



Bigel inks for 3D food printing: rheological and extrusion behavior

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3D printing technology presents an enormous potential to be applied in the food field since it enables personalized and intricately shaped designs, offers personalized nutrition, simplifies the supply chain, and enables the use of non-conventional food materials, among others. The extrusion method is the most favored technique in 3D food printing, as it allows food, in paste or liquid form, to be shaped. However, precise ink properties are needed to meet printing process requirements and product expectations. In this sense, bigels (BG) are semisolid gels constituting an oleogel (OG) and a hydrogel (HG) typically formed by mechanical mixing at certain temperatures and conditions of gel setting. Compared with OG and HG, BGs possess the advantages of both phases, allowing them to transport hydrophilic and hydrophobic nutritional compounds. Furthermore, their physicochemical properties can be manipulated by adjusting the composition and amount of each phase. This allows the tailoring of their rheological properties, making BGs potentially suitable materials for 3D printing. Therefore, the objective of this work was to prepare different mixtures of BG in order to test their rheological properties and their potential as 3D printing materials. Regarding the HG phase, three different hydrogelators in different proportions were tested: Xanthan Gum (XG), Guar Gum (GG), and Carrageenan (CR). The ability of HGs to incorporate 10% of vegetable (beetroot) dried powder (BP) was also tested. Thus, HGs were formulated with 1, 5 and 10% of XG, GG and 1% of CR, either with or without 10% of BP. The OG phase was prepared using high oleic sunflower oil and 10% of monoglycerides. BGs were produced by mixing different HG and OG ratios (HG:OG of 80:20, 50:50 and 20:80) at 80°C, followed by rapid cooling. The rheological properties, elastic (G') and viscous (G'') modulus, of HGs, OG and BGs were analyzed using frequency and strain sweep tests. In addition, to evaluate the printability of BGs, a forward extrusion test was performed using a texture analyzer. The extrusion cell consisted of a sample container with a 3D printer nozzle (2mm of diameter) on its base and a piston disc. It was found that the optimum hydrogelator concentration was 5% for XG and GG, and the strength of HGs increased with the incorporation of BP. The G' values for the HGs containing BP were 7.3E3, 6.3E3, and 1.2E3 Pa for GG, CR, and XG, respectively. Successful bigels were obtained using the 80:20 ratio, which exhibited the same G' trend as their corresponding HGs (GG>CR>XG). The results also showed that the BGs were stronger as the ratio of OG increased. Regarding the forward extrusion test, the mean force required for extrusion of 80:20 BGs was 125.6, 110.7, and 32.2 N for XG, GG, and CR, respectively; being BGs from XG and GG the ones with the best self-supporting ability. Overall, the results show that BGs presented suitable properties to be used as inks for extrusion 3D food printing.



Palabras Clave: Additive manufacturing, rheology, printability, printing materials, extrusion.



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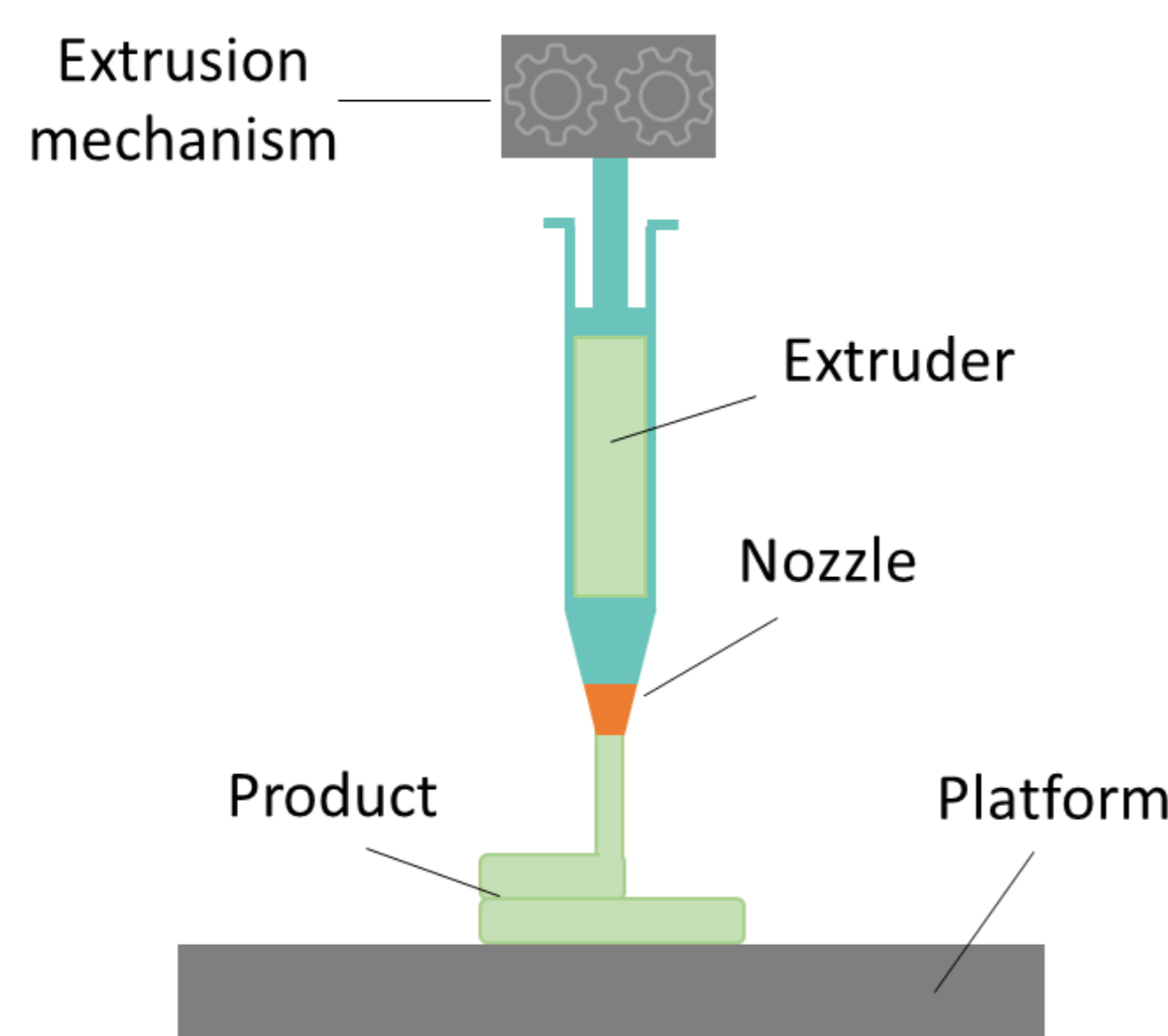
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ABSTRACT

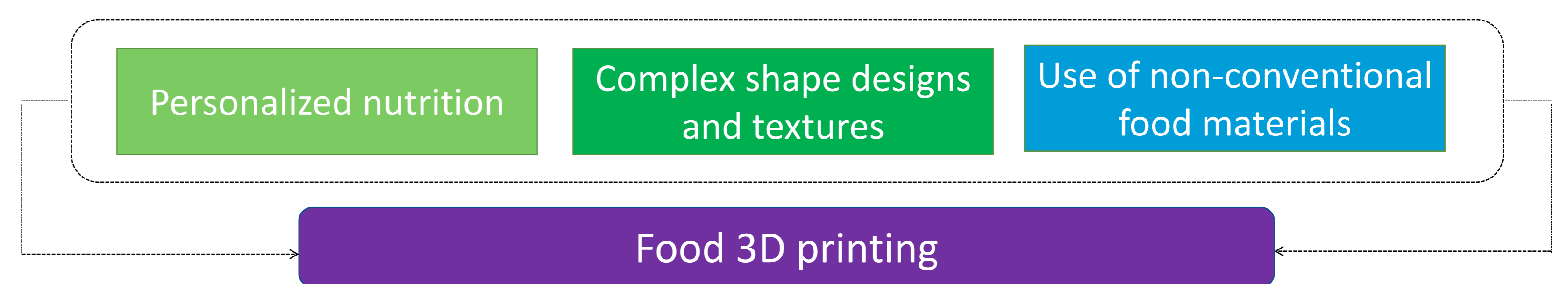
In this work, bigels (BGs) were prepared using three types of hydrogelators in the hydrogel and different oleogel/hydrogel ratios. The feasibility of these gels for their use as printing materials for extrusion 3D printing was tested. BGs were evaluated in terms of their rheological properties (elastic and viscous modulus) and their ability to be extruded through nozzles of different diameters (extrusion test).

Extrusion 3D printing (EXT)

- Layer-by-layer deposition of semi-solid forms (gels or pastes) of suitable viscosity.
- Extrusion through a syringe with/without temperature control.
- It may or may not require a post-processing stage.



INTRODUCTION



BIGELS (BGs): Semisolid gels of oleogel (OG) + hydrogel (HG)

- Allows the transport of both, **hydrophilic** and **hydrophobic** nutritional compounds.
- Physicochemical properties can be manipulated by adjusting the **composition** and **amount** of each phase (OG and HG).
- Tailoring** of rheological properties → **BGs potentially suitable materials for 3D printing.**

OBJECTIVE: to prepare different BGs containing vegetable powder in order to test their rheological properties and their potential as 3D printing materials.

MATERIALS AND METHODS

BGs preparation

COMPOSITION

- HG of Xanthan Gum (XG):** 1, 5 or 10 %wt/wt XG + 0 or 10 %wt/wt beetroot powder (R).
- HG of Guar gum (GG):** 1, 5 or 10 %wt/wt XG + 0 or 10 %wt/wt beetroot powder (R).
- HG of Carrageenan (CR):** 1 %wt/wt + 0 or 10 %wt/wt beetroot powder (R).
- OG:** high-oleic sunflower oil + 10 %wt/wt monoglycerides (MG).
- Bigels:** 80:20, 70:30, 50:50 and 20:80 ratio of HG:OG.

PREPARATION

Oleogel Hydrogel



- MIXING** T = 80°C, 15000 rpm, 5 min
- COOLING** ice bath
- STORAGE** @ 4°C for 48h

Characterization techniques

RHEOLOGY

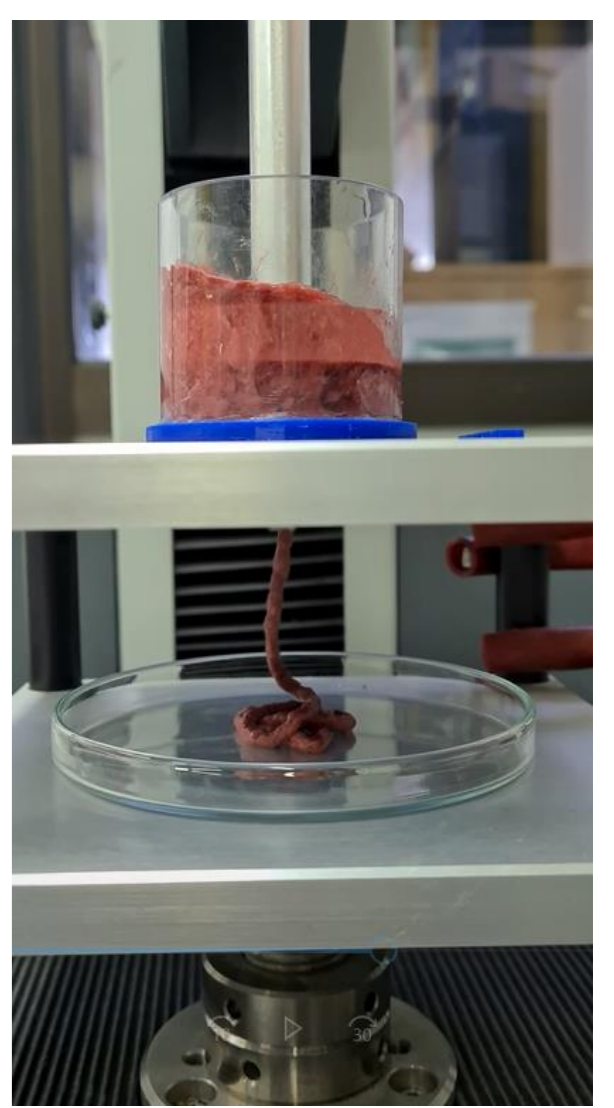
- Frequency and strain sweep tests.
- Elastic (G') and viscous (G'') modules.

FORWARD EXTRUSION TEST

- Texture analyzer with a special cell.
- Nozzle diameter: 1,2 and 2 mm.

IMAGE ANALYSIS

- Pictures for assaying BG homogeneity.
- Microscope for microstructure.



RESULTS AND DISCUSSIONS

RHEOLOGY

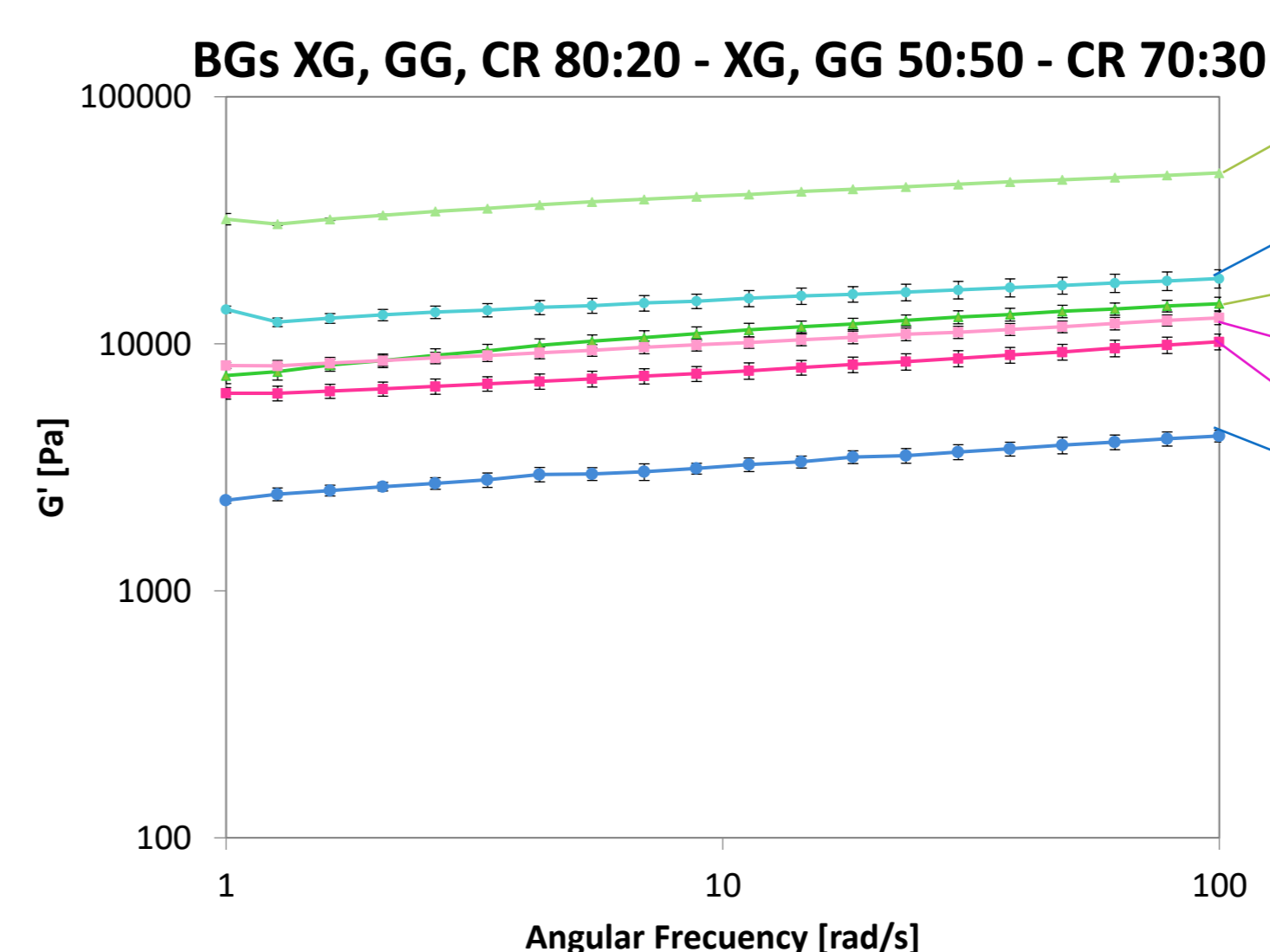
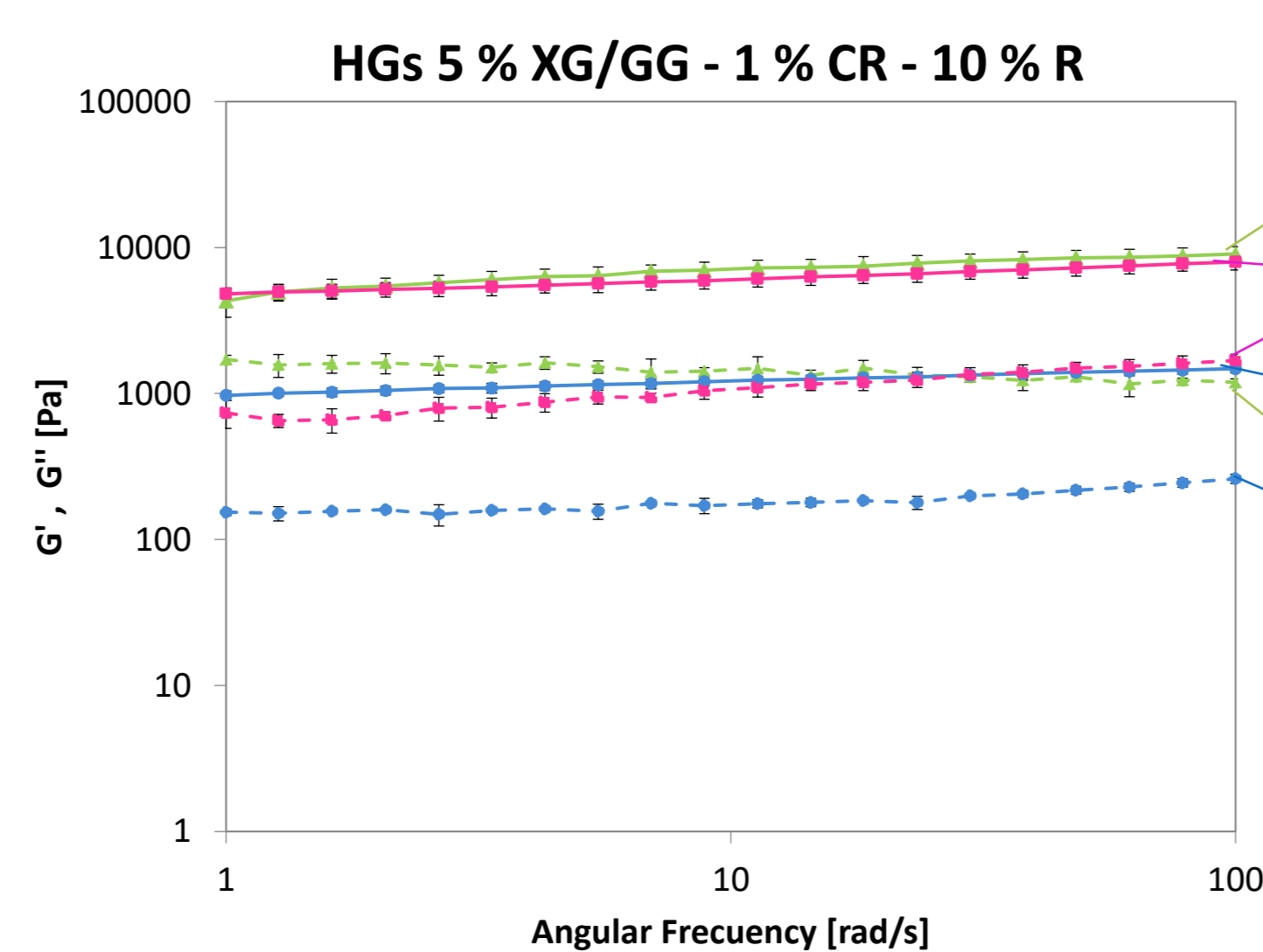
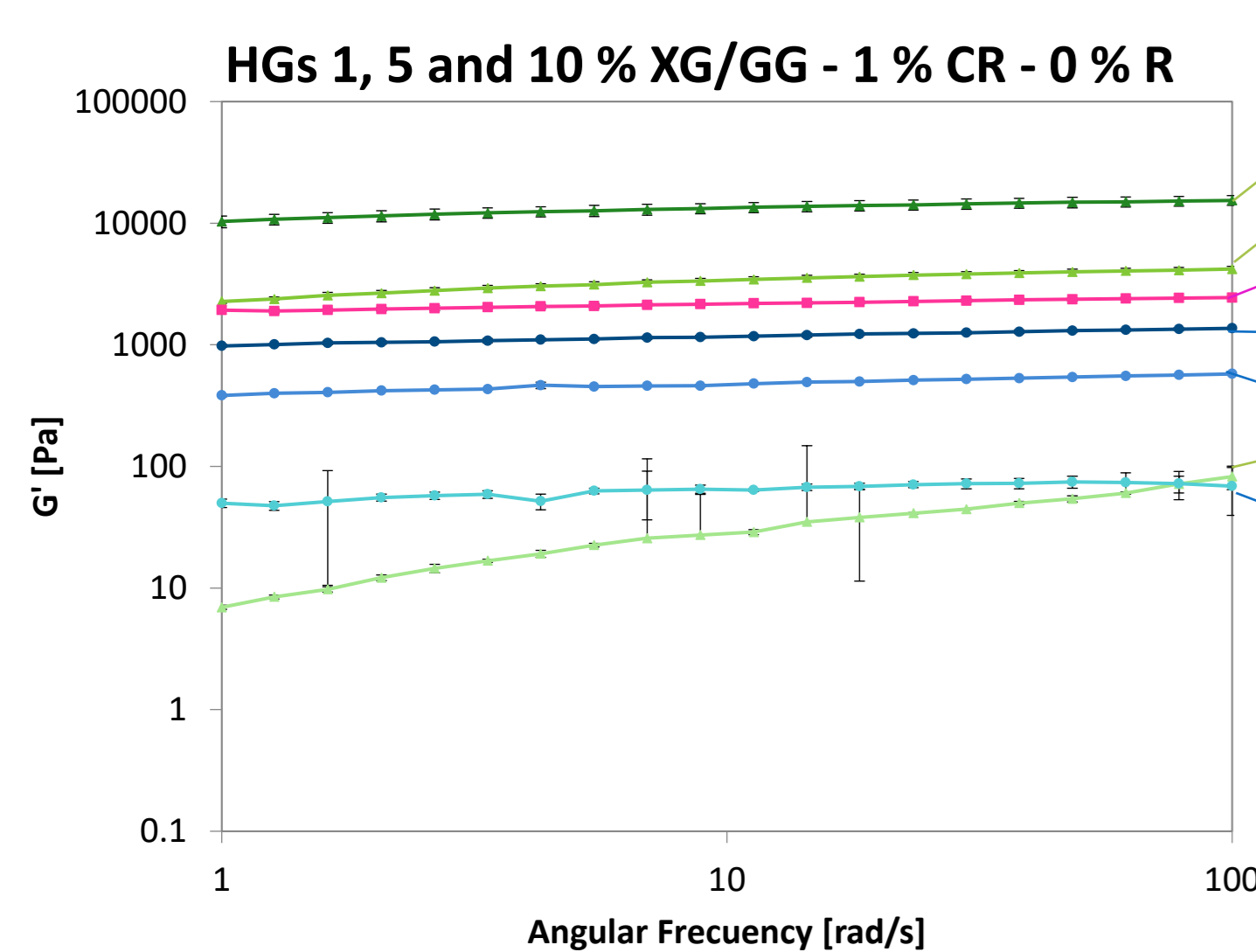
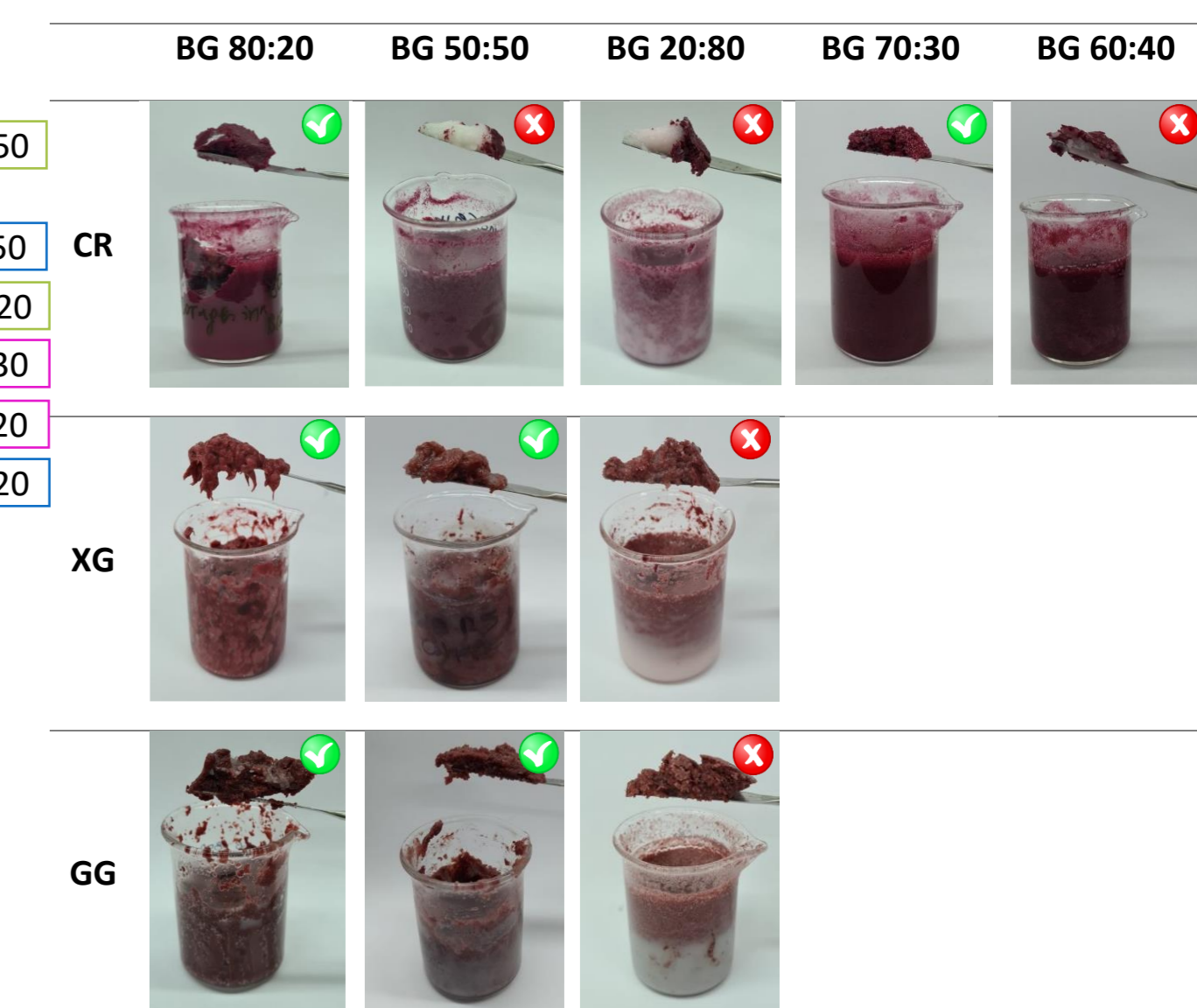
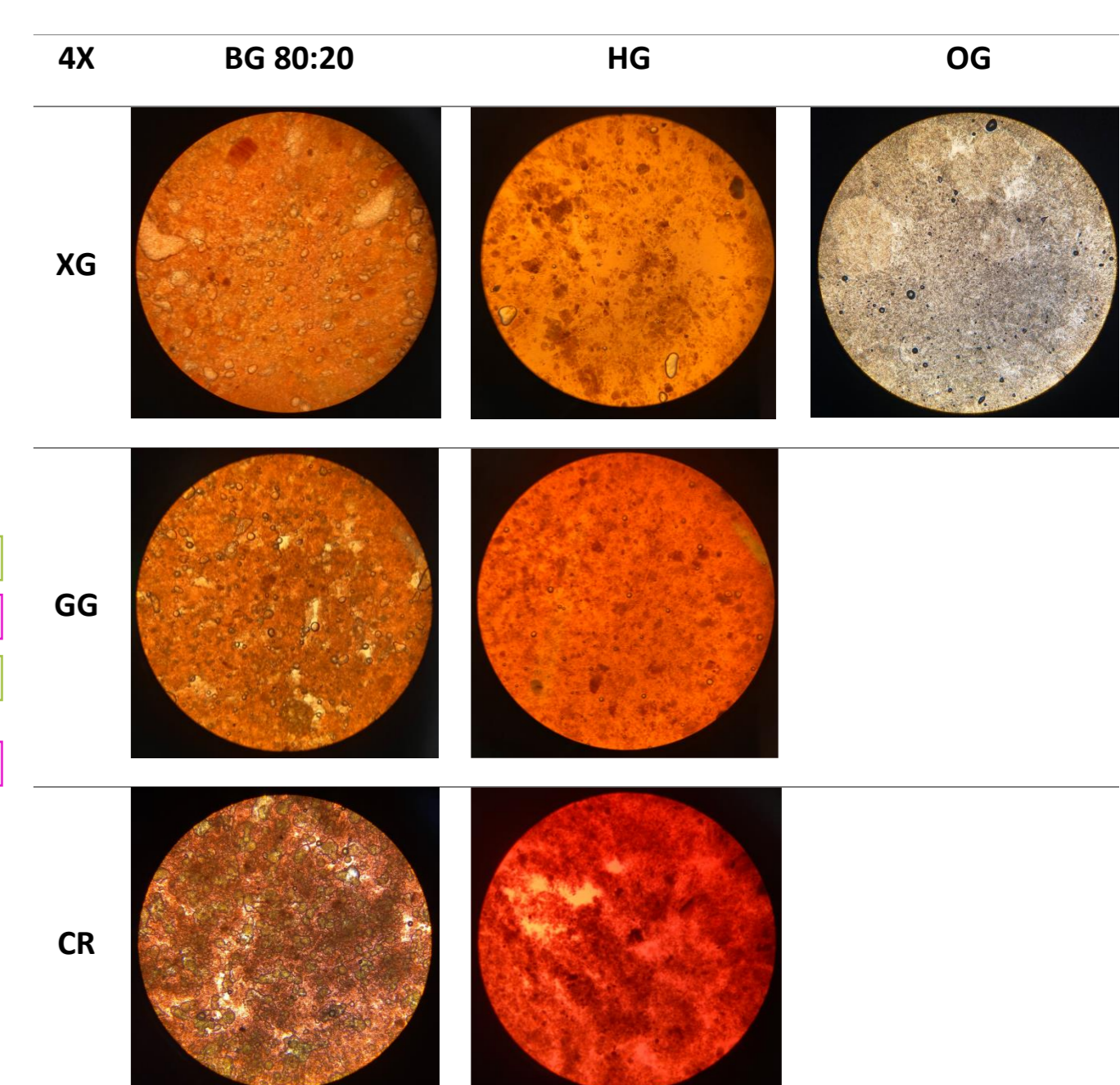
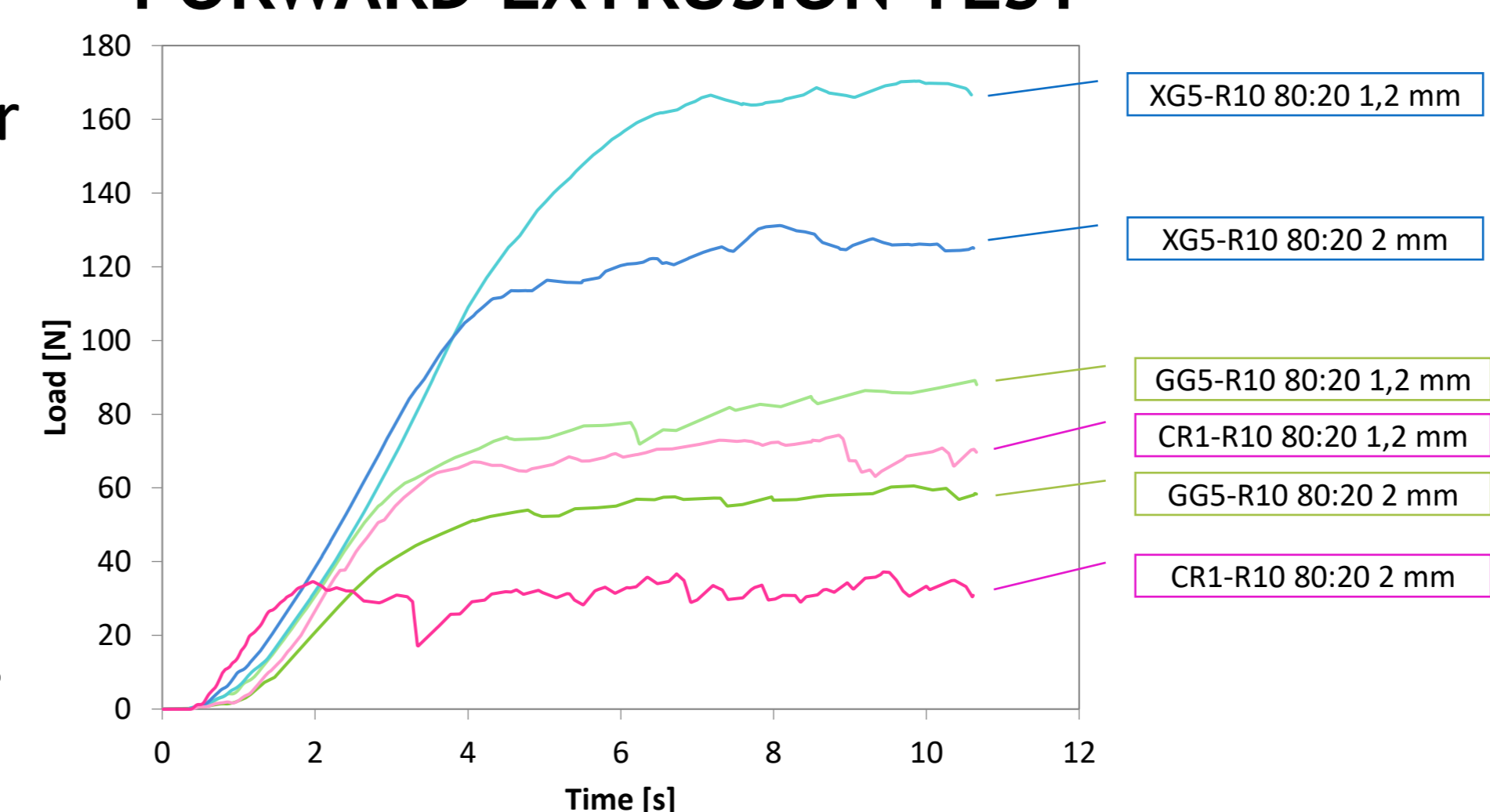


IMAGE ANALYSIS



- The optimum hydrogelator concentration was 5 % for XG and GG.
- The strength of HGs increased with the incorporation of R.
- The G' trend for the HGs containing R were $GG > CR > XG$.
- Successful BGs were obtained using the 80:20 ratio for all hydrogelators, 50:50 for XG and GG and 70:30 for CR.
- BGs exhibited the same G' trend as their corresponding HGs ($GG > CR > XG$).
- BGs were stronger as the ratio of OG increased.
- The mean force for extrusion of 80:20 BGs increased with decreasing the nozzle diameter.
- BGs of XG presented the highest mean force of extrusion, followed by GG and CR.
- Microscope images of BGs show combination of OG and HG individual structures.

FORWARD EXTRUSION TEST



CONCLUSIONS

- We find the proportion of HG:OG that resulted in homogeneous bigels.
- All homogeneous bigels presented potential to be used as inks for printing.
- Bigels from GG and CR (20:80) were stronger than the ones formulated with XG. The higher the OG fraction, the strongest the bigel.
- When testing the ability to be extruded out of a nozzle, bigels from XG presented the highest resistance, probably due to its rubbery nature against the brittle-type behavior of GG and CR bigels.