional" formative methods by the absence of molds. As a result, objects may collapse during the manufacturing process. Considering that the material resistance is initially low relative to its self-weight, the 'buildability' [58, 59] of a mortar is an important property in assessing its suitability for printing. The term, however, refers to a range of processes and properties that require elaboration in order that it becomes a meaningful concept.

- 456 At present, two mechanisms have been recognized as causes of collapse in extrusion-based,
 457 layered 3D concrete printing during manufacturing:
- material failure (Figure 6a), and
- loss of stability (Figure 6b).
- 460 Material failure occurs when the material strength is exceeded, resulting in yielding, flow, or
- 461 fracture, whereas stability loss is defined by a loss of equilibrium of forces and moments, initi-462 ating uncontrolled deformations or displacements.
- 463



464 465

466

a)

b)

467 Figure 6. Collapse in extrusion-based additive manufacturing: a) material failure, b) stability468 failure [60].

469

470 Either mechanism is triggered by a combination of support, load, and geometry conditions - of 471 which the former is generally constant, but the latter do change over time due to the gradual 472 buildup of the object during manufacturing. In the hitherto limited number of developed pre-473 diction models for object failure during printing of cementitious materials, the centered self-474 weight of the material is considered as the sole load condition exerted on the print object. This 475 allows the formulation of relatively simple relations between material strength and build height; 476 see Section 6.1.2, Eqs. 2 and 3. Several additional types of loading are conceivable, such as 477 kinematic pressure from the deposition of filament, vertically or horizontally, when layers are 478 squeezed together, non-vertical pressures from supporting filler materials as in Figure 7, eccen-479 tric loading due to accidental misalignment of layers, or purposefully created cantilevering ge-480 ometries.

481



482
483 Figure 7. Structural failure of an object during 3D printing due to horizontal loading by a highly
484 fluid infill material

485

In any specific case, 'buildability' – or the resistance to failure during printing – is not only
dependent on the material's characteristics but, also on the object design, e.g. size, geometry,
and process parameters such as print speed. For example, at otherwise identical print settings,

a straight wall may fail sooner than a curved wall of the same path length due to the increased

490 stability provided by the curved wall's bend(s). In another case, a short wall could topple over 491 at fewer layers than a long wall, even though buckling is determined by the perpendicular sec-

491 tion, which is identical for both, because of the slower buildup and longer time for the material

493 to develop strength and rigidity [61].

494 To define the initiation of material failure, i.e., fracture, yielding, etc., many criteria have been

495 proposed [62], but it is not immediately clear which one(s) is (are) suitable for the mortars that 496 are used in extrusion-based concrete printing. This is primarily due to the transition that print-

497 able cementitious mortars undergo from a non-Newtonian fluid state to a solid state during the

498 manufacturing process. This process, which usually takes several hours but can be as short as

- 499 several minutes when fast-setting cements or accelerators are applied, includes reversible phys-
- 500 ical, e.g., flocculation-induced thixotropy, as well as irreversible chemical phenomena (hard-
- 501 ening due to cementing reactions). As a result, both strength and rigidity increase over time,
- 502 which in rheology is often labelled as the structuration rate of the yield stress, A_{thix} .
- 503 The selection of a suitable criterion to characterize material failure is further complicated by
- the fact of printable mortars having considerably varying levels of initial yield stress and rigid-
- ity, associated sheared or un-sheared flow regimes, and different structuration and hardening
- rates as well while the level of material development in an individual print depends on the print

507 strategy and object design. Thus, the appropriate material failure criterion may depend on the

- 508 mortar, process and design characteristics in any specific case further increasing the chal-
- 509 lenges in developing generic prediction models.
- 510 Considering the fluid-to-solid transition, the material could be assumed to behave as a highly
- 511 viscous fluid or as an initially very compliant visco-elastic or elasto-plastic solid. In the case of
- the former, concepts from the field of fluid mechanics and rheology should be applied, whereas
- 513 solid mechanics should be applied in the case of the latter. The approach selected governs the
- 514 definition of material properties and the experimental methods required to determine them. Vice
- versa, it should be recognized that not all experimental methods are suitable for all ages of print
- 516 mortar; for example, some mortars are too stiff to perform rotational rheometry without plug
- 517 forming, while others may be too fluid to perform unconfined compression tests. Indeed this
- 518 could serve as an indicator regarding the applicability of certain theoretical approaches.

519 The approach that is to be taken also determines the types of analysis that may be performed to 520 quantify 'buildability'. A solid approach governed by mechanics allows recognition of material 521 failure under anisotropic, multi-axial stress states as well as failure due to loss of stability. In 522 fluid mechanics, only isotropic stress states are considered, and stability analyses are highly

- 523 unusual. However, it is more appropriate for flow analyses.
- 524

525 **6.1.2** Approaches in literature

526 In the literature, a range of different approaches has been suggested to quantify buildability, 527 with the lack of unanimity being caused by the sheer number of aspects discussed above. The 528 summary provided in Table 2 shows that the spectrum of models not only stems from the fun-529 damental approach to the material state, but also from other associated issues related to exper-530 imental methods, validation, time dependency, and so on. In the elaboration provided below, 531 some quantitative data as well are presented for comparative purposes. However, it should be 532 noted that these values have been obtained from significantly different experimental methods,

533 the validity of which may still be under debate.

Publication	Failure mode(s)	Theory	Material failure cri- terion	Experimental Proce- dure	Time effect	Geometrical Model	Validation
[63]	Material	Solid mechanics	Drucker-Prager	Unconfined uniaxial compression	Constant	2D in vertical section plan	Unconfined uniaxial compression
[15]	Material	Rheology	Yield stress of non- Newtonian fluid	Rotational rheometer	Linear & exponential	vertical stack, 1D	Unconfined uniaxial compression
[1]	Material	Rheology	Yield stress of non- Newtonian fluid	n/a*	Linear	vertical stack, 1D	n/a
[17]	Material, stability	Solid mechanics	Mohr-Coulomb	Shear box + uncon- fined uniaxial com- pression	Linear	vertical stack, 2D in vertical section plan	Cylindrical print trial
[64]	Material, stability	Solid mechanics	Mohr-Coulomb	Shear box + uncon- fined uniaxial com- pression**	Linear & exponential decaying	Linear wall struc- tures, 2D and 3D	Wall print trials
[10]	Material, stability	Mixed	Yield stress of non- Newtonian fluid	Rotational rheome- ter*	linear & exponential	vertical stack, 1D	n/a
[65]	Material	Rheology	Bulk yield stress from Benbow-Bridgewater model	Ram-extruder	Constant	vertical stack, 1D	Cylindrical print trial
[66]	Material	Solid mechanics	Shear stress	n/a**	Linear	Vertical stack, 2D along print path length	n/a
[61]	Material, stability	Solid mechanics	Mohr-Coulomb	Tri-axial compression	Linear	vertical stack, 2D in vertical section plan	Wall print trials
[67]	Material	Rheology	Yield stress of non- Newtonian fluid	Rotational rheometer	Bi-linear	vertical stack, 1D	Cylindrical print trial

Table 2. App	roaches to	the q	quantificatio	n of	'buildability'	' in	literature.

theoretical, based on volume fraction and an average interparticle force.
analytical paper, material failure properties based on data from other study.
analytical paper, no direct relation to experimental work.

537 Di Carlo *et al.* [63] investigated the compressive strength of a fresh print mortar using a uniaxial 538 compression test and found compressive strengths of 5.52 kPa to 88.3 kPa and moduli of elas-539 ticity of 77.9 kPa to 1241 kPa, for mortars at ages of 11 to 288 minutes (strength 10.7 kPa after 540 47 minutes). They recreated the experiments in a Finite Element Model (FEM) in which a 541 Drucker-Prager failure criterion was applied.

- 542 Alternatively, Perrot *et al.* [15] presented a buildability approach based on the rheological yield 543 stress τ_c of the print mortar as the material failure criterion, which was relatively easily obtained 544 from rotational rheometry and validated against uniaxial compression tests on shallow samples. 545 Eq. (2) presents the basis of their approach in slightly rewritten form, with α_{geom} a geometrical
- 546 factor, $\tau_0(t)$ the time dependent yield stress of the mortar, ρ its density, g the gravitational
- 547 acceleration, and h(t) the time dependent object height:

$$\alpha_{geom}\tau_0(t) \ge \rho gh(t) \tag{2}$$

- 548 The yield stress of the mortar applied in that study developed linearly over time until reaching
- 549 an approximate age of 40 minutes, from 4.13 to 6.29 kPa ($A_{thix} = 54$ Pa/min), but it developed

550 exponentially when considering a time frame of 0 to 80 minutes. The validation experiments fit

- 551 the predicted failure moments very well. Further experimental rheological data on printable
- 552 mortars can be found in [68, 69, 70].
- 553 Wangler et al. [1] adopted a very similar approach, but applied a von Mises plasticity criterion,
- by introducing a factor $\sqrt{3}$ into the buildability equation. On the other hand, a geometry factor
- 555 was not applied. Rewritten, this yields:

$$\pi_0(t) \ge \rho g h(t) / \sqrt{3} \tag{3}$$

- 556 The first method to take both material and stability failure into account was presented by Wolfs 557 et al. in [17], and further developed in [61, 71]. This method is based on FEM expanded with 558 time-dependent properties, and the adoption of the Mohr-Coulomb failure criterion, including 559 the development of experimental procedures to determine the full failure envelope. The dual 560 failure criterion of plastic yielding and elastic buckling requires the experimental determination 561 of five time-dependent material properties: the apparent Young's modulus E(t), Poisson's ratio 562 v(t), the cohesion C(t), angle of internal friction $\varphi(t)$, and dilatancy angle $\psi(t)$. For two com-563 mercially available mortars this yielded linearly developing compression strengths from 5 to 90 minutes of 7.1 kPa to 18.9 kPa (rate: 139.6 Pa/min) and 2.8 kPa to 29.7 kPa (rate: 317.2 Pa/min), 564 565 and moduli of elasticity of 80.2 kPa to 186 kPa (rate: 1244 kPa/min) and 35.9 kPa to 325 kPa 566 (rate: 3400 Pa/min). Suiker [64] introduced a parametric mechanistic model which focuses on 567 the competition between elastic buckling and plastic collapse, and can be used to predict struc-568 tural failure of straight, free-standing walls during 3D printing.
- A mixed methodology combining the rheology material failure criterion with the stability considerations from solid mechanics was suggested by Roussel [10]. Besides (3), this yields (4), again, slightly rewritten, representing the material and stability failure criteria, respectively. Through the relation E = 2G(1 + v) between Young's modulus *E*, shear modulus *G* and Poisson's ratio *v* on the one hand, and the relation $\tau_c = G\gamma_c$ between yield stress, shear modulus and critical shear strain γ_c on the other, the transition height can be determined according to (5), at which one failure criterion becomes dominant over the other.

$$E_c(t) \ge 3\rho g h(t)^3 / 2\delta^2 \tag{4}$$

$$h_t = 2\delta \sqrt{\frac{1+\nu}{3\sqrt{3}\gamma_c}} \tag{5}$$

- 576 Continuing from the material assumptions by Perrot *et al.* [15], Wangler *et al.* [1], and Roussel
- 577 [10], Kruger *et al.* [67] suggested, rather than a linear structuration rate, a bilinear rate, which
- 578 yielded a shear strength development of 1.15 kPa to 2.73 kPa at a re-flocculation rate of $R_{thix} =$
- 579 413 Pa/min over the first 230 s, and further up to 6.37 kPa at 60 minutes at $A_{thix} = 64.8$ Pa/min.
- 580 A rheological approach with a Benbow-Bridgwater model rather than a Bingham model was
- adopted by Chaves Figueiredo *et al.*, [65]. This yields the material parameters bulk yield stress
- and shear yield stress, of which the former seems to be primarily responsible for the aspect ofbuildability.
- 584 Finally, an analytical model was proposed by Jeong *et al.* [66], who took the age of the material 585 along the print path length into account. The shear strength (solid mechanics) was adopted as 586 failure criterion without extensive elaboration.
- 587

588 6.1.3 Research needs

589 Many aspects surrounding in-print failures are still unclear. Globally three topics seem to be 590 particularly in need of research.

First, little is known about the actual loads acting on in-print objects. Although self-weight, the
most important load, can be established relatively easily, the effects of kinematic pressures,
support materials, and eccentricities are not quantitatively known.

594

595 For material failures, the appropriate failure criterion or criteria need(s) to be established. A 596 fundamental underlying question is in which cases and whether at all it is relevant to allow for 597 anisotropic stress states for such a criterion, which requires considerably more complicated 598 analyses and experimental procedures. The accuracy of buildability predictions based on dif-599 ferent methods and associated material property experiments should be rigorously tested 600 through extensive printing trials. Rather than focusing on a single case, such trials should en-601 compass a range of different geometries, print speeds, print resolution, and object sizes as well 602 as their being performed on different printing facilities to eliminate equipment-dependent bias. 603 The development of a range of benchmark print geometries for comparative studies would be

604 beneficial.

605 606 Once the theoretically most satisfying approach has been found, the correlations between vari-607 ous (experimental) methods may be studied to determine which approach is to be preferred 608 from a practical standpoint. In this campaign it should be recognized that the appropriateness 609 of a certain approach may depend on the properties of the individual print mortar or its rela-610 tionship to the physical condition. For instance, it should be expected there are thresholds of yield stress or viscosity above or below which solid or fluid mechanics are clearly more appli-611 612 cable, but it is conceivable that transitional stages exist in which each approach may work sim-613 ilarly well and the preferred approach may depend on other factors such as practical arguments. 614 Only a few multi-modal material characterizations have yet been performed [72, 73], and cor-

- 615 relation attempts to link different approaches are still minimal.
- 616

617 Finally, the transitory characteristics of print mortars should be more comprehensively studied.

- 618 Current buildability models only consider properties such as strength and modulus of elasticity
- 619 as a linear or non-linear function of time. It is, however, likely that such developments are a
- 620 function of multiple parameters, the most obvious of which is temperature. The temperature
- 621 dependency of buildability has tentatively been established [74], which indicates that time-de-
- 622 pendent development functions must be derived for a multitude of temperatures, or through a
- 623 combined parameter such as maturity, indeed a combination of time and temperature, as has
- already been defined for concrete of ages beyond the setting time. Considering printing's not

always taking place under fully conditioned environmental conditions, this should be consid ered relevant. Possibly other parameters, such as the shearing history of the mortar may also
 need to be included.

628

629 6.2 Deformation behavior

630

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631 6.2.1 Deformation mechanisms

The actual deformations which occur in cementitious mortars during selective material deposition by extrusion *before* failure have hardly been studied. Two mechanisms should be distinguished. First, in some cases, depending on the solidification strategy of the process in question (cf. Sections 3 and 4), the mortar flows briefly upon deposition before a rapid increase in strength initiated by viscosity modifiers or accelerators stops the visco-plastic flow and the material assumes a stable shape.

638 Other processes deposit mortars in an unsheared condition, directly in a shape-stable state,
639 which can last from minutes to hours. The visco-elastic deformations that may occur during
640 this period may influence both the geometrical conformity and the object stability during print641 ing.

642 To determine the elastic deformations, the apparent Young's modulus E may be determined

643 from unconfined compression experiments directly; cf. Section 6.1.2. Obtaining E through the 644 shear modulus G taken from rheometric tests does not seem feasible, as this requires the as-

- 645 sumption of a value for the Poisson's ratio v.
- 646 Depending on which of the deformation mechanisms, flow or (visco-)elastic deformation, is
- 647 being studied, different modelling approaches can be adopted: Computational Fluid Dynamics
- 648 (CFD) for the former [75] or Finite Element Modelling (FEM) for the latter. A combination of
- both may be required to capture the full deformation and displacement behavior. The Discrete
- Element Method (DEM) might also be suggested, as it is suitable for granular materials and
- theoretically capable of modelling both flow and deformations [76, 77].
- 652

653 6.2.2 Research needs

654 The accurate prediction of deformations and displacements is relevant to geometry-related fail-655 ure behavior, i.e., primarily stability failure and, to a lesser extent, material failure, but to issues 656 of the geometric conformity of the finished product as well. Because little data on fresh mortar 657 deformation behavior is currently available, more research into the related material properties 658 is required. This includes the development of suitable experimental methods, for which some 659 initial suggestions have been made in [8]. Modelling methods need to be developed and validated, which also calls for the establishment of suitable means of measurement during or after 660 printing. The interaction between in-print failure and deformation behavior should be further 661 662 explored.

663

664 7. Physical properties and their evolving over time

665 **7.1 Viscosity**

666 Viscosity is the measure of the internal resistance of a fluid to its being deformed by shear 667 stresses. In this context the less viscous a fluid is, the easier it flows. Due to the flocculation 668 and stiffening of the cement-based materials, the apparent viscosity of the cement-based mate-669 rial increases with time at rest. However, for yield stress this behavior is reversible in time since 670 the mixing power is sufficient to break down the links between particles. Here it is important 671 to note that the viscosity of the cement-based materials can evolve due to process-induced ma-672 terial variation during pumping or extrusion. Such variation can be associated, for example,

with particle migration under shear flow. 673

674 7.2 Yield stress

675 Yield stress is the material property that denotes the transition between solid-like and fluid-like 676 behaviors. On the microscale, interparticle forces between the solids in a suspension result in a 677 yield stress that must be overcome to start flow. An applied stress lower than the yield stress will result in deformation behavior like that of a solid. In other words, yield stress is the mini-678 679 mum stress that makes the fluid flow as a viscous material. This minimum level of solicitation 680 depends on the stress history of the cementitious materials. Consequently, static and dynamic yield stresses can be distinguished. Static yield stress is an increasingly important parameter 681 682 that depends on shearing and resting periods and is related to microstructural buildup and is 683 defined as the shear stress required to make the material flow, while dynamic yield stress can 684 be defined as the yield stress measured under the steady-state flow of material with unstructured 685 cement paste.

686 The major challenge associated with the 3D printing of cement-based materials is the determi-687 nation of increase in static yield stress after a layer has been deposited. Since such measure-688 ments are not trivial; see e.g. [78]. And there are numerous activities on developing adequate

689 test methods and protocols; see e.g. [79, 80].

690 7.3 Thixotropy parameter / rate of structural buildup / structural breakdown

691 Fresh cement-based materials undergo cement hydration, which impacts their physical and rhe-692 ological behavior. It is this change in mechanical characteristics that allows the fresh material 693 to build up and be able to support the increasing load generated by the successive deposits of the layers of the printed structure. Therefore, it is necessary to rely on the kinetics of the me-694 695 chanical structural buildup of the material as they relate to the activity of cement in water [1, 696 15].

697 To understand why the cement material becomes rigid, it is necessary to take into account the 698 organization of the network of cement grains in suspension in the water. After intense mixing 699 culminating in de-structuring, the cement grains begin to flocculate under the influence of col-700 loidal interactions. This flocculated lattice structure induces an increase in rigidity and strength, 701 over a time period of several tens of seconds [10, 81]. Then, over longer periods of time, the 702 rapid formation of C-S-H linkages in the contact zones between the grains goes on to form the 703 origin of the continued mechanical structural buildup of the cement material left at rest [81]. 704 This phenomenon of the nucleation of the cement grains occurs during the period known as the 705 dormant period, before the cement setting. These nucleations are a chemically irreversible phe-706 nomenon. However, if the power of the pumping and/or mixing system suffices, these links 707 may be broken. It is important to note that in the 3D printing of cement-based materials, accel-708 erators are often added to the cementitious materials leading to the early formation of other 709 hydrates such as ettringite, monosulfate, and aluminate hydrates, which are responsible for the 710 early increase in static yield stress.

- 711 Therefore, the parameters of mechanical behavior are subject to the structural buildup kinetics
- 712 over time when the cement materials are left to set after being deposited in layers. To illustrate
- 713 and describe this phenomenon, the changes in the stiffness and the yield stress values have often
- 714 been reported by several researchers [81, 82, 83].

- 715 The increase in the progression of yield stress over time is often considered to be linear and, as
- such, allows a structural buildup rate to be defined as A_{thix} in Pa·s⁻¹ [81, 82, 83]. In this case,
- 717 the equation is written in this way:

$$\tau_0(t) = \tau_{0,t=0} + A_{thix}t$$
(6)

718 with $\tau_{0,t}=0$, the shear yield stress of the material in a de-structured state and t being the duration 719 of the resting period of the material.

The linear modelling of shear yield stress is generally valid during the first hour of the resting period of the cement-based material [84]. Beyond that the change accelerates and the kinetics of the structural buildup become exponential [85, 86]. This change in rates can be explained by the beginning of the setting, the increase of the solid volume fraction with the possible interpenetration of the C-S-H crystals created. Perrot *et al.* [85] proposed a law for the exponential progression of shear yield stress, tending towards the linear model over short time periods. This model is given by (7):

$$\tau_0(t) = A_{thix} t_c \left(e^{t/t_c} - 1 \right) + \tau_{0,t=0}$$
(7)

727 with t_c as a characteristic time over which the behavior can be considered linear. The model of

(7) can be used to describe the progression of the shear threshold over longer periods. Other
more sophisticated models have recently been reported in the literature [17, 18, 86]. Some authors have even shown that a Von Mises-type plasticity criterion may ultimately not be wellsuited to account for the breakage of the material. Effectively, at a certain stage in the development of the setting, the behavior of the material displays a pressure-sensitive, granular type

of behavior, probably related to the interconnection of the hydrates, then becoming sensitive to

- pressure. The mechanical behavior thus represents a progressive dissymmetry, with a resistance
- that is always higher in compression than in tension. Hence, the transition to hardened concrete
- behavior can be seen to start at this time.

Mantellato *et al.* [87] recently reported that the increase in the yield stress associated with an
irreversible flow loss can be associated with an increase in the solid particles' specific surface
induced by chemical cement hydration. At the same time the critical deformation decreases
slightly with the hardening of the material [10, 17, 18].

Additionally, this increase in stiffness over time is reflected in an increase in the elastic modulus. This increase in stiffness and strength allows the material deposited to withstand the increased loads associated with the printing of the structure. In this way it is possible to calculate and predict the optimal manufacturing speeds to guarantee the stability of the structure printed and to ensure the compensation for the elastic deformation.

746

747 **7.4** Measurement procedure of rheological and early age properties

748 To assess the stability of the in-print cementitious materials, it is necessary to follow the evo-749 lution of the materials' early-age properties after being deposited. Then effort must be made to 750 monitor inline the evolution of stiffness and strength of the cementitious material.

- 751 If oscillatory rheometry and ultrasound test measurements are reliable methods to estimate the
- revolution of a material's elastic modulus over time, they require expensive and sensitive de-
- vices that are not easy to implement in a printing environment [50].
- 754 To relieve this situation, instantaneous or continuous penetration tests or gravity-induced flow
- tests are being developed in order to monitor, using simple tools, the evolution of the materials'
- properties over time. For example, the penetration test (cone plunger) was used by filling the
- cone mold with mortar in two layers, then releasing the cone plunger, thus allowing the plunger

to penetrate into mortar under its own weight [88]. Good relationships ($R^2 = 0.86 - 0.89$) between the penetration and slump flow were established. It was reported that the increase in the penetration values followed the same trend with the increase in the slump flow of mortar.

761 8. Summary and conclusions

3DCP offers great potential to facilitate development towards a sustainable Construction Industry 4.0, tackling existing problems such as low productivity and shortage of skilled labor in the process. Among 3DCP approaches, those based on material extrusion seem to be particularly promising at this stage with respect to both overall technological readiness level and economic viability. This article focuses on this specific 3DCP approach. The explanations and statements are the result of collective research performed by the authors in the framework of the RILEM Technical Committee 276 "Digital fabrication with cement-based materials".

769 Three main categories of extrusion-based 3DCP are identified as: i) extrusion of stiff material, 770 similar to conventional extrusion, ii) extrusion of flowable material with or without addition of admixture(s) in the printhead, and iii) extrusion of material using additional energy input, e.g. 771 772 vibration to facilitate delivery and deposition of stiff mixtures. To each category and each pro-773 duction step, i.e., transportation of build material, printhead process, deformation of build ma-774 terial during deposition, and behavior of build material after deposition, relevant processes and 775 corresponding physical fundamental are presented. In particular, gravitational flow, pressureinduced flow during pumping and extrusion, and dispersion of admixture in the printhead are 776 777 considered. Also, attention is paid to the load-bearing and deformation behavior of the mate-778 rial/printed object after material deposition. Two major failure modes are defined for object 779 failure during manufacturing, both crucial to buildability: i) material failure, where the material 780 strength is exceeded, and ii) stability failure due loss of the equilibrium of forces and moments. 781 Various models to estimate buildability are presented and critically discussed. Deformation 782 mechanisms are explained as well. Finally, key physical properties of cement-based materials 783 in the fresh state – viscosity, yield stress, and thixotropy – as well as their evolution in time are 784 identified, followed by brief remarks on measurement procedures of rheological and early-age 785 properties.

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787 In summarizing the knowledge as presented, it can be concluded that underlying physics are 788 well understood for most processes of large-scale additive manufacturing by material extrusion. 789 This understanding can and should be utilized for the purposeful design of 3DCP systems rather 790 than trying to use a trial-and-error approach in shaping the 3DCP process. Purposeful, system-791 atic approaches based on the associated physics should facilitate material development and me-792 chanical engineering design as well as optimize process regimes and process control. For some 793 processes analytical, scientifically based formulas already offer reasonable predictions with re-794 spect to material flow in the case of relatively simple geometries. Nevertheless, further research 795 is needed in order to enable the development of reliable tools for quantitative process analysis 796 and for predictions based on the underlying physics. The major challenges in analyzing 3DCP 797 systems arise out of the complexity of flow regimes and patterns in various production steps as 798 well as the time- and shear-history-dependent behavior of cement-based materials, which are 799 inherently complex multiscale, multiphase, densely packed suspensions. Much effort needs to 800 be invested in studying and describing specific flow behavior and developing adequate testing 801 technics to quantify key material parameters. Numerical simulation can contribute greatly to 802 analyze flow processes under consideration of complex geometric boundaries; this has the po-803 tential to be developed into a powerful design tool for shaping the 3DCP processes. The deri-804 vation of model parameters is the main issue here, requiring appropriate experimentation. This 805 deliberation holds true as well for the estimation of buildability. Material behavior, geometry

- of the printed element, particularities of printing process, and other aspects need to be consid ered collectively. While analytical formulas may deliver reasonable predictions for relatively
 simple cases, numerical simulation constitutes a promising approach for analysis of more com plex cases.
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