The authors would like to express gratitude to the reviewers of Polymers for their review of our submission entitled "Controlling morphology and physio-chemical properties of stimulus-responsive polyurethane foams by altering chemical blowing agent content". We understand and appreciate the time spent on the review and sincerely appreciate the thoughtful comments and the constructive suggestions for improvement.

We have revised the manuscript as described in the responses (in red) below.

Reviewer 1

The manuscript by Hasan et al. reports on a study investigating the influence of the concentration of the chemical blowing agent water upon the physical, physio-chemical and functional properties of a polyurethane foam. It is a very nice fundamental work.

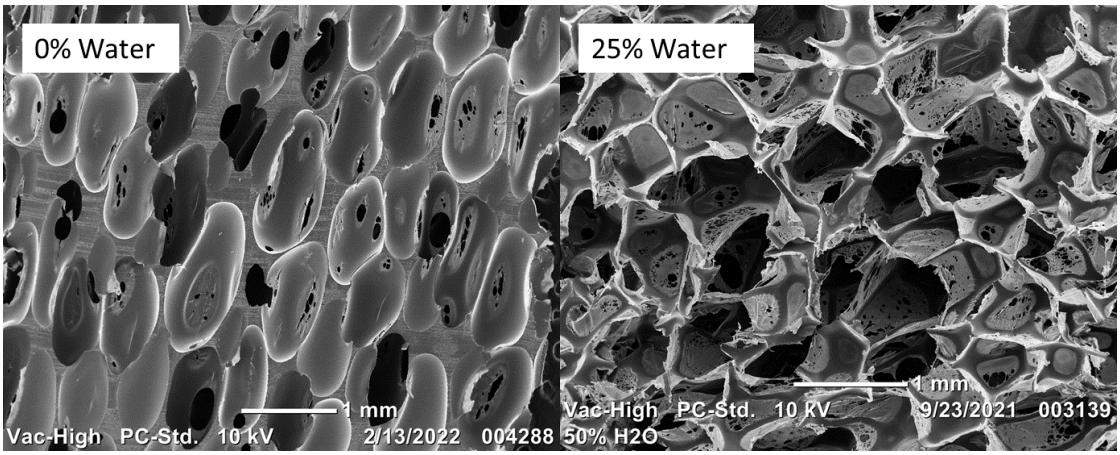
Here are my comments as suggestions for possible improvements:

- Abstract: Better use another expression for "aqueous actuation".

  An alternate word for "aqueous" was used on line 13.
- In terms of the foam synthesis, could you please specify the water content, above which water will no longer serve as a reactant?

  This is a great question. Water molecules will continuously react with free isocyanate molecules until they are fully consumed or there are no more isocyanate groups available for reaction. Theoretically, we could replace all hydroxyl (OH) groups in the reaction vessel with water and the isocyanate groups will continue to react until they are fully consumed. For foaming purposes, however, it is important to balance the concentration of water compared to other hydroxyl groups (TEA, HPED) to facilitate gas formation alongside network growth. While there is no specific water concentration above which the reaction will stop to move forward, we can hypothesize that above 50% water concentration, the stoichiometry will be unbalanced, and a viable polyurethane foam will not form.
- Are there any hints in how far water content influences the open cell content?

  Thank you for allowing us to clarify. Increased water content results in higher gas formation due to the water molecules reacting with free isocyanate groups. Increased gas formation will result in a more open cell foam due to membranes rupture as the gas escapes. At 25% water, the foam membranes were highly populated with pinholes compared to the foam membranes at 0% water. Please refer to the SEM images below for qualitative evidence of increased pinhole formation in 25% water foams compared to 0% water foams.



- Some recent work on shape memory polyurethane foams is not mentioned (exp Polym Lett 6 (2012) 63-69, Polymer Chemistry 8 (2017) 5039, Polymers 12 (2020) art. no. 1914.).

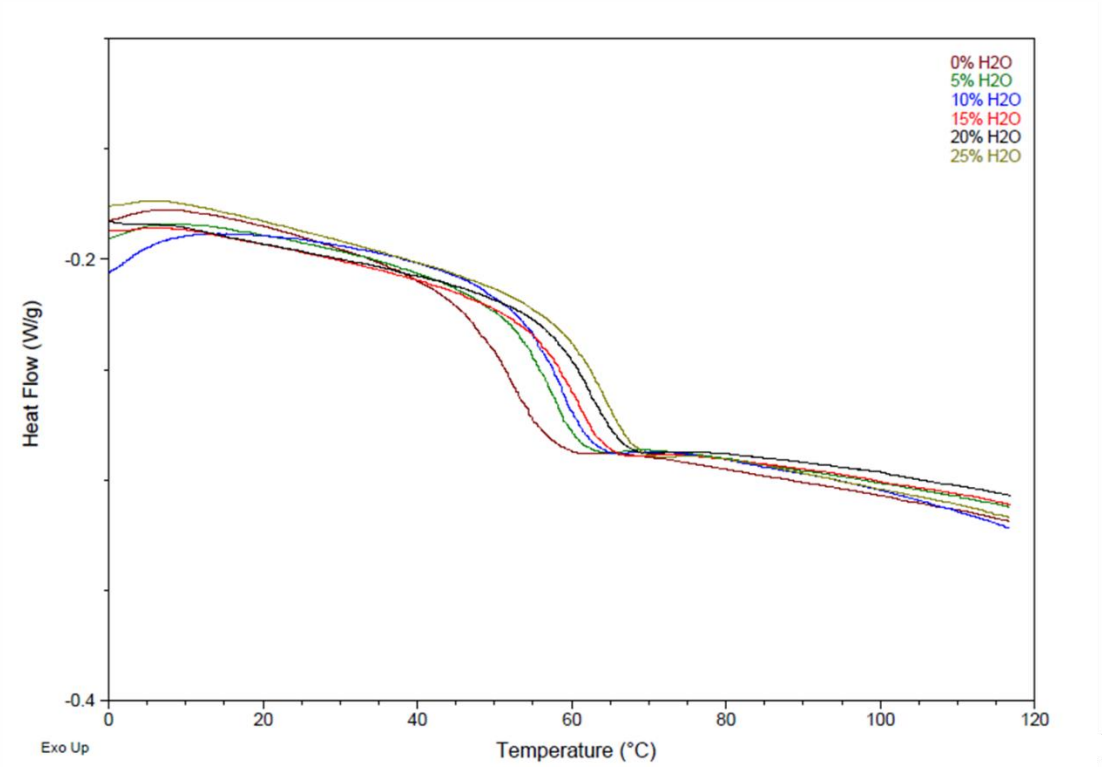
Thank you for providing these references to strengthen our introduction section. We have included these papers in our reference list.

Polymers 12 (2020) art. no. 1914 has been included as reference #5

Polym Lett 6 (2012) 63-69 has been included as reference #20

- The different thermal stabilities of the SMP foams shown in Figure 3 are not comprehensible. Do they suggest changes in the ratio of hard to soft segments? Please comment.

The thermal degradation profiles of SMP foams, in Figure 3, are overlapping to indicate that minimal to no changes in thermal degradation temperature was observed when modifying the water content. These SMP foams are amorphous, as confirmed by the lack of melting temperature in the DSC profiles (see figure below), therefore there are no hard or soft segments within the polymer network. Based on the thermal degradation profiles, changes in urea concentrations within the polymer network were not significant enough to shift the thermal degradation temperature of the various SMP foams. Furthermore, based on a comment by Reviewer 2, we have separated the TGA curves to aid interpretation.



Reviewer 2

1. In the literature analysis, the authors presented a modest study of the works on changes in the structure and properties of viscoelastic foams (or shape memory foams) caused by the difference in the amount of the blowing agent. Materials recently presented two papers on viscoelastic foams with different cell structures:

Okrasa, M.; Leszczyńska, M.; Sałasińska, K.; Szczepkowski, L.; Kozikowski, P.; Majchrzycka, K.; Ryszkowska, J. Viscoelastic Polyurethane Foams for Use in Seals of Respiratory Protective Devices. Materials 2021, 14, 1600. https://doi.org/10.3390/ma14071600

Grzęda, D.; Węgrzyk, G.; Leszczyńska, M.; Szczepkowski, L.; Gloc, M.; Ryszkowska, J. Viscoelastic Polyurethane Foams for Use as Auxiliary Materials in Orthopedics. *Materials* 2022, *15*, 133. https://doi.org/10.3390/ma15010133

1. Moreover, many works on SMP made of viscoelastic polyurethane foams were presented, which can be used in the preparation of the introduction.
   Thank you for providing us with the abovementioned articles. We believe that addition of these works into the introduction section will help solidify our proposal for tuning the pore morphology of our SMP systems by modifying water content. We have included the references in our introduction section (lines 85-89), as references 23 and 24.
2. In the quantitative analysis of the foam images, too few points were used for analysis. Such an analysis should perform for a minimum of one hundred points to be reliable.
   Thank you for allowing us to improve upon our methods. The methods for pore size evaluation were updated on page 3. We re-evaluated the original images and measured 25 pores for each of the 4 samples that were cut from a bulk foam. This was done for each foam formulation, thereby yielding 100 axial and 100 transverse pore size measurements for each foam. The results in Table 2 were updated to reflect the remeasured pore sizes (page 6). With increased data points, we still observe a trend of increasing pore sizes with increasing water content.
3. There are no DSC thermograms. Without them, one can have doubts about the interpretation presented by the Authors. The introduction of more urea bonds, caused by the introduction of more water to the reaction mixture increases the stiffness of the macromolecules of the foams, causing an increase in Tg. The hydrogen bonding between urea groups is stronger compared to that between urethane groups; therefore, more energy is required to disrupt this intermolecular force in polymers with a higher urea concentration.
   We have included DSC thermograms overlaid in Figure 3, on page 9 line 275. Overall, no significant changes were observed in the thermograms for the foams with increasing urea concentration. All foam formulations had an onset thermal degradation temperature of approximately 250°C.
4. The TGA analysis results should supplement DTG curves and their interpretation.
   Figure 4 (previously Figure 3) has been updated to include the DTG curves along with the TGA curves. Additionally, we separated each of the thermograms so that they are easy to interpret.
5. IMPEDE and IMPEDE-FX embolisation plugs use the self-expanding properties of SMP foam to seal diseased blood vessels. Therefore, to describe their properties, it is worth using the designation of properties during compression and permanent deformations after reduction.
   We believe the reviewer is asking for shape fixity and shape recovery properties of the SMP foams upon being compressed into their crimped shape. Table 3 provides volume recovery (%) of the SMP foams post-actuation. Most formulations experienced complete recovery (>100%), indicating that there was minimal to no permanent deformation after programming. Furthermore, the foams cylinders were crimped to a diameter of 1 – 2 mm, depending on the strut thickness. All foams maintained a small, crimped diameter, as shown in Figure 6, at t=0 The t=0 crimped foam values were collected 24 hours after the foams were conditioned into a crimped state. This indicates that the SMP foams also have a high shape fixity because the foam cylinders did not passively re-expand under ambient conditions. Exposure to a 37°C aqueous environment triggered the SMP foams to actuate from their compressed to recovered state.