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How fluorine minimizes density fluctuations of silica glass: Molecular dynamics study with machine-learning assisted force-matching potential

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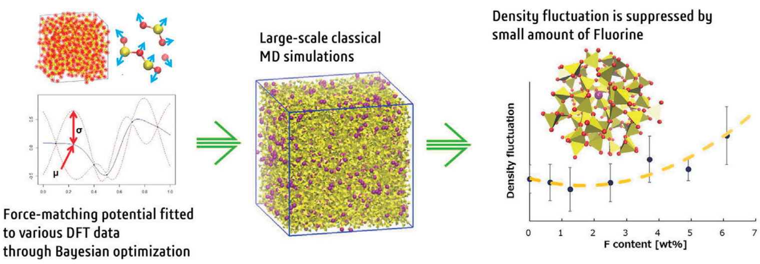
HIGHLIGHTS

- Accurate force-matching potential for F-doped silica was developed based on DFT calculations using Bayesian optimization.
- The new potential successfully reproduced anomaly of density change of silica and effect of F-doping to anomaly attenuation.
- Large-scale MD simulations unraveled how fluorine minimizes density fluctuation of silica glass.

GRAPHICAL ABSTRACT



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ABSTRACT

Transparent silica glass is an indispensable material for today's information technology society as an ultra-low loss optical fiber. Recently, further low-loss optical fibers are demanded due to rapidly expanding information. A small amount of fluorine doping is known to reduce the Rayleigh scattering further by reducing the density fluctuations of silica glass, even though the fluorine is an impurity for the pristine silica glass. This study devoted to answer the riddle of F-doping and an accurate force-matching potential optimized by machine-learning enables us to understand how fluorine remedies the disorders in silica glass. It was found that fluorine mainly replaces an oxygen in tetrahedral SiO_4 unit, but also forms five-fold Si, such as SiO_4F and SiO_3F_2 units. The former disrupts silica network, while the latter attenuates the network rigidity by loosening the five Si-O(F) bonds. Since the two local effects of fluorine spreads through the glass network, F-doping prompts silica glass to relax further even at temperature lower than the glass transition (fictive) temperature. Consequently, fluorine minimizes the density fluctuation (thus, Rayleigh scattering) of silica glass at around 1 wt%, although fluorine itself is an impurity for silica glass.

1. Introduction

Silica glass (a-SiO_2), one of the simplest oxide glasses, changes the physical and chemical properties by a variety of additives [1]. For instance, rare-Earth ions (such as erbium and ytterbium), and bismuth in a-SiO_2 induce emissions, and the doped glasses have been practically used as an optical fiber amplifier [2,3]. When TiO_2 is added into a-SiO_2, the doped a-SiO_2 exhibits low thermal expansion in a wide range of temperature [4,5]; thus the modified a-SiO_2 is suitable for photomask blanks and telescope lens. Fluorine is also a useful dopant that adds new functions to a-SiO_2 and glasses [6]. A small amount of fluorine

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