

Hydrogenation-Controlled Mechanical Properties in Graphene Helicoids: Exceptionally Distribution-Dependent Behavior

Ali Sharifian,^a Hamid Hassanzadeh Afrouzi,^{*b} Abouzar Moshfegh,^{c,d} Ashkan Javadzadegan,^{c,d} Mostafa Baghani^a and Majid Baniassadi^{*a,e}

^a *School of Mechanical Engineering, College of Engineering, University of Tehran, P.O. Box 11155-4563, Tehran 1417466191, Iran.*

^b *Faculty of Mechanical Engineering, Babol Noshirvani University of Technology, P.O. Box 484, Babol, Iran. E-mail: Hamidhasanzade@yahoo.com.*

^c *Faculty of Medicine and Health Sciences, Macquarie University, Sydney, NSW 2109, Australia.*

^d *ANZAC Research Institute, The University of Sydney, Sydney, NSW 2139, Australia*

^e *University of Strasbourg ICube/CNRS, 2 Rue Boussingault, 6700 Strasbourg, France. E-mail: m.baniassadi@ut.ac.ir*

S1. Comprehensive study on potential and non-dimensional RMSD diagrams

There also exists a comprehensive study on potential and non-dimensional RMSD diagrams that represents the final equilibrium state. As shown in Figure S 1, all samples reached their equilibrium level of up to 100 times. Should the graphene helicoids have a more symmetric structure, it will reach sooner reach to the equilibrium state like the pristine or the complete functionalized structures. It can also be seen from the diagrams that the samples which have been only functionalized in their inner and outer parts do have higher equilibrium fluctuations. In addition, the bond configuration for samples 3 and 4 in the functionalization percentages of 30 and 50 for the C-C and C-H bonds is shown in Figure S 2, which illustrates similarity of the C-H bond configuration to the samples. Moreover, C-C bonds also have a general similarity, but for each configuration, it is unique based on the hybridization of carbon atoms.

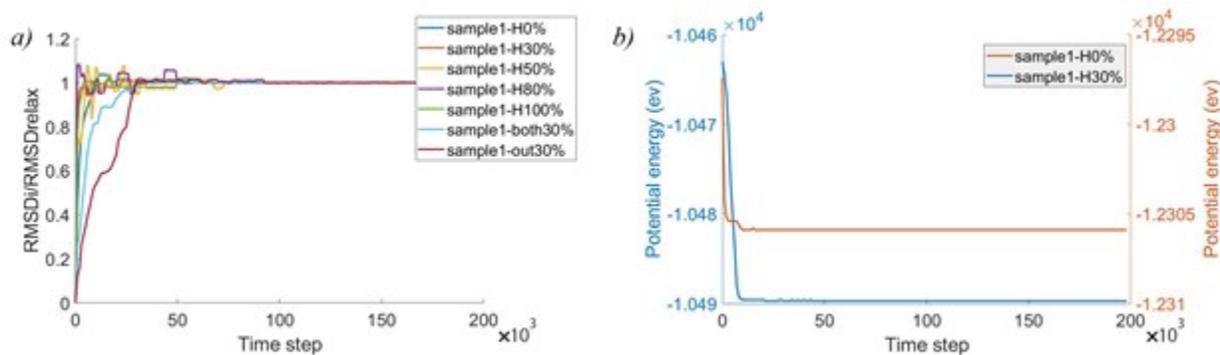


Figure S 1. a) Dimensionless RMSD diagram versus time step for sample 1 at percentages of 0, 30, 50, 80 and 100, as well as 30 percent functionalized inner and outer parts. b) Potential energy diagram versus time step for Sample 1 in functionalized percentages of 0 and 30.

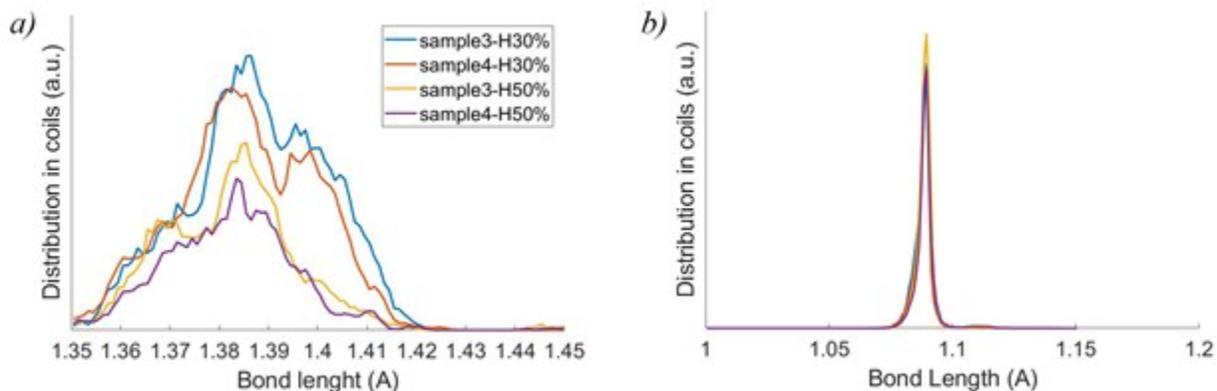


Figure S 2. Bond configuration for a) C-C bond and b) C-H bond for samples 3 and 4 in functionalized percentages of 30 and 50.

S2. The tensile behavior in pristine and 30% hydrogenated sample 2.

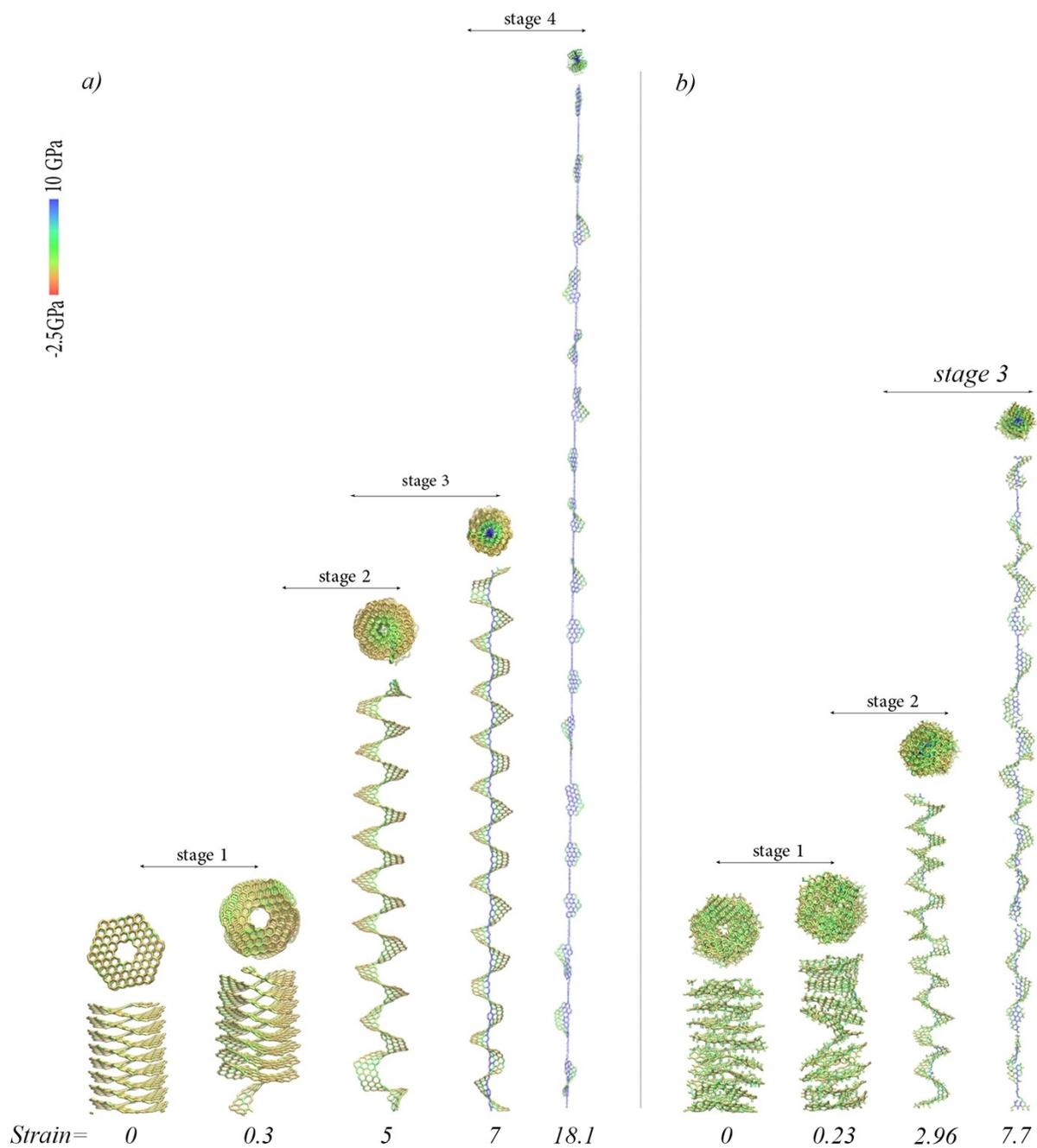


Figure S 3. Comparison of the tensile behavior in: a) pristine GH of sample 1. b) hydrogenated GH for samples 2 at percentages of 30. The color of atoms is related to stress in the tension direction.

S3. The tensile behavior in pristine and 30% hydrogenated sample 3.

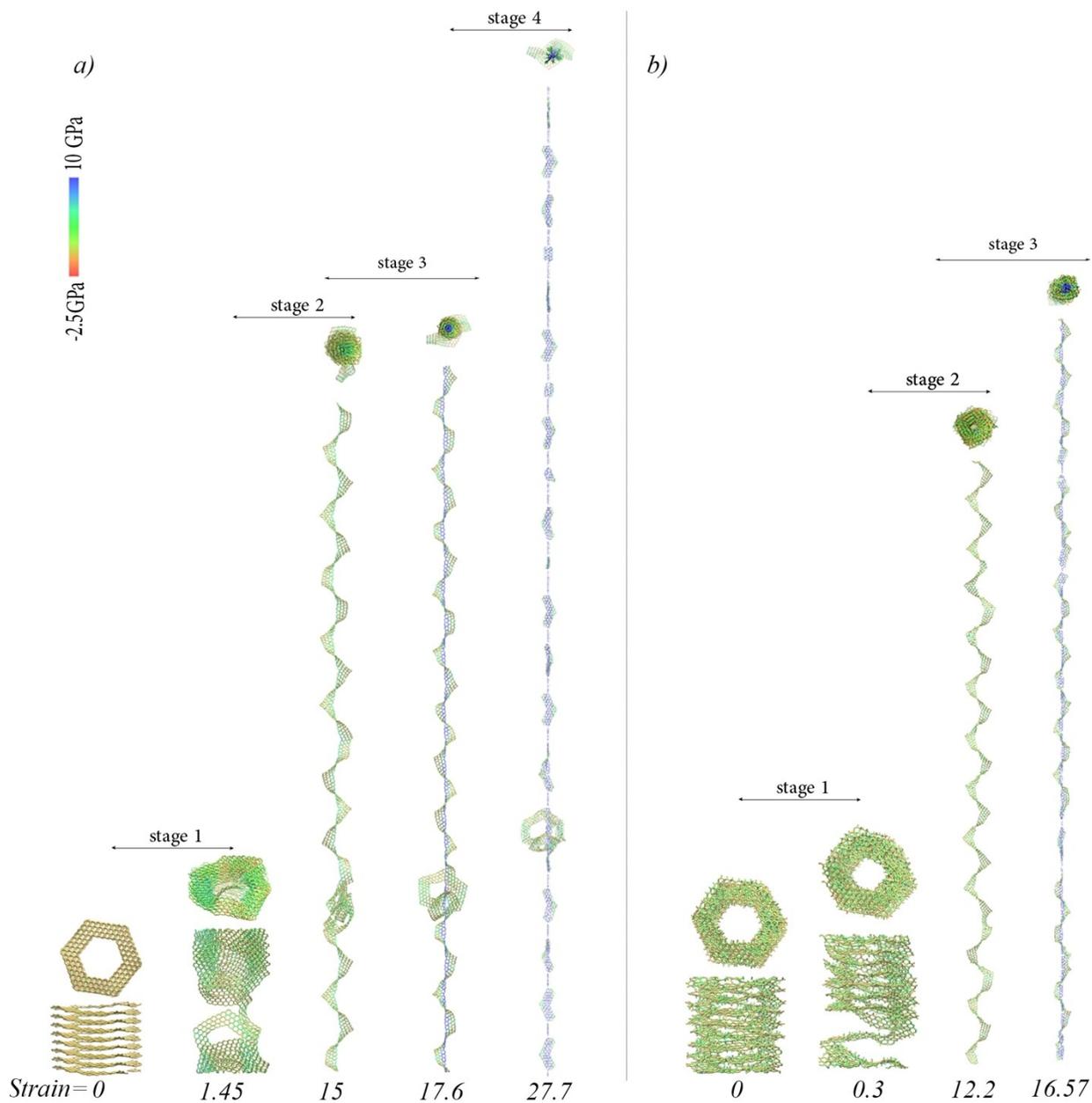


Figure S 4. Comparison of the tensile behavior in: a) pristine GH of sample 1. b) hydrogenated GH for samples 3 at percentages of 30. The color of atoms is related to stress in the tension direction.

S4. Comparing the remaining alongside each other of the adjacent layers for pristine and hydrogenated samples 1,2 and 3.

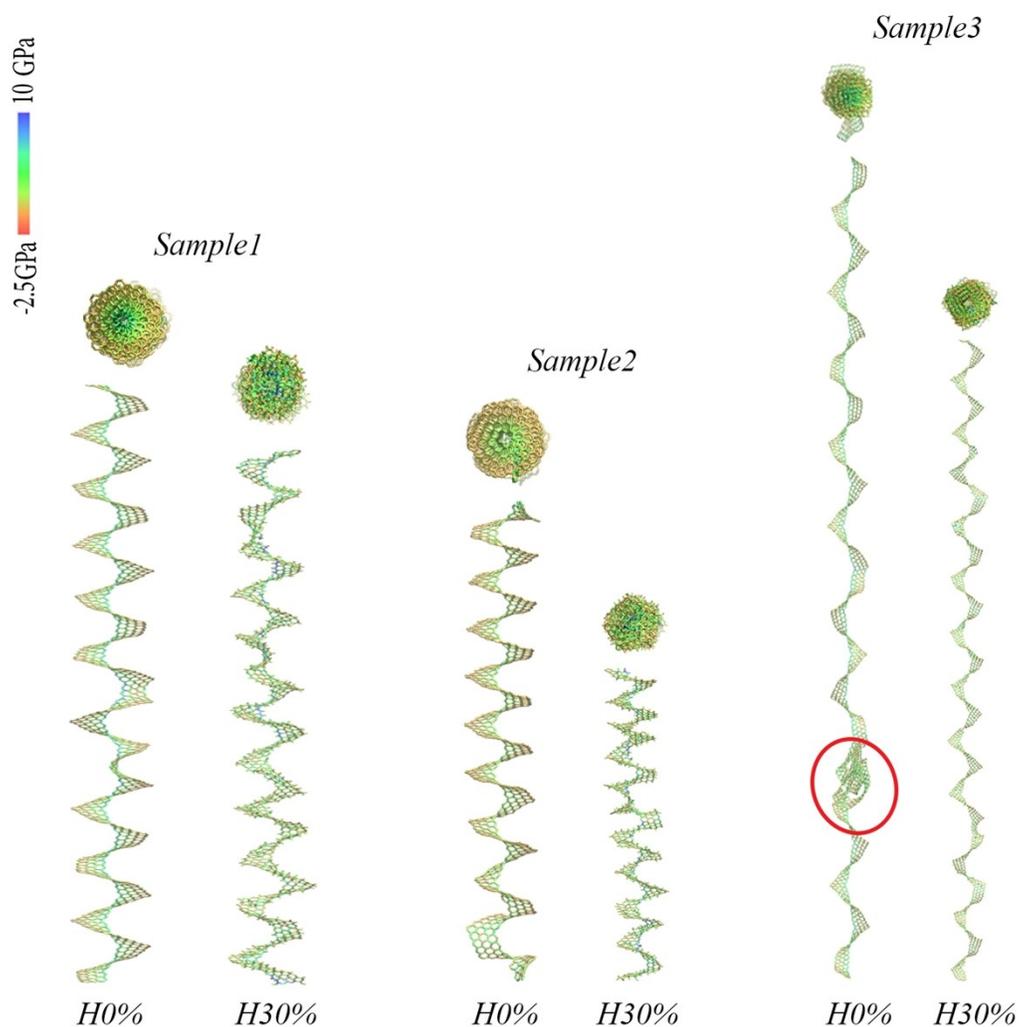


Figure S 5. Comparing the remaining alongside each other of the adjacent layers for pristine and hydrogenated samples 1,2 and 3.

S5. Investigation of the tensile behavior of samples 1,2 and 3 in H-coverage at 10% and 20%.

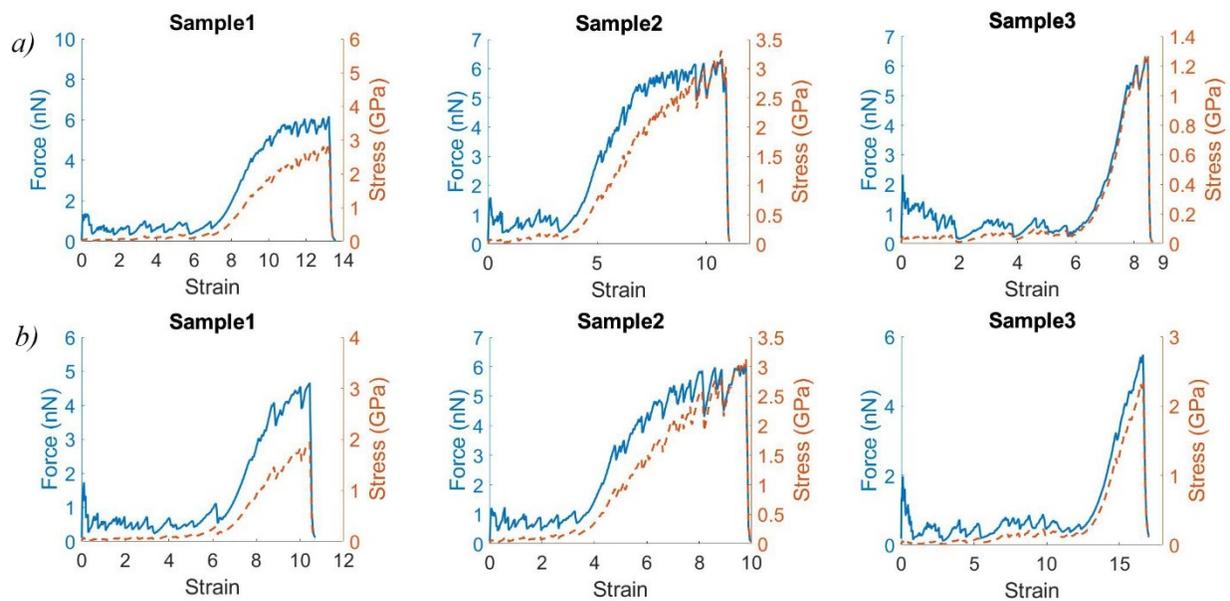


Figure S 6. Investigation of the tensile behavior of samples. 1,2 and 3 in hydrogen coverage at: a) 10%. b) 20%

S6. Examining the difference in tensile behavior of pristine GHs for Sample 4 and Sample 5

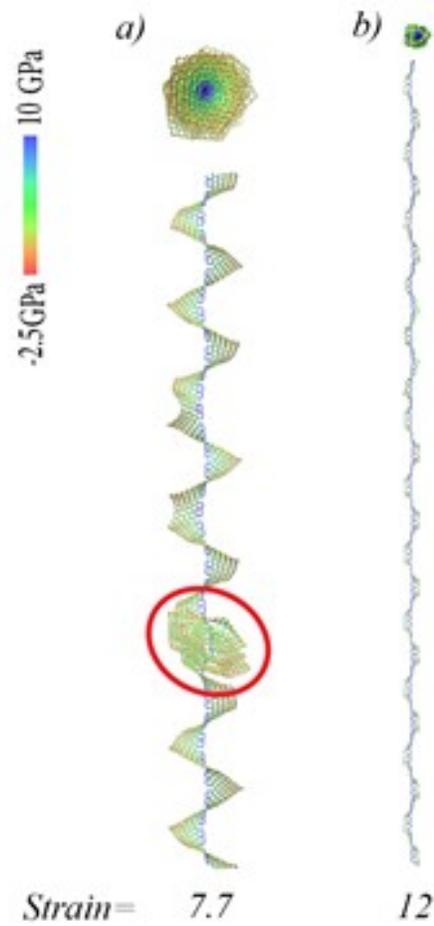


Figure S 7. Examining the difference in tensile behavior of pristine GHs a) Sample 4. b) Sample 5 in the third stage. The color of the atoms is related to stress in the direction of tension.

in the third stage.

S7. Comparing the remaining alongside each other of the adjacent layers for pristine and hydrogenated samples 4 and 5.

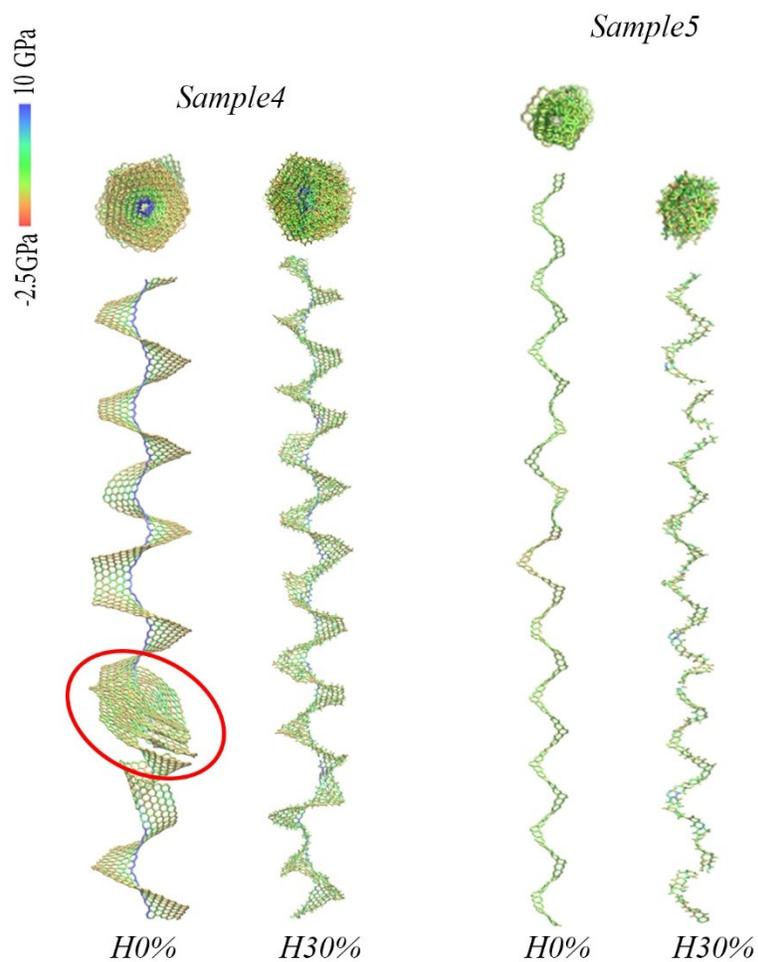


Figure S 8. Comparing the remaining alongside each other of the adjacent layers for pristine and hydrogenated samples 4 and 5.

S8. The tensile behavior in pristine and 30% hydrogenated sample 4.

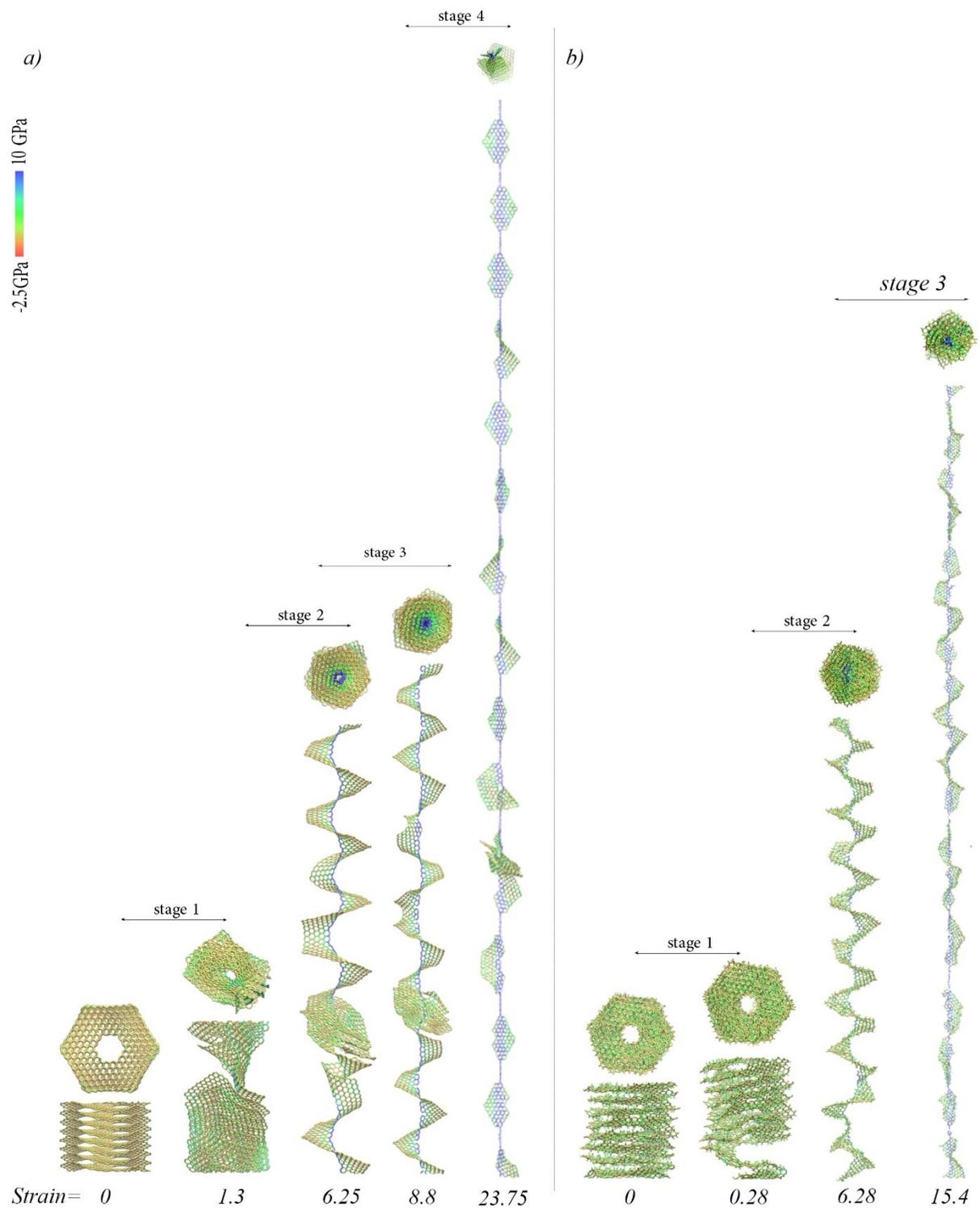


Figure S 9. Comparison of the tensile behavior in: a) pristine GH of sample 1. b) hydrogenated GH for samples 4 at percentages of 30. The color of atoms is related to stress in the tension direction.

S9. The tensile behavior in pristine and 30% hydrogenated sample 5.

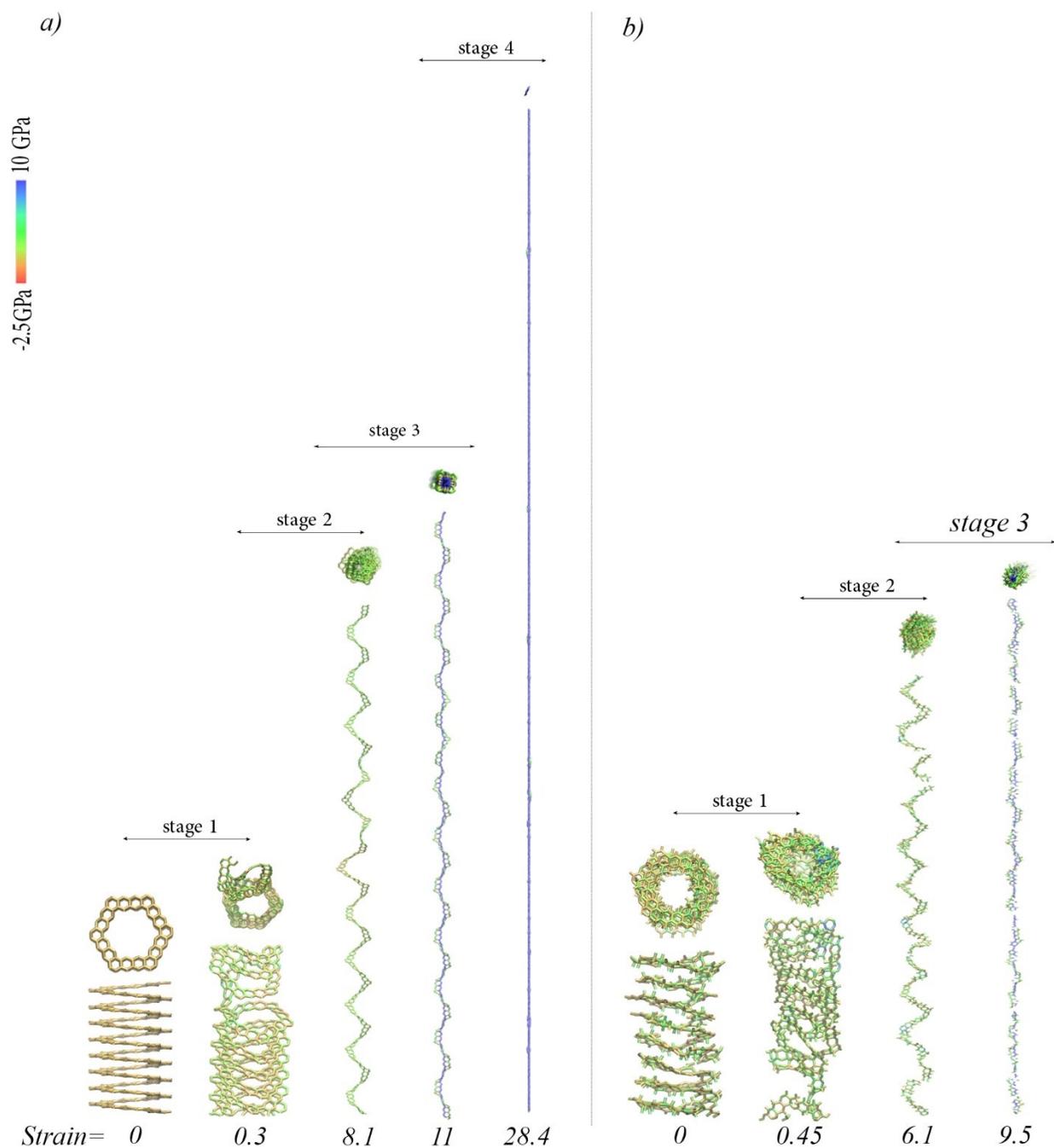


Figure S10. Comparison of the tensile behavior in: a) pristine GH of sample 1. b) hydrogenated GH for samples 5 at percentages of 30. The color of atoms is related to stress in the tension direction.

S10. Comparing the vdW, REBO and torsion energies for samples 4 and 5.

Upon comparing the vdW, REBO and torsion energies, one can obtain interesting information about performing stages of the tensile test. As shown in Figure S 11, this process was performed for functionalized and pristine samples of 4 and 5. By studying the LJ energy region, a significant leap in the initial strain range can be observed. Nonetheless, this leap in the strain range of 0 to 0.8 percent, which is the computational criterion for the Young's modulus and spring constant, is for the functionalized samples that are greater than the pristine GH. Investigating the REBO energy in this small initial strain range indicates a larger energy difference for pristine GH compared to the functionalized one. In fact, upon implementing the strain to the functionalized GH, a very small tensile stress will form and the layers will separate. On the other hand, concerning the pristine sample, some tensile stress is created in the layers, and upon more strain, one can overcome the vdW forces of the adjacent layer. In larger strains and in stage 2, the increase of LJ energy is still observed, while a slight change in REBO and TORSION energies is seen. Indeed, upon comparing the pristine and functionalized sample 4, a slight difference in torsion energy has been occurred, while this phenomenon does not exist for sample 5. This phenomenon can be attributed to the arduous separating of the layers and eventually remaining some layers alongside each other in the pristine sample 4. In the third stage, the increasing trend of LJ energy has been cut and slight change can be observed. However, a considerable change in REBO and TORSION is seen that arise from increasing the tensile stress and the displacement of the atoms relative to each other. Also, the breakdown of some bonds has been observed in a frequent way for the amount of REBO and TORSION energies. This process continues in the fourth stage.

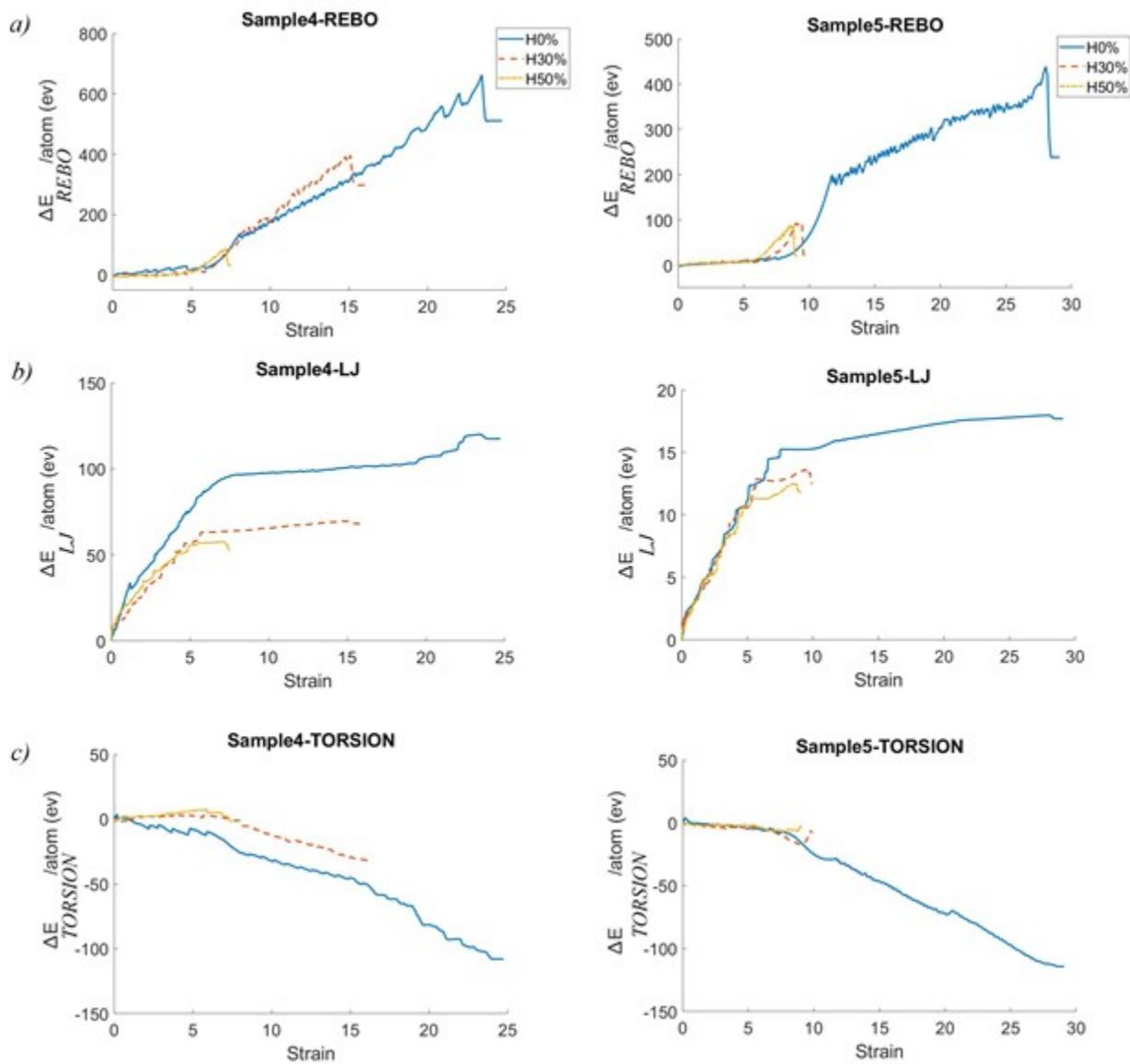


Figure S 11. Examining the energy changes during the tensile test of pristine and H-coverage GHs at percentages of 30 and 50 for samples 4 and 5. a) REBO energy variations, b) LJ energy variations and c) TORSION energy variations for the aforementioned samples.

S11. Patterned H-coverage GHs.

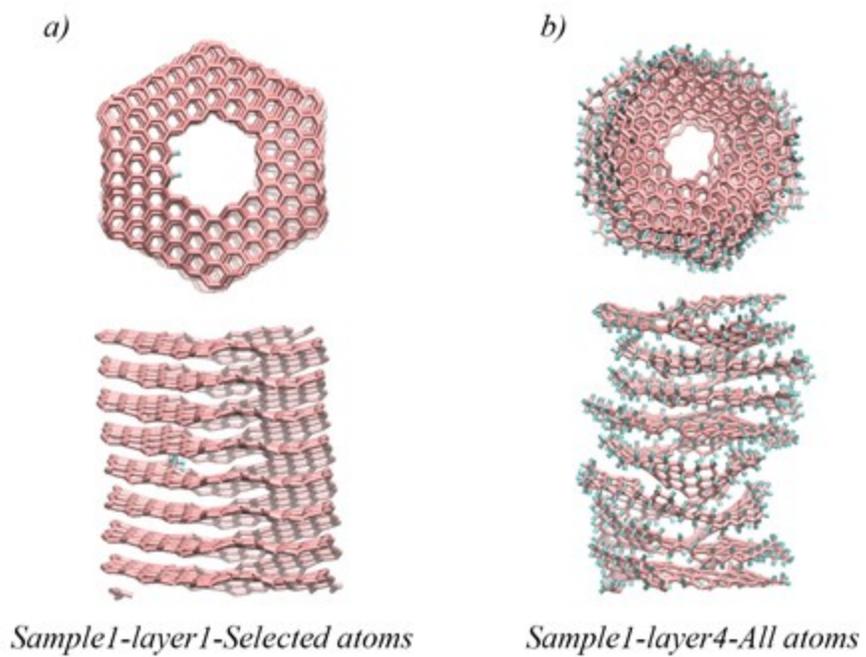


Figure S 12. Regular H-coverage on: a) innermost layer (layer 1) in a selective pattern. b) the outermost layer (layer 4) for all atoms. The color of the atoms is based on their type.