DATA-DRIVEN MODELING AND PHYSICS-INFORMED MACHINE LEARNING FOR GLASS DISCOVERY

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Data-driven Modeling and Physics-informed Machine Learning for Glass Discovery

by

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Dedicated to my family

Certificate

This is to certify that the thesis entitled *Data-driven Modeling and Physics-informed Machine Learning for Glass Discovery* is being submitted by Mr. Ravinder to the Indian Institute of Technology Delhi for the award of the degree of Doctor of Philosophy. This is a record of the research work and is entirely carried out by him under my supervision and guidance. The research report presented in this thesis has not been submitted for the award of any other degree or diploma.

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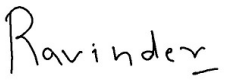
Declaration

I hereby declare that the thesis contains the work carried out in fulfillment of the requirements for the award of the degree of Doctor of Philosophy under the guidance and supervision of Prof. N. M. Anoop Krishnan, Associate Professor, Department of Civil Engineering, IIT Delhi. The contents of the dissertation, including text, tables, figures, etc., have not been reproduced from other reports, theses, etc. Wherever reproduction has been made from books, journals, reports, manuals, websites, etc., the sources have been duly acknowledged and referenced appropriately.



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Abstract

The progress of human civilizations has been dependent on material discovery throughout history. With the discovery of iron, it replaced most farming tools such as sickle, plow tips, etc., which were earlier made from bronze and stones, leading to efficient farming. Also, the early use of iron in weapons made some civilizations dominant in warfare. Similarly, glasses have been significantly affecting human civilizations since their first use as an ornament. Today, inorganic glasses are being used in numerous industries as they provide unique properties with an astronomical number of possible glass compositions (*glasses* and *inorganic glasses* are being used interchangeably throughout the whole work). Glasses find their applications in computer screens, optical lenses, bio-active implants, optical fiber, the automotive industry, nuclear waste management, laboratory glassware, electrolytes, etc. The glass composition is not fixed, unlike crystalline materials, and can be formed in almost any stoichiometry. The glass property can significantly change with a slight change in glass composition or the addition and removal of glass components. Hence, finding an optimum glass composition for a targeted property is a challenging task. A naive trial and error method or heuristic approach with experiments can get easily overwhelmed with the number of possibilities of a glass composition. Therefore, we use machine learning (ML) methods to model the composition–property relationship for glasses. These relationships can be used with optimization algorithms to find the optimum glass composition given some constraints.

The research work use machine learning methods to model numerous glass properties (25 glass properties, namely (a) Abbe number, (b) annealing point, (c) bulk modulus, (d) compressive strength, (e) crystallization temperature, (f) density, (g) dielectric constant, (h) electrical conductivity, (i) Vickers hardness, (j) refractive index, (k) shear modulus, (l) softening point, (m) strain point, (n) liquidus temperature, (o) Littleton temperature, (p)

thermal expansion coefficient (TEC), (q) tensile strength, (r) glass transition temperature, (s) thermal conductivity, (t) thermal shock resistance, (u) infrared (IR) transmittance, (v) solar transmittance, (w) visible transmittance, (x) ultraviolet (UV) transmittance, and (y) Young's modulus). The research work introduces glass selection chats which can be used to identify optimum glass compositions by optimizing two-glass properties simultaneously.

Further, these ML models are then used with explainable ML techniques (SHAP) to understand the compositional control of particular glass properties. The research capture the already known composition–property relationships such as Lowestein's rule, boron anomaly, etc. It also elucidates the known phenomenon, such as the increase in thermal expansion coefficient for low network-former glasses, decrease in liquidus temperature for high network-modifier glass, etc. Finally, the models were tested on a new glass family $0.5P_2O_5$–(1–x)CaO–xMgO, for which ML models predict an accurate trend for density, hardness, and refractive index. The effect of MgO mol% is captured by SHAP values elucidating the effectiveness of SHAP analysis in understanding the composition–property relationship.

The composition–property relationships in inorganic glasses are complex, which ML methods can capture and explain with significant accuracy. The mechanisms behind these relationships can not be captured with black-box ML models, and therefore molecular dynamics or DFT simulations are needed to reveal these mechanisms. The DFT simulations are costly, and interatomic potential for MD simulations is not always available. Therefore, the research introduces two frameworks, namely MCLNN, and LGNN, which can be used to approximate and machine learning interatomic potentials from DFT simulation trajectories. MCLNN can learn simple systems with pairwise interactions such as LJ systems, harmonic spring systems, etc., while LGNN can learn systems with complex interactions such as liquid silica systems. LGNN is able to capture structural features such as pair distribution function, bond angle distribution, etc., with high accuracy. The MD simulations can only simulate systems with nanometer size therefore, an upscaling of material property is required to

simulate the system at mesoscale (such as peridynamics simulations) with a large system size. The research introduces a software package, PeriDyn, which provides an interface to write arbitrary peridynamics workflow with machine learning frameworks. The package can be used to learn the peridynamics material model parameters from MD trajectories to upscaling the material property to mesoscale.

Overall, the research explores the use of machine learning methods in modeling composition–property relationships for inorganic glasses, explaining composition dependence of inorganic glass properties, understanding mechanisms with MD simulations, and up-scaling of material response to mesoscale using MD or DFT simulation trajectories.

Keywords: Inorganic glass, glass, machine learning, explainable ML, PINN, Physics-informed ML, ML interatomic potential, upscaling material model, peridynamics

सार

मानव सभ्यताओं की प्रगति पूरे इतिहास में नए पदार्थ की खोज पर निर्भर रही है। लोहे की खोज के साथ अधिकांश खेती के औजार जैसे हंसिया, हल की नोक आदि बदल गए, जो पहले कांसे और पत्थरों से बनाए जाते थे, जिससे कुशल खेती हुई। हथियारों में लोहे के शुरुआती उपयोग ने कुछ सभ्यताओं को युद्ध में प्रभावी बना दिया जिससे लोहे को प्राप्त करना आवश्यक हो गया। इसी तरह, शीशा मानव सभ्यताओं को एक आभूषण के रूप में पहली बार उपयोग करने के बाद से महत्वपूर्ण रूप से प्रभावित कर रहा है। आज, कई उद्योगों में शीशे का उपयोग किया जा रहा है क्योंकि शीशा अशंख्य संरचनाओं के साथ प्राप्त किया जा सकता है एवं हर संरचना अलग गुण प्रदान करती है। शीशा कंप्यूटर स्क्रीन, ऑप्टिकल लेंस, बायो-एक्टिव इम्प्लांट्स, ऑप्टिकल फाइबर, ऑटोमोटिव उद्योग, परमाणु अपशिष्ट प्रबंधन, प्रयोगशाला परखनली, इलेक्ट्रोलाइट्स इत्यादि में प्रयोग किया जाता है। शीशे की संरचना तय नहीं होती है जिससे यह लगभग किसी भी अनुपात में बनाया जा सकता है। शीशे की संरचना में मामूली बदलाव या शीशे के घटकों को जोड़ने और हटाने पर शीशे के गुण महत्वपूर्ण रूप से बदल सकते है। इसलिए, लक्षित गुण के लिए शीशे की इष्टतम संरचना खोजना एक चुनौतीपूर्ण कार्य है। शीशे की इष्टतम संरचना खोजने के लिए एक सहज ट्रायल–एंड–एरर विधि के साथ प्रयोग शीशे की संरचना की संभावनाओं की संख्या से आसानी से अभिभूत हो सकते है। इसलिए, हम शीशे के लिए रचना–गुण–संबंध को मॉडल करने के लिए मशीन लर्निंग विधियों का उपयोग करते हैं। शीशे की इष्टतम संरचना को खोजने के लिए इन मॉडल्स का उपयोग एक अनुकूलन एल्गोरिदम के साथ उपयोग किया जा सकता है।

अनुसंधान कार्य कई शीशे के गुणों (25 शीशे के गुणों (a) एब्बे नंबर, (b) एनीलिंग पॉइंट, (c) बल्क मॉड्यूलस, (d) कंप्रेसिव स्ट्रेंथ, (e) क्रिस्टलिज़ेशन तापमान, (f) घनत्व, (g) परावैद्युत स्थिरांक, (h) विद्युत चालकता, (i) विकर्स हार्डनेस, (j) अपवर्तक सूचकांक, (k) शियर मॉड्यूलस, (l) सॉफ्टनिंग पॉइंट, (m) स्ट्रेन पॉइंट, (n) लिक्विडस तापमान, (o) लिटलटन तापमान, (p) ताप विस्तार गुणांक (TEC), (q) टेंसिल स्ट्रेंथ, (r) ग्लास ट्रांजीशन तापमान, (s) थर्मल चालकता, (t) थर्मल शॉक प्रतिरोध, (u) अवरक्त (IR) संप्रेषण, (v) सौर संप्रेषण, (w) विज़िबल संप्रेषण, (x) पराबैंगनी (UV) संप्रेषण, और (y) यंग का मापांक) को मॉडल करने के लिए मशीन लर्निंग के तरीकों का उपयोग करता है। यह शोध कार्य शीशा-चयन-चार्ट पेश करता है जिसका उपयोग शीशे के दो गुणों को एक साथ अनुकूलित करके इष्टतम शीशे की पहचान करने के लिए किया जा सकता है।

इसके अलावा, शीशे के विशेष गुणों के संरचनागत नियंत्रण को समझने के लिए इन मशीन लर्निंग मोडलों का उपयोग SHAP जैसे व्याख्यात्मक मशीन लर्निंग तकनीकों के साथ किया जाता है। अनुसंधान पहले से ही ज्ञात संरचना–गुण संबंधों जैसे की लोवेस्टीन के नियम, बोरोन विसंगति आदि को पकड़ने में सक्षम है। यह ज्ञात नियमो को भी प्रकट करता है, उधारणतः कम नेटवर्क–फॉर्मर के होने पर ताप विस्तार गुणांक में वृद्धि, उच्च नेटवर्क–मॉडिफिएर के लिए लिक्विड्स तापमान में कमी आदि। अंततः, मशीन लर्निंग मॉडलों का परीक्षण एक नए शीशे ($0.5P_2O_5$-(1-x)CaO-xMgO) पर किया गया, जिसके लिए मशीन लर्निंग मॉडल्स घनत्व, कठोरता और अपवर्तक सूचकांक के लिए सटीक अनुमान लगाते हैं। इसमें MgO का प्रभाव SHAP मूल्यों द्वारा सटीक दर्शाया जाता है, जो संरचना–गुण सम्बन्ध को समझने में SHAP विश्लेषण की प्रभावशीलता को स्पष्ट करता है।

शीशे में संरचना–गुण सम्बन्ध जटिल होता हैं, जिन्हें मशीन लर्निंग विधियां सटीकता के साथ पकड़ और समझ सकती है। इन संबंधों के पीछे के तंत्र को ब्लैक–बॉक्स मशीन लर्निंग विधियों के साथ नहीं समझा जा सकता है। इसलिए इन तंत्रों को समझने के लिए MD सिमुलेशन या DFT सिमुलेशन की आवश्यकता होती है। DFT सिमुलेशन काफी समय लेते हैं और MD सिमुलेशन के लिए इंटरटोमिक पोटेंशियल हमेशा उपलब्ध नहीं होता है। इसलिए, अनुसंधान दो रूपरेखाओं को पेश करता है, MCLNN एवं LGNN, जिसका उपयोग DFT सिमुलेशन प्रक्षेपवक्र से मशीन लर्निंग इंटरटोमिक पोटेंशियल को अनुमानित करने के लिए किया जा सकता है। MCLNN सरल प्रणालियों जैसे युग्मानुसार स्थितिज ऊर्जा, LJ प्रणाली, हार्मोनिक स्प्रिंग प्रणाली आदि सीख सकता है, जबकि

LGNN तरल सिलिका प्रणाली जैसे जटिल इंटरैक्शन वाली प्रणाली सीख सकता है। LGNN संरचनात्मक विशेषताएं जैसे युग्म बंटन–फलन, कोण बंटन–फलन इत्यादि को उच्च सटीकता के साथ प्राप्त करने में सक्षम है। MD सिमुलेशन केवल नैनोमीटर आकार की प्रणालियों का ही अनुकरण कर सकते हैं, इसलिए बड़े आकार (मेसोस्केल) की प्रणालियों (उधारणतः peridynamics सिमुलेशन) का अनुकरण करने के लिए पदार्थ की गुणता की अपस्केलिंग की आवश्यकता होती है। अनुसंधान एक सॉफ्टवेयर पॅकेज, PeriDyn पेश करता है, जो मशीन लर्निंग फ्रेमवर्क के साथ मनमानी कार्य प्रणाली लिखने के लिए एक इंटरफेस प्रदान करता है। सॉफ्टवेयर पॅकेज का उपयोग एमडी प्रक्षेपवक्र से peridynamics मटेरियल मोडल मापदंडों को सीखने के लिए किया जा सकता है, जिससे पदार्थ की गुणता की अपस्केलिंग की जा सकती है।

कुल मिलाकर, अनुसंधान शीशे के संरचना–गुण संबंध, शीशे के गुणों की संरचना निर्भरता की व्याख्या करना, MD सिमुलेशन से तंत्र को समझना और पदार्थ के गुणों की अपस्केलिंग करने इत्यादि में मशीन लर्निंग विधियों के उपयोग की पड़ताल करता है।

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Nomenclature

Roman Symbols

F	interatomic force

Acronyms / Abbreviations

ACE	atomic cluster expansion
ALE	accumulated local effects
CART	classification and regression tree
CGCNN	crystal graph convolution neural network
CNN	covariant compositional networks
DeepLIFT	Deep Learning Important Features
DFT	density function theory
DNN	deep neural network
FFNN	feed forward neural network
FSDP	first sharp diffraction peak
GAP	gaussian approximation potential

GNNFF graph neural network force field

GNN graph neural network

GPR gaussian process regression

LGNN lagrangian graph neural network

LIME local interpretable model-agnostic explanations

LJ Lennard-Jones

MCLNN momentum conserving lagrangian neural network

MD molecular dynamics

MEGNet MatErials graph network

ML machine learning

MPGNN message passing graph neural network

MTP moment tensor potential

NN neural network

NNP neural network potential

PCA principal component analysis

PDF pair distribution function

PES potential energy surface

PINN physically informed neural network

RF random forest

SHAP shapely additive explanations

SNAP spectral neighbor analysis potential

SOAP smooth overlapping atomic positions

SVM support vector machine

List of publications

Papers featuring in the thesis work:

1. Ravinder, R; Sridhara, Karthikeya H; Bishnoi, Suresh; Grover, Hargun Singh; Bauchy, Mathieu; Jayadeva, J; Kodamana, Hariprasad; Krishnan, NM Anoop; Deep Learning Aided Rational Design of Oxide Glasses, Materials Horizons, 2020, Royal Society of Chemistry
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