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Molecular Dynamics Study of the Ni Content-Dependent Mechanical Properties of NMC Cathode Materials

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Abstract: Lithium nickel manganese cobalt oxides (NMCs) are widely used as cathode materials in commercial batteries. Efforts have been made to enhance battery energy density and stability by adjusting the element ratio. Nickel-rich NMC shows promise due to its high capacity; however, its commercial viability is hindered by severe capacity fade, primarily caused by poor mechanical stability. To address this, understanding the chemo-mechanical behavior of Ni-rich NMC is crucial. The mechanical failure of Ni-rich NMC materials during battery operation has been widely studied through theoretical approaches to identify possible solutions. The elastic properties are key parameters for structural analysis. However, experimental data on NMC materials are scarce due to the inherent difficulty of measuring the properties of electrode active particles at such a small scale. In this study, we employ molecular dynamics (MDs) simulations to investigate the elastic properties of NMC materials with varying compositions (NMC111, NMC532, NMC622, NMC721, and NMC811). Our results reveal that elasticity increases with nickel content, ranging from 200 GPa for NMC111 to 290 GPa for NMC811. We further analyze the contributing factors to this trend by examining the individual components of the elastic properties. The simulation results provide valuable input parameters for theoretical models and continuum simulations, offering insights into strategies for reducing the mechanical instability of Ni-rich NMC materials.



Academic Editors: Zhongren Zhou
and Robert F. Klie

Received: 15 February 2025

Revised: 12 March 2025

Accepted: 13 March 2025

Published: 15 March 2025

Citation: Haq, I.U.; Lee, S. Molecular Dynamics Study of the Ni Content-Dependent Mechanical Properties of NMC Cathode Materials. *Crystals* **2025**, *15*, 272. <https://doi.org/10.3390/cryst15030272>

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Keywords: NMC; Ni-rich NMC materials; elastic properties; molecular dynamics simulation; Li-ion batteries

1. Introduction

The high energy density, fast charging, excellent reversibility, high power output, long lifespan, and environmental benefit of Li-ion batteries have resulted in their wide use in electric vehicles, portable devices, and energy storage systems [1–3]. As their role in daily life evolves, significant advances are being made in cycle life, energy density, and charging efficiency [4–6]. However, to accelerate the widespread adoption of electric vehicles and expand the applications of mobile technologies, further improvements need to be made [7,8]. Recent advancements in lithium-ion batteries have focused on enhancing energy density, cycle stability, and safety through material innovations and structural optimizations [9,10].

Due to its high stability and long lifespan, lithium cobalt oxide (LCO) has been widely used in the early stages of battery use, particularly in the cathodes of small portable devices [11,12]. However, the high cost and environmental concerns associated with LCO have resulted in the exploration of alternative materials [13,14]. Transition metal oxides,

such as LiMn_2O_4 [15], LiFePO_4 [16], and LiNiO_2 , have emerged as promising candidates. Each cathode material offers distinct advantages and limitations [17,18], e.g., LiNiO_2 has been found to be difficult to develop and offers poor cyclability [19], while LiMnO_2 , after charge–discharge cycling, undergoes crystallographic transitions to a spinel structure [20].

There is significant potential to enhance performance through the doping of additional elements, leading to the development of ternary materials, such as $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$ (NMC) [21,22] as a versatile alternative. Charge redistribution results in the oxidation states of transition metals in the NMC system differing from those in single-element transition metal oxides, like LiMO_2 (where $\text{M} = \text{Ni}^{3+}$, Co^{3+} , or Mn^{3+}). In the NMC cathode, each transition metal plays a distinct role; for example, nickel (Ni) participates in redox reactions to act as a double redox-active center that cycles between Ni^{2+} , Ni^{3+} , and Ni^{4+} [23–25]. Manganese (Mn) facilitates the reduction of some Ni^{3+} ions to Ni^{2+} , which increases reversible capacity while enhancing structural stability during cycling [26,27]. Cobalt (Co) helps maintain a highly ordered and stable structure, remaining in the Co^{3+} state until all Ni has transitioned to Ni^{4+} during charge and discharge.

The capacity of NMC batteries is known to increase with higher Ni content. However, as the Ni content increases, the ratios of Mn and Co decrease, which decreases the structural stability compromise of the material. Although their superior capacity prompts Ni-rich NMC materials to be considered promising next-generation batteries, their commercialization is hindered by a rapid capacity fade, which is primarily caused by mechanical degradation [28]. Mechanical degradation leads to a loss of lithium storage space and an increase in side reactions, making it a key contributor to capacity fade [29–31].

Theoretical approaches, including the development of chemo-mechanical models, have been undertaken to address the mechanical degradation in Ni-rich NMC. A chemo-mechanical model considering the anisotropic feature of Ni-rich NMC has been proposed [32]. The numerical study on the coating effect of Ni-rich NMC particles showed that by accommodating the volume change in the particles, the coating effectively reduces stress [33]. The effects of grain size and the morphology of NMC811 have been investigated [34]. Particle damage is predicted from the effects of particle sizes and operating conditions [35]. The fracture behavior of single and polycrystalline NMC811 particles has been examined by introducing the cohesive zone model [36]. A chemo-mechanical damage model has been developed to study cracking inside polycrystalline NMC particles [37]. Mechanical properties are essential in numerical studies that use continuum models.

The wide range of transition metal combinations and doping strategies presents significant challenges for experimental studies that aim to determine the mechanical properties of NMC materials. Experimental setups are often difficult to manage and time-consuming. In contrast, computational approaches offer a viable solution to explore various phenomena across different material combinations and doping effects [38]. First-principle studies, which are commonly referred to as *ab initio* studies and often use density functional theory (DFT), offer a key computational technique for these investigations [39,40]. However, the limited number of atoms that can be modeled makes this approach less practical, particularly for studying dynamic behavior [41].

Molecular dynamics (MD) simulation is another computational approach that provides insights into atomic and molecular interactions within Li-ion batteries. This method can offer detailed information on key mechanisms that govern battery performance, including ion diffusion, phase transformations, and structural stability under various operating conditions. Unlike first-principle studies, MD simulations can deal with a large number of atoms, making them well-suited to capture dynamic phenomena at a larger scale.

Several MD simulation studies have been conducted to determine the mechanical properties of battery cathode materials. Lee et al. investigated the state of charge (SOC)-

dependent elasticity of lithium manganese oxide (LMO) materials and showed that competing pair interactions modify the elasticity [42]. Tyagi et al. reported SOC-dependent Young's modulus for LMO ranging (140 to 170) GPa [43]. The Lee group developed the second-nearest-neighbor-modified embedded-atom method (2NN MEAM) potential with charge equilibration (Qeq) for LMO and calculated the bulk modulus and SOC-dependent diffusivity [44]. They also developed 2NN MEAM potential with Qeq for LCO and calculated the bulk modulus and diffusivity of layered and spinel LCO [45]. Asadi et al. explored the SOC-dependent properties of LMO, reporting that as SOC decreases from (1 to 0), C_{11} increases from (145 to 170) GPa [46]. Classical MD simulations have been used to explore the structural properties and defect effect of layered NMC [47]. DFT studies have been performed to calculate the mechanical properties of NMC materials [48,49].

Notwithstanding that atomic simulation approaches have been widely used to investigate the material properties of battery materials, the data on NMC materials remain scarce. Cheng et al. measured the mechanical properties of $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ using nanoindentation [50]. Xu et al. measured the elastic modulus and hardness of $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ using nanoindentation on the polished surface of the synthesized NMC pellets [51]. Sharma et al. measured anisotropic mechanical properties of different composition NMC materials using nanoindentation [52]. The experimental measurements of electrode materials mostly use nanoindentation techniques, which indirectly assess elasticity by indenting the polished surface of sintered pellets.

This study seeks to calculate the elastic properties of NMC materials with a varying Ni content ratio. Understanding composition-dependent elasticity, particularly in Ni-rich NMC materials, provides essential data for higher-scale simulation studies that include finite elements simulations with continuum models.

2. Method

The NMC materials are transition metal oxides with Li-doping, encompassing both electrostatic and pairwise interactions. The total energy of the system can be described by the short-range interatomic interactions and the electrostatic energy due to charges. The Morse potential is employed to calculate the pairwise interactions [53,54]. The Morse function is commonly used for covalent bond interactions; its mathematical representation is written as follows:

$$E = D_0 \left[e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)} \right] \quad (1)$$

where D_0 indicates the potential well depth, α denotes the width parameter, r is the distance between atoms, and r_0 is the distance at equilibrium. Table 1 summarizes the parameters used in this study. The energy contribution for Coulombic interactions is calculated by

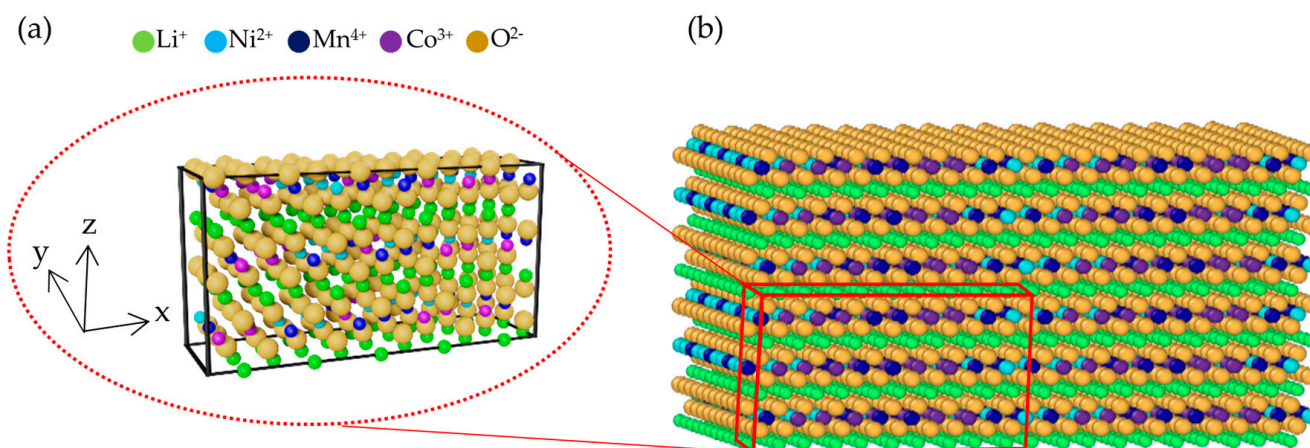
$$E = \frac{Cq_iq_j}{\epsilon r^2} \quad (2)$$

where C represents the energy-conversion constant, q_i and q_j are the charges of two atoms, and ϵ denotes the dielectric constant. Since the battery operates through the intercalation and deintercalation of lithium ions, which involve significant electrostatic forces between the lithium and surrounding electrode material, the charge interactions are crucial.

Table 1. Morse potential function parameters [55].

Interaction Pair	D_0 (eV)	r_0 (Å)	α (Å ^{−1})
Li ⁺ – O ^{2−}	0.001114	2.681360	3.429506
Ni ²⁺ – O ^{2−}	0.029356	2.500754	2.679137
Ni ³⁺ – O ^{2−}	0.029356	2.500754	2.679137
Mn ⁴⁺ – O ^{2−}	0.029658	2.440000	3.012000
Co ³⁺ – O ^{2−}	0.010958	2.400628	3.461272
O ^{2−} – O ^{2−}	0.042395	3.358701	1.659316

The layered structure of NMC belongs to the R3m space group and features a hexagonal crystal structure [56–58]. This layered structure is thermodynamically stable and is usually obtained in the synthetic process. Figure 1a shows the unit cell made up of 360 atoms that we consider. The unit cell includes 90 lithium atoms, 180 oxygen atoms, and 90 transition metals of nickel, manganese, and cobalt, the latter of which are randomly placed in the unit cell. Figure 1b shows the super structure with 5760 atoms obtained for MD simulations by expansion of the unit cells through duplicating four units on the *x*-axis, two units on the *y*-axis, and two units on the *z*-axis. To remove the random effect of transition metals, five different structures where the transition metals are randomly mixed are generated, and the simulations are carried out for the five structures; all simulation results are then obtained by averaging the five random structures.

**Figure 1.** (a) Unit cell of the NMC111 containing 360 atoms and (b) the corresponding superstructure.

Five composition ratios of NMC111, NMC532, NMC622, NMC721, and NMC811 are considered in the LiNi_xMn_yCo_{1−x−y}O₂ family. Table 2 summarizes the atomic composition ratios for the five materials. For nickel, both Ni²⁺ and Ni³⁺ are considered. As the ratio of nickel increases, the Ni³⁺ content is increased to maintain the system neutrality.

Table 2. Composition ratios of the LiNi_{1−x−y}Mn_xCo_yO₂ samples.

NMC	Ni ²⁺	Ni ³⁺	Mn ⁴⁺	Co ³⁺
111	0.33	0	0.33	0.33
532	0.3	0.2	0.3	0.2
622	0.2	0.4	0.2	0.2
721	0.2	0.5	0.2	0.1
811	0.1	0.7	0.1	0.1

We employed LAMMPS [59], an open source simulation tool, for the MD simulations. First, the structures were equilibrated for 50 picoseconds (ps). Figure 2 shows that the

potential energy of the structures is minimized and shows a stable state through the equilibrium process. After equilibration, a strain rate of 0.005 is applied to the structure in the x-, y-, and z-direction to calculate the stiffness of C_{11} , C_{22} , and C_{33} , respectively.

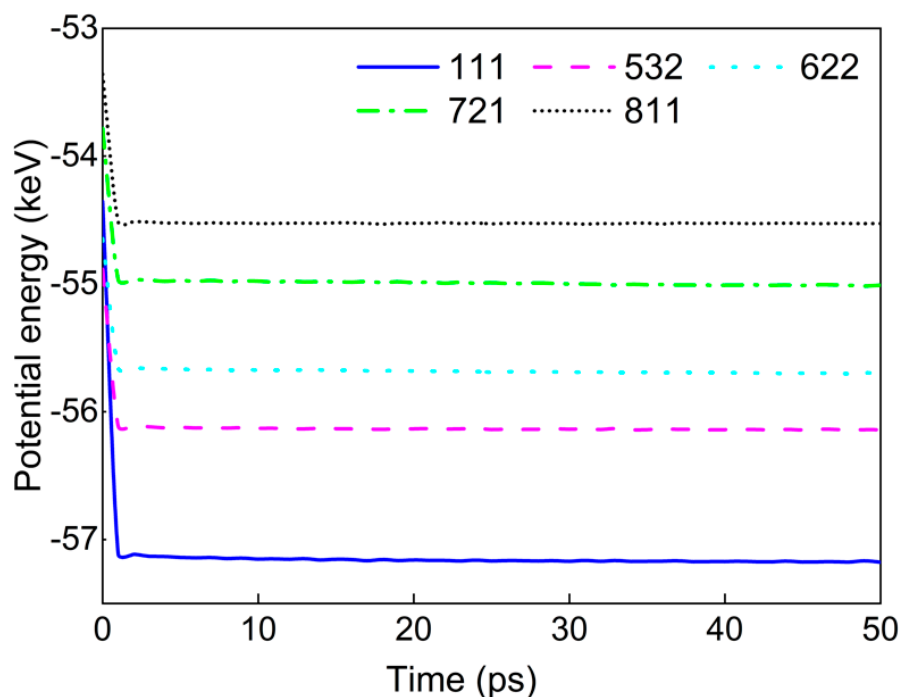


Figure 2. Energy curves of the five NMC compositions during equilibration.

Since long-range Coulombic force is typically stronger than short-range interactions, the atomic charges need to be scaled down to balance the long- and short-range interactions [60]. We calculated the C_{11} of NMC111 by changing atomic charges, then compared them with the available values in the literature; Table 3 summarizes the results. The stiffness calculated with 60% of formal charges shows good agreement. Therefore, all simulations in this study were carried out with 60% of formal charges. The partial charges used for the calculation of Coulombic interactions are summarized in Table 4.

Table 3. Change in stiffness of the NMC111 with different charges.

NMC	Charge Weight	C_{11} (GPa)	
		Simulation Result	Reference Result [Ref.]
111	50%	142.0	199 [50]
	60%	195.7	
	80%	326.5	
	100%	490.4	

Table 4. Partial charges used for Coulombic interactions.

Ions	Partial Charges (q_i)
Li^+	+0.6
Ni^{2+}	+1.2
Ni^{3+}	+1.8
Mn^{4+}	+2.4
Co^{3+}	+1.8
O^{2-}	−1.2

3. Results and Discussion

3.1. Model Validation

To validate the accuracy of the model, we compare the lattice constants with the available values in the literature. The lattice constants were calculated by averaging the size of the equilibrated system over time. Table 5 summarizes the results. Although an excess of 3.6% at maximum is shown, they demonstrate good agreement overall. As the Ni ratio increases, the calculated lattice constants show a decreasing trend, most notably along the z-axis, in agreement with previous studies [48]. As the Ni ratio increases, the proportion of Ni^{3+} ions also increases, leading to stronger electrostatic interactions with neighboring O^{2-} . This enhanced attraction draws the oxygen ions closer, resulting in a contraction of the lattice. Since nickel and oxygen ions are arranged in layers, this effect is pronounced along the z-axis.

Table 5. Comparison of lattice constants between the simulation results and reference.

NMC	Simulation Result (Å)		Reference Result (Å)		
	x-Axis	z-Axis	x-Axis	[Ref.]	z-Axis
111	2.955	14.703	2.862	[61]	14.238
			2.865	[62]	14.249
			2.865	[63]	14.25
			2.868	[47]	14.213
			2.8125	[64]	14.42
532	2.951	14.685	2.925	[64]	14.42
622	2.936	14.610	2.8683	[65]	14.2241
			2.91	[64]	14.39
721	2.942	14.638	2.8565	[66]	14.1576
811	2.932	14.590	2.83	[67]	14.3

3.2. Elastic Modulus

Figure 3a–c shows the stress–strain curves of all the selected compositions within the elastic range; the stiffness is determined by measuring the slope of the curves. Figure 3d then shows the calculated elasticities. C_{11} exhibits the largest value across all compositions, ranging from approximately 200 to 290 GPa for NMC111 to NMC811. C_{22} follows a similar trend with slightly lower values, ranging from 190 to 260 GPa for NMC111 to NMC811. C_{33} is significantly lower, which is expected, since atomic interactions in the layer direction are weaker than in the plane directions. As the Ni content increases, C_{33} also shows an increasing trend from (130 to 145) GPa for NMC111 to NMC811, although the difference is not significant.

The increased stiffness with higher Ni contents raises more possibility of the generation of larger stress in materials upon volume change due to lithiation/delithiation. Therefore, the simulation results suggest that the increase in stiffness with higher Ni contents can be a factor that contributes to the poor mechanical stability of Ni-rich NMC. In addition, the calculated elasticity ranges from 186.3 to 293.2 GPa, showing significant variation upon different compositions; this indicates that compositional tuning, particularly the manipulation of nickel content, can be an effective strategy to tailor the mechanical properties of NMC materials.

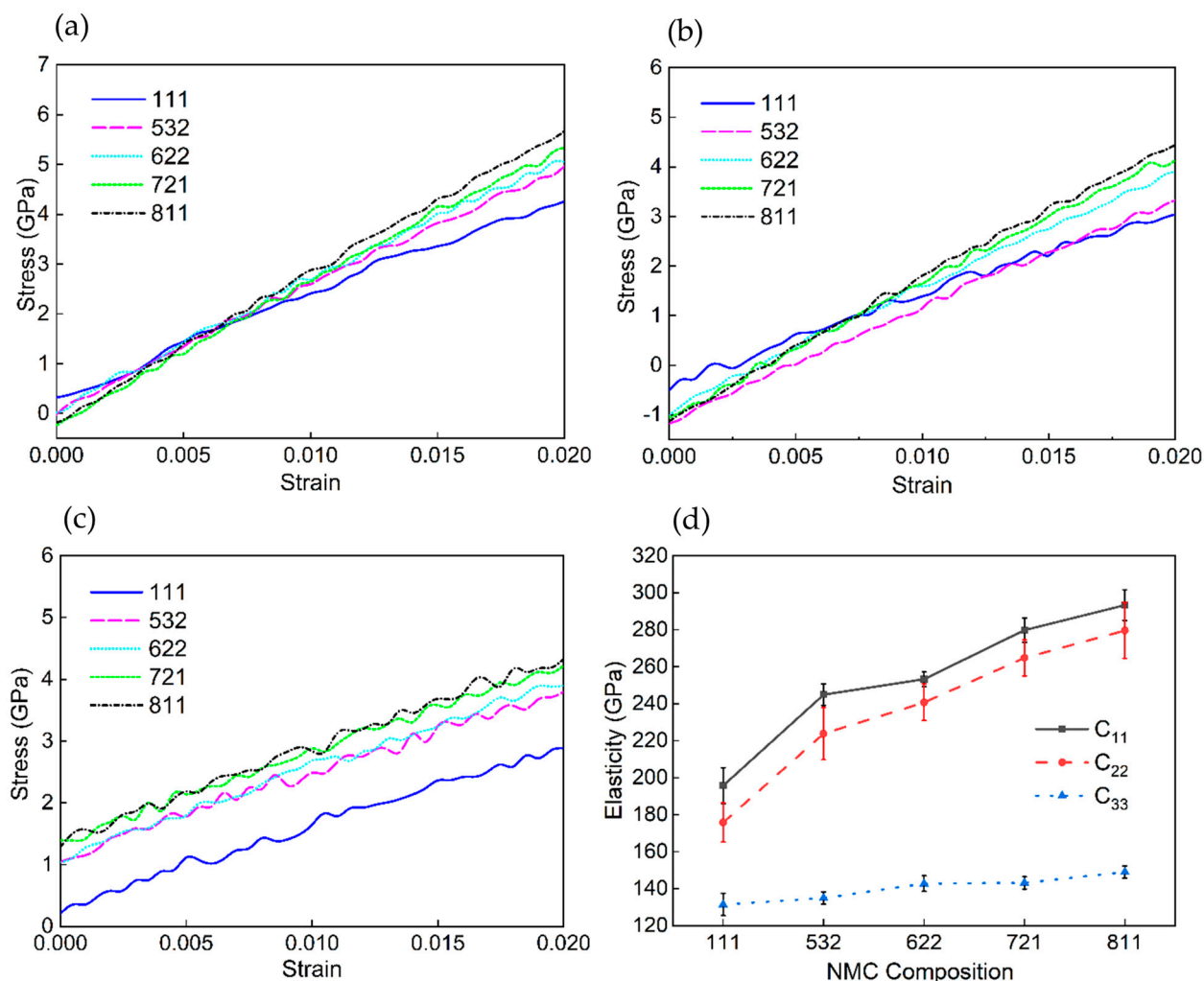


Figure 3. Stress–strain curves of different NMC compositions with stretching in the (a) x-direction, (b) y-direction, and (c) z-direction; and (d) calculated stiffness of the C_{11} , C_{22} , and C_{33} for NMC111, NMC532, NMC622, NMC721, and NMC811.

3.3. Component Analysis

The calculated stresses are decomposed to investigate the factors influencing the change in elasticity of different compositions. Figure 4 shows the three main contributions to stress of kinetic, long-range electrostatic, and pairwise interactions. The total stresses represent the stress values at 0.02 strain. Since the stress–strain curves start from zero, the component breakdown of specific stress can be used for that of stiffness. The decomposition shows the pairwise energy to be the predominant contributor to stiffness.

As the Ni^{3+} content increases, the Ni–O bond length shortens, strengthening local interactions. This increased bonding localization enhances charge neutralization and increases electrostatic screening over longer distances, thereby reducing long-range electrostatic interactions. As a result, the overall electrostatic stress contribution decreases from NMC111 to NMC811, while short-range pairwise interactions become more dominant.

The pairwise energy is further decomposed into atomic contributions of Li^+ , Ni^{2+} , Ni^{3+} , Mn^{4+} , Co^{3+} , and O^{2-} . Figure 5 shows the contribution of each ion to stiffness. As the nickel content increases, the green and cyan curves increase, indicating that the two species of Ni^{3+} and O^{2-} are primarily responsible for the increase in stress. As the nickel content increases, the charge of nickel increases, which enhances the repulsive forces between nickel ions and neighboring oxygen ions, resulting in an increase in the overall energy and consequently, the stress within the system. Although not as much as Ni^{3+} , the contribution

of Li^+ also increases as Ni content increases. The shortened Ni–O bond length compacts the overall crystal structure, slightly reducing the spacing of Li^+ and surrounding oxygen ions. As a result, Li^+ experiences stronger short-range repulsions from surrounding Ni and O ions. This leads to a higher pairwise interaction contribution from Li^+ , even though its absolute number remains unchanged. This analysis highlights the intricate interplay between particle interactions and the resultant mechanical properties of these materials.

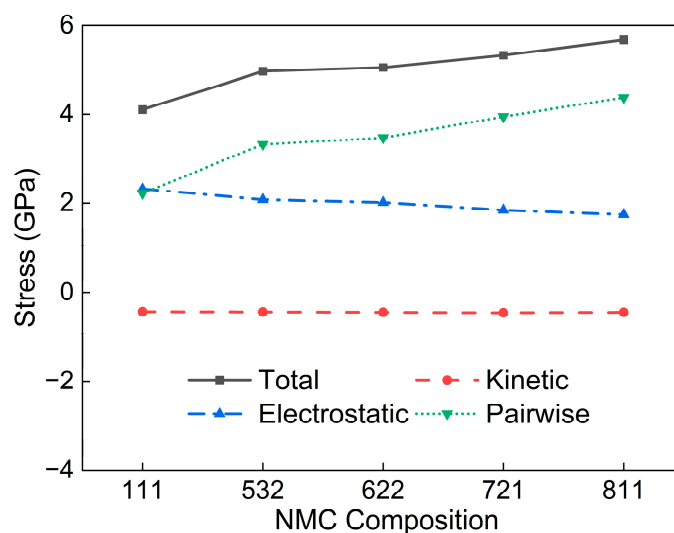


Figure 4. Stress decomposition into the kinetic, electrostatic, and pairwise energy contribution.

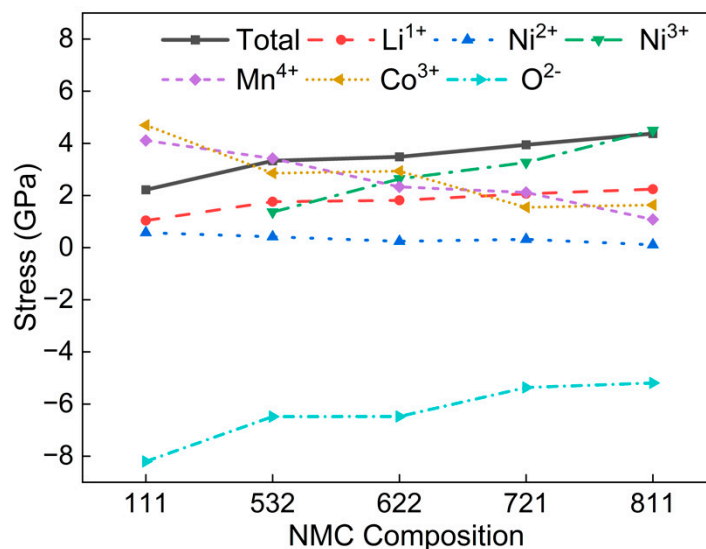


Figure 5. Decomposition of pairwise energy into individual atom contribution of the Li^+ , Ni^{2+} , Ni^{3+} , Mn^{4+} , Co^{3+} , and O^{2-} .

4. Conclusions

In this study, we investigated the change in the mechanical properties of NMC materials with Ni content. The elastic constants C_{11} , C_{22} , and C_{33} were computed for five different NMC compositions. NMC811, which contains the largest nickel content, exhibits the largest stiffness. Our decomposition analysis identified that the primary contributor to stress is the pairwise interaction, which is particularly influenced by the charge states of nickel and oxygen ions. As the nickel content increases, changes in ionic interactions result in the increase in repulsive forces, elevating the overall energy and stress in the system. The results provide useful data of the mechanical properties of NMC materials for continuum

models of Li-ion batteries, while also emphasizing the effect of the composition on the mechanical properties.

Author Contributions: Conceptualization, S.L. and I.U.H.; methodology, I.U.H.; software, I.U.H.; validation, I.U.H.; formal analysis, I.U.H.; investigation, I.U.H.; resources, S.L.; data curation, I.U.H.; writing—original draft preparation, I.U.H.; writing—review and editing, S.L.; visualization, I.U.H.; supervision, S.L.; project administration, S.L.; funding acquisition, S.L. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Ministry of Science and ICT, grant number 2022R1A2C1003003.

Data Availability Statement: The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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

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