

**EFFECT OF IRRADIATION ON MECHANICAL AND
STRUCTURAL PROPERTIES OF BOROSILICATE
GLASSES**

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**DEPARTMENT OF CIVIL ENGINEERING
INDIAN INSTITUTE OF TECHNOLOGY DELHI
JULY 2023**

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STRUCTURAL PROPERTIES OF BOROSILICATE
GLASSES**

By

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DEPARTMENT OF CIVIL ENGINEERING

Submitted

in fulfilment of the requirements of the degree of Doctor of Philosophy

to the



INDIAN INSTITUTE OF TECHNOLOGY DELHI

JULY 2023

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.

Rajesh Kumar

2017CEZ8528

July 2023

Certificate

This is to certify that the thesis entitled *Effect of Irradiation on Mechanical and Structural Properties of Borosilicate Glasses* is being submitted by Mr. Rajesh Kumar 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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July 2023

Acknowledgements

First and foremost, it is a pleasure to express sincere thanks to my supervisor, Prof. N. M. Anoop Krishnan, for his constant support, availability, dedication, patience, and passion for research. Prof. N. M. Anoop Krishnan has always been very keen on finding new opportunities for me to explore different research avenues and facilitated me to attend various training sessions, workshops, seminars to improve my skills and knowledge. He allowed me to enjoy my academic freedom to the fullest and taught me how to read, write, think, and present confidently as a scientific researcher.

I would like to express my gratitude to my research committee: internal expert Prof. Shashank Bishnoi, external expert Prof. Saswata Bhattacharya, and the Chairman Prof. D. R. Kaushal for their valuable input at different stages of my research work. I also acknowledge the Civil Engineering Department and Ministry of Education, Department of Higher Education, Government of India for the financial support for lab equipment, workshops, and conferences.

I would like to thank my mother, Subhadra Kumari, and father, Joginder Singh, who stood with me all the time and for their unconditional love, encouragement, and support throughout my life. The major credit for this degree belongs to them. Thanks to my loving wife, Preeti Soni, who always stood by my side and tolerated my absence at most times.

I acknowledge the computational resources and support provided by HPC, IIT Delhi. I would like to thank the members of the M3RG lab, especially Ravinder, Tanu Pittie and Amreen Jan, for their love and support. I would also like to thank Dr. B. N. Tiwari for friendly and philosophical discussion during a part of this work. Finally, I thank the almighty, who has granted me countless blessings, opportunities, knowledge, and energy to write this thesis.

Abstract

Nuclear waste glasses are used to contain and dispose of the waste generated in nuclear power plants and other similar facilities. Most of the radioactive waste emits large amounts of radiation and has long half-life. The management of high-level waste is carried out using a process called vitrification. Borosilicate glasses are used for vitrification as the base glass due to their high-temperature resistance and chemical stability. One more prominent advantage of borosilicate glasses is that they maintain their formability across large compositions. While various experiments and simulation studies have been conducted to predict damage due to irradiation in a particular class of borosilicate glasses used in nuclear waste immobilization, the effect of irradiation on the structural and mechanical properties of different variants of borosilicate glasses is poorly understood. Thus, it is essential to understand the effect of glass compositions and irradiation on the structure and properties of a series of such glasses.

The central aim of this thesis is to estimate the severity of irradiation and its effect on the mechanical and structural properties of borosilicate glasses of varying compositions. The mechanical properties investigated in the thesis are the density, fracture toughness, brittleness index, and fracture energy. Also, the study aims to develop composition-independent potential parameters for studying the sodium boroaluminosilicate (NABS) glasses.

Molecular modeling played a central role in the discovery of new materials and in studying the properties of these glasses at atomic scales. To achieve this, molecular dynamics MD-based studies were conducted to calculate the aforementioned structural and mechanical properties of borosilicate glass variants. To fully harness the power of modern parallel computers, a Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) was used, which is a scalable molecular dynamics code including long-range Coulomb interactions. BKS potential parameters have been specifically calibrated in-house to simulate the effect of radiation on glasses and their mechanical and structural properties. We simulated glasses using the melt-quench method and run fracture simulation on both irradiated and pristine glass structures.

The results of the simulations revealed a composition-dependent transition in the volumetric response of borosilicate glasses under irradiation. It was observed that the borate-rich compositions tend to swell, whereas the silica-rich glasses tend to densify. Also, the structural and mechanical properties of irradiated and pristine glasses were compared, and the simulated results were validated against published experimental results and simulations from the existing literature. The effect of cooling rate and irradiation on the structure and glass-forming ability

of glasses was also studied. The analysis showed that glass transition in forbidden and allowable regimes depends on the method of glass formation. As mentioned before, fracture simulation on pristine and irradiated glasses showed that the irradiation could be a factor in the increase of ductility in irradiated glasses relative to their pristine counterpart. For the development of NABS potential parameters, we examined the mechanical properties, such as the density and elastic modulus. The developed potential parameters were also validated against both experiments as well as previously simulated results.

सार

परमाणु अपशिष्ट कांच का उपयोग परमाणु ऊर्जा संयंत्रों और अन्य समान प्रकार के सुविधाओं में उत्पन्न होने वाले अपशिष्ट को संग्रहित और निपटान करने के लिए किया जाता है। अधिकांश विकर्णीय अपशिष्ट बहुत अधिक विकिरण उत्पन्न करते हैं और उनकी अर्धायु लंबी होती है। उच्च स्तरीय अपशिष्ट का प्रबंधन एक प्रक्रिया जिसे विट्रीफिकेशन कहा जाता है के द्वारा किया जाता है। बोरोसिलिकेट कांच उच्च तापमान प्रतिरोध और रासायनिक स्थिरता के कारण विट्रीफिकेशन के लिए मूल कांच के रूप में उपयोग किया जाता है। बोरोसिलिकेट कांच का एक और प्रमुख लाभ यह है कि वे अपनी बनावटीयता को बड़े पैमाने पर बरकरार रखते हैं। हालांकि, परमाणु अपशिष्ट संबंधीकृत एक विशेष प्रकार के बोरोसिलिकेट कांच के नुकसान की पूर्वानुमान और यांत्रिकीय गुणों पर विपरीत प्रभाव को प्रभावित करने के लिए विभिन्न प्रयोग और सिमुलेशन अध्ययन किए गए हैं, लेकिन अपशिष्ट के विभिन्न प्रकार के बोरोसिलिकेट कांच की संरचनात्मक और यांत्रिक गुणों पर परमाणु विकिरण के प्रभाव को समझना मुश्किल है। इसलिए, ऐसे कई कांचों की संरचना और गुणों पर कांच के संयोजन और परमाणु विकिरण के प्रभाव को समझना महत्वपूर्ण है।

इस थीसिस का मुख्य लक्ष्य विभिन्न कंपोजिशनस के बोरोसिलिकेट शीशों की संरचना और यांत्रिक गुणों पर विकिरण प्रभाव का आकलन करना है। थीसिस में जांचे गए यांत्रिक गुण हैं: घनत्व, टूटने की कठोरता, भंजनता सूचकांक और टूटने की ऊर्जा। साथ ही, अध्ययन का उद्देश्य सोडियम बोरोआल्यूमिनो सिलिकेट (NABS) शीशों की अध्ययन के लिए बनावट (कंपोजिशन)-स्वतंत्र पोटेंशियल पैरामीटर्स विकसित करना है।

आणविक मॉडलिंग ने नए सामग्री की खोज में और इन शीशों के गुणों का परमाणु स्तर पर अध्ययन करने में महत्वपूर्ण भूमिका निभाई। इसे प्राप्त करने के लिए, मोलेक्युलर डायनामिक्स (MD) पर आधारित अध्ययनों को संचालित किया गया, जिससे बोरोसिलिकेट शीशों की उपरोक्त संरचनात्मक और यांत्रिक गुणों का गणनन किया गया। आधुनिक पैरलल कंप्यूटरों की शक्ति का पूरी तरह से उपयोग करने के लिए, लार्ज-स्केल अटॉमिक/मोलेक्युलर मैसिव्ली पैरलल सिमुलेटर (LAMMPS) का उपयोग किया गया, जो दीर्घ-सीमा कूलम्ब अंतराक्रियाओं को शामिल करने वाला एक मॉलेक्युलर डायनामिक्स कोड है। BKS पैरामीटर्स विशेष रूप से शीशों पर विकिरण के प्रभाव की सिमुलेशन करने और उनके यांत्रिक और संरचनात्मक गुणों की सिमुलेशन करने के लिए आंतरगृहीत रूप से नियमित किए गए हैं। हमने मेल्ट-क्वेंच विधि का उपयोग करके शीशों की सिमुलेशन की और विकिरण से प्रभावित और प्राकृतिक शीशा संरचनाओं पर टूटने की सिमुलेशन चलाई।

आंकलनों के परिणामस्वरूप, विकिरण के तहत बोरोसिलिकेट ग्लास की वॉल्यूमेट्रिक प्रतिक्रिया में बनावट (कॉम्पज़िशन)-निर्भर संक्रमण प्रकट हुआ। यह पाया गया कि बोरेट-समृद्ध संयोजन स्वेल्(बढ़ जाना) करता है, जबकि सिलिका-समृद्ध शीशे सघन (डेन्सिफ़ायी) होते हैं। साथ ही, प्राकृतिक शीशों की संरचनात्मक और यांत्रिक गुणों की तुलना की गई, और प्रकाशित प्रयोगात्मक परिणामों और मौजूदा साहित्य से सिमुलेशन के खिलाफ तार्किक परिणामों की मान्यता प्राप्त की गई। शीशों की संरचना और काँच बनाने की क्षमता पर ठंडाई दर(कूलिंग रेट) और किरणों के प्रभाव(इरेडीएशन) का अध्ययन भी किया गया। विश्लेषण ने दिखाया कि फ़ॉर्बिडेन और अलावेबल रेज़ीम्ज़ में काँच का संक्रमण काँच गठन की पद्धति पर निर्भर करता है। पहले ही बताया गया था कि प्राकृतिक और किरणों से प्रभावित शीशों पर टूटने की सिमुलेशन ने दिखाया कि किरण प्राकृतिक-संबंधित किरणांतर में शीशों में डकटिलीटी में वृद्धि का कारक हो सकती है। NABS पोटेंशियल पैरामीटर्स के विकास के लिए, हमने घनत्व और प्रतिस्थापनीय कोशल(इलास्टिक मोडुलस) जैसे यांत्रिक गुणों की जांच की। पोटेंशियल पैरामीटर्स को पूर्ववत सिमुलेशन परीक्षण के साथ-साथ प्रयोगों के साथ भी मान्यता प्राप्त की गई।

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Publication related to thesis

- 1) **Kumar, R.**, Jan, A., Bauchy, M., & Krishnan, N. M. A. (2021). Effect of irradiation on the atomic structure of borosilicate glasses. *Journal of the American Ceramic Society*, 104(12), 6194–6206. <https://doi.org/10.1111/jace.18013>
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- 5) **Kumar, R.**, Jan, A., & Krishnan, N. M. A. Irradiation-induced forbidden borosilicate glasses. (*Submission pending*)
- 6) **Kumar, R.**, Ravinder, R., & Krishnan, N. M. A. Development of a transferable inter-atomic potential for boroaluminosilicate glasses. (*Submission pending*)
- 7) **Kumar, R.**, Ravinder, R., & Krishnan, N. M. A. Irradiation-induced ductility in fracture simulation due to irradiations. (*Manuscript in preparation*)

Other publications from PhD

- 1) Bhaskar, P., **Kumar, R.**, Maurya, Y., Ravinder, R., Allu, A. R., Das, S., et al. (2020). Cooling rate effects on the structure of 45S5 bioglass: Insights from experiments and simulations. *Journal of Non-Crystalline Solids*, 534, 119952. <https://doi.org/10.1016/j.jnoncrysol.2020.119952>
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Abbreviations and acronyms

Beest Kramer van Santen

(BKS), 5

Bridging oxygen

(BO), 57

Brittleness index

(B), 72

Coordination Number

(CN), 28

First Sharp Diffraction Peak

(FSDP), 30

Four coordinated boron

(B4), 82

Fracture energy

Gc, 72

Full Width at Half Maximum

(FWHM), 29

High-level waste

(HLW), 1

International Simple Glass

(ISG), 15

Large-scale Atomic/Molecular Massively Parallel

Simulator

(LAMMPS), 21

Linear Elastic Fracture Mechanics

(LEFM), 65

Low and intermediate-level waste

(LILW), 1

Medium range order

(MRO), 3

Molecular dynamics

(MD), 2

Non-bridging oxygen

(NBO), 57

Pair Distribution Function

(PDF), 26

Sodium aluminoborosilicate

(NABS), 10

Structure Factor

(S(Q)), 26

The American Society for Testing Materials

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Three coordinated boron

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