

Supplementary Information

Effect of structural disorder induced by irradiation on the alteration of a four-oxide borosilicate glass

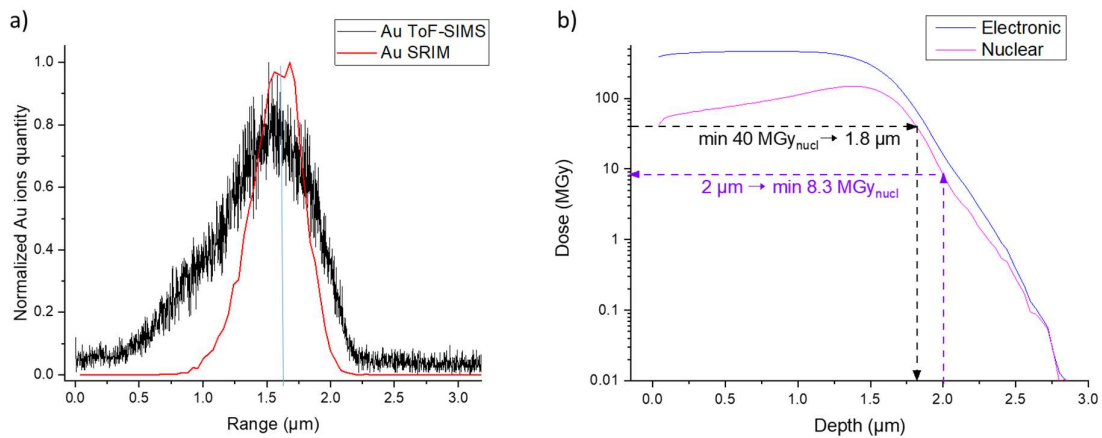
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Supplementary Information 1

The implantation of Au ions due to irradiation was measured by ToF-SIMS and calculated using with the Stopping and Range of Ions in Matter (SRIM) software¹. The comparison given in Supplementary Figure 1a shows a good agreement with a maximum Au concentration around 1.6 μm , followed by a decrease until a depth of about 2 μm . Under these irradiation conditions, the cumulative electronic and nuclear doses as a function of CJ2 glass depth were calculated using SRIM software (Supplementary Figure 1b). Above a nuclear dose of 40 MGy_{nuc} the saturation of the effects of irradiation on the material properties was reported. Here, the depth impacted by a minimum dose of 40 MGy_{nuc} is about 1.8 μm . Then, between 1.8 and 2 μm , the nuclear dose decreases until 8.3 MGy_{nuc} . According to Peugeot et al.², even if this dose is below the saturation one, there is still a high impact on ISG glass properties, including its leaching behavior. Therefore, for the purpose of this study, an irradiated depth of about 2 μm was considered.



Supplementary Figure 1 : a) Au ions implantation as a function of depth, measured by ToF-SIMS and calculated by SRIM software ; b) electronic and nuclear doses as a function of depth, calculated by SRIM software.

Supplementary Information 2

The four following tables are related to initial dissolution rate measurements conducted at 90°C and pH 9.

Non-irradiated sample

Concentrations (mg/L)	Time (day)	Si	B	Na	Al	Li
Cj2 t0	0.00	1.38	0.00	0.09	0.05	9.18
Cj2 t1	0.33	2.54	0.38	0.73	0.24	8.95
Cj2 t2	1.00	6.79	0.96	1.72	0.61	9.04
Cj2 t3	1.33	6.88	1.15	2.00	0.71	9.02
Cj2 t4	2.00	8.79	1.55	2.67	0.94	9.36
Cj2 t5	2.33	10.42	1.78	3.06	1.08	9.29
CJ2 t6	3.00	11.26	2.05	3.51	1.23	9.23
CJ2 t7	3.33	12.38	2.24	3.87	1.34	8.81
Normalized mass loss (g.m ⁻²)	Time (day)	Si	B	Na	Al	Li
Cj2 t0	0.00	1.9	0.0	0.4	0.6	-
Cj2 t1	0.33	1.5	2.5	2.6	2.0	-
Cj2 t2	1.00	7.4	6.4	6.5	6.3	-
Cj2 t3	1.33	7.4	7.7	7.6	7.3	-
Cj2 t4	2.00	9.9	10.2	10.1	9.8	-
Cj2 t5	2.33	12.0	11.6	11.5	11.2	-
CJ2 t6	3.00	12.9	13.1	13.1	12.6	-
CJ2 t7	3.33	14.3	14.3	14.3	13.7	-

Irradiated sample

Concentrations (mg/L)	Time (day)	Si	B	Na	Al	Li
CJ2 Irr t0	0.00	0.76	0.00	0.00	0.00	5.62
CJ2 Irr t1	0.08	1.96	0.10	0.21	0.07	5.64
CJ2 Irr t2	0.17	1.77	0.24	0.61	0.16	5.67
CJ2 Irr t3	0.25	2.93	0.37	0.71	0.24	5.62
CJ2 Irr t4	0.33	3.08	0.50	0.94	0.31	5.72
CJ2 Irr t5	1.00	6.22	1.08	1.79	0.66	5.57
CJ2 Irr t6	1.17	6.69	1.08	1.77	0.65	5.85
CJ2 Irr t7	1.33	6.87	1.22	2.04	0.75	5.63
Normalized mass loss (g.m ⁻²)	Time (day)	Si	B	Na	Al	Li
CJ2 Irr t0	0.00	1.07	0.00	0.00	0.00	-
CJ2 Irr t1	0.08	1.62	0.68	0.86	0.81	-
CJ2 Irr t2	0.17	1.38	1.59	2.46	1.84	-
CJ2 Irr t3	0.25	2.95	2.50	2.82	2.66	-
CJ2 Irr t4	0.33	3.12	3.33	3.72	3.50	-
CJ2 Irr t5	1.00	7.34	7.13	7.05	7.31	-
CJ2 Irr t6	1.17	7.85	7.03	6.87	7.13	-
CJ2 Irr t7	1.33	8.02	7.85	7.85	8.09	-

Supplementary Information 3

The following table is related to residual dissolution rate measurements conducted at 90°C and pH 9.

Non Irradiated CJ2 sample			Concentrations					Normalized mass loss		Equivalent thickness	
Date	Time	Sol. Vol.	C(Si)	C(B)	C(Na)	C(Al)	C(Li)	NL(B)	NL(Na)	ETH(B)	ETH(Na)
	day	cm ³	mg/L	mg/L	mg/l	mg/L	mg/L	g/m ²	g/m ²	nm	nm
NIR-0	19/04/2022	0	20.5	286	0.10	2.08	0.01	42.0	0	-	-
NIR-1	02/06/2022	44	20.0	281	0.62	2.42	0.01	29.4	1.02	422	0
NIR-2	20/07/2022	92	19.0	272	1.02	2.80	0.01	29.0	1.84	765	0
NIR-3	22/09/2022	156	18.0	268	1.37	3.48	0.01	28.7	2.37	984	637
NIR-4	15/11/2022	210	17.2	273	1.75	4.84	0.01	29.0	3.02	1253	1204
NIR-5	17/01/2023	273	16.6	261	1.55	4.37	< dl	30.8	2.70	1119	1015
NIR-6	09/03/2023	324	16.0	240	1.51	4.48	< dl	30.4	2.63	1092	1056
NIR-7	03/05/2023	379	15.5	251	1.59	5.03	< dl	32.0	2.76	1145	1268
NIR-8	03/11/2023	563	15.1	166	1.51	5.1	< dl	38.8	2.50	1038	1281
7 MeV Au Irradiated sample											
7M-0	26/05/2023	0	23.5	257	<dl	1.67	< dl	na	-	-	-
7M-1	17/07/2023	52	23.5	270	26.94	40.79	< dl	na	8.85	7.67	3672.2
7M-2	14/09/2023	111	22.5	269	31.30	45.67	< dl	na	10.54	8.84	4371.5
7M-3	06/10/2023	133	22.5	266	32.92	46.33	< dl	na	11.04	8.97	4582.5

Supplementary Information 4

The gel maturation time, t_m , corresponds to the duration during which a given gel layer has been in contact with the solution. Since the gel forms gradually over time, meaning as the glass alters, this maturation time depends on the z-coordinate (gel depth). Calculation of t_m , is performed as follow.

For two consecutive solution analysis, n and $n+1$ (Figure 4), equivalent thicknesses of altered glass, $Eth(B)_n$ and $Eth(B)_{n+1}$, were calculated (Supplementary Information 3). These numbers correspond to the thickness of gel at the two corresponding durations, n and $n+1$, assuming that glass alteration is isovolumetric. This assumption is supported by the good agreement between solution data which provide the thickness of altered glass and ToF-SIMS data which provide a good estimation of the gel thickness. Then we assumed that the increase $Eth(B)$ follows a linear evolution between t and $t+1$. This gives:

$$Eth(B)_{n+1} - Eth(B) = a_n \cdot t_n + b_n$$

The maturation time of the gel, t_m , is simply calculated from the difference between the duration of the experiment and t_n .

Supplementary References

- 1 Ziegler, J. F., Ziegler, M. D. & Biersack, J. P. SRIM – The stopping and range of ions in matter (2010). *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **268**, 1818-1823, doi:<https://doi.org/10.1016/j.nimb.2010.02.091> (2010).
- 2 Peugeot, S., Tribet, M., Mougnaud, S., Miro, S. & Jegou, C. Radiations effects in ISG glass: from structural changes to long-term aqueous behavior. *Npj Materials Degradation* **2**, doi:10.1038/s41529-018-0044-3 (2018).