New Constraints for the Binary Phase Diagram of N_2-H_2 Mixtures under High Pressures

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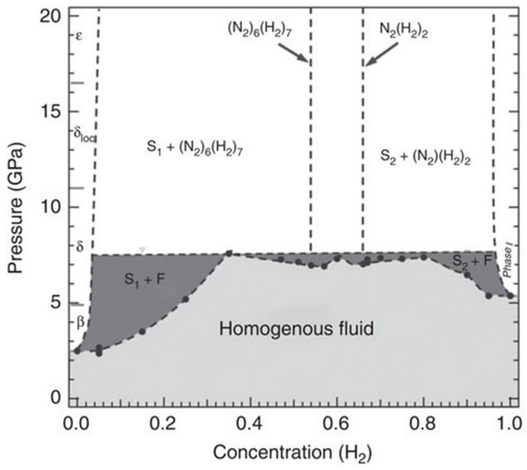
Motivation

N_2-H_2 mixtures can produce several compounds such as ammonia, hydrazine, ammonium azides and more, but not all are stable at ambient condition. Previous studies have examined this simple molecular system at varying concentrations up to 20 GPa. A binary phase diagram was constructed and indicated the formation of several van der Waal compounds formed at high pressure.



Limited amount of data is known for the phase diagram of N_2-H_2 in the pressure range of interest (< 20 GPa). Thus, we examine the existing data to determine what questions remain in the current phase diagram and expand to higher pressures to observe how pressure can alter the systems of simple molecular compounds and mixtures, in hopes to recover high pressure phases at ambient condition.

Approach

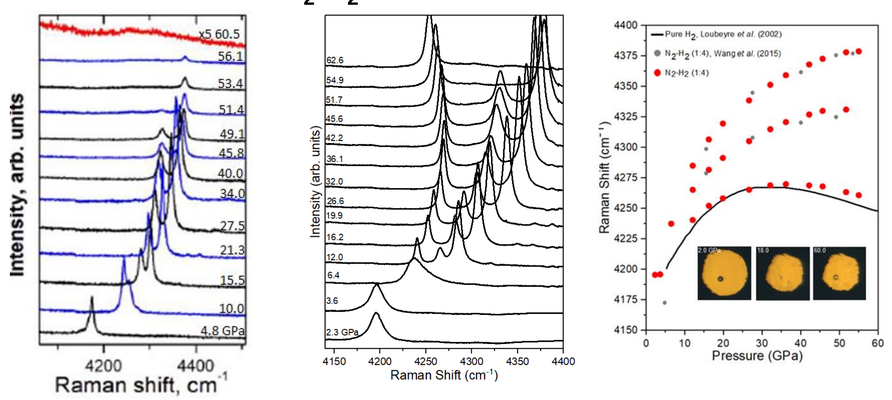
1) Study various mixtures of N_2-H_2 up to 60 GPa and expand the current phase diagram to higher pressure ranges

2) Compare and contrast data sets previously published, with the intent to better define regions on current phase diagram and gain further understanding into the chemistry between nitrogen and hydrogen

3) Expand research further by conducting experiments with D_2-N_2 to better understand the isotope effect on the phase diagram

Phase Behavior of N_2-H_2

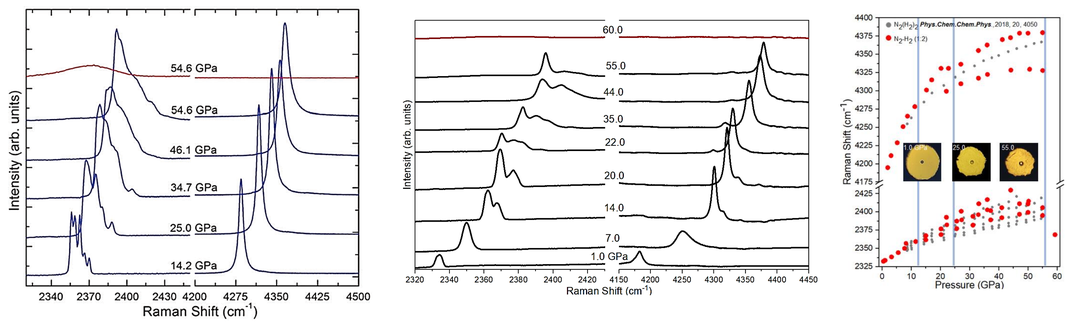
N_2-H_2 1:4 Mixture



- A clear phase transformation occurs at 12 GPa indicated by the H_2 peak splitting into 3
- A new peak is observed at ~4230 cm^{-1}, which follows closely with pure hydrogen as the pressure increases
- Above 55 GPa only one peak remains and is observed to 64 GPa
- The microphotographs display a change in texture, and at 18 GPa a significant change in the refractive index was observed

Chemical Behavior of N_2-H_2

N_2-H_2 1:2 Mixture



- At low pressure, the N_2 peak forms a doublet
- Above 22 GPa both sets of data suggest disorder within the structure indicated by the broadening shoulder peak at ~2380 cm^{-1}
- Above 55 GPa a new broad peak forms at ~ 2360 cm^{-1}
- A chemical reaction can be observed taking place at ~45 GPa and completes near 60 GPa. The disappearance of the H_2 peak followed by the appearance of a new broad peak indicates N-H bonding
- The microphotographs display the change in texture as the pressure is slowly increased to 60 GPa
- Comparing the peak position with pressure three significant phases were observed

Acknowledgements

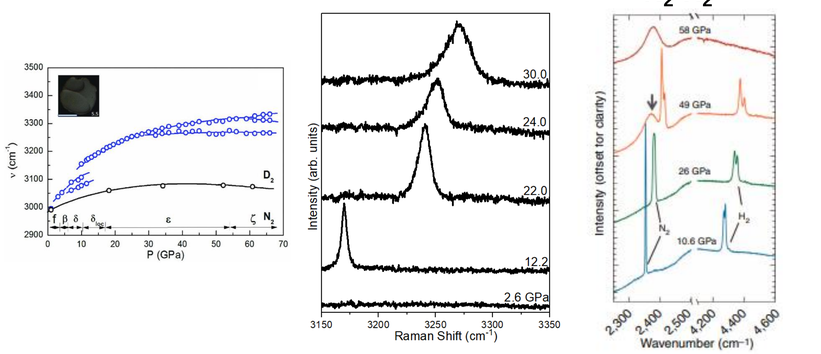
The present work has been funded by DOE-NNSA (DE-NA 0003342), NSF (DMR - 1203834) and ARO (W911NF-17-1-0468)

We acknowledge Dr. Minseob's earlier efforts which have motivated the present study

Isotope Effect

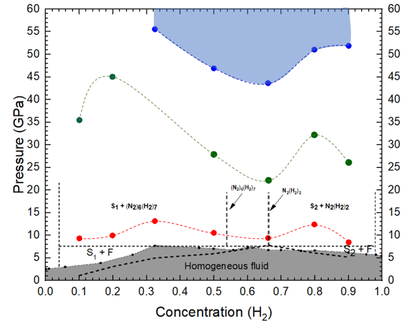
N_2-D_2 9:1

N_2-H_2



- Previous data has suggested significant chemical differences between N_2-H_2 and D_2-N_2 mixtures

Summary



- The first phase, represented in red, was observed by the splitting of the H_2 vibron that significantly blue shifted, suggesting the hydrogen is mixing with the nitrogen
- Phase two is indicated in green. The second phase generally shows characteristics of the N_2 peak broadening and beginning to split. The left side of the phase diagram behaved very different from what was observed at higher pressures in the mixtures with high hydrogen concentrations. The nitrogen peak in the nitrogen heavy mixtures split 3 times and remains stable through 60 GPa
- The third phase is very well defined and outlined in blue. A chemical reaction slowly takes place from 45 GPa to 60 GPa. This reaction ends with the appearance of a broad peak around ~ 2360 cm^{-1} and the disappearance of the H_2 vibron

References

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