

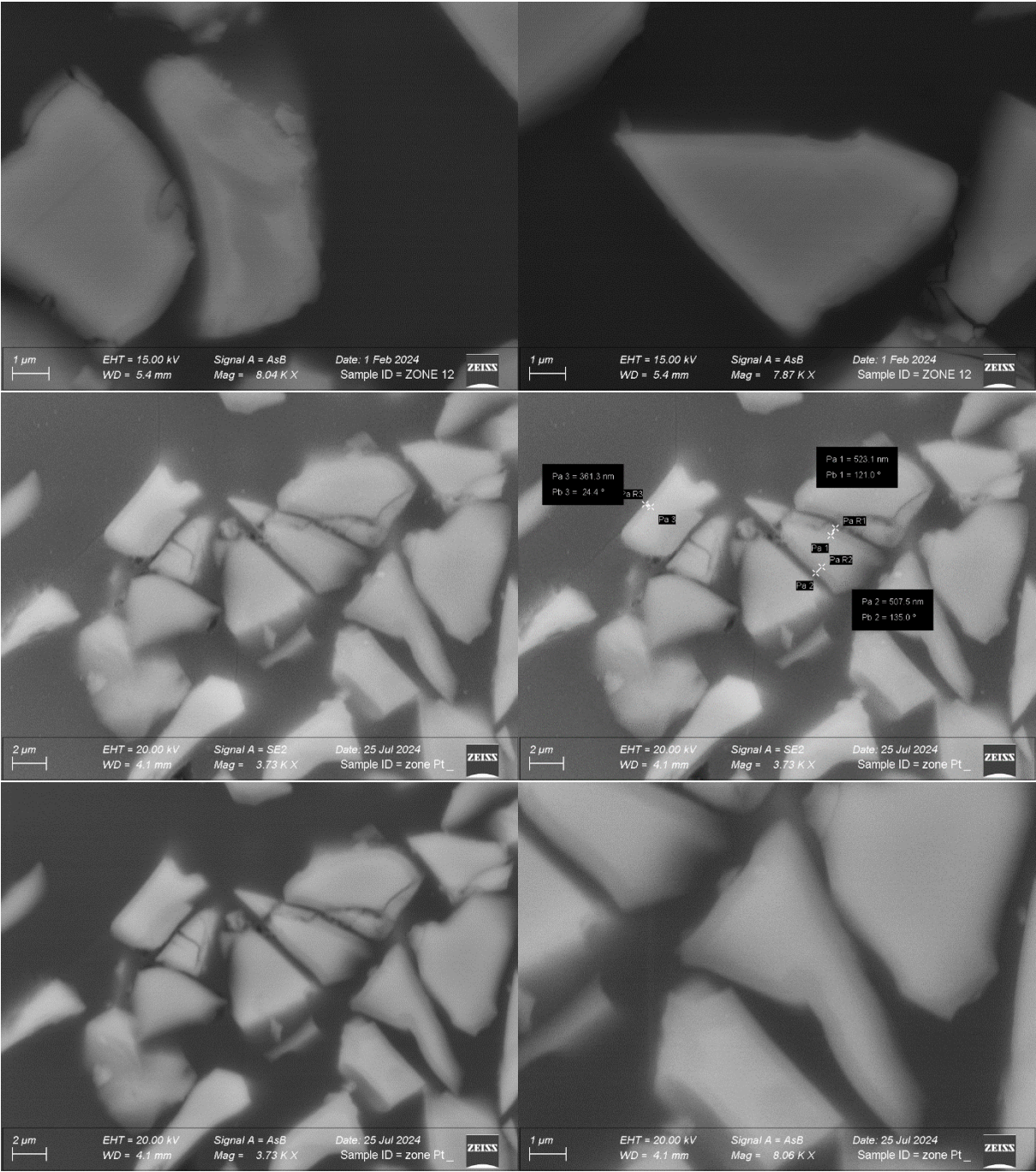
Supplementary data: NMR study of a gel layer formed on an irradiated Na-aluminoborosilicate glass during aqueous alteration

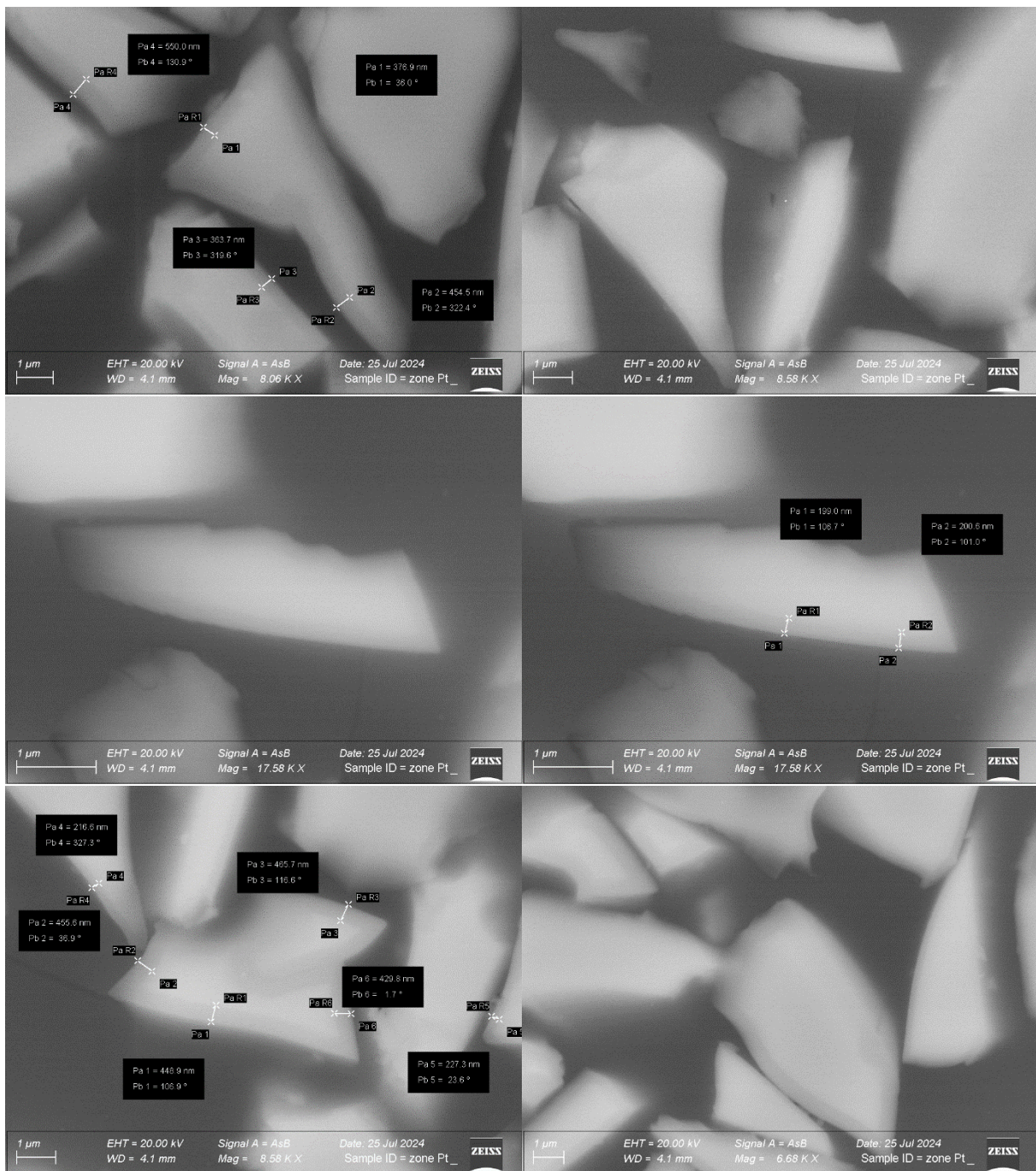
Authors: Sathya Narayanasamy¹, Thibault Charpentier², Amreen Jan¹, Mélanie Moskura², Matilde Benassi², Jean-Marc Delaye¹, Stéphane Gin¹

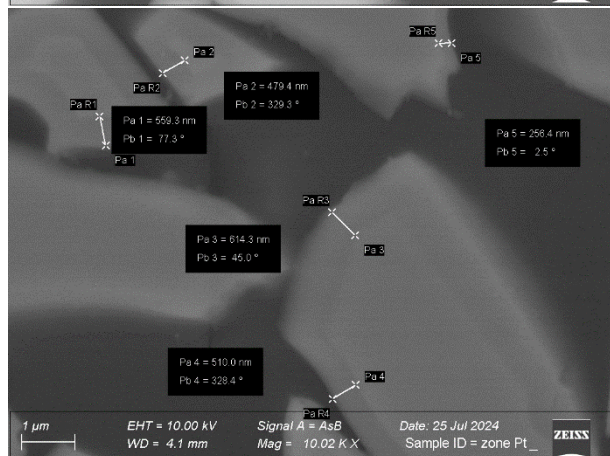
Affiliations: ¹ CEA, DES, ISEC, DPME, SEME, University of Montpellier, Marcoule, Montpellier F-30207 Bagnols-sur-Cèze, France

² Université Paris-Saclay, CEA, CNRS, NIMBE, 91191 Gif-sur-Yvette, Cedex, France

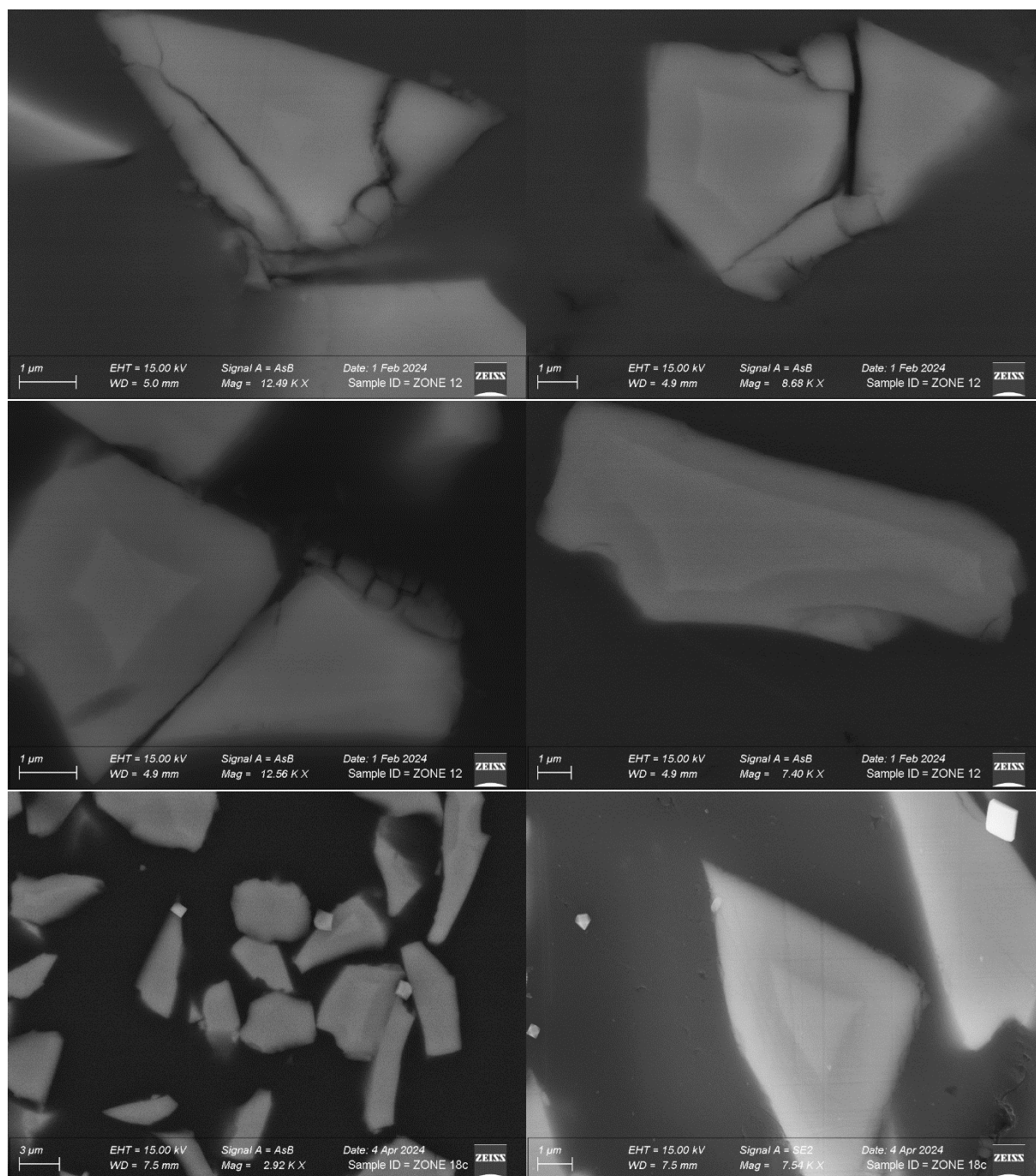
Supplementary Table 1: SEM images of the altered CJ2 glass powders (non-irradiated)

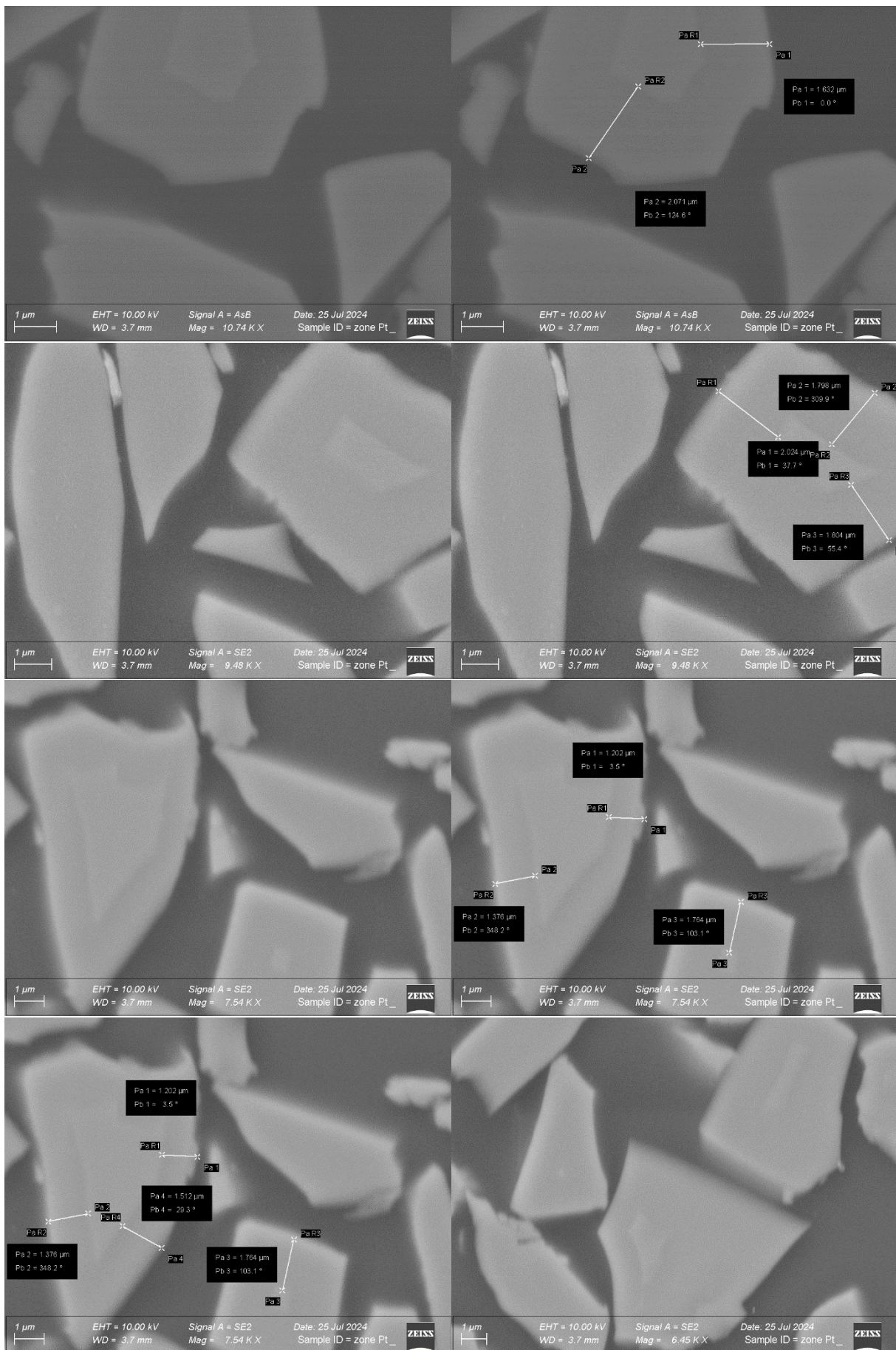


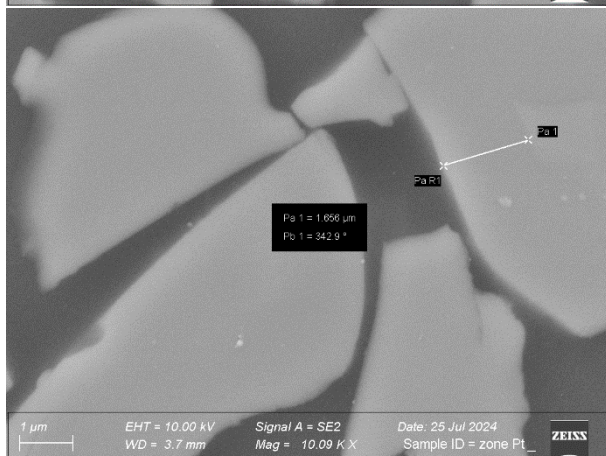
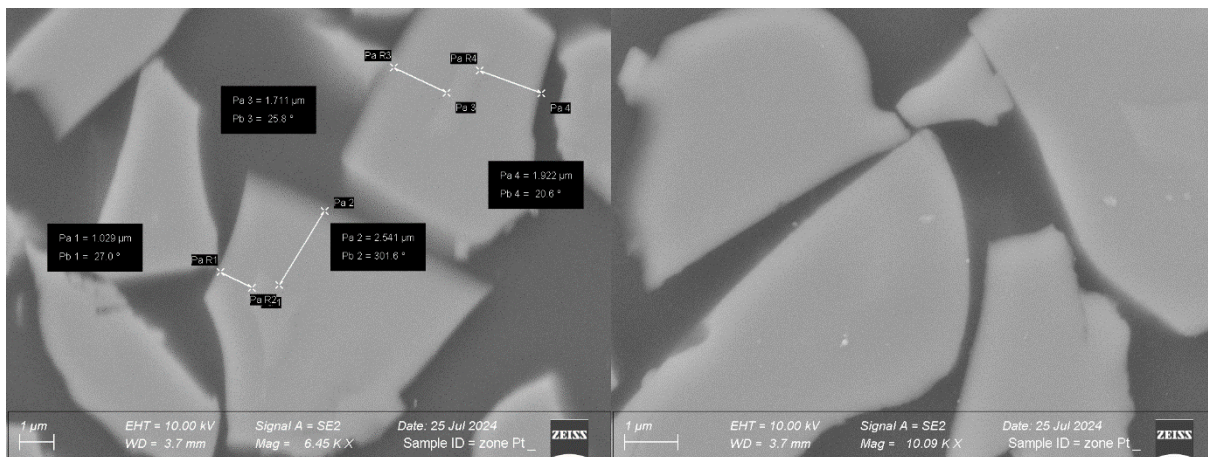




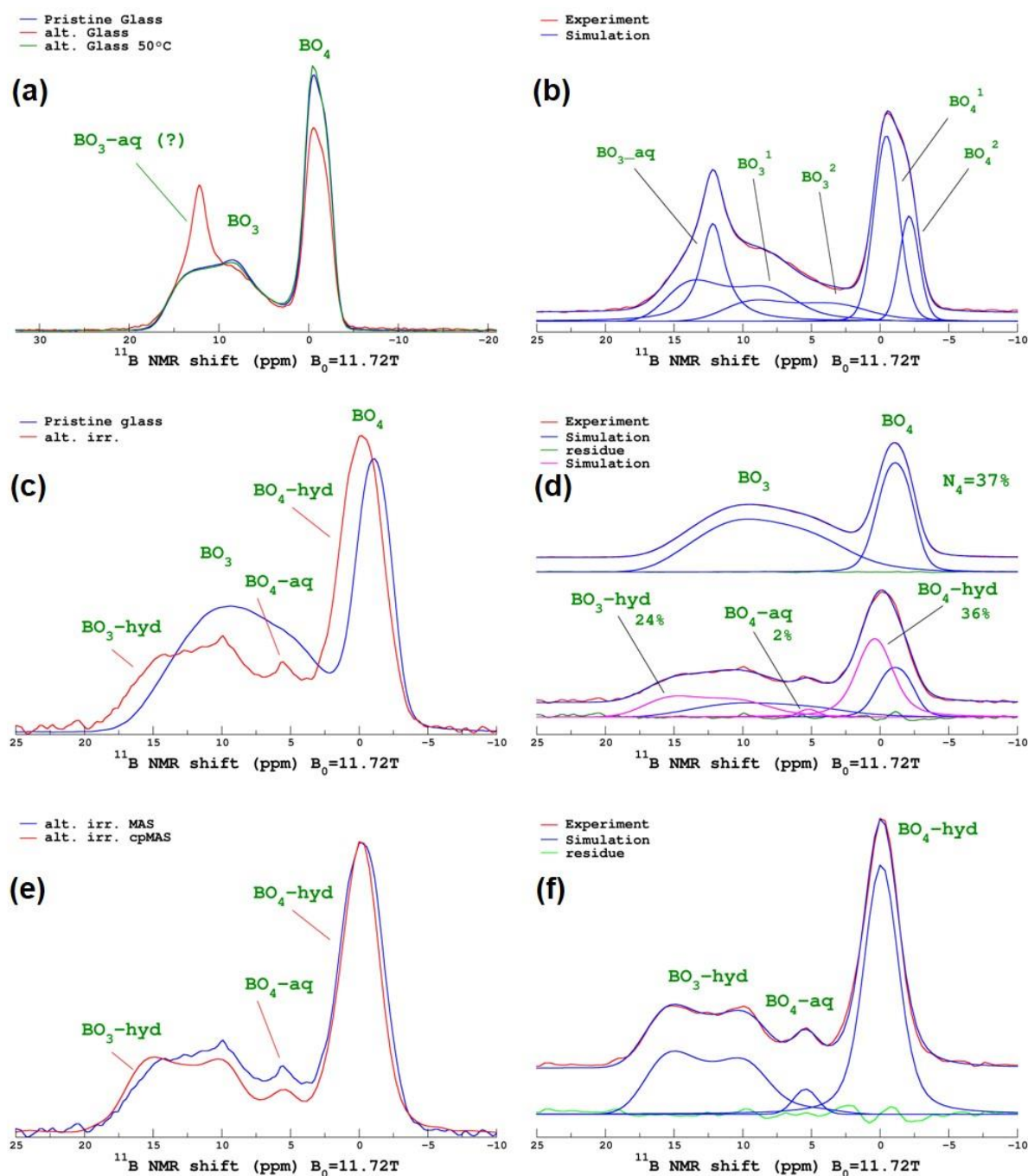
Supplementary Table 2: SEM images of the irradiated and altered CJ2 glass powders



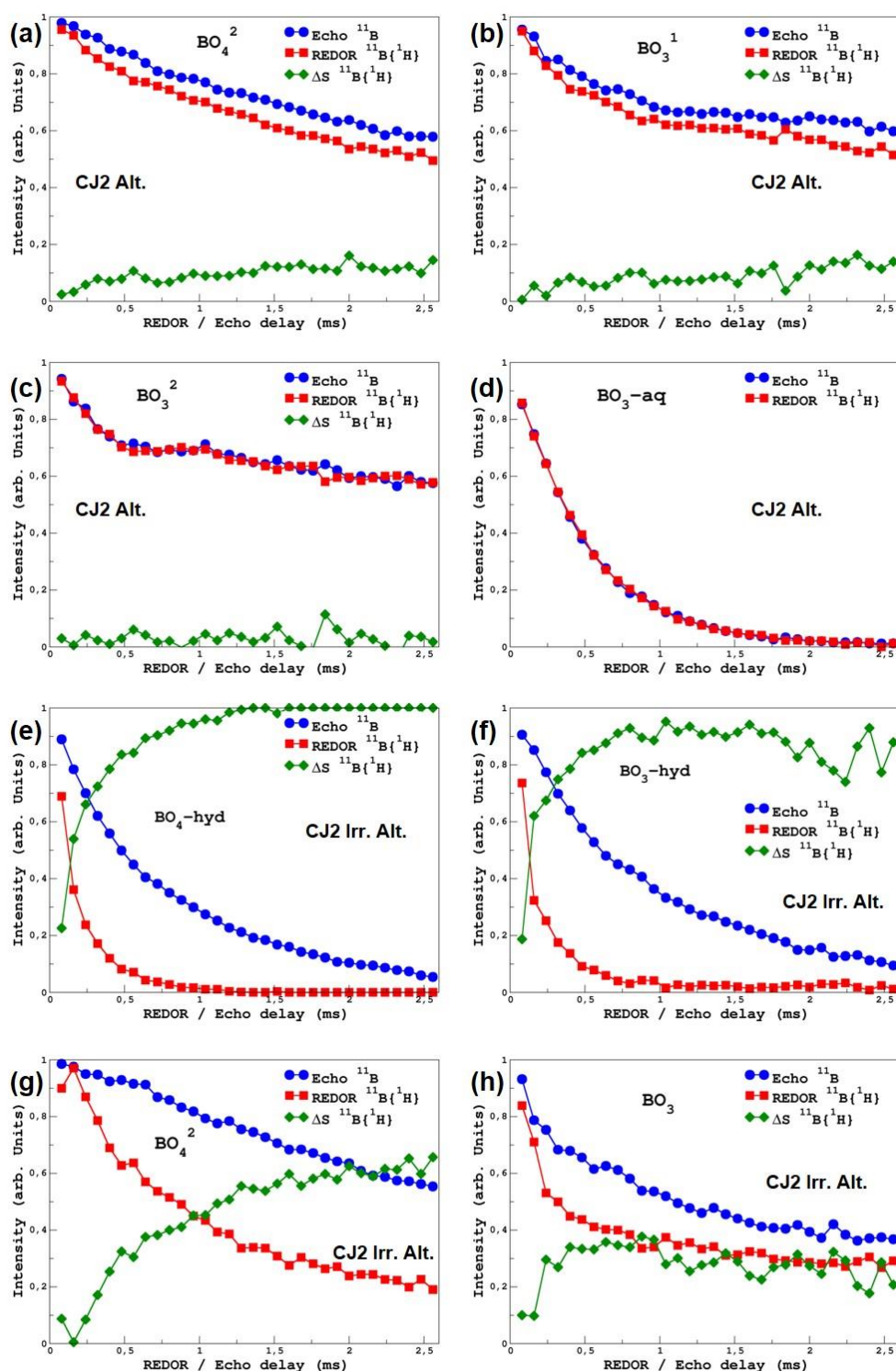




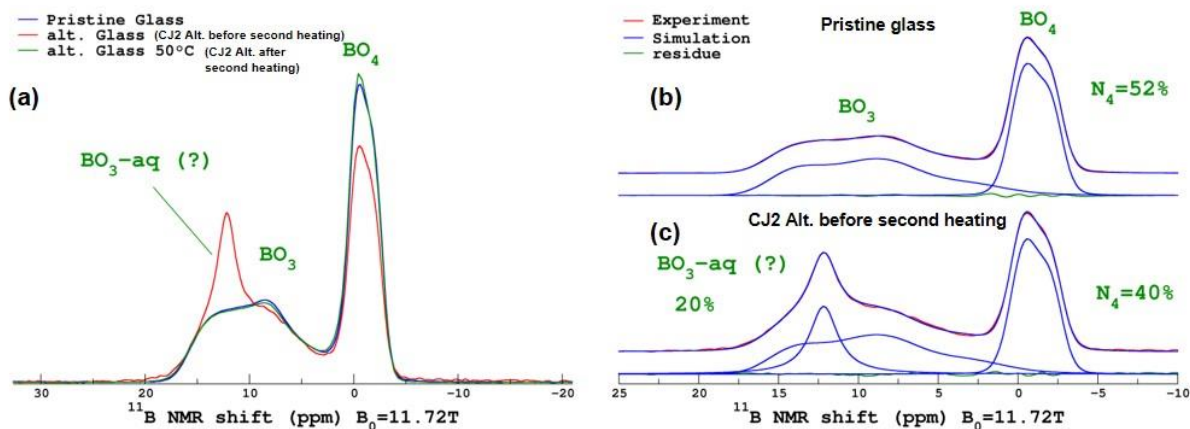
Part 2 : NMR data



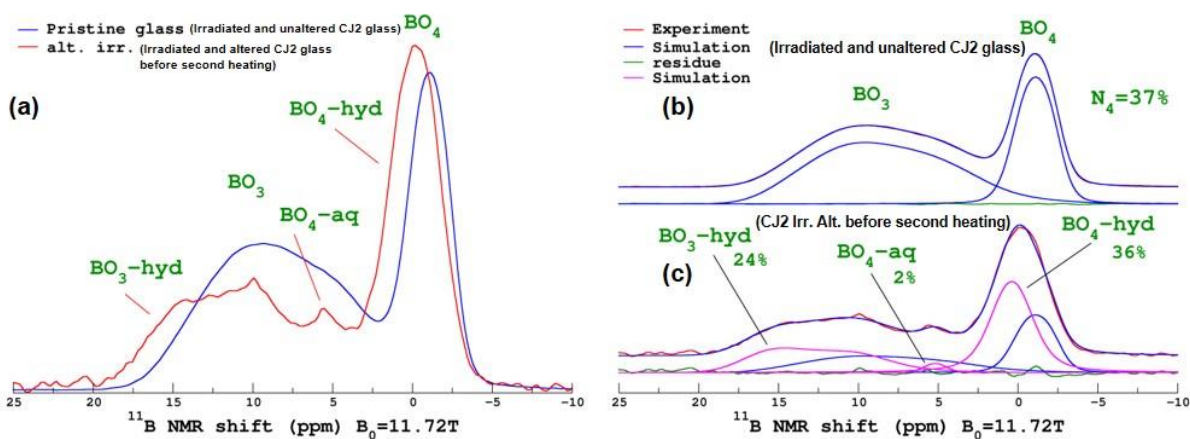
Supplementary Figure 1 (a) ^{11}B MAS NMR spectra of the CJ2 Alt. sample before the second heating at 50°C compared to the ^{11}B NMR spectra after the second heat and the pristine CJ2 glass. (b) ^{11}B MAS spectra fitted with five components. (c) ^{11}B MAS NMR spectra of the unaltered CJ2 irradiated sample compared with the ^{11}B MAS NMR spectra of the CJ2 Irr. Alt. sample. (d) (Top) ^{11}B MAS NMR spectra of the unaltered CJ2 irradiated sample fitted with a BO_3 component and a BO_4 component (Note that both the $\text{BO}_3^{1\&2}$ components have been merged into a single BO_3 component and likewise for the merged BO_4 component). (Bottom) ^{11}B MAS NMR spectra of the CJ2 Irr. Alt. sample. Fitted with the glass components as above and the hydrated BO_3 and BO_4 components identified through cpMAS experiments. (e) ^{11}B MAS NMR spectra of the CJ2 Irr. Alt., compared with the ^{11}B cpMAS spectra of the sample. (f) ^{11}B cpMAS NMR spectra of the CJ2 Irr. Alt. fitted with the hydrated BO_3 and BO_4 components.



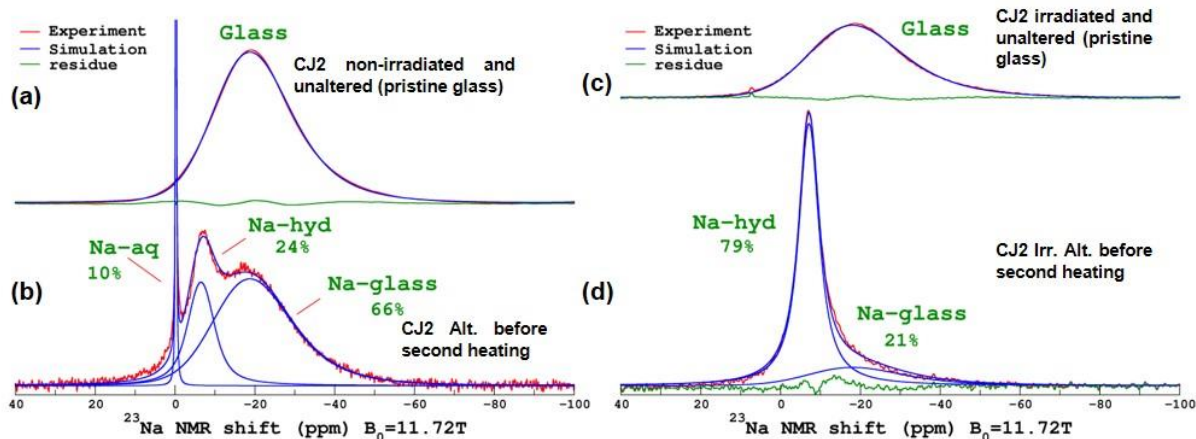
Supplementary Figure 2 $^{11}\text{B} \{^1\text{H}\}$ REDOR corresponding to different components of the non-irradiated and altered and the irradiated and altered sample. For the non-irradiated and altered sample: (a) BO_4^{2-} component. (b) BO_3^{1-} component. (c) BO_3^{2-} component. (d) $\text{BO}_3\text{-aq}$ component. For the irradiated and altered sample: (e) $\text{BO}_4\text{-hyd}$ component. (f) $\text{BO}_3\text{-hyd}$ component. (g) BO_4^{2-} component. (h) BO_3 component.



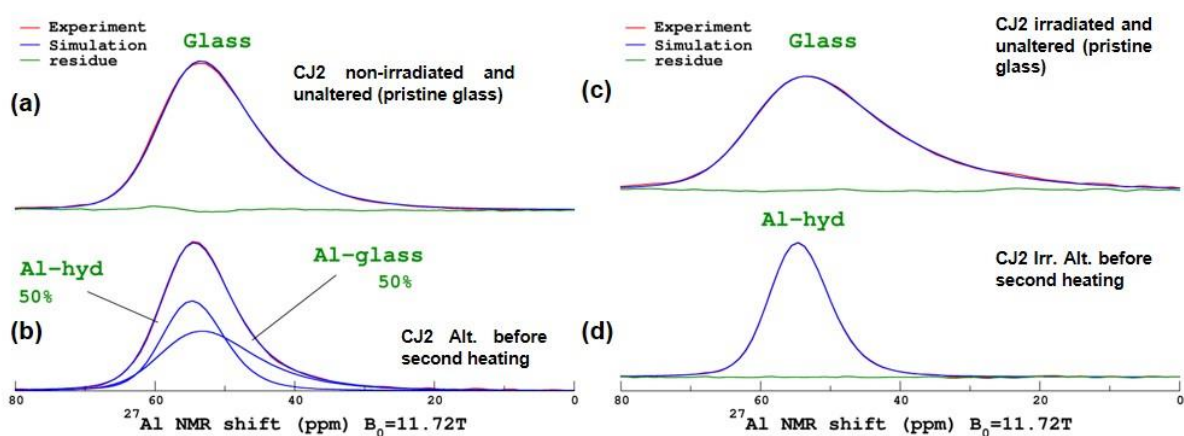
Supplementary Figure 3 (a) ^{11}B MAS NMR spectra of the non-irradiated CJ2 unaltered (pristine) glass, non-irradiated and altered glass before the second heating and non-irradiated and altered glass after the second heating. (b) Deconvolution of the ^{11}B MAS NMR spectra of the unaltered glass. (c) Deconvolution of the ^{11}B MAS NMR spectra of the altered glass before the second heating.



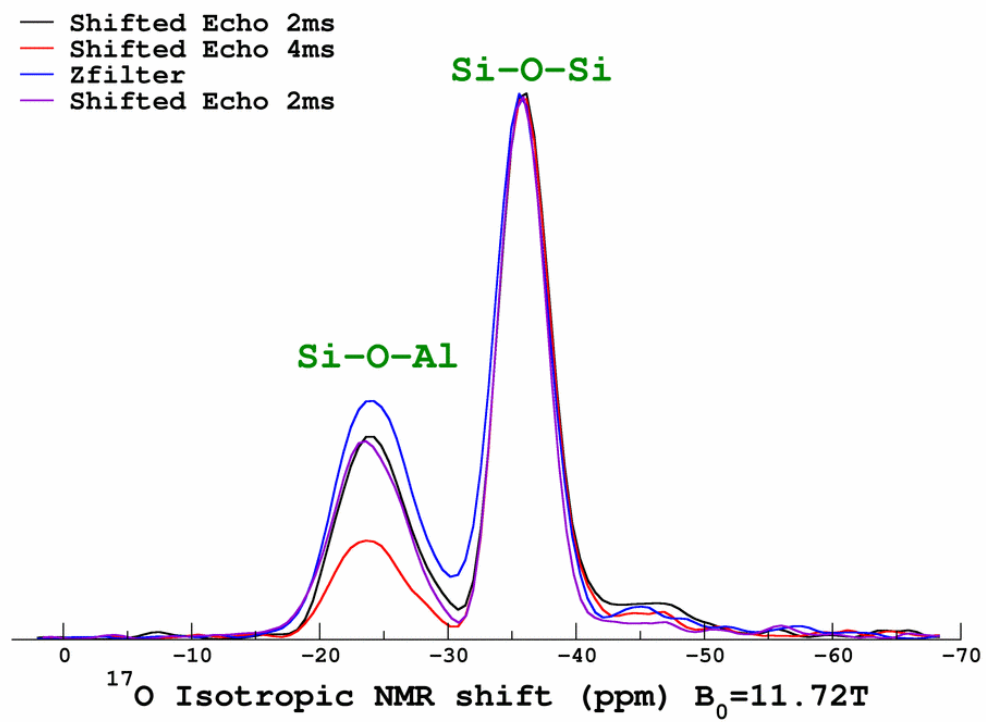
Supplementary Figure 4 (a) ^{11}B MAS NMR spectra of the irradiated CJ2 unaltered (pristine) glass, irradiated and altered glass before the second heating. (b) Deconvolution of the ^{11}B MAS NMR spectra of the unaltered glass. (c) Deconvolution of the ^{11}B MAS NMR spectra of the altered glass before the second heating.



Supplementary Figure 5 (a) ^{23}Na MAS NMR spectra (experimental and simulated) of the non-irradiated and unaltered CJ2 glass. (b) ^{23}Na MAS NMR spectra (experimental and simulated) of the non-irradiated and altered sample before the second heating. (c) ^{23}Na MAS NMR spectra (experimental and simulated) of the irradiated and unaltered CJ2 glass. (d) ^{23}Na MAS NMR spectra (experimental and simulated) of the irradiated and altered sample before the second heating.



Supplementary Figure 6 (a) ^{27}Al MAS NMR spectra (experimental and simulated) of the non-irradiated and unaltered CJ2 glass. (b) ^{27}Al MAS NMR spectra (experimental and simulated) of the non-irradiated and altered sample before the second heating. (c) ^{27}Al MAS NMR spectra (experimental and simulated) of the irradiated and unaltered CJ2 glass. (d) ^{27}Al MAS NMR spectra (experimental and simulated) of the irradiated and altered sample before the second heating.



Supplementary Figure 7 Isotropic project of ^{17}O MQMAS spectra acquired at different pulse sequences for the CJ2 irradiated and altered sample after the second drying