SUPPLEMENTARY INFORMATION

**Unlocking the Potential of Open-Tunnel Oxides: DFT-Guided Design and Machine Learning-Enhanced Discovery for Next- Generation Industry-Scale Battery Technologies**

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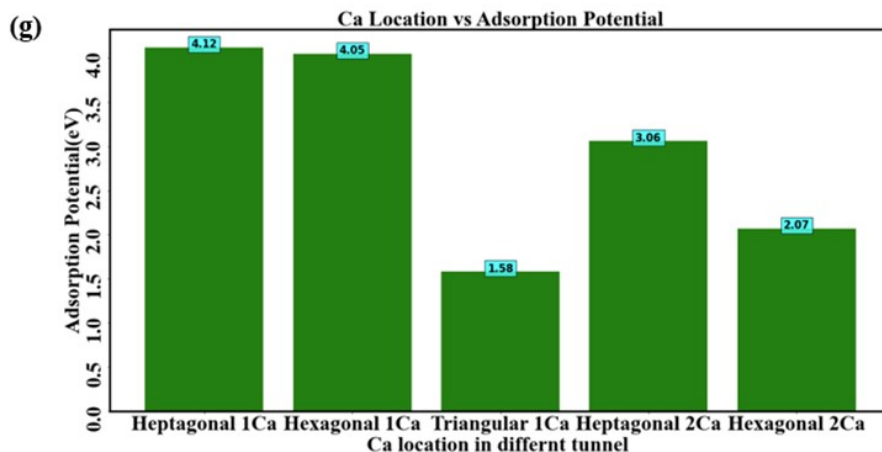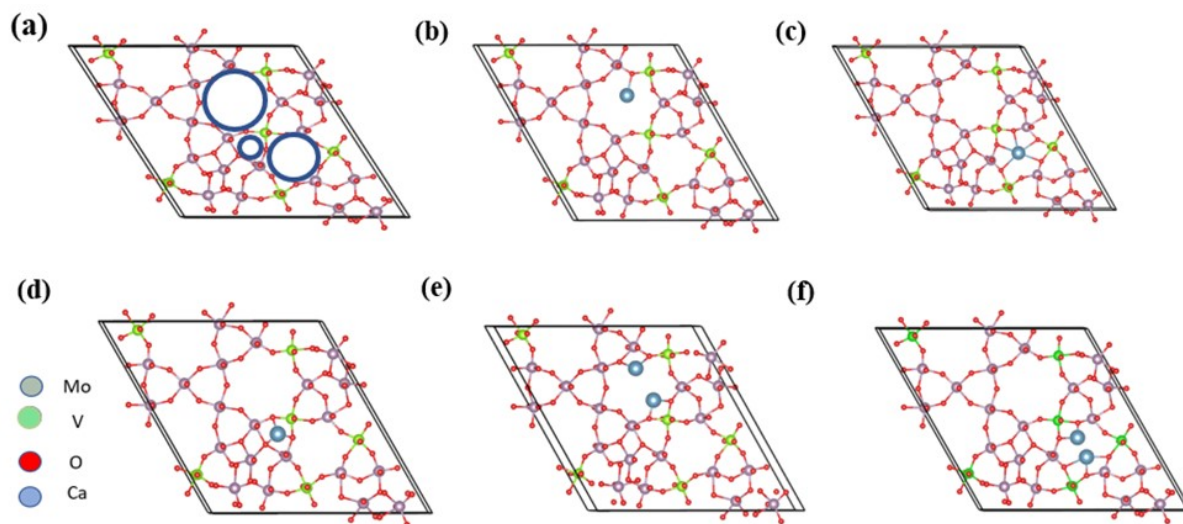
Figure S1. Incorporating Ca into the Triangular MoVO Structure: (a) The original Trigonal MoVO structure without Ca. (b) Placement of a single Ca atom within the heptagonal channel. (c) Positioning a single Ca atom within the hexagonal channel. (d) Positioning a single Ca atom within the trigonal channel. (e) Accommodating two Ca atoms within the heptagonal channel. (f) Accommodating two Ca atoms within the hexagonal channel. (g) Determining the Ca atom's location in relation to its adsorption potential. The heptagonal channel emerges as the most favorable site for Ca, outperforming other channels. The triangular channel is incapable of accommodating two Ca atoms due to its limited dimensions.
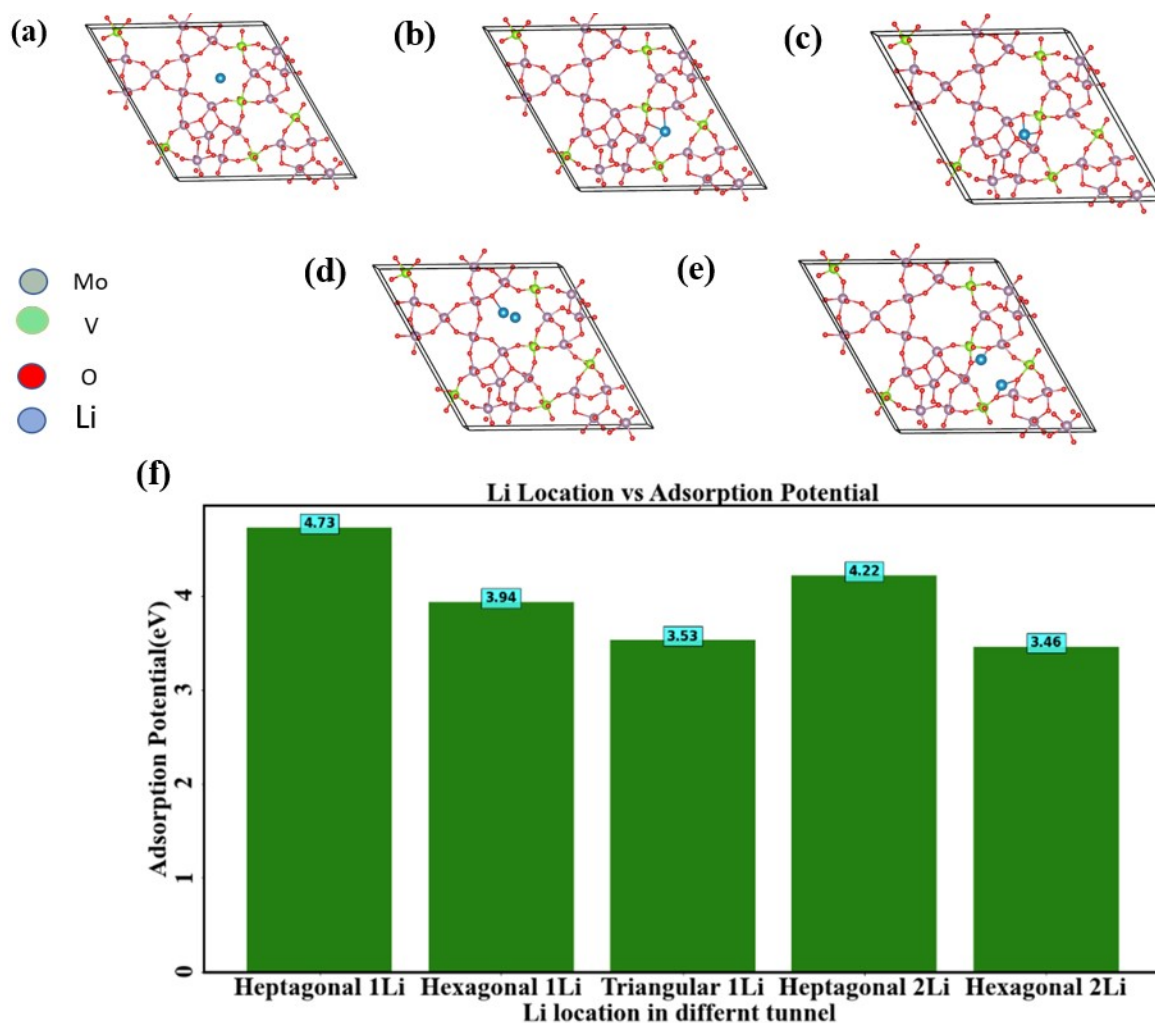
Figure S2. Introducing Li into the triangular MoVO structure: (a) Placement of a single Li atom within the heptagonal channel. (b) Positioning a single Li atom within the hexagonal channel. (c) Inserting a single Li atom into the trigonal channel. (d) Accommodating two Li atoms within the heptagonal channel. (e) Incorporating two Li atoms within the hexagonal channel. (f) Determining the location of Li ions with respect to their adsorption potential. The adsorption favorability of Li ions within the channels varies based on the dimensions of the respective channels.
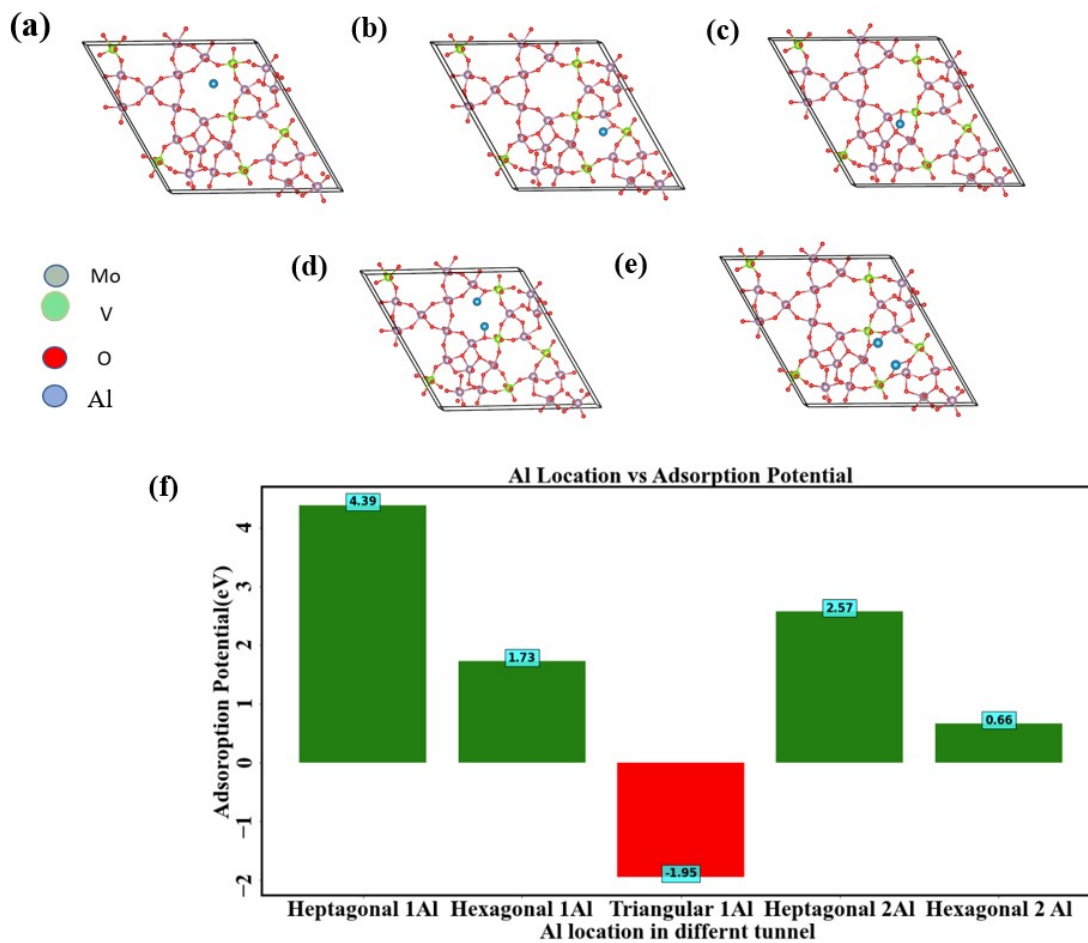
Figure S3. Introducing Al into the Triangular MoVO Structure: (a) Placement of a single Al atom within the heptagonal channel. (b) Positioning a single Al atom within the hexagonal channel. (c) Inserting a single Al atom into the trigonal channel. (d) Accommodating two Al atoms within the heptagonal channel. (e) Incorporating two Al atoms within the hexagonal channel. (f) Determining the location of Al ions with respect to their adsorption potential. It is noteworthy that Al, due to its high charge density, exhibits a negative adsorption potential when inserted into the triangular channel (refer to Table 1).
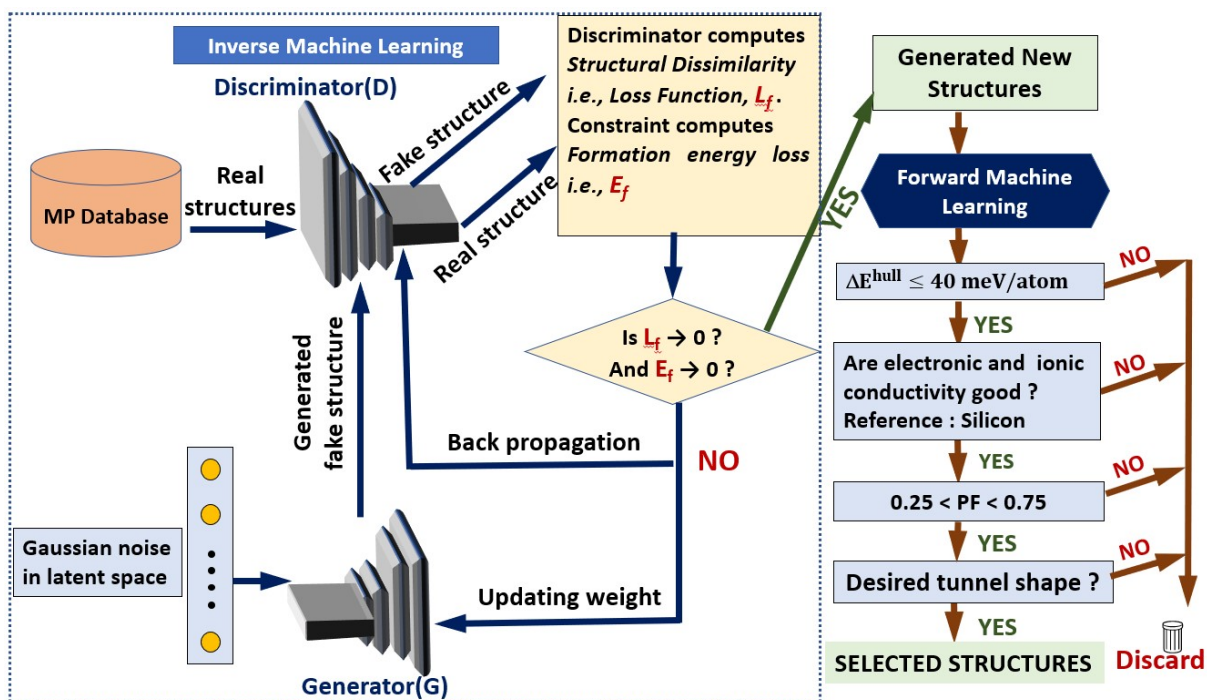
Figure S4: Illustration of the methodology for discovering novel open-tunnel oxide materials through a ML approach. The process involves two stages. Firstly, inverse machine learning utilizes a GAN architecture to generate practical test configurations of open-tunnel oxide materials. Secondly, the generated test structures are evaluated for specific traits using a traditional forward ML model. This facilitates the identification of promising candidates for multivalent-ion batteries. Reproduced with permission[1].Copyright 2023, Wiley Online Library Limited.

References

(1)   Lakhnot, A. S.; Panchal, R. A.; Datta, J.; Mahajani, V.; Bhimani, K.; Jain, R.; Datta, D.; Koratkar, N. Intercalation Hosts for Multivalent-Ion Batteries. *Small Struct.* **2022**, *2200290*, 2200290. https://doi.org/10.1002/sstr.202200290.