Ab Initio Simulation Beryllium in Solid Molecular Hydrogen: Elastic Constant

Carlo L Guerrero and Jose M Perlado
Instituto de Fusión Nuclear, Universidad Politécnica de Madrid.
Madrid 28006, Spain

E-mail: guerrerocarlo@yahoo.es

Abstract. In systems of inertial confinement fusion targets Deuterium-Tritium are manufactured with a solid layer, it must have specific properties to increase the efficiency of ignition. Currently there have been some proposals to model the phases of hydrogen isotopes and hence their high pressure, but these works do not allow explaining some of the structures present at the solid phase change effect of increased pressure. By means of simulation with first principles methods and Quantum Molecular Dynamics, we compare the structural difference of solid molecular hydrogen pure and solid molecular hydrogen with beryllium, watching beryllium inclusion in solid hydrogen matrix, we obtain several differences in mechanical properties, in particular elastic constants. For $C_{11}$ the difference between hydrogen and hydrogen with beryllium is 37.56%. This may produce a non-uniform initial compression and decreased efficiency of ignition.

1. Introduction
The interest of the study of solid molecular hydrogen (SMH) includes different fields of science and technology, essentially in models of giant planets [1] and exoplanets, also in the construction of future aerospace systems where it is proposed as fuel [2]. The study of the SMH phase transitions is very important in the material compression model [3,4], which in turn are applied to systems of inertial confinement fusion (ICF). The ICF Systems are based on a target compression of deuterium tritium solid (DT ice) by irradiation with high power lasers. The DT ice content in the target must have a well-defined characteristics and properties [5]. These properties may be modified depending on the capsule container. The interest of this paper is to submit a first approximation of the effects of possible dissolution of beryllium atoms from the target capsule with DT ice. This approximation is achieved by inserting a Be atom in a molecular matrix solid H. The SMH serves as a model since this study has been more studied than T or D, and can compare some features with our simulations [4]. Our general hypothesis is that the initial conditions of the target configuration with respect to the properties of the DT ice and capsule container determine the efficiency of ICF ignition system [3]. In this paper we propose to study through the static model of phase transitions occurring in a dynamic compression. Static model called us to compression simulated by Quantum Molecular Dynamics (QMD) for a given temperature and pressure, so that it represents a pressure ramp we modeled a large area of the phase diagram of H and an approximation of dynamic compression system that occurs in ICF. We focus on the moments before or on the same ICF initial compression where low temperatures between 15 K and 300 K, which in turn can be compared to experimental results of compression systems diamond anvil cells (DAC), where the maximum pressure that can be reached is approximately 300 GPa.
The problem of segregation and dissolution of metals in contact with SMH is currently under discussion because it has presented a report which shows evidence conductivity of SMH at pressures above 200 GPa and 300 K [6]. A model that can explain this change in properties with the possible immersion of metals in the SMH crystal structure in the experimental case [6] technique they use DAC with copper coated diamonds to measure conductivity, this is where different authors have hypothesized that this copper can be dissolved into the structure and change the H properties [7]. Taking this hypothesis we have seen immersing a metallic light element as Be and changes in the structural and mechanical properties of SMH. Fuel contamination with material of the capsule to been linked to effects on the efficiency of ignition in previous works [8-10].

2. Methodology
Through simulation we could play a series of properties for SMH as lattice constant, crystal structure, phase changes [3] and mechanical properties such as modulus Bulk [4], in particular in this work we study the elastic constants. For which changes occur in the structure and mechanical properties of DT ice, use arrays we have obtained for a wide range of temperatures and pressures, in the case of SMH from 1 GPa to 200 GPa and temperatures of 5 K to 300 K, in this work we shows results for 300 K and pressure range 10 GPa to 25 GPa for hydrogen and hydrogen with beryllium. SMH simulations is for 576 atoms, using the methodology applied in previous works [3,4]. Every \( H_2 \) molecule occupies a hexagonal close-packed (HCP) lattice point, the structure consists 576 atoms in 288 H molecules. We use periodic boundary conditions with which approximates volume representation. The simulation Be in H solid is performed with 574 H atoms comprising 287 molecules, and adding a Be atom at the centre position, which should be occupied by one molecule of H in original structure H 288 molecules, this new configuration is named H_Be. With this initial structure we evolve the system to reach again the temperature and pressure at which we want to compare the structure obtained.

QMD simulations have been carried out using SIESTA code [11] which is based on Density Functional Theory (DFT) calculations with plane waves. We use exchange-correlations functional Local-Density Approximation (LDA) based on functional Cerpeley-Alder (CA) with DZP basis, MeshCutoff 100 Ry. The pseudopotential is Troullier-Martin type. For testing Generalized Gradient Approximation (GGA) functional use in revised Perdew-Burker-Ernzerhof (revPBE), these tests showed no difference with LDA calculations for pressures below 180 GPa. Our QMD exploration consists of a combination of well founded individual stages. The process is initiated by the search for more stable structural configuration, we used the method of Conjugated Gradients (CG).With configurations obtained by simulation for QMD defined temperature and pressure, we calculated the elastic constants \( C_{11}, C_{33}, C_{12} \) and \( C_{13} \) for H and H_Be structure in a range of 10 GPa to 25 GPa, and temperature of 300 K. The initial setting is obtained using QMD with implementation the Nose-Hoover thermostat and Parrinello-Rahman barostat. To calculate the elastic constants we realize the deformation of the structure defined in the three axes X, Y and Z, obtaining the relationship between stress and strain experienced by the structure. The relationship is \( C = \sigma / \epsilon \), where \( \sigma \) is the stress and \( \epsilon \) strain. In order to estimate each constant use transverse isotropic model applied to the SMH-HCP structure, where \( C_{22} = C_{11} \) and \( C_{23} = C_{13}, C_{44} \) no show in this work.

3. Results
Figure 1 show a comparison between experiment SMH [12] and our results for \( C_{11}, C_{33}, C_{12} \) and \( C_{13} \), the experiment uses DAC, for pressure of 6 GPa to 23 GPa. We study phase I, where \( H_2 \) is solid state. In this pressure, figure 2, our results show a maximum difference of 18.65 \%, for the constants \( C_{33} \), and \( C_{11} \), to \( C_{12} \) and \( C_{13} \) the difference is smaller. This is our starting point for interpreting the changes to add the Be atom in the crystal structure. The introduction of Be atom within a volume of 576 atoms allows us to calculate the changes in the SMH, which may well be related to changes in the properties of the DT ice. As a first step evaluate the crystal structure, to compare in figure 3 the position of SMH alone and with Be introduced. Both cases are structures obtained after thermal stabilization and pressure. For SMH pressure range 10 GPa to 25 GPa and 300 K has a hexagonal configuration
recognizable, compatible with phase I for SMH [13]. In figure 3 is displaying projections of all the
atoms in different planes XY, ZY and XZ. In the case of Be immersed in H\textsubscript{2} - HCP, there is no change
in the crystal structure, but are observed significant changes in the mechanical properties. Figure 2
shows the comparison between the simulation results for H and H\textsubscript{Be}, concerning the calculation of
the elastic constants.

![Figure 1. Simulated SMH [4] and experimental results [12]. The difference between the elastic constant varies according to axis. The mean difference: C\textsubscript{11} 16.37 %, C\textsubscript{33} 18.65 %, C\textsubscript{12} 3.68 % and C\textsubscript{13} 9.97 %.](image1)

![Figure 2. Comparison between the elastic constants calculated for H and H\textsubscript{Be}. C\textsubscript{12} and C\textsubscript{13} are similar in both cases, but C\textsubscript{11} and C\textsubscript{33} mean difference is more than 37.56 % and 30.84 % respectively.](image2)

With a linear regression we can compare point to point each of the results. The greatest differences
occur between the C\textsubscript{11} and C\textsubscript{33}, for C\textsubscript{12} and C\textsubscript{13} the difference is minimal. In figure 1 C\textsubscript{12} mean
difference is 16.37 %, C\textsubscript{33} 18.65 %, C\textsubscript{12} 3.68 % and C\textsubscript{13} 9.97 %, with respect to the experimental result.
The inclusion of Be atom by two of H produces a marked change in the magnitude of the elastic constant,
and hence in its mechanical properties, for figure 2 C\textsubscript{11} mean difference is 37.56 %, C\textsubscript{33} 30.84 %,
C\textsubscript{12} 5.65 % C\textsubscript{13} 4.73 %, with respect to the hydrogen simulation. Although the structure as
shown in figure 3 does not change, its mechanical properties change. This study is a first step to obtain
and study the mechanical properties of the phase change system, in this paper we carry only the Phase
I study, eventually we see the relation between changes in the mechanical properties and crystal
structure of the system to thereby characterize either the compression process in the field of DT ice in
the case of ICF and turn get a good representation of the phase diagram of H isotopes. This method of
study allows us to compare our results with the DAC compression systems or Laser, with additional
value that our method allows a window of atomic structural information.

4. Conclusions
We show a comparison between the elastic constants of H and H\textsubscript{Be}. Beryllium inclusion causes an
increase in the amount of C\textsubscript{11} and C\textsubscript{33}, this effect is equivalent to increasing the pressure of the SMH,
this due to the formation of beryllium hydride, which may explain the appearance of solid hydrogen
properties presented higher pressure such as metallization [7].

This method allows have a view of the early stages of compression from 15 K to 300 K in a
pressure range from 0.1 GPa to 200 GPa as we have shown in previous work [3,4]. This type of
analysis can be applied to DAC compression or by laser compression, which are of interest in systems
for inertial confinement fusion.

We have observed that fuel contamination with Be (Z-low) of the capsule changes the elastic
properties of solid materials by 30%, then the deformation or strain induced by the initial shockwave
differs at each point with Be in 30%, this may cause a non uniform initial compression. A way to
reduce the contamination effect after the initial compression in ICF experiment is the so-called "high
foot\textsuperscript{th} method [10]. Should be studied the storing and delivering times to ICF target to know whether the SMH layer fuel contamination with material of the capsule occurs before the initial compression.

Figure 3. Planes XY, ZY, XZ simulation boxes for the pressures of 10 GPa at H and H(Be), and 25 GPa for H. They can be seen as setting the crystal structure suffers few changes in the presence of Be atom (big sphere), in agreement with that observed experimentally for phase I of H. HCP Crystal Structure and H\textsubscript{2} molecules in lattice point.

Acknowledgment
To S. Cuesta. The author acknowledges the computer resources and technical assistance provided by the Centro de Supercomputación y Visualización de Madrid (CeSViMa). The authors thank the Spanish Ministry of Science and Innovation for economical support via the ACI-PROMOCIONA program 2009 (ACI2009-1040).

References
[1] Duffy T, Vos W, Zhang Ch, Hemley R J, and Mao H, 1994 Science, 263, 5153, 1590-1593
[2] Silvera I and Cole J, 2010 J. of Phys.: Conf. Series, 215 012194. Bennitong S, Lovell A, Headen T, Royse D, Nathanson A and Voller S, 2013 US Patent 2013/0095307A1
[3] Guerrero C, Cuesta S and Perlado J M, 2013 Eur. Phys. J. Web of Conferences 59, 16004
[4] Guerrero C, Cuesta S and Perlado J M, 2014 Europhys. Lett. 108, 26001. Guerrero C and Perlado J M 2013, 25th Symposium on Fusion Engineering (SOFE), pp.1,4 doi: 10.1109/SOFE.2013.6635457.
[5] Aleksandrova I V et al 2007 J. of Russian Laser Research, 28, 3
[6] Eremets M I and Troyan A I 2011 Conductive dense hydrogen Nat. Mater. 10, 927
[7] Nellis W J, Ruoff A and Silvera I F 2012 Has Metallic Hydrogen Been Made in a Diamond Anvil Cell? Preprint arXiv:1201.0407
[8] Caruso A and Strangio C 2003 J. Exp. Theor. Phys. 97, 5, 948-957
[9] Yi S A, et al 2014 Phys. Plasmas 21, 092701
[10] Hurricane O A, et al 2014 Nature 506, 343
[11] Soler J M, Artacho E, Gale J D, Garcia A, Junquera J, Ordejon P and Sanchez-Portal D 2002 J. Phys.: Condens. Matter 14, 2745-2779.
[12] Zha C, Duffy T, Mao H and Hemley R J 1993 Phys. Rev. B. 48, 13, 9246-9255
[13] Toledano P, Katzke H, Goncharov A F and Hemley R J 2009 Phys. Rev. Lett. 103, 105301