



# Structural study of lanthanum aluminoborate glasses combining molecular dynamics, nuclear magnetic resonance and neutron diffraction

Erwan Chesneau, Thibault Charpentier, Daniel Caurant, Rodolphe Pollet, Odile Majérus, Laurent Cormier

## ► To cite this version:

Erwan Chesneau, Thibault Charpentier, Daniel Caurant, Rodolphe Pollet, Odile Majérus, et al.. Structural study of lanthanum aluminoborate glasses combining molecular dynamics, nuclear magnetic resonance and neutron diffraction. Joint Meeting of DGG – USTV, May 2019, Nürnberg, Germany. cea-02329346

**HAL Id: cea-02329346**

**<https://cea.hal.science/cea-02329346>**

Submitted on 23 Oct 2019

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

# Structural study of lanthanum aluminoborate glasses combining molecular dynamics, nuclear magnetic resonance and neutron diffraction

Erwan Chesneau<sup>1</sup>, Thibault Charpentier<sup>1</sup>, Daniel Caurant<sup>2</sup>, Rodolphe Pollet<sup>1</sup>, Odile Majérus<sup>2</sup>, Laurent Cormier<sup>3</sup>

1) NIMBE, CEA, CNRS, Université Paris-Saclay, CEA Paris-Saclay – 91191 Gif-sur-Yvette – France

2) IRCP, Chimie ParisTech – 75231 Paris – France

3) IMPMC, Université Pierre et Marie Curie – 75252 – Paris

\*erwan.chesneau@cea.fr

**Key words:** solid state NMR, molecular dynamics, borate glasses, DFT, Structure factor

In this work, we investigate the structure and properties of  $(1-x)\text{LaB}_3\text{O}_6-x\text{Al}_2\text{O}_3$  glasses ( $x \leq 0.375$ ) using solid state NMR and neutron diffraction combined with molecular dynamics simulations. This study aims at understanding the mechanism of aluminium oxide incorporation in lanthanum metaborate glass ( $\text{LaB}_3\text{O}_6$ ) [1] and its impact on structural features.

$^{11}\text{B}$  and  $^{27}\text{Al}$  NMR experiments have been performed to obtain the atomic speciation of boron ( $\text{BO}_3$  and  $\text{BO}_4$ ) and aluminium ( $\text{AlO}_{4,5,6}$ ). Two samples were enriched with oxygen-17 to obtain the BO, NBO population and advanced 2D through-bond heteronuclear correlation ( $^{17}\text{O}$ - $^{11}\text{B}$ ,  $^{17}\text{O}$ - $^{27}\text{Al}$ ) was performed to highlights the distribution of NBO on the  $\text{BO}_x$  and  $\text{AlO}_x$  structural units, whereas  $^{11}\text{B}$ - $^{27}\text{Al}$  2D NMR revealed their connectivity.

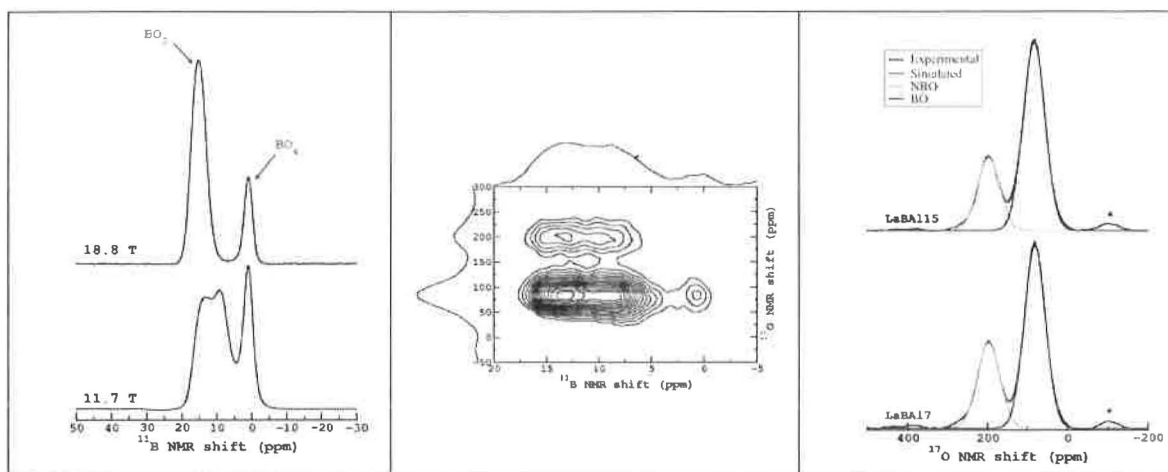


Figure 1 : Left panel:  $^{11}\text{B}$  MAS NMR spectra at 18.8T and 11.7T; middle panel: J-HMQC  $^{11}\text{B}\{^{17}\text{O}\}$  spectra at 11.7T; right panel:  $^{17}\text{O}$  MAS NMR spectra at 11.7T

However, concerning superstructural units, more precise information are difficult to extract because of broad NMR shift distribution due to the amorphous structure of the glass. We therefore perform Molecular Dynamics (MD) simulations. As shown in previous studies [2], classical MD (based on empirical potential) cannot reproduce superstructural units in contrast to ab-initio MD (aiMD). Nevertheless, aiMD is an expensive computational time method and is therefore limited in the quenching rate so that too much liquid like structures are generated. In order to overcome these limitations, a Hybrid Reverse Monte Carlo modelling approach has been developed accounting for all constraints provided by NMR, in addition to the neutron and X-ray structure factors to converge into stable structures answering to all experimental data.

- [1] D. S. Pytalev, D. Caurant, O. Majérus, H. Trégouët, T. Charpentier, and B. N. Mavrin, « Structure and crystallization behavior of  $\text{La}_2\text{O}_3 \cdot 3\text{B}_2\text{O}_3$  metaborate glasses doped with  $\text{Nd}^{3+}$  or  $\text{Eu}^{3+}$  ions », *J. Alloys Compd.*, vol. 641, p. 43–55, 2015.
- [2] A Takada, C R A Catlow and G D Price, « Computer modeling of  $\text{B}_2\text{O}_3$  . II. Molecular dynamics simulations of vitreous structures », *J. Phys. Condens. Matter*, vol. 7, n° 46, p. 8693, 1995.