



Article Asphaltene Behavior during Thermal Recovery: A Molecular Study Based on Realistic Structures

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Abstract: Asphaltene precipitation and deposition can occur at both the surface and subsurface levels, leading to the formation of organic-based scales. Asphaltene precipitation can also lead to changes in petrophysical properties such as wettability, which affects the ultimate recovery. Asphaltene precipitation is linked to changes in fluid composition driven by pressure drawdown and temperature variation across the reservoir. Thus, asphaltene deposition can adversely influence the ultimate recovery. Thermal recovery methods are invoked to mitigate the adverse effects of asphaltene precipitation. The behavior of asphaltene under thermal recovery along with the link between the asphaltene molecular structure and its response to the increase in temperature during thermal recovery are not fully understood. In this paper, realistic asphaltene structures based on actual crude samples were recreated on a computational platform, and several characteristics of the asphaltene structures (density, viscosity, and interfacial tension) were evaluated during the heating process. The density of asphaltene was correlated with the percentage of aromatic carbon in its structure. The viscosity and interfacial tension decreased substantially as the temperature increased. The IFT reduced by approximately 30 mN/m as the temperature was increased from 300 K to 450 K. Moreover, the mechanical stability of asphaltene was found to be highly influenced by heating. The findings provide nanoscale insights into the behavior of asphaltene during thermal recovery, which can be used to improve the design of thermal recovery processes.

Keywords: asphaltene; thermal recovery; heavy oil; petrophysics; molecular simulation

1. Introduction

Petroleum typically includes four components: aliphatic hydrocarbon saturates (e.g., n-alkanes), polycyclic aromatics and their alkylated derivatives, resins, and asphaltenes, which are collectively known as SARA (saturates, aromatics, resins, and asphaltenes). During field production, along with secondary and tertiary recovery phases, crude oil is subject to continuous changes in pressure and temperature, which in turn trigger asphaltene deposition [1]. Asphaltene precipitation leads to severe plugging formation and alters the wettability, which consequently reduces the ultimate recovery. Moreover, asphaltene deposition causes enormous increases in operational expenses and difficulties in field maintenance. Asphaltene deposition can be mitigated through chemical soaking, mechanical scraping, microbial, and dissolution approaches. Physical cleaning includes processes such as scratching and pigging, which are costly. A proactive approach involves the addition of crystal modifiers and asphaltene stabilizers such as sodium dodecyl sulfate, triton X-100, and benzoic acid to inhibit aggregation [2,3]. These treatments are commonly used in surface facilities but are rarely applied to the reservoir itself because of technical difficulties.

The above methods rely on either shifting the thermodynamic equilibrium to reverse precipitation or inhibiting aggregation by chemically altering the bonding between asphaltene units. However, the formation of asphaltene is inevitable, especially under subsurface



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Copyright: © 2022 by the author. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). conditions where rock–oil interactions, along with the temperature and pressure, are difficult to control. Hence, enhanced recovery methods such as thermal recovery are invoked to mitigate the adverse effect of asphaltene deposition on the ultimate recovery [4]. The primary aim of thermal recovery is to reach a temperature that provides favorable asphaltene transport and interfacial properties.

Asphaltenes are high-molecular-weight compounds containing nitrogen (N), sulfur (S), and oxygen (O) heteroatoms as minor skeletal components. Asphaltenes possess island–ocean, rosary, or archipelago molecular structures [5,6]. The presence of N, S, O, and other non-hydrocarbon ingredients such as iron, nickel, manganese, and copper affect the chemical and physical behaviors of asphaltene. Generally, asphaltenes consist of carbon (80%–86%), hydrogen (6%–8%), sulfur (2%–3%), nitrogen (0.5%–2%), oxygen (0.5%–2%), and trace metals (0.1%–0.2%) [7]. Asphaltenes are soluble in aromatic solvents such as benzene, toluene, and xylene, while they are insoluble in liquefied gases such as ethane, methane, and propane [7]. The solid parts of asphaltenes are soluble in toluene and insoluble in n-heptane.

The precipitation of asphaltenes is initiated by the shifting of the thermodynamic equilibrium of the crude oil. Disturbed asphaltenes regain their equilibrium state by associating with other surrounding hydrocarbon molecules. Association with other asphaltene molecules leads to aggregation and eventually precipitation from the oil phase onto the rock surface [8,9]. The asphaltene deposition envelope (ADE) highlights the effects of pressure and temperature changes on asphaltene formation (as shown in Figure 1). The typical ADE diagram shows that asphaltene precipitation increases as the pressure decreases to the bubble point, with the most precipitation occurring at the bubble point [10]. When the pressure drops below the bubble point, precipitation decreases [7]. The rate of precipitation is also affected by changes in temperature. For heavy crude oils, asphaltene precipitation increases with increasing temperature. In contrast, for light oil, precipitation decreases with increasing temperature. Many studies have focused on the nonlinear relationships between asphaltene precipitation and changes in pressure and temperature [3]. Typically, asphaltene precipitation is correlated with the presence of heavy hydrocarbons, and reducing the density of the crude oil will reduce precipitation [11,12]. The deposition of asphaltene on rock grains, which is induced by the polarity of asphaltenes, is linked to alterations in wettability and early water breakthrough during secondary recovery [13-18]. The interactions between rock and crude oil are augmented by the polarity of asphaltene and acid/base reactions. The asphaltene content was found to be a key factor in such interactions [19,20]. The experimental findings are consistent with microscopic investigations based on molecular simulation [21–30]. Molecular simulations have linked the asphaltene aggregation rate to the structure of the asphaltene model [31-36] and the type of organic solvent [37]. In general, molecular simulations complement the experimental results and reveal asphaltene aggregation in an organic solvent followed by thin-film formation [38–40]. However, a comprehensive description of asphaltene film dynamics throughout a typical production process and different enhanced oil recovery schemes has not been reported. Thermal recovery processes are applied to heavy oil reservoirs [41]. Despite the differences among these thermal methods, they are all based on increasing the temperature of the heavy oil to improve its flow parameters and interfacial properties [42]. Asphaltene is a primary component of heavy oil, and a thorough assessment of its behavior during thermal processes is crucial for understanding the overall effect of heating on heavy oil [43,44]. In this article, the behavior of asphaltene and asphaltene-water interactions were microscopically studied under a range of temperatures reflective of typical heating profiles encountered in real reservoirs. Realistic asphaltene structures derived from an actual oil sample were used to generate the asphaltene phase for comprehensive molecular simulations. The remainder of the paper is organized as follows: Section 2 outlines the methodology, including the recreation of asphaltene within a computational platform, the governing potential function, the characteristics of the asphaltene film, and asphaltene–water interactions under a range



of temperatures. Section 3 then presents the results and discussion, and concluding remarks are provided in Section 4.

Temperature

Figure 1. Typical asphaltene deposition envelope (ADE).

2. Methodology

Representative asphaltene molecular units served as the building blocks in the simulations. Alqam et al. [5] derived realistic asphaltene structures based on the detailed characterization of oil samples collected from a carbonate formation; from these asphaltene structures, three models were selected (Table 1). The selected models vary in the percentage of aromatic carbon, molecular weight, and carbon-hydrogen ratio. Moreover, the third model has two island-like fused benzene rings connected by a sulfur atom [45].

The variation in the aforementioned characteristics provides an opportunity to elucidate the effects of aromaticity, polarity, and the presence of heteroatoms on the thermodynamics and volumetric properties of asphaltene. The interactions within the asphaltene molecules and between asphaltene and other molecules such as water (i.e., in the case of hot water/steam injection) were captured through the non-bonding 9-6 potential function given by:

$$U_{LJ} = \epsilon \left[2 \left(\frac{\sigma}{r_{ij}} \right)^9 - 3 \left(\frac{\sigma}{r_{ij}} \right)^6 \right] + \frac{q_i q_j}{4\pi\epsilon_0 r_{ij}}.$$
 (1)

Equation (1) models the interactions between two atomic centers with charges q_i and q_j and separated by a distance r_{ij} . The potential exhibits a minimum at a distance greater than σ , and the maximum attraction is given by ϵ . In the second term of the right side of Equation (1), ϵ_0 is the dielectric constant.

For unalike centers, a proper mixing rule is needed. The sixth-power averaging rule has been shown to be sufficient for approximating σ and ϵ , as given in Equations (2) and (3) [46]:

$$\sigma = \left(\frac{\sigma_i^6 + \sigma_j^6}{2}\right)^{1/6},\tag{2}$$

$$\epsilon = 2\sqrt{\epsilon_i \epsilon_j} \frac{\sigma_i^3 \sigma_j^3}{\sigma_i^6 + \sigma_i^6}.$$
(3)

The σ and ϵ values for different atomic centers were parametrized using a predefined forcefield. Forcefields such as COMPASS and CFF93 have been extensively used for organic-based materials [47]. These forcefields have been complimented by the launch of the polymer-consistent forcefield PCFF+ [32], which was used to configure the parameters appearing in Equations (1)–(3). The same molecular modeling approach has been applied in similar systems and achieved decent results that reasonably match the experimental results [48–55].

Table 1. Asphaltene structures with different percentages of aromatic carbon adopted for use in this study.

Asphaltene	Molecular Structure	Molecular Formula	Percentage of Aromatic Carbon
1	HH HH	C ₃₇ H ₃₃	73%
2	HH HH	C ₃₁ H ₃₅	58%
3	HAR HA	C ₃₈ H ₃₅ S	73%

2.1. Asphaltene Film

The asphaltene films were constructed from the respective asphaltene macromolecules given in Table 1 by starting from some number of macromolecular units and condensing them to yield the final structures. For this purpose, molecular dynamics simulations were run using LAMMPS. First, 100 units of asphaltene were initialized in a low-density threedimensional periodic boundary cell (i.e., asphaltene was initialized at a low density by placing the molecular units in a relatively large cell to avoid any molecular instability). Subsequently, the energy was minimized, the molecular velocity was initialized, and a simulation run was conducted in an isochoric-isothermal NVT ensemble at 900 K. The high temperature helped to speed up the molecular relaxation (i.e., 250 ps in this study). A 200 ps run was then conducted in an isobaric-isothermal ensemble at 20.67 MPa and the same temperature. The temperature was then gradually decreased to 700 K, 500 K, and finally to 350 K through three consecutive runs in the NPT ensemble. In this study, a final temperature of 350 K and a final pressure of 20.67 MPa were selected to represent typical conditions encountered in the reservoir. The final density of the asphaltene 1, asphaltene 2, and asphaltene 3 films were 1.14, 1.02, and 1.13 g/cm³, respectively, and the three cells had respective sizes of $41.08 \times 41.08 \times 41.08$ Å, $40.55 \times 40.55 \times 40.55$ Å, and $42.52 \times 42.52 \times 42.52$ 42.52 A. These cell sizes were sufficient to yield representative volumes, as demonstrated in previous studies [49,54]. The final structures are shown in Figure 2. The structures were further analyzed in terms of their ability to host molecules by characterizing the intermolecular spaces. This was carried out by the continuous insertion of nonoverlapping



Figure 2. Visual representations of the final asphaltene structures.



Figure 3. Intermolecular space (in radius) for the asphaltene films, revealing similar pore size distributions.

2.2. Viscosity Calculations

The viscosity, which is a crucial parameter for assessing the effect of heating on mobility, was calculated from the velocity and molecular trajectories of the equilibrated asphaltene films during a simulation run under the NVT ensemble with an extended simulation time of 5 ns [57,58]. The viscosity package of MedeA software was used to complete the calculations. Since one of the primary objectives of this study was to model the viscosity of asphaltene as a function of temperature, the viscosity calculations were repeated under increasing temperature. An equilibration molecular dynamic simulation consisting of runs under the NPT and NVT ensembles was conducted prior to the viscosity calculations for the considered range of temperatures.

spheres with a predefined threshold [56]. The distributions of intermolecular space in the asphaltene structures are shown in Figure 3.

The interactions between water at elevated temperatures and asphaltene were quantified using two approaches. In the first approach, the water molecules hosted by asphaltene at a given temperature were assessed by running equilibrium Gibbs ensemble Monte Carlo simulations. These simulations mimicked the water molecules crossing the interface from the aqueous phase to the asphaltene film. The Gibbs ensemble Monte Carlo simulations were run using the water fugacity as an input for the calculations (see Table 2).

Table 2. Fugacity of water as a function of temperature calculated using the Peng–Robinson equation of state at 20.67 MPa.

Т (К)	Fugacity (MPa)	
300	0.004	
350	0.052	
375	0.137	
400	0.311	
450	1.149	

To calculate the interfacial tension (IFT), the boundary of asphaltene was adjusted in the z-direction to allow for the stacking of asphaltene by water. The water layer consisted of 600 water molecules with a density of 1.0 g/cm^3 . The simulation workflow consisted of an equilibration stage for the changes in temperature and an extended NVT simulation, with the last 300 ps used to calculate IFT. This simulation time was sufficient for the IFT value to converge, as shown in Figure 4. The calculations were carried out using the IFT package in MedeA software.



Figure 4. Example of an IFT calculation showing the convergence of the IFT value to the final calculated value.

2.4. Mechanical Stability of the Asphaltene Film

The asphaltene film is subject to forces during primary and enhanced recovery. During thermal recovery, the increase in temperature may also affect film stability. In this study, the mechanical properties of the asphaltene films were assessed under continuous strain

in concert with energy quantification. The relationship between the strain and change in energy is governed by Equation (4):

$$U = \frac{E_{tot} - E_0}{V_0} = \frac{1}{2} \sum_{i=1}^{6} \sum_{j=1}^{6} C_{ij} e_i e_j,$$
(4)

where *U* is defined as the change in energy $(E_{tot} - E_0)$ resulting from the applied strain *e* divided by the initial volume V_0 . The subscripts *i* and *j* yield a tensor with 36 elements across the structure, and *C* is a stiffness matrix component. The number of elements is reduced to three as the asphaltene film forms a cubic cell. The energy was quantified by running equilibration molecular dynamics simulations before and after deformation. The simulation workflow consisted of a minimization stage followed by an extended NVT run. This approach has been applied extensively to investigate the mechanical stability of different materials [59].

3. Results and Discussion

3.1. Asphaltene Physical Properties

Asphaltene density is used to macroscopically distinguish asphaltene based on type. In this study, three asphaltene models with different chemical structures were constructed from single units through molecular dynamics simulation. The density of each structure was obtained as a function of temperature based on equilibration molecular dynamics simulations (Figure 5).



Figure 5. Density profiles of the asphaltene structures as a function of temperature.

The density of the first and third structures (asphaltene 1 and 3) was higher than that of the second structure (asphaltene 2). This could be attributed to the higher fraction of sheet-like carbon (i.e., aromatic carbon) in the molecular structure of asphaltene 1 and 3, which allowed for more condensed stacking compared to asphaltene 2. For all structures, the density decreased as temperature increased. Interestingly, the density of asphaltene 2 decreased to slightly below 1 g/cm^3 at high temperatures.

Next, the viscosity was analyzed for the same range of temperatures, as shown in Figure 6. At a low temperature (i.e., 300 K), the viscosities of the asphaltene 1, 2, and 3 structures were 963, 390, and 149 cP, respectively. These values are within the range anticipated for waxy and heavy organic-based materials. Asphaltene 1 had a higher

viscosity than the other two, which can be attributed to its structure. Asphaltene 1 has eight fused benzene rings while asphaltene 2 has five. Asphaltene 3 has two separate three and four fused benzene rings. Interestingly, the viscosity at lower temperatures followed the order of the size of fused benzene rings.



Figure 6. Viscosity profiles for the asphaltene structures as a function of temperature. In all structures, the viscosity decreases drastically as the temperature increases.

As the temperature increased from 300 to 350 K, the viscosity reduced drastically to 60.3 and 19.6, and 57.0 cP, respectively, for the three structures. These changes suggest that the pour point (i.e., the temperature above which a material is considered to behave like a liquid) was in the range of 300–350 K. As the temperature continued to increase to 450 K, the viscosity continued to reduce to 19.4 cP for asphaltene 1 and 1.3 cP for asphaltene 2. Asphaltene 3, however, showed less sensitivity to a further decrease in the temperature as the viscosity was maintained almost the same with increasing the temperature. This suggests that the intermolecular forces induced by the polarity of sulfur restrict further reduction in the viscosity.

3.2. Interactions with Water and Interfacial Behavior

Thermal recovery often involves the injection of hot water/steam. Hence, it is vital to understand the interactions between water and asphaltene as a function of temperature. For this purpose, the asphaltene films were molecularly stacked by a water layer, and IFT calculations were performed for the three structures (Figure 7).

The initial IFT values for asphaltene 1, 2, and 3 were 75.7, 72.0, and 67.0 mN/m, respectively. Similar to the density and viscosity, the IFT decreased with increasing temperature, reaching 35.4, 27.0, and 24.0 mN/m for the three asphaltenes, respectively, without a noticeable influence of sulfur. The visual representations of the stacked asphaltene–water systems (Figure 8) show that the dispersion effect of water molecules increased as the temperature increased, suggesting that heating can instigate emulsion. Notably, the effect of temperature was more pronounced in asphaltene 2 and 3 compared to asphaltene 1, consistent with the reduction in IFT.



Figure 7. Calculated IFT values of the asphaltene structures as a function of temperature.



Figure 8. Visual representations of the water–asphaltene stacked systems upon the completion of the IFT calculations.

To provide more insight into the effect of temperature on asphaltene–water interactions, Gibbs ensemble Monte Carlo simulations were employed for the three structures. These simulations aimed to quantify the capacity of asphaltene to disperse the water layer by hosting water molecules. As outlined in the methodology section, water fugacity was evaluated as a function of temperature, and the number of water molecules entrapped by asphaltene was determined (Figure 9). Asphaltene became able to host water molecules when the temperature exceeded 350 K, and the capacity to host water increased drastically as the temperature increased further. Compared to asphaltene 1, asphaltene 2 had a stronger capacity for hosting water molecules. These results are consistent with the IFT calculations and the visual representations of the asphaltene–water systems are given in Figures 7 and 8.



Figure 9. Asphaltene–water interactions (presented as the millimoles of water hosted per kilogram of asphaltene) as a function of temperature.

3.3. Mechanical Integrity

The molecular simulation framework provides an opportunity to assess the mechanical stability of the asphaltene films using the approach outlined in Section 2.4. The asphaltene films were continuously deformed to a final strain of 0.4 with a strain increment of 0.02. The stress–strain relationships of the three films were then obtained and plotted at temperatures of 300 and 450 K (Figures 10–12).

The stress–strain relationships for the asphaltene structures revealed solid-like characteristics at 300 K with elastic deformation followed by mechanical failure via strainsoftening behavior. The asphaltene 3 structure had a slightly higher yield strength than the other two structures. This could be attributed to the structural difference between the three models. Upon heating to 450 K, the behavior completely shifted, and the three films deformed plastically without a clear elastic region. Notably, the toughness (i.e., the area enclosed by the stress–strain curve) decreased from 0.0457 to 0.0312 GPa for the asphaltene 1 model. The toughness of the asphaltene 2 model decreased from 0.0390 to 0.0256 GPa. Similarly, the toughness of asphaltene 3 decreased from 0.0498 to 0.0331 GPa. The reduction in the toughness of asphaltene 3 was lesser than the other two structures, which was also observed in the viscosity trend of the same model. Again, resistance to heating might be attributed to the presence of sulfur.



Figure 10. Stress–strain relationships for the asphaltene 1 structure at 300 and 450 K.







Figure 12. Stress–strain relationships for the asphaltene 3 structure at 300 and 450 K.

3.4. Wettability Alteration

The water/asphaltene interface showed high sensitivity to the increase in temperature, as discussed in Section 3.2. The overall impact of thermal recovery, however, is also governed by the rock/fluid interactions (i.e., wettability). In general, it is well-documented that maximum recovery is anticipated at intermediate to strong water-wetness conditions. Upon deposition, the thin layer of asphaltene coating the rock surface acts to shift wettability toward oil-wet characteristics [19,54]. The deposited asphaltene film is, however, subject to deterioration during thermal recovery (see Figures 8–12). To cast more light on wettability alteration, asphaltene, water, and substrate (calcite) were stacked and equilibrated at two temperatures to elucidate the wettability mode. The calcite structure is recreated in a similar manner as in other molecular simulation studies [60,61]. The structures were constructed as calcite coated with a thin layer of asphaltene interacting with water. Water and asphaltene layers of equal thickness were selected (i.e., 1 nm) while 4 nm of calcite was used. Snapshots of the system were taken during an extended NVT stage (i.e., 5 ns) to qualitatively assess the wettability, as shown in Figure 13.



Figure 13. Stacked system of calcite/asphaltene 3/water showing the time evolution of fluid-fluid, fluid-rock interactions at 300 and 450 K. The red layer represents water, while the black is asphaltene 3 stacked on calcite.

It can be seen that the system at the higher temperature showed a drastic change where water molecules caused the detachment and replacement of the asphaltene film with time. These observations are consistent with the results of IFT and mechanical integrity analyses.

3.5. Consistency with Experimental Observations

This study provided in-depth nanoscale investigations of the changes in the physical and interfacial properties of asphaltene during a thermal recovery scheme. Some efforts have been done experimentally to unfold the underlying mechanisms of thermal recovery enhancement. These studies vary in their scale from simple core scale to full reservoir scale. Table 3 provides a summary of some studies highlighting the primary property under investigation.

Reference	Type of Formation	Property under Investigation	Observation
Rao [62]	Carbonate	Wettability	Upon hot water injection, the wettability shifted from oil to water wet
Tang et al. [63]	Carbonate	Viscosity	The viscosity of the oil reduced to 2 cP as the temperature reached 240 °C
Al-Hadrami and Blunt [64]	Carbonate	Interfacial tension	The interfacial tension reduced significantly as the temperature increased which resulted in reduced capillary forces
Roosta et al. [65]	Carbonate/Quartz	Wettability	The wettability shifted toward water wet system as the temperature increased
Bardon and Longeron [66]	NA	Interfacial tension	The interfacial tension reduced which influenced the relative permeability positively

Table 3. Summary of some experimentally reported investigation of thermal recovery effects on oil physical and interfacial properties.

The molecular simulation investigation reported in this study showed that increasing the temperature could reverse the wettability by washing out the precipitated asphaltene layer (see Figure 13) which is consistent with the experimental observations reported by Rao [62] and Roosta et al. [65]. Moreover, the increase in the temperature reduces the interfacial tension significantly for all asphaltene types considered in this study which is also consistent with the experimentally reported studies [64,66].

4. Conclusions

Thermal recovery methods have been applied to enhance recovery from heavy oil reservoirs. The effects of heating during thermal recovery on the characteristics of crude oil have been actively investigated. However, the behavior of asphaltene and its interactions with the aqueous phase is not well understood. In this study, realistic asphaltene models were created in a computational platform to assess the characteristics of asphaltene upon heating. The following conclusions can be drawn:

- The density of asphaltene was influenced by the chemical structure and temperature. Asphaltene containing a higher percentage of aromatic carbon had a higher density. The density was inversely correlated with temperature.
- Similar to the density, the viscosity of asphaltene increased with increasing temperature. The asphaltene structure containing the largest share of fused benzene rings had the highest viscosity at lower temperatures. At higher temperatures, however, the presence of sulfur seemed to restrict further reduction in the viscosity. The pour points of the asphaltene models were between 300 and 350 K.
- The temperature affected the interfacial tension, which decreased from more than 70 mN/m at 300 K to approximately 24 mN/m at 450 K. This can be explained by the enhanced ability of asphaltene to disperse the water phase under increasing temperature.
- Mechanical analysis revealed the solid-like behavior of asphaltene with both elastic and plastic deformation at 300 K. The mechanical behavior changed drastically upon heating, and the toughness of the asphaltene films decreased by approximately 60%.
- The findings provide in-depth insights into asphaltene behavior as a function of temperature. These insights can be used to optimize the design of thermal recovery

processes. The findings of this study should be experimentally validated and repeated by extending this study to other asphaltene models.

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Data Availability Statement: The molecular simulation data, including the constructed asphaltene structures used to support the findings of this study, are available from the corresponding author upon request.

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Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

- T Temperature, K
- *P* Presssure, psi, MPa, or Pa
- V_P Pore volume, m³
- *E* Young's modulus, GPa
- *B* Bulk modulus, GPa
- G Shear modulus, GPa
- ν Poisson's ratio, dimensionless parameter
- r_{ij} Separation distance between two center forces
- $q_{i,j}$ Atomic charge
- ϵ_0 Dielectric constant
- σ Distance at which the interaction potential is zero
- Maximum amplitude of the potential well
- E_0 Energy of the structure, J
- *E*_{tot} Energy after deformation, J

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