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Development of Cement Mortar Incorporating a Natural Biopolymer for Potential Use in Extrusion-Based 3D Printing

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Abstract

This study developed and evaluated cement mortar with a natural biopolymer Ceratonia siliqua seed gum, which could be used in extrusion-based 3D printing. A total of 9 formulations included a plain control, chemical-admixture control and seven dosages of biopolymer, 0.1 to 0.8% by cement mass. Cement:sand ratio and nominal added-water-cement were about 1:1.25 and 0.32, respectively. Flow-table observations and manual mass extrusion through a 30 mm outlet were made; the chemically modified compression batches contained an accelerator. The rate of mass extrusion was 2.92 g/s for plain mortar and 6.54 g/s for the admixture control and reached its maximum with 0.3% biopolymer (7.08 g/s). The corresponding three-day strengths were 20.53, 28.83 and 30.25 MPa. The extrusion rate and strength of the 0.3% formulation were 8.28% and 4.93% higher than the admixture control, respectively. Increased dosages raised recorded flow-table D2 values but decreased extrusion rate and strength. The dosage of 0.3% is thus a candidate to be tested further and not a proven optimum. The limitations of interpretation include manual actuation, missing replicate data, and missing specimen documentation, whereas the pumpability, multilayer buildability and interlayer performance are not verified.

Keywords: cement mortar; Ceratonia siliqua; early compressive strength; extrusion-based 3D printing; manual extrusion; natural biopolymer

1 Introduction

In extrusion-based three-dimensional printing cementitious substances are required to flow through gear, deposit, and maintain their shape upon deposition. These requirements are regulated by mixture composition, equipment and construction sequence. According to Buswell et al. [1] and Roussel [2] this is one of the key challenges: simple discharge fails to create a stable layer and hardened strength fails to characterize resistance throughout the deposition process.

Laboratory screening has the capability of reducing the mixtures to be used in controlled printing trials. Le et al. [3] and Kazemian et al. [4] employed complementary fresh-state analysis to form printing mixtures. In cases where a printer is not available, manual tests provide convenient initial comparisons, but with apparatus-specific responses must be separated out of pumpability and multilayer performance.

Ceratonia siliqua seed gum, commonly called locust bean gum (LBG), is a galactomannan derived as a by-product of carob seed endosperm [5]. Its application as an admixture is not the same as that of cement replacement with African locust-bean pod ash. In their research, Gonzalez-Aviina et al. [6] examined LBG and other biopolymers in cement paste taking into account the preparation, dosage, rheology, hydration, stability and strength. The next paper they published looked at biopolymer-

superplasticiser blends [7]. These experiments encourage the experimentation of the gum in the targeted mortar and chemical-admixture system.

The present study develops and evaluates biopolymer-modifier cement mortar at predetermined nominal water cement and cement:sand ratios. The base chemical treatment is distinguished by a plain control and a chemical-admixture control with the incremental response to seven dosages of gum. Flow and quantitative extrusion involved the use of unaccelerated mortar, but the modified compression batches included the use of accelerator.

The aim was to develop cement mortar that utilizes a natural biopolymer that can be used in 3D printing by extrusion. The experiment recorded the materials of constituents and initial ratios, plain and chemically modified mortar, and biopolymer dosage using flow-table records, mass extrusion rate, filament size and compressive strength after three days. These answers were applied in determining the most promising formulation that had been tested to be further printed. The manual laboratory programme offers rough material development as opposed to the ultimate printing specification or statistically validated optimum.

2 Materials and methods

2.1 Materials and formulations

The programme was conducted in the Materials Laboratory, Department of Civil Engineering, University of Ibadan, Nigeria. The base batches had 2,220 g Portland cement, 2,770 g sand and 710.4 g added tap water. There was no additional cementitious material, coarse aggregate or fibre. The ratio of cement to sand-mass was 1:1.25. For mortar batching, the sand was sieved using a 2.36 mm sieve.

The biopolymer was packaged *Ceratonia siliqua* seed-endosperm gum. Dosages were 0.1, 0.2, 0.3, 0.4, 0.5, 0.6 and 0.8% of cement mass, equivalent to 1–8 g/kg of cement. Table 1 gives the corresponding batch quantities. C0 contained neither chemical admixture nor gum. C1 contained the chemical admixtures appropriate to each test route but no gum.

Table 1. Formulation quantities on the recorded base-batch basis.

Mix	Gum (g)	WR/R (mL)	ACC* (mL)
C0	0.00	0	0
C1	0.00	20	18
LBG0.1	2.22	20	18
LBG0.2	4.44	20	18
LBG0.3	6.66	20	18
LBG0.4	8.88	20	18
LBG0.5	11.10	20	18
LBG0.6	13.32	20	18
LBG0.8	17.76	20	18

WR/R = water reducer/retarder; ACC = accelerator. *ACC applies only to activated tests; scaling to individual specimens and portions was not documented.

The admixture of water also possessing a retarding action was the water-reducing type and was found to be a sulphonated system of naphthalene formaldehyde. It had a quantity of 20 mL per modified base batch listed. The activated formulation basis had 18 mL labeled accelerator. No mass percentage of either liquid was prepared due to lack of density and solids content. The grade of gum and the strength category of cement used and the elaborate process of dispersion of gum was not documented in the experiment.

2.2 Water basis and preparation

The nominal added-water–cement ratio was 710.4/2,220 = 0.32. Aggregate moisture and absorption and water added by liquid admixtures were not measured; 0.32 is thus an added-water ratio and not an effective ratio that was validated. Mini mixer was applied. Ambient, water and fresh-mortar temperatures were recorded at about 28-29 °C, 26 °C and 27-28 °C respectively. Relative humidity and the actual test-age reference instant were not recorded, nor was mixing duration.

Batched and mixed mortar on the base was separated into a small amount to be deposited to explore the deposition. That was mixed with accelerator and put into a piping bag. This was pre-loading manual activation, as opposed to in-line nozzle injection. Accelerator was initially mixed with water in the Vicat procedure and the mixture was subsequently added to cement. There was also accelerator that was added to the batch that was to be used in each activated compression cube. These paths were distinct of non-accelerated flow and quantitative extrusion. Figure 1 shows the Vicat apparatus and cast cubes in their moulds.



Figure 1. Laboratory photographs: (left) Vicat apparatus and (right) cast cement-mortar cubes in their moulds.

2.3 Fresh and deposition measurements

Approximately five minutes after mixing the flow was observed under no accelerator using 20 table drops. D1 and D2 were recorded on the record as well as a field identified as average spread. D1 was

100 mm across and the third field was D2 -D1. Since the record was not conclusive to establish the diameter convention, then D2 is reported as a recorded diameter entry and the difference is not reported as a proven percentage flow.

At about twenty minutes quantitative extrusion commenced. A 2,220 g charge was placed in a manual plunger device with a 30 mm circular outlet, and was discharged during 240 s. The charge mass refers to mortar, rather than cement. The rate of average mass extrusion was determined by means of the formula, as shown in (1) where m is the discharged mass in grams, and t is the seconds of collection.

$$q = \frac{m}{t} \tag{1}$$

The entry in the record was a 300 N force entry and the method of application and measurement was not set. This means that there are no constant-force conditions. The mass record was accompanied by observations of blockage, tearing and segregation, but there was no history of continuous filament length or pressure.

A recession of filament width and height five minutes following extrusion was made relative to nominal values of 30 and 10 mm. The record was silent whether these readings were obtained by unaccelerated discharge of plunge and by piping-bag deposition or by the action of piping-bag deposition. They are then reported as geometry at a given time, without a state of acceleration or interpretation of differences in targets as settlement. Vicat observations were duly made in minutes at ca. 28 °C; constitution of the specimen, origin of time and penetration criteria was not fully recorded.

2.4 Compression and data analysis

At three days of compression, nominal 70.7 mm cubes were used to measure compression. C0 was not accelerated, C1 and LBG batches of cubes were. The strength was re-calculated by using the reported P failure load in kN and nominal loaded area in mm^2 as in Equation (2). The casts were made in moulds (Figure 1).

$$f_c = \frac{1000P}{A} \tag{2}$$

where A = 70.7^2 = 4,998.49 mm^2 and f_c is expressed in MPa. There were no individual measured dimensions, specimen formation and compaction details, curing regime and loading rate. The results are therefore considered as nominal-area three-day cube strengths, as opposed to printed directional strengths or results compared to a given standard method.

Averages were reported as formulation-level values, and replicate counts, as well as individual observations, were not available. Analysis was limited to recalculation, relative changes and numerical rankings. There were no error bars, confidence intervals and significance tests built. Relative changes were based on unrounded values. C1 was given an index of 100 of extrusion and strength. No fitted weighted score or continuous optimisation model was fitted.

3 Results and discussion

3.1 Sand grading and comparison basis

Corrected source-sand retained masses summed to 500.00 g. Percentages passing were 93.89% at 4.75 mm, 80.49% at 2.36 mm, 47.47% at 1.18 mm and 15.06% at 0.600 mm. The 1.18 and 0.600 mm sieves together retained 65.43% of the recovered sample, while only 0.77% passed 0.300 mm. These values characterise the original source sand. An individual grading curve was not obtained of the fraction prepared with the 2.36 mm sieve.

The source grading cannot thus be termed as the grading of the batched mortar sand which has been measured. Specifically, the source material stored at a level of 2.36 mm and above does not determine the oversize fraction in the batch prepared. The experiment keeps the sand preparation constant and is unable to isolate a grading effect. The available mixture-development literature [3], [4] gives some background on the need to limit aggregate size, although they do not support the current outlet and sand mix with alternative equipment.

C0-to-C1 comparisons: The chemical treatment that is appropriate to the test. This was the water reducer/retarder, alone, in flow and extrusion. It provided the activated treatment in compression. C1-LBG comparisons indicate the response to the incremental dosage in that background. This difference allows the difference between gum-containing mortar and C0 not to be fully ascribed to the biopolymer.

Table 2. Unaccelerated flow-table D2 entries and extrusion results.

Mix	D2 (mm)	m (g)	q (g/s)
C0	150.0	700	2.92
C1	161.0	1570	6.54
LBG0.1	161.2	1584	6.60
LBG0.2	162.0	1650	6.88
LBG0.3	162.0	1700	7.08
LBG0.4	163.0	1685	7.02
LBG0.5	164.0	1623	6.76
LBG0.6	165.0	1600	6.67
LBG0.8	165.0	1584	6.60

D1 = 100 mm throughout. Flow age ≈ 5 min; extrusion age ≈ 20 min. Collection time = 240 s; loaded mortar = 2,220 g.

3.2 Flow response

Table 2 shows an increase in D2 from 150.0 mm for C0 to 161.0 mm for C1. The 11.0 mm difference in control was greater than the following 4.0 mm difference between C1 and the highest LBG entries. Within the gum series, D2 increased from 161.2 mm at 0.1% to 165.0 mm at 0.6 and 0.8%. LBG0.3 recorded 162.0 mm, only 1.0 mm above C1.

The major differentiation in the flow record was thus linked with the chemical-admixture control. The minor variations among gum doses are incapable of creating a significant enhancement of flowability in the absence of measurement error and replication. Such a visual examination did not measure stability: no observed bleeding or segregation was noted during the flow evaluation.

Figure 2 is a comparison of D2 to the calculated discharge rate. Beyond 0.3% gum, D2 went on rising whereas the extrusion rate fell. Another difference between the two tests was based on age and imposition of deformation. The entry in the flow-table was therefore inadequate to determine the mixture which had the greatest discharge under the conditions that were adopted. This lends credence to the application of complementary tests in extrusion based development of mixtures [2]–[4].

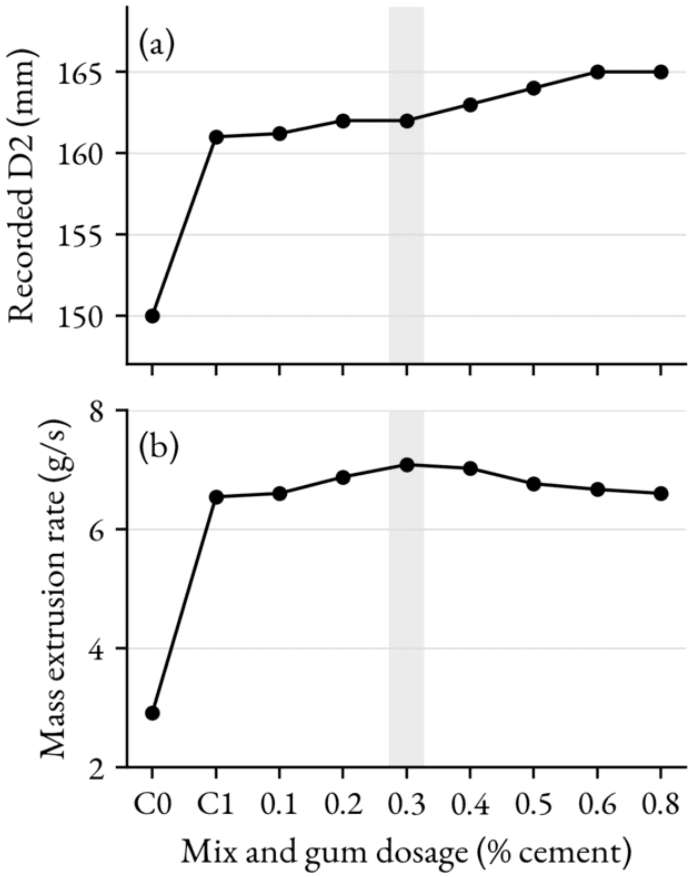


Figure 2. (a) Recorded flow-table D2 and (b) average mass extrusion rate. Both tests excluded accelerator; lines connect tested formulations, and no fitted model or uncertainty estimate is implied.

3.3 Manual extrusion performance

C0 discharged 700 g in 240 s, giving 2.92 g/s and 31.53% of the loaded mass. C1 discharged 1,570 g, giving 6.54 g/s and 70.72%. The increase in rate from C0 to C1 was 124.29%. There was no accelerator and this difference cannot be because of acceleration upon discharge.

Rate rose to 6.60, 6.88 and 7.08 g/s at 0.1, 0.2 and 0.3% gum, then declined through 7.02, 6.76 and 6.67 to 6.60 g/s at 0.8%. LBG0.3 discharged 1,700 g, or 76.58% of the charge. Its rate exceeded C1 by 8.28%, but exceeded LBG0.4 by only 0.0625 g/s, or 0.89%. Replication is required to confirm whether the small neighbour difference can be reproduced.

There were no observed blockages, tearing or segregation in the available extrusion observations. Mass accumulated does not automatically create continuous flow and mortar that is left after 240 s may not necessarily be treated as waste or obstruction.

3.4 Three-day compressive strength

Table 3 gives 20.53 MPa for C0 and 28.83 MPa for C1; the latter is 40.45% (unrounded) higher. A large part of the strength difference between plain mortar was thus already in the chemical-admixture control. This analogy is not in isolation of an accelerator effect since the treatment was a combination of chemical admixtures.

Table 3. Three-day failure loads, nominal-area strengths and relative differences from C1.

Mix	P (kN)	f_c (MPa)	ΔC1 (%)
C0	102.6	20.53	-28.80
C1	144.1	28.83	+0.00
LBG0.1	143.1	28.63	-0.69
LBG0.2	146.3	29.27	+1.53
LBG0.3	151.2	30.25	+4.93
LBG0.4	144.5	28.91	+0.28
LBG0.5	135.3	27.07	-6.11
LBG0.6	130.0	26.01	-9.78
LBG0.8	123.9	24.79	-14.02

C1 and LBG cube batches contained accelerator; C0 did not. A = 4,998.49 mm^2.

LBG0.3 recorded the highest load of 151.2 kN of 30.25 MPa which was recorded in the gum series. Its strength was 4.93% above C1 and 47.37% above C0. LBG0.2 and LBG0.4 were also above C1, albeit lower than LBG0.3. The strength declined with all dosages tested in excess of 0.3%, and to 24.79 MPa at 0.8%. The latter remained above C0 but was 14.02% below C1 and 18.06% below LBG0.3.

The results are indicative of a middle dosage in this series of formulations. They fail to develop a microstructural or hydration mechanism. It could be due to differences in hydration, air content, homogeneity or compaction, but calorimetry, porosity, microscopy and fresh-density could not be done to differentiate between them. An assertion of pore filling or matrix densification would thus be in excess of the evidence.

3.5 Joint formulation assessment

Figure 3 shows that LBG0.3 is the joint numerical maximum with extrusion index of 108.28 and strength index of 104.93. Its measured filament size measured 31.0 x 9.8 mm after five minutes. Based on this, 0.3% gum which will correspond to 3.0 g/kg cement or 6.66 g in the batch being listed is the most promising formulation for further development.

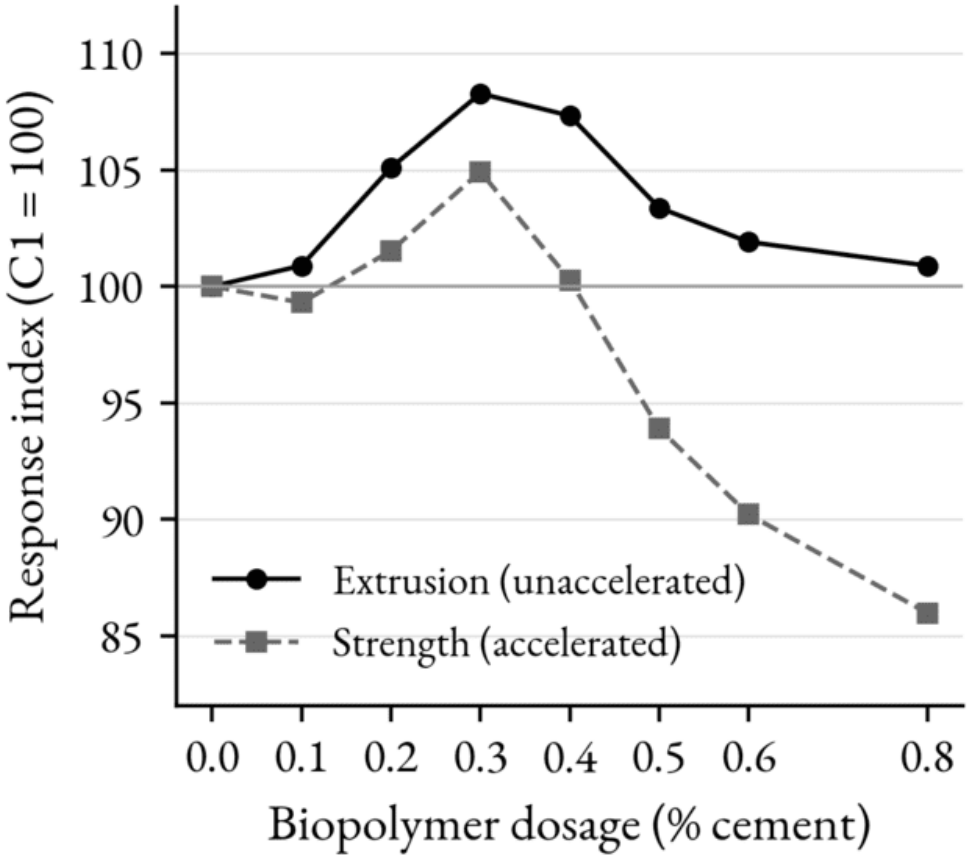


Figure 3. Extrusion and strength indices relative to C1. The responses describe separate unaccelerated and accelerated tests; connecting lines indicate tested dosage order, not a continuous optimum.

Such a dose does not imply that this was the dose that maximised all responses. The highest D2 values were at 0.6 and 0.8% and there were other gum mixtures which were closer to the nominal filament width. LBG0.4 was near the peak discharge, whereas LBG0.2 had more discharge and strength than C1. The range of 0.2–0.4% is thus a convenient range to revisit as opposed to the evidence of a well-defined continuous optimum.

The divergence of the flow and extrusion rankings is also practically applicable. When choosing the highest D2, the high dosage and three-day strength and discharge would be preferred compared to LBG0.3. Selecting candidates is useful with the resultant retention of separate responses as opposed to observing them as a single arbitrary score. However, extrusion and strength indices should not be understood as the measurements of a single state of a material: the acceleration history was not the same during the two tests.

The recommendation is specific to the recorded cement, prepared sand, nominal water basis and chemical treatment. It is not a transferable construction-printing specification, but rather a candidate dosage to be validated on a controlled basis.

3.6 Filament geometry and setting

C0 and C1 filaments measured 30.0 × 9.9 mm and 32.0 × 10.0 mm, respectively, at five minutes. The LBG filaments were either 30.0 or 31.0 mm wide and all had a recorded height of 9.8 mm. Width was 31.0 mm at 0.1, 0.2, 0.3 and 0.6% gum, and 30.0 mm at 0.4, 0.5 and 0.8%. Relative to the nominal targets, the LBG width deviations were 0 or +3.33%, with a height deviation of −2.00%.

These observations are an account of closeness to nominal geometry at the time of measurements. The difference in the height of 0.2 mm is not a measured settlement since the initial height deposited was not measured. The unproved association between dimension readings and deposition path also leaves the attribution to quicker or slower material on an acceleration or unacceleration level. The dimensions give supporting observations, but cannot give a buildability limit or accept an acceptance threshold carried over to a different apparatus. Figure 4 shows the stratified nature of deposition without an extra measured buildability result.



Figure 4. Illustration of layered cement-mortar deposition. The image is not used to establish specimen dimensions, layer stability or a formulation-specific result.

The Vicat initial and final times were recorded as 30 and 240 min of C0, and 2 and 3.5 min of C1 and all LBG formulations. Thus, there was a big control to treatment difference and there was no solved dosage depended difference between accelerated formulations. The same entries could not demonstrate that gum was not hydrating; there were no observation intervals or histories of penetration available.

Accelerator was added to the water in the Vicat preparation prior to being introduced into cement, but with piping-bag deposition a part of a previously mixed mortar was brought into action. These are alternative histories. Setting-on-demand research elucidates the viable supply condition and the triggered material around deposition [8]. In the current experiment, the accelerated Vicat times would not determine the usable open time of the unaccelerated extrusion mortar, and would not determine the rate at which a deposited layer would acquire load-bearing resistance.

3.7 Interpretation against prior work

Extrusion involves both deformation of the material and interaction with the apparatus. Perrot et al. [9] address equipment effects, wall contact and potential liquid movement in cement-based extrusion. These are some of the reasons why manual mass rate cannot be directly converted into viscosity or yield stress. In this case, the variation in the flow observations that are made after five minutes and twenty minutes of extrusion also restricts a direct comparison of rheology.

Preparation and compatibility can be investigated using studies of biopolymers [6] and biopolymer-superplasticiser mixtures [7]. The current sand-based mortar, though, has not been tried in the mechanisms that were being examined in those paste experiments. The numerical peak of 0.3% is not indicative of a specific interaction between the polymer and cement. Independent variation of components and repeated treatments would be necessary in the chemical synergy.

Three-day strength deals with a later phase than the deposition stability. The build-up in structures is time-dependent and load sequence-dependent [10], and the structures printed may fail by plastic collapse or by elastic buckling [11]. The Hardened-property [12] and interface studies [13] also show that in the case of layer support, interlayer bond and directional strength, the tests must be in excess of the current cube measurements.

3.8 Limitations and validation priorities

The absence of replicate data does not allow uncertainty estimates, particularly of the slight difference between 0.3 and 0.4% extrusion and the 4.93% strength difference between LBG0.3 and C1. Both differences are not considered as statistically significant.

Driving action was not automated or checked and the barrel geometry and outlet length was not completely documented. The test then filters out discharge under the installed arrangement as opposed to pumpability within a delivery line. A repeat programme must be able to measure applied pressure or force, to record mass continuously and record interruptions with a constant test age and geometry of the apparatus.

Reproducibility involves full material grades, gum preparation, mixing, moisture and accelerator-scaling records, cube formation, curing, dimensions and loading rate. The filament activation path and Vicat composition and endpoints have to be settled as well before these observations can be used to justify stronger process claims.

The comparison of C0, C1 and 0.2–0.4% gum should be performed with independent batches, repeated-age extrusion, deposition of multiple layers and interlayer tests should be conducted. Separate measurements are needed of seven- and twenty-eight-day strengths. No performance or environmental gain on a construction-scale is implied here.

4 Conclusions

A cement mortar that uses a natural biopolymer was created and tested in extrusion-based 3D printing. Nine formulations were filtered with a nominal added-water-cement ratio of 0.32, and

cement:sand ratio of about 1:1.25. The conclusions below refer to the materials and laboratory procedures tested.

1. The chemical-admixture control raised the unaccelerated rate of mass extrusion from 2.92 to 6.54 g/s in comparison to plain mortar. Nominal area cube strength (3 days) rose from 20.53 to 28.83 MPa in the separate activated compression route.
2. The highest recorded rate of extrusion, 7.08 g/s, and strength, 30.25 MPa were recorded with the 0.3% Ceratonia siliqua gum dosage. These were 8.28 and 4.93% above the corresponding admixture-control values, respectively.
3. Increasing dose of gum caused observed flow-table D2 to rise but decreased discharge rate and strength. Flow-table ranking in itself was thus not able to identify the formulation with the most combined response.
4. Filament measurements characterized geometry at five minutes, and did not measure settlement or multilayer stability. Accelerated Vicat times recorded were the same with the modified formulations and failed to set extrusion open time.
5. The most promising candidate tested towards further development towards extrusion-based 3D printing is the 0.3% dosage, and the 0.2–0.4% range is appropriate to be tested further with focus on repeat testing. The lack of replicate data, hand actuation and incomplete specimen records, forbid an approved optimum or a general printability claim.

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