

Structurally controlled Mg isotope fractionation between talc and fluid: implications for the oceanic and mantle Mg cycle

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ABSTRACT

Talc and structurally related Mg-phylosilicates are ubiquitous products of hydrothermal alteration and metamorphism, and play an important role in the global magnesium cycle. However, their Mg isotope fractionation behavior remains poorly constrained, and previous studies on Mg-bearing layered minerals have yielded apparently conflicting results. Here we present the first experimental determination of Mg isotope fractionation between talc and aqueous fluids under hydrothermal conditions (130–188°C). Our closed-system experiments demonstrate that talc preferentially incorporates heavy Mg isotopes relative to coexisting fluid, with $\Delta^{26}\text{Mg}_{\text{talc-soln}}$ increasing from values +0.12‰ at early stages to $\sim +0.80\text{‰}$ as reaction proceeds. This temporal evolution correlates with structural maturation of talc, indicating that crystallographic ordering and progressive stiffening of Mg–O bonds are primary controls on isotope partitioning. This finding provides a unifying mechanism to reconcile discrepancies in published fractionation factors for other Mg-bearing layered minerals (e.g., smectite, brucite), suggesting that the variability in natural and experimental data likely reflects different degrees of structural ripening. More broadly, our experiments provide an experimental reference for Mg-phylosilicates and highlight the sensitivity of the marine Mg-silicate sink to mineral identity and structural maturity. In subduction-zone settings, talc-forming reactions may generate heavy-Mg signatures in talc-bearing metasomatic lithologies at the slab-mantle interface, and these signatures may be retained and redistributed during subsequent talc breakdown, fluid-rock interaction and metamorphic replacement.

1. Introduction

Talc, with an ideal chemical formula of $\text{Mg}_3\text{Si}_4\text{O}_{10}(\text{OH})_2$, is a ubiquitous magnesium phyllosilicate in the Earth's crust (Bailey, 1988). It commonly forms through low- to medium-temperature hydrothermal alteration of (ultra)mafic rocks and silica-rich metasomatism of dolomite and calcite, and can also precipitate directly from the mixing of seawater and hydrothermal fluids (e.g., Cuadros et al., 2008; Dekov et al., 2008; Hodgkinson et al., 2015; Peacock and Hyndman, 1999; Schroeder et al., 2015). Due to its high Mg content, low frictional strength and chemical stability, talc has long been used as an indicator of specific fluid-rock interaction regimes, (de)carbonation reactions, mechanically weak shear zones, and the cycling of volatiles in subduction environments (e.g., Hodgkinson et al., 2015; Hoover et al., 2022;

Papeschi et al., 2025; Peacock and Wang, 2021; Spandler et al., 2007). Its occurrence and composition provide important constraints on the thermal and chemical evolution of hydrothermal, metasomatic and subduction-related systems.

Stable oxygen and hydrogen isotopes in talc have been widely applied as tracers of fluid sources and temperatures (e.g., Saccocia et al., 2009; Sharp et al., 1993; Shin and Lee, 2003; Spandler et al., 2007; Tornos and Spiro, 2000). In such applications, talc is treated as a robust recorder of the isotopic composition of its parent fluids and of the thermal regime during its formation. However, O and H isotopes primarily track the H_2O and CO_2 components of the fluid and are less sensitive to the reaction pathways and exchange of Mg during fluid-rock interaction. Magnesium isotopes can provide a complementary view because they directly trace the sources, transfer and partitioning of Mg.

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For instance, a recent study observed the significant enrichment of heavy Mg isotopes in talc compared to protolith serpentinite and adjacent sedimentary rocks at subduction interface, and proposed that talc formed via the loss of isotopically light Mg from serpentinite rather than by addition of Si (Easthouse et al., 2025). This suggests that talc can systematically fractionate Mg isotopes relative to its protolith and coexisting fluids.

To broadly apply Mg isotope to talc-related geological problems, it is essential to calibrate the Mg isotope fractionation between talc and dissolved Mg^{2+} . In addition, talc shares the same the MgO_6 octahedral sheet (“O”) with other Mg-phylosilicates, such as clay minerals (Bailey, 1988). Therefore, a well-constrained Mg isotope fractionation between talc and aqueous solution would not only underpin the interpretation of $\delta^{26}\text{Mg}$ values in talc-bearing rocks, but also provide insights for understanding Mg isotope fractionation across Mg-phylosilicates more generally, as Mg-phylosilicates formed during seafloor alteration represent a major long-term sink for seawater Mg, and their isotope systematics exert a first-order control on the global Mg budget and on the secular evolution of seawater $\delta^{26}\text{Mg}$ (Higgins and Schrag, 2015; Hu et al., 2022; Shalev et al., 2019).

Despite this potential, there are currently no direct experimental constraints on Mg isotope fractionation between talc and coexisting fluids. Here we provide the first experimental determination of Mg isotope fractionation between talc and aqueous solution under hydrothermal conditions. We performed closed-system hydrothermal experiments at 130, 150, and 188°C, in which calcite and fumed silica react with MgCl_2 solutions to produce talc. The reaction temperatures were chosen between the low (<100°C) and high-temperature (>400°C) hydrothermal conditions to (1) accelerate the reaction rate in laboratory timescale, (2) avoid limited Mg isotope fractionation at high temperature, as the magnitude of isotope fractionation decreased with increasing temperatures (Schauble, 2004). Our results show that talc preferentially incorporates heavy Mg isotopes relative to aqueous solution, and that the apparent Mg isotope fractionation between talc and fluid increases with reaction time, reaching values of +0.80‰ by the end of the experiments. These observations point to kinetic isotope effects during the onset of talc formation and provide experimental support for significant depletion of heavy Mg isotopes in fluids during their reaction with oceanic lithosphere. More broadly, our study confirms that talc carries heavy Mg isotopic signatures into the mantle during the subduction of oceanic lithosphere and refines the use of talc Mg isotopes as a geochemical indicator in talc-bearing systems.

2. Experimental methods

The experiments were conducted by reacting calcite, fumed silica, with MgCl_2 solutions at hydrothermal conditions (130, 150, and 188°C) in a closed system. All experiments and analyses were conducted at State Key Laboratory of Critical Earth Material Cycling and Mineral Deposits, Nanjing University. The analytical reagent calcite and fumed silica powders were purchased from Sigma-Aldrich® and the powder XRD analyses confirmed the nearly 100% purity. In addition, the ICP-OES analyses showed that the calcite contained 0.89 mol% Mg (Mg/Mg + Ca). The reagent grade $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ salt from Sigma-Aldrich® was dissolved in the deionized water ($=18.2 \text{ M}\Omega \cdot \text{cm}^{-1}$) to prepare the stock solution (2 M of MgCl_2).

2.1. Hydrothermal experiments

The hydrothermal reactions were proceeded in the fused glass tubes (Appendix Fig. S1). First, 22 mg of fumed silica, 25 mg of calcite, 0.25 mL of 2 M MgCl_2 , and 0.5 mL deionized water loaded into the single port glass tubes (5.5 mm inner diameter and 7.5 mm outer diameter), to simulate the reaction between silica rich solution and carbonate minerals. Then the open end of the reaction tubes was sealed using an oxyhydrogen flame. Finally, the tubes were placed into the stainless-

steel bombs filled with deionized water to equilibrate the air pressure difference between the inner and outside the reaction tubes under elevated temperatures. This configuration also allowed any gas phase generated during reaction to be retained. The initial reactant of fumed silica is spherical with the size ranging from 50 nm to 5000 nm (Fig. 3A), while the initial reactant calcite is column-like with around 1000 nm in length (Fig. 3B).

Experiments were conducted at 130, 150, and 188°C (Experiment series: Syn130, Syn150 and Syn188, respectively). Each set of hydrothermal experiments involved multiple reaction tubes and were sampled in a time series. Upon removal from the oven and quenched to room temperature in the air within 2 h, the Raman spectra for the vapor phases of the reaction tubes were collected. Afterwards, the products were transferred to a clean tube, where the solid and fluid contents were separated by centrifuging. The fluid product was centrifuged at 12000 rpm for 1 min three times to remove any possible suspended solids, while the solid product was washed with de-ionized water and centrifuged three times, and dried. This step may remove minor amounts of weakly adsorbed or exchangeable Mg from particle surfaces, but is not expected to dissolve significant structural Mg from talc-dominated products over the short duration of rinsing. The potential effect of Mg loss during rinsing and sample transfer was evaluated using isotope mass-balance calculations in Section 4.1.

2.2. Raman spectra collection

Raman spectra were collected by a high-resolution Raman spectrometer (LabRAM HR800 JY/Horiba) using a 532.11 nm laser from an air-cooled, frequency-doubled Nd: YAG laser excitation. A 50x Olympus objective was applied to focus the laser on the vapor phases in the reaction tubes. The 1800 groove/mm grating was used, and the resultant spectral resolution was $\sim 1 \text{ cm}^{-1}$. The spectra were collected from 1200 to 1500 cm^{-1} for the coverage of the main vibration modes of CO_2 . The accumulation time was set to 120 s to obtain a relatively high signal-to-noise ratio spectrum. The Raman spectral analyses were conducted by LabSpec 5 software.

2.3. Mineralogical analysis

X-ray diffraction measurements were conducted on a Rigaku Rapid II X-ray diffractometer that was equipped with a rotating $\text{Mo K}\alpha$ source, running at 50 kV and 90 mA. The XRD pattern for the products was collected using a two-dimensional image plate with a 5-minute exposure time. Data treatment and interpretation were performed using a Rigaku 2DP program and Jade 6.5 software. The morphology of the solid samples was characterized on a Carl Zeiss Supra 55 Field Emission Scanning Electron Microscope, the chemical compositions of the solids were characterized using Energy-Dispersive X-ray Spectroscopy (EDS) with the SEMs.

2.4. Solid and solution chemistry analysis

Part of the solid products (approximately 8 mg) was reacted with a neutral-pH solution of 1 M NH_4Cl for 24 h to extract the exchangeable Mg (Hindshaw et al., 2020; Wimpenny et al., 2014), which occurred at the surface or interlayers of the mineral. These are referred to as the “Exchangeable Mg” in the study. Afterwards, the mixture was centrifuged, and the supernatant was transferred to the clean Teflon beaker and dried down for further elemental and isotopic analyses. The remained solids were then treated with 1 M HCl for 24 h to dissolve the Mg in the carbonate and easily leachable sites of clay minerals. Then the same centrifuge operation was conducted to separate the residual solid and supernatant. The supernatant was transferred and dried down for further elemental and isotopic analyses. This part is referred to as the “HCl-leachable Mg” in this study. The last residual solids were reacted with the mixture solution consisting of concentrated HNO_3 and

concentrated HF in the Teflon beakers at 95°C for 24 h and then dried down for further elemental and isotopic analyses. This part is referred to as the “Residual Mg” in this study.

Elemental analyses of the solution, “Exchangeable Mg”, “HCl-leachable Mg”, and “Residual Mg” were conducted using inductively-coupled plasma optical emission spectroscopy (ICP-OES) with reference to a series of gravimetrically prepared multi-element standard solutions.

2.5. Magnesium isotope analysis

Purification of Mg in the samples was achieved using a one-stage ion-exchange column chemistry procedure (Hu et al., 2017; Li et al., 2019). An aliquot of the sample was first converted into the nitrate form and dissolved in 0.5 mL 1.5 N HNO₃. Then the solution was loaded onto a fumed silica column with pre-cleaned 1 mL Biorad® cation exchange resin (AG50W-X12, 100 to 200 mesh). The 1.5 N HNO₃ and 6 N HCl were used to elute the column to remove Na, Ca, and other cations from Mg.

Magnesium isotope ratio measurements were performed on a Nu

1700 Sapphire MC-ICP-MS at Nanjing University (Liu and Li, 2023). The instrument was operated in high energy (no collision cell) and wet plasma mode. The standard-sample-standard bracketing method was applied to correct for the instrument drift and mass bias. An in-house Mg solution (A-Mg), which has a $\delta^{26}\text{Mg}$ value of -3.25‰ relative to DSM3, was used as the bracketing standard. The Mg concentration of the samples matched that of the standard at a level of $100 \pm 10\%$. Solutions were introduced into the instrument using a 100 $\mu\text{L}/\text{min}$ Glass Expansion nebulizer. Each isotope analysis consisted of forty 4-second integrations, which yielded a typical internal precision of $< 0.04\text{‰}$ for $^{26}\text{Mg}/^{24}\text{Mg}$ ratios.

Magnesium isotope data are reported using the standard delta notation (‰) of $\delta^{26}\text{Mg}$ and $\delta^{25}\text{Mg}$ for the $^{26}\text{Mg}/^{24}\text{Mg}$ and $^{25}\text{Mg}/^{24}\text{Mg}$ ratios relative to the DSM3 Mg isotope standard, where

$$\delta^x\text{Mg} = \left[\left(\frac{^x\text{Mg}/^{24}\text{Mg}_{\text{sample}}}{^x\text{Mg}/^{24}\text{Mg}_{\text{DSM3}}} \right) - 1 \right] \times 1000 \quad (1)$$

and the apparent Mg isotope fractionation factor between two phases A and B is expressed as:

Table 1

Mineralogy of solid products (XRD), Mg partitioning among the three operational Mg pools (“Exchangeable Mg”, “HCl-leachable Mg” and “Residual Mg”), and chemical and Mg isotopic compositions of coexisting aqueous solutions.

Experimental ID	Temp (°C)	Duration (hours)	Mineralogy by XRD	Exchangeable Mg Mg(%)	$\delta^{26}\text{Mg}$ (‰)	HCl-leachable Mg Mg(%)	$\delta^{26}\text{Mg}$ (‰)	Residual Mg Mg(%)	$\delta^{26}\text{Mg}$ (‰)	Solution Mg(M)	Ca(M)	$\delta^{26}\text{Mg}$ (‰)
130Syn	130	24	Qz + Cal	39.26	-2.17	15.76	-1.38	44.98	-0.68	0.65	0.05	-0.98
	130	48	Qz + Cal	30.10	-2.16	17.39	-1.37	52.51	-0.54	0.64	0.07	-0.90
	130	96	Qz + Cal	27.53	-2.15	18.25	-1.47	54.21	-0.54	0.63	0.09	-0.95
	130	192	Qz + Cal	26.03	-2.15	16.02	-1.44	57.95	-0.49	0.63	0.08	-1.01
	130	384	Qz + HMC + Dol	9.61	-2.14	57.66	-2.05	32.73	-0.41	0.55	0.20	-0.94
	130	720	Qz + HMC + Dol	9.64	-2.14	60.02	-2.04	30.34	-0.43	0.52	0.20	-0.85
	130	960	Qz + HMC + Dol	10.74	-2.12	55.49	-2.05	33.77	-0.42	0.57	0.19	-0.93
	130	1320	Qz + HMC + Dol	9.17	-2.06	55.26	-2.11	35.57	-0.38	0.51	0.18	-0.93
	130	1800	Qz + HMC + Dol	9.83	-2.11	57.02	-2.11	33.15	-0.38	0.64	0.24	-0.89
	130	2760	Qz + HMC + Dol + Talc	10.19	-1.94	51.58	-2.10	38.24	-0.40	0.57	0.22	-0.93
	130	4320	Qz + HMC + Dol + Talc	10.46	-1.89	51.04	-2.01	38.50	-0.43	0.51	0.22	-0.94
	130	8232	Qz + HMC + Dol + Talc	9.82	-1.73	58.21	-1.92	31.97	-0.42	0.48	0.29	-0.88
	130	11,856	Qz + HMC + Dol + Talc	16.69	-1.83	37.02	-1.84	46.29	-0.39	0.53	0.24	-1.03
150Syn	150	12	Qz + Cal	29.99	-2.53	34.77	-1.58	35.23	-0.39	0.64	0.06	-1.08
	150	24	Qz + Cal	19.88	-2.01	29.03	-1.37	51.08	-0.62	0.62	0.09	-1.04
	150	48	Qz + Cal	18.43	-2.26	23.61	-1.83	57.95	-0.63	0.61	0.10	-0.98
	150	72	Qz + Cal	39.27	-2.11	11.58	-1.61	49.16	-0.59	0.65	0.07	-0.99
	150	96	Qz + Cal	27.89	-2.34	33.02	-1.47	39.10	-0.59	0.64	0.09	-1.00
	150	120	Qz + Cal	18.53	-2.23	25.60	-1.72	55.87	-0.56	0.63	0.11	-1.02
	150	144	Qz + Cal	39.81	-2.49	27.23	-1.66	32.96	-0.50	0.64	0.08	-0.97
	150	216	Qz + Cal + Dol	22.03	-2.33	39.74	-1.96	38.23	-0.49	0.62	0.10	-0.95
	150	240	Qz + Cal + Dol	13.06	-2.10	40.28	-2.10	46.66	-0.57	0.59	0.15	-1.00
	150	312	Qz + Cal + Dol	18.75	-2.36	43.10	-1.83	38.15	-0.46	0.59	0.11	-0.93
	150	360	Qz + HMC + Dol	11.34	-1.97	55.85	-2.21	32.81	-0.51	0.54	0.19	-0.88
	150	480	Qz + HMC + Dol	17.67	-2.29	61.41	-1.97	20.93	-0.42	0.57	0.20	-0.75
	150	720	Qz + HMC + Dol + Talc	10.33	-2.35	51.91	-2.07	37.77	-0.53	0.53	0.17	-0.92
	150	1080	Qz + HMC + Dol + Talc	14.70	-1.87	53.99	-2.05	31.31	-0.46	0.53	0.21	-0.92
	150	1680	Qz + HMC + Dol + Talc	13.36	-1.90	51.63	-2.10	35.00	-0.48	0.51	0.20	-0.92
	150	2880	Qz + HMC + Dol + Talc	13.31	-1.95	46.71	-1.99	39.97	-0.45	0.53	0.20	-0.92
	150	3840	Qz + HMC + Dol + Talc	13.59	-1.87	50.74	-2.08	35.67	-0.45	0.50	0.22	-0.82
	150	5040	Qz + HMC + Dol + Talc	14.80	-1.89	49.21	-2.06	35.99	-0.41	0.52	0.21	-0.92
	150	14,400	Qz + HMC + Dol + Talc	14.55	-1.88	47.96	-1.83	37.49	-0.36	0.47	0.27	-0.82
188Syn	188	3	Qz + Cal	15.97	-2.19	35.12	-1.45	48.90	-0.89	0.60	0.09	-1.02
	188	6	Qz + Cal	12.76	-1.86	26.05	-1.39	61.19	-0.76	0.54	0.15	-1.05
	188	9	Qz + Cal + Talc	10.27	-1.84	24.63	-1.25	65.10	-0.74	0.54	0.15	-1.01
	188	12	Qz + Cal + Talc	8.56	-1.74	17.12	-1.25	74.32	-0.76	0.51	0.22	-1.07
	188	24	Qz + Cal + Talc	10.71	-1.51	17.96	-1.16	71.34	-0.71	0.46	0.23	-1.06
	188	72	Qz + HMC + Talc	11.28	-1.56	13.97	-1.89	74.76	-0.62	0.57	0.24	-1.13
	188	96	Qz + HMC + Dol + Talc	10.87	-1.66	14.38	-1.78	74.75	-0.63	0.52	0.20	-1.14
	188	144	Qz + Talc	10.65	-1.85	15.65	-1.85	73.71	-0.60	0.52	0.23	-1.16
	188	216	Qz + Talc	14.64	-1.21	13.20	-1.70	72.16	-0.60	0.50	0.25	-1.20
	188	312	Qz + Talc	6.85	-1.42	15.55	-0.98	77.59	-0.70	0.46	0.33	-1.29
	188	480	Qz + Talc	12.25	-0.95	16.24	-0.86	71.51	-0.70	0.48	0.26	-1.29
	188	960	Qz + Talc	7.82	-1.38	11.50	-1.38	80.68	-0.60	0.49	0.26	-1.26
	188	1920	Qz + Talc	11.55	-1.26	10.92	-1.16	77.53	-0.63	0.49	0.28	-1.29
	188	3000	Qz + Talc	9.75	-1.18	10.27	-0.87	79.97	-0.64	0.45	0.32	-1.29
	188	4320	Qz + Talc	9.13	-0.74	9.47	-0.81	81.40	-0.59	0.40	0.36	-1.43

Qz: quartz; Cal: Calcite; HMC: High-magnesium calcite; Dol: Dolomite.

$$\Delta^x \text{Mg}_{\text{A-B}} = \delta^x \text{Mg}_{\text{A}} - \delta^x \text{Mg}_{\text{B}} \approx 10^3 \ln \alpha_{\text{A-B}}^{x/24} \quad (2)$$

where $x = 25$ or 26 .

The error in Mg isotope fractionation factors was calculated following the error propagation function:

$$\text{Err} \Delta \text{Mg}_{\text{A-B}} = \left[(\text{Err} \delta \text{Mg}_{\text{A}})^2 + (\text{Err} \delta \text{Mg}_{\text{B}})^2 \right]^{1/2} \quad (3)$$

The accuracy of Mg isotope measurements was verified by analyzing the pure Mg international standards (Cambridge1 and DSM3). Moreover, DTS-2b and two matrix-matching synthetic standard solutions (mixture of NH_4Cl and in-house Mg standard HPS932001, 20:1 mol ratio for NH_4Mg) were processed as the unknown samples in the ion-exchange procedure. The measured Mg isotope compositions for both pure Mg standard solutions and standard samples (DTS-2 and matrix-matching synthetic standard) match well with published values (i.e., $\delta^{26}\text{Mg}_{\text{DTS-2}} = -0.33 \pm 0.04\%$, Appendix Table S1).

3. Results

The mineralogy of the solid products, the relative proportions, and Mg isotopic compositions of the three types of Mg-phases, and the chemical and Mg isotopic composition of the aqueous solution are summarized in Table 1.

3.1. Vapor phase characterization

The CO_2 molecule has four vibrational modes: a symmetric stretching mode (ν_1), two bending modes (ν_{2a} and ν_{2b}), and an antisymmetric stretching mode (ν_3). The interference between the ν_1 and $2\nu_2$ leads to two prominent Raman peaks, including a lower band at $\sim 1285 \text{ cm}^{-1}$ and an upper band at $\sim 1388 \text{ cm}^{-1}$, due to a Fermi resonance (Fermi, 1931) (Fig. 1).

Before the reaction, no signal of CO_2 was observed (Fig. 1), because the vapor phase in the tubes was air, which only contained around 440 ppm of CO_2 , and it was below the detection limit of the Raman. At the first sampling of the reaction tubes (1 day and 12 h for experiments at 130 and 150°C , respectively), the clear CO_2 Fermi bands appeared at $\sim 1286 \text{ cm}^{-1}$ and $\sim 1389 \text{ cm}^{-1}$ (Fig. 1), indicating generation of CO_2 by the reactions between the reactants (i.e., decarbonation of calcites).

3.2. Mineralogy

Based on the XRD analyses, the solid phase in experiments at 130°C and 150°C only consisted of fumed silica and calcite at the early stage of experiments (8 and 6 days of reaction, respectively). This indicates that any newly formed Mg-Si phase at these stages was either poorly crystalline or below the detection limit of XRD. However, flake-like minerals appeared at the time of first sampling (1 day for experiments at 130°C and 12 h for experiments at 150°C) under SEM (Fig. 3D), and the EDS analyses confirmed this mineral only contained Mg, Si, and O (Appendix Fig. S2), consistent with the formation of Mg-bearing silicate phases. These Mg-Si-O flakes may be poorly crystalline talc-like Mg-phyllsilicates or hydrated Mg-silicate precursors. With increasing reaction time, talc reflections became detectable in the XRD patterns (Fig. 2), indicating progressive development of talc-bearing products. Both high-magnesium calcite (HMC) and dolomite can be identified from XRD with longer reaction time (Fig. 2). The newly-formed HMC/dolomite exhibits as the cubic morphology (Fig. 3C), which is significantly different from that of the starting calcite.

In experiments at 188°C , the XRD results showed the appearance of talc after 3 days of reaction (Fig. 2C), while the SEM revealed that the flake-like talc formed after 3 h of reaction (Appendix Fig. S3). The HMC and dolomite first appeared after 3 and 4 days of reaction, respectively. They both disappeared with longer reaction time (after 6 days) and the solid phase consisted only of talc and quartz. (Table 1).

3.3. Solid and fluid chemistry

In all experiments, the proportion of “Exchangeable Mg” decreased with reaction time (Table 1, Fig. 4). At 130°C and 150°C , the proportion of “HCl-leachable Mg” increased rapidly to $\sim 50\%$ within 16 days (Fig. 4), then decreased gradually with longer reaction time. The proportion of “Residual Mg” showed an initial increase over the early experimental period (up to 12 days), followed by a sharp decrease and then a slower increase with continued reaction (Fig. 4).

In contrast, at 188°C the proportion of “HCl-leachable Mg” exhibited a slight but continuous decrease throughout the experiments (Fig. 4), whereas the proportion of “Residual Mg” rose sharply, reaching $>70\%$ within 12 h (Fig. 4). In all experiments, Mg^{2+} concentrations in the aqueous phase decreased with time, while Ca^{2+} concentrations increased (Fig. 4D, Appendix Fig. S4), consistent with progressive uptake of Mg into solid phases and concurrent decarbonation/dissolution

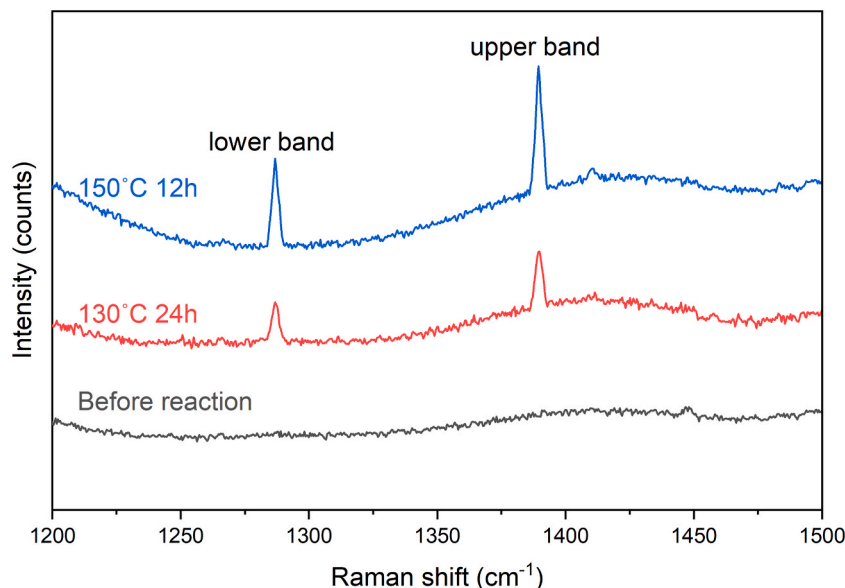


Fig. 1. Raman spectra of vapor phases in sealed reaction tubes.

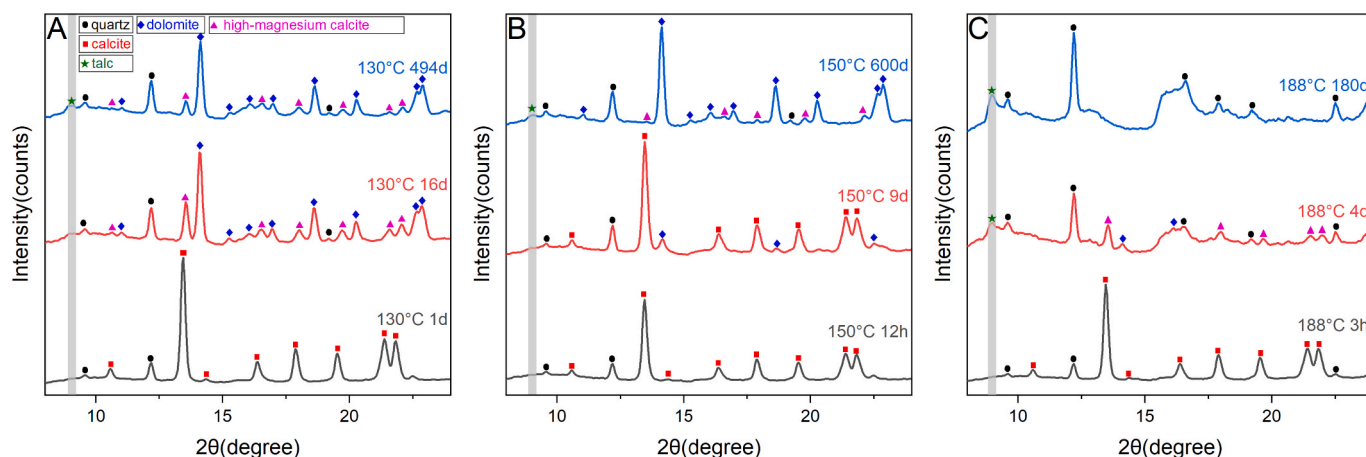


Fig. 2. XRD patterns of representative solid products from experiments at (A) 130°C, (B) 150°C, and (C) 188°C. Circles, squares, triangles, rhombi and stars denote diffraction peaks of quartz, calcite, high-magnesium calcite, dolomite and talc, respectively. The grey shaded band marks the 2θ range of talc peaks.

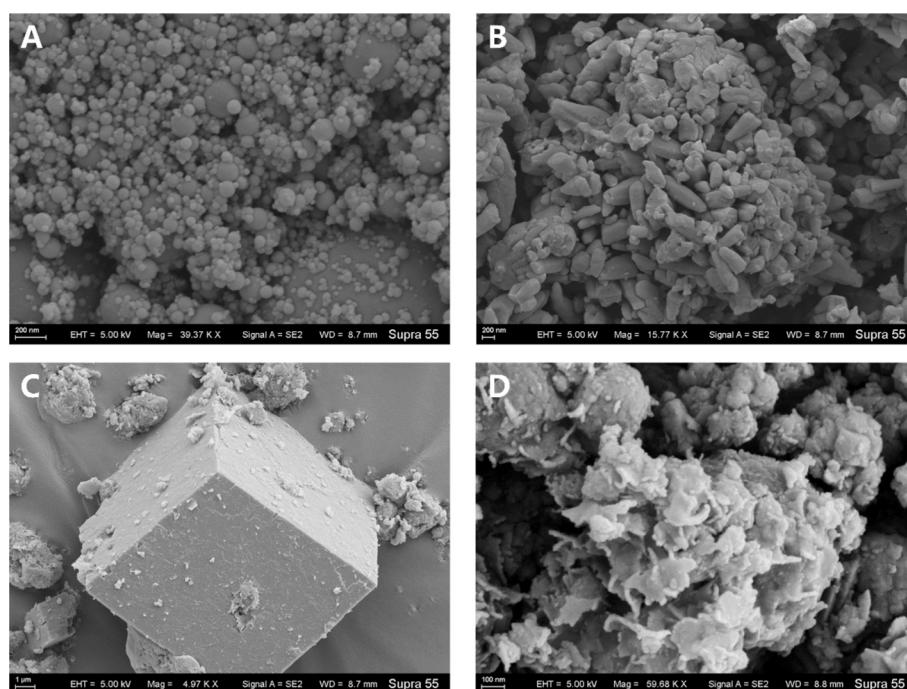


Fig. 3. Representative SEM images of solid reactants and products: (A) initial fumed silica, (B) initial calcite, (C) newly formed dolomite from the 130°C experiment after 16 days, and (D) newly formed Mg-phylosilicate from the 150°C experiment after 12 h.

of calcite.

3.4. Magnesium isotope data

The initial calcite and MgCl_2 solution had $\delta^{26}\text{Mg}$ values of $-1.48 \pm 0.10\text{‰}$ and $-1.05 \pm 0.09\text{‰}$, respectively (Appendix Table S1). The $\delta^{26}\text{Mg}$ value of the fluids in experiments evolves differently at the three experimental temperatures (Table 1). At 130°C, solution $\delta^{26}\text{Mg}$ value remained relatively constant at around -1.0‰ (Fig. 5A), while it showed a slightly increased trend from -1.0‰ to -0.8‰ with increasing reaction time at 150°C (Fig. 5B). In contrast, the $\delta^{26}\text{Mg}$ value of solution decreased to -1.4‰ by the end of experiments at 188°C (Fig. 5C).

The “Exchangeable Mg” tended to be enriched in ^{24}Mg at early stages, and the corresponding $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$ increased with reaction time at all temperatures (Fig. 5D, E), which became positive at 188°C (Fig. 5F). The Mg isotope fractionation between “HCl-leachable Mg” and

aqueous solution ($\Delta^{26}\text{Mg}_{\text{Di-soln}}$) was around -0.4‰ at the first sampling time for experiments at all three reaction temperatures, which reflected the isotopic composition difference between the initial MgCl_2 solution and starting calcite. With increasing reaction time, the $\Delta^{26}\text{Mg}_{\text{Di-soln}}$ showed a sudden decrease coincident with the onset of HMC/dolomite formation, and remained negative throughout the experiments at 130°C and 150°C (Fig. 5G, H), while the $\Delta^{26}\text{Mg}_{\text{Di-soln}}$ value increased to around $+0.4\text{‰}$ at 188°C by the end of experiments (Fig. 5I). The “Residual Mg” was enriched in heavy Mg isotope relative to the solution, and the $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ increased with increasing reaction time (Fig. 5J, K, L), which ranged from $+0.12\text{‰}$ to $+0.83\text{‰}$ for all experiments.

4. Discussion

4.1. Mg isotope mass balance calculation

A strict elemental recovery of Mg based on the recovered dry solid

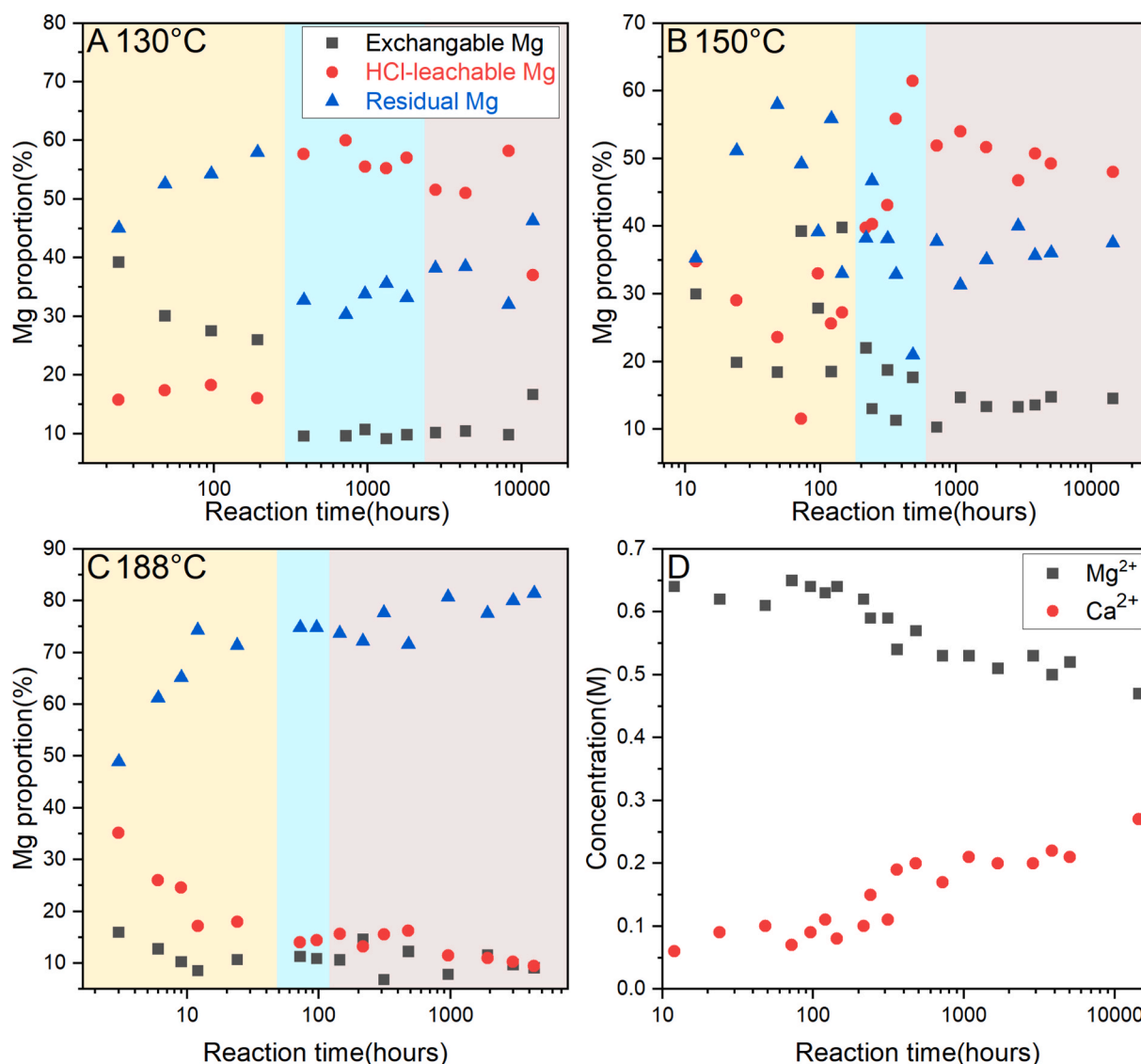


Fig. 4. Evolution of Mg partitioning among solid and solution composition with reaction time. (A–C) Proportions of “Exchangeable Mg”, “HCl-leachable Mg” and “Residual Mg” in solids from experiments at (A) 130°C, (B) 150°C, and (C) 188°C. Black squares, red circles and blue triangles represent “Exchangeable Mg”, “HCl-leachable Mg” and “Residual Mg”, respectively. Light yellow, light cyan and light pink shaded fields indicate the three reaction stages discussed in Section 4.1. (D) Concentrations of Mg^{2+} and Ca^{2+} in solution for the 150°C experiments.

mass is difficult for these experiments because minor physical loss of fine-grained solids may occur during centrifugation, washing and transfer. Therefore, the amount of Mg incorporated into solids was calculated from the difference between the initial Mg input and the Mg remaining in the final solution, and the relative proportions of “Exchangeable Mg”, “HCl-leachable Mg” and “Residual Mg”.

Although this approach does not provide an independent gravimetric recovery of all solid material, it allows a direct Mg isotope mass-balance test. We calculated the bulk $\delta^{26}\text{Mg}$ of each experiment by mass-weighted mixing of the final solution and the three operational solid Mg pools. The calculated bulk $\delta^{26}\text{Mg}$ values are generally consistent with the initial Mg isotopic composition of the system within uncertainty, except for a few minor deviations (Fig. S5). This agreement indicates that there was no isotopically significant unaccounted Mg reservoir.

4.2. The reaction pathways

The reaction process of this system can be divided into three stages, (1) the induction period, (2) the dolomite-forming period, and (3) the talc-ripening period. This subdivision is based on the combined

evolution of mineralogy (XRD and SEM-EDS), the distribution of Mg in solid phase, and the aqueous chemistry (Fig. 4).

In the first stage, no dolomite and HMC is detected by XRD, consistent with the “induction period” originally defined for the calcite-dolomite transformation (Sibley et al., 1987), and subsequently recognized as a fundamental feature of dolomitization kinetics (Kaczmarek and Sibley, 2007, 2011, 2014; Kell-Duivestijn et al., 2019). In this study, the first appearance of dolomite occurred after 16, 9, and 4 days at 130°C, 150°C, and 188°C, respectively, consistent with that the duration of the induction period decreases with increasing temperature (Kaczmarek and Thornton, 2017). During this stage, the dissolution of calcite increased Ca^{2+} concentration and alkalinity, driving the solution toward oversaturation with respect to the dolomite. Raman spectra showed appearance of CO_2 Fermi bands, demonstrating that decarbonation of calcite, in addition to simple dissolution, contributes to CO_2 generation in the closed tubes. Meanwhile, the XRD and SEM-EDS revealed that the newly formed flake-like phase was talc. The overall reaction capturing early talc formation from silica, calcite, and Mg^{2+} can be expressed as:

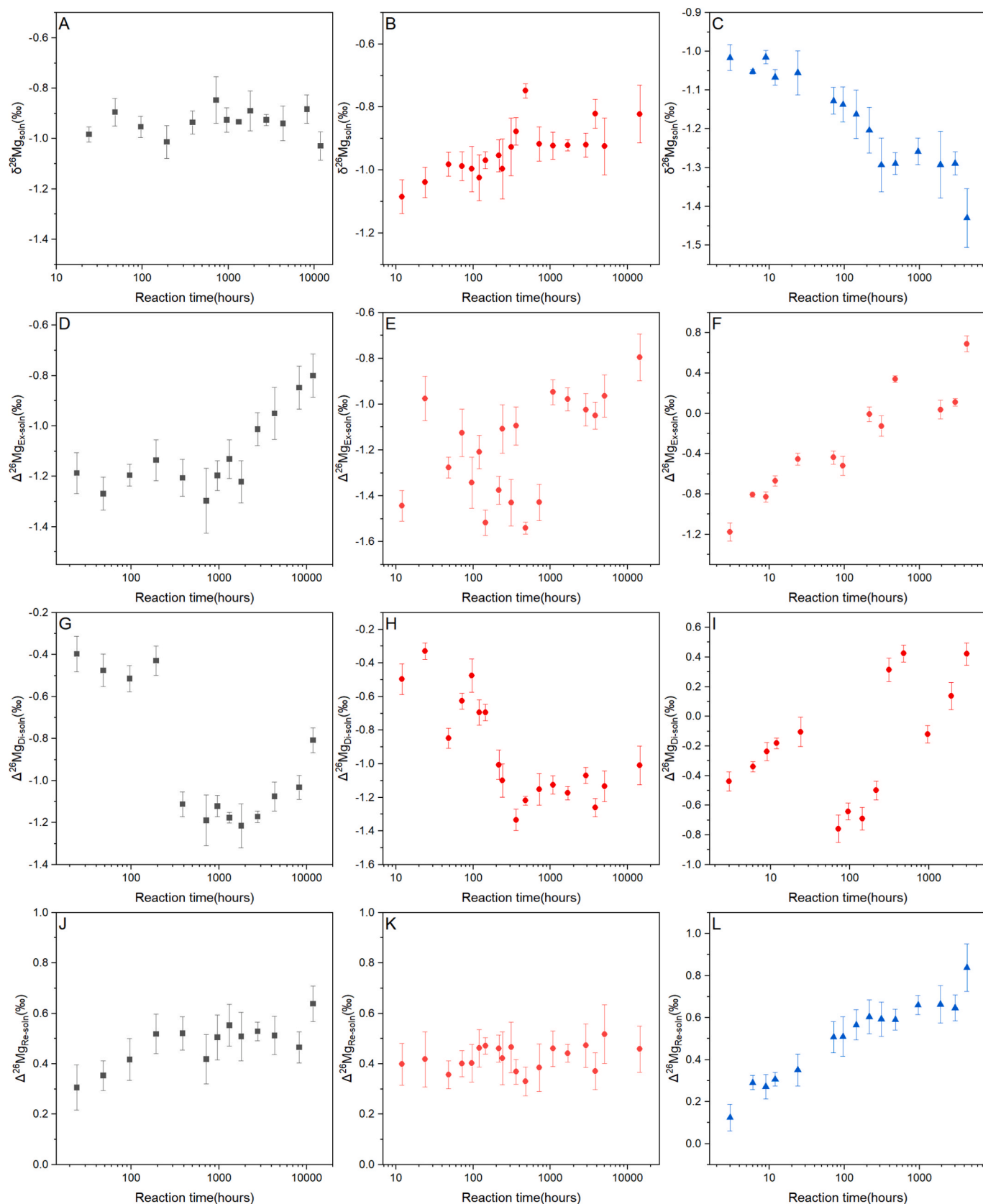
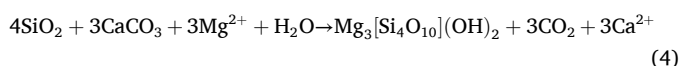


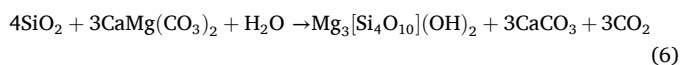
Fig. 5. Time evolution of Mg isotopic composition of solution and Mg isotope fractionation between operational Mg pools and coexisting solution. (A, B, C) $\delta^{26}\text{Mg}$ of solution for experiments at 130, 150, and 188°C, respectively. (D, E, F) $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$ between “Exchangeable Mg” and solution at 130, 150, and 188°C. (G, H, I) $\Delta^{26}\text{Mg}_{\text{Di-soln}}$ between “HCl-leachable Mg” and solution at 130, 150, and 188°C. (J, K, L) $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ between “Residual Mg” and solution at 130, 150, and 188°C.



Consistent with this reaction, the proportion of “Residual Mg” increases markedly during the induction period, indicating the early formation of talc is the dominant sink for aqueous Mg^{2+} .

The second stage is characterized by the appearance and growth of dolomite and/or HMC, as recorded by XRD and SEM. At 130 and 150°C, the onset of dolomite/HMC formation coincided with a steep increase in the proportion of “HCl-leachable Mg” (Fig. 4B), reflecting incorporation of Mg into carbonate phases. The significant different morphology between the starting columnar calcite and the newly formed euhedral to subhedral carbonate rhombs supports a dissolution-reprecipitation mechanism (Kaczmarek and Sibley, 2014). In contrast, at 188°C the proportion of “HCl-leachable Mg” continued to decrease, while the proportion of “Residual Mg” increased gradually (Fig. 4C). This behavior indicated that, at higher temperature, talc remained the principal sink for aqueous Mg^{2+} , and dolomite-forming period at 188°C was short-lived and strongly overprinted by rapid talc growth.

The third stage is marked by a gradual increase in the proportion of “Residual Mg”, indicating continued Mg-bearing layered products growth and “ripening” of the talc-dominated assemblage (Fig. 4C). At 130 and 150°C, this stage likely involves progressive transformation of talc-like Mg-silicate precursors and carbonate-hosted Mg into talc-bearing residual products, given that the early Mg-Si-O flakes are poorly crystalline and that talc reflections are resolved only after longer reaction times. At 188°C, dolomite disappeared from the XRD patterns at longer reaction time, implying dolomite decomposition and conversion into talc. Previous studies have shown that dolomite and silica can react with water to generate talc, calcite, as well as CO_2 (Gordon and Greenwood, 1970; Wan et al., 2017):



In addition, Wan et al. (2017) interpreted this reaction as a metamorphic decarbonation process, where CO_2 was produced by breakdown of carbonate minerals rather than by decomposition of dissolved CO_3^{2-} , and showed that it can proceed at temperatures above 100°C. Our observations were consistent with this mechanism not only at 188°C, where dolomite was fully consumed, but also at 130 and 150°C, where it contributes to the progressive transfer of Mg from dissolved and carbonate fractions into the Mg-bearing layered product “Residual Mg”.

4.3. Magnesium isotope fractionation between solid and solution

4.3.1. Exchangeable Mg

At early reaction stage, the “Exchangeable Mg” was depleted in ^{26}Mg , and the $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$ value increased with time. Notably, positive $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$ values were obtained at 188°C. This behavior cannot be explained solely by the decrease in solution $\delta^{26}\text{Mg}$, because the $\delta^{26}\text{Mg}$ of “Exchangeable Mg” itself increased from -2.19‰ to -0.74‰ (Table 1). We therefore attribute the temporal evolution of $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$ to a progressive transition in the dominant mineral hosts of the “Exchangeable Mg”.

Throughout the experiments, the solid comprised quartz, calcite, talc, dolomite and HMC in varying proportions. The “Exchangeable Mg” thus should include Mg sorbed on mineral surfaces and in interlayers, and can be not only on talc surfaces/interlayers (Evans and Guggenheim, 1988), but also on calcite and fumed silica. Calcite typically carries a negative ζ -potential at neutral to basic pH and can attract cations from solution (Al Mahrouqi et al., 2017; Li et al., 2016). In addition, fumed silica also has a negative ζ -potential (Kosmulski, 2009; Yuxselen-Aksoy and Kaya, 2011), and its very small grain size (~ 100 nm on average) yields a large specific surface area, providing abundant sorption sites for aqueous Mg^{2+} . Therefore, at the early stage, the

“Exchangeable Mg” was dominated by Mg^{2+} adsorbed on calcite and fumed silica surfaces, and was correspondingly enriched in light Mg isotopes.

With increasing reaction time, fumed silica and calcite were progressively consumed to form talc and dolomite/HMC with larger crystal sizes. As consequence, an increasing fraction of the “Exchangeable Mg” became associated with talc surfaces and interlayers, leading to progressively increasing $\delta^{26}\text{Mg}$ values and eventually positive $\Delta^{26}\text{Mg}_{\text{Ex-soln}}$, especially at 188°C where talc dominated the residual solids. It is consistent with the observations of natural weathering profiles, where the kaolin minerals and gibbsite preferentially sorbed heavy Mg isotopes (Huang et al., 2012; Liu et al., 2014). At the same time, previous experimental studies highlight that Mg isotope behavior during sorption is not universal. Hindshaw et al. (2020) found that smectite, despite having a similar TOT layer structure to talc, preferentially adsorbs light Mg isotopes, and little or no Mg isotope fractionation has been reported during Mg uptake onto illite, montmorillonite and kaolinite (Wimpenny et al., 2014). These contrasting results emphasize that Mg isotope fractionation during sorption is sensitive to mineral structure, surface speciation and coordination environment, and may differ substantially among phyllosilicates.

4.3.2. HCl-leachable Mg

The Mg isotope fractionation between the “HCl-leachable Mg” and the solution was initially controlled by the isotopic differences between the starting calcite and MgCl_2 solution during the induction period. Then, the formation of dolomite and HMC led to a significant decrease in $\Delta^{26}\text{Mg}_{\text{Di-soln}}$, which is consistent with previous experiments that dolomite/calcite are depleted in ^{26}Mg compared to solution (Chen et al., 2020a; Li et al., 2015; Li et al., 2012; Liu and Li, 2023; Mavromatis et al., 2025; Mavromatis et al., 2013; Zhang et al., 2024). At 130 and 150°C, the $\Delta^{26}\text{Mg}_{\text{Di-soln}}$ during the dolomite-forming stage was more negative than the equilibrium Mg isotope fractionation between dolomite and aqueous solution (Fig. 6A). This is consistent with the combined effect of HMC and early, poorly ordered dolomite, both of which preferentially incorporate isotopically light Mg relative to ordered dolomite (Li et al., 2015).

At 188°C, $\Delta^{26}\text{Mg}_{\text{Di-soln}}$ behaves differently: values increased with time and eventually became positive. We infer that, in this case, the “HCl-leachable Mg” was not composed solely of carbonates. After dolomite and HMC disappeared from the XRD patterns, any remaining “HCl-leachable Mg” must come from partial dissolution of talc. Because talc itself is isotopically heavy relative to solution (Section 4.3.3), inclusion of talc in the “HCl-leachable Mg” can plausibly account for the higher and positive $\Delta^{26}\text{Mg}_{\text{Di-soln}}$.

4.3.3. Residual Mg

The “Residual Mg” predominantly represented structural Mg in talc and enriched in ^{26}Mg relative to solution. Because the HCl-leaching step can partially dissolve talc, as discussed above, we first evaluated whether this effect influences the Mg isotopic composition of the “Residual Mg”. The 188°C experiments after 312 h provide the best test case, because carbonate phases are absent or minimal in these late-stage products and the $\delta^{26}\text{Mg}$ values of the HCl-leachable fraction increase abruptly. Under these conditions, the HCl-leachable Mg is unlikely to be dominated by carbonate-derived Mg and instead mainly reflects minor acid dissolution of talc or talc-like Mg-phyllosilicate. In these samples, the HCl-leachable Mg accounts for approximately 10–20% of the combined HCl-leachable plus residual Mg pool. We therefore recalculated a corrected residual Mg isotope composition by combining the HCl-leachable and residual pools according to their Mg abundances:

$$\delta^{26}\text{Mg}_{\text{Re,corr}} = (\delta^{26}\text{Mg}_{\text{Di}} \cdot f_{\text{Di}} + \delta^{26}\text{Mg}_{\text{Re}} \cdot f_{\text{Re}}) / (f_{\text{Di}} + f_{\text{Re}}) \quad (7)$$

Where f_{Di} and f_{Re} are the proportions of Mg in the “HCl-leachable Mg” and “Residual Mg”, respectively. The corrected $\delta^{26}\text{Mg}_{\text{Re,corr}}$ values are

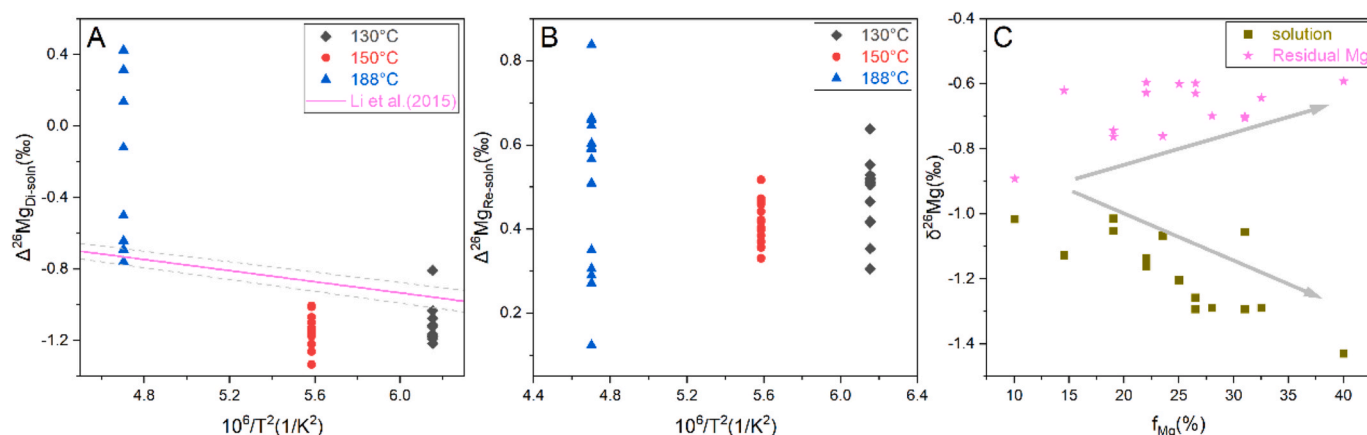


Fig. 6. (A) Mg isotope fractionation between “HCl-leachable Mg” and solution ($\Delta^{26}\text{Mg}_{\text{Di-soln}}$) as a function of reaction temperature during the dolomite-forming stage; the solid line shows the equilibrium dolomite-fluid fractionation from Li et al. (2015). Data from the induction period are excluded. (B) Mg isotope fractionation between “Residual Mg” and solution ($\Delta^{26}\text{Mg}_{\text{Re-soln}}$) versus reaction temperature. (C) $\delta^{26}\text{Mg}$ of solution (squares) and “Residual Mg” (stars) as a function of f_{Mg} (fraction of Mg removed from solution).

very similar to the uncorrected residual values and preserve the same temporal trend of increasing $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ with reaction time (Fig. S6). Thus, partial dissolution of talc during HCl leaching affects the operational distribution of Mg between the HCl-leachable and residual pools, but has only a minor influence on the Mg isotopic composition of the talc-dominated reservoir. This correction confirms that the enrichment of heavy Mg isotopes in the residual pool is not an artifact of the sequential extraction procedure.

In talc, Mg occupies octahedral sites with an average Mg-O bond length of 2.071 Å (Perdikatsis and Burzlaff, 1981), whereas in the aqueous Mg hexa-aquo complex $[\text{Mg}(\text{OH}_2)_6]^{2+}$ the Mg-O bond length is 2.08 Å (Pavlov et al., 1998). This observation is consistent with equilibrium isotope fractionation theory, which predicts that phases with shorter, stiffer Mg-O bonds preferentially incorporate heavy isotopes (Schauble, 2004). A striking feature of our experiments is that the largest magnitudes of $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ occurred at the highest reaction temperature (188°C) (Fig. 6B), opposite to the expectation for equilibrium fractionation, which typically decreases with increasing temperature (Schauble, 2004). Because Mg removal from solution is also greatest at 188°C, progressive precipitation of Mg into the solid phase could, in principle, produce Rayleigh-type isotope evolution of the residual solution (Johnson et al., 2004). However, a simple Rayleigh process cannot explain the observed opposite temporal trends between the residual pool and solution. In such a process, newly formed or cumulatively preserved solids are expected to broadly track the isotope evolution of the solution from which they form, even if early-formed talc becomes isolated from later fluid–solid exchange. In contrast, $\delta^{26}\text{Mg}_{\text{Re}}$ increases with time whereas $\delta^{26}\text{Mg}_{\text{soln}}$ decreases (Fig. 6C). This divergence indicates that the increasing $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ is not controlled solely by progressive Mg removal from solution, but reflects an evolving effective fractionation factor during reaction.

A plausible interpretation for the evolving effective fractionation factor is the progressive maturation of the residual Mg-bearing layered products during reaction, although this maturation does not necessarily follow an identical pathway at all temperatures. In the 130 and 150°C experiments, the earliest flake-like Mg-Si-O products are visible by SEM-EDS but are not resolved as crystalline talc by XRD until longer reaction times, indicating that the residual pool may initially include poorly crystalline talc-like Mg-phylosilicates or hydrated Mg-silicate precursors. Continued reaction would promote dehydration, recrystallization, and layer ordering of these precursor phases, thus modifying the local Mg-O bonding environment and changing the apparent fractionation between the residual solid and solution. While in the 188°C experiments, where talc appears early and becomes the dominant Mg-

bearing phase, aging promotes higher crystallinity and more ordered talc layer stacking, as evidenced by the sharpening of talc XRD peaks (Fig. 2). The increasing crystallinity is expected to shorten and stiffen the average Mg-O bonds, thereby enhancing the preference of the solid phase for heavy Mg isotopes. However, Mg-O bond lengths were not directly measured in our experimental products. Direct verification of this interpretation will require future structural measurements. For example, synchrotron X-ray diffraction, pair distribution function analysis, and Mg K-edge EXAFS could quantify Mg-O bond lengths, octahedral distortion, and short-range ordering in products formed at different reaction times and temperatures. These analyses would provide direct tests of whether increasing Mg isotope fractionation is linked to Mg-O bond stiffening during structural maturation. Similar couplings between structural ordering or crystallinity and Mg isotope fractionation have been reported for dolomite, smectites, and sepiolite, where more ordered or better-crystallized phases exhibit higher Mg isotope fractionation factors (Baldermann et al., 2025; Hindshaw et al., 2020; Li et al., 2015).

Overall, our experiments show that talc can be significantly enriched in heavy Mg isotopes relative to hydrothermal fluids, with $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ reaching $\sim +0.8\text{‰}$ in the most crystalline, late-stage 188°C products. We therefore treat $\sim +0.8\text{‰}$ as the largest fractionation observed for the talc in our experiments, rather than as a temperature-dependent equilibrium fractionation factor. This value provides an experimental reference for relatively crystalline talc formed under the present hydrothermal conditions. Natural talc and related talc-like Mg-phylosilicates may record a range of fractionation factors depending on temperature, precursor phases, hydration state, defect density, layer ordering, reaction progress, and structural maturity. The strong time dependence of $\Delta^{26}\text{Mg}_{\text{Re-soln}}$ nevertheless indicates that newly formed Mg-phylosilicates are unlikely to record instantaneous isotope equilibrium with coexisting fluids, and that their isotope signatures can evolve substantially during structural ordering and mineral maturation.

4.4. Controls on Mg isotope fractionation in Mg-bearing layered minerals

Our experimentally constrained talc-fluid Mg isotope fractionation can be directly compared with Mg isotope partitioning among other Mg-phylosilicates and brucite, which all share the same fundamental MgO_6 octahedral sheet structure. Previous work has shown that these phases span a wide range of $\Delta^{26}\text{Mg}$ relative to aqueous solution, with both positive and negative mineral-fluid fractionations depending on structure and bonding environment. Brucite, chrysotile, and smectite tend to be depleted in ^{26}Mg relative to aqueous solution (Hindshaw et al., 2020;

Li et al., 2014; Stamm et al., 2022; Wimpenny et al., 2010), whereas kerolite, lizardite, sepiolite and now talc are enriched in ^{26}Mg (Baldermann et al., 2025; Ryu et al., 2016). The first-order control on the mineral-fluid Mg isotope fractionation is the Mg-O bond length within MgO_6 octahedra. Among these mineral phases and $[\text{Mg}(\text{OH}_2)_6]^{2+}$ complex, the average Mg-O bond length increases in the order of lizardite < talc $\approx$ kerolite < sepiolite < $[\text{Mg}(\text{OH}_2)_6]^{2+}$ < chrysotile < brucite (Appendix Table. S2), demonstrating that minerals with shorter, stronger Mg-O bonds preferentially incorporate heavy Mg isotopes.

However, even for the same mineral, the reported Mg isotope fractionation factors are not always consistent. Li et al. (2014) used three-isotope exchange experiments at 22°C to constrain brucite-Mg aquo ion isotope exchange, and also reported brucite synthesis experiments at 7, 22 and 40°C. The obtained apparent brucite-Mg aquo ion fractionation increased from negative values at 7 to 22°C toward near-zero values at 40°C. Stamm et al. (2022) also applied the three-isotope method at 25°C, and obtained low-temperature equilibrium brucite-Mg $^{2+}$ fractionation factors broadly consistent with the low-temperature results of Li et al. (2014). In contrast, Wimpenny et al. (2014) reported a positive brucite-solution fractionation factor of $\sim +0.5\text{‰}$ for brucite synthesized at 80°C. Because equilibrium isotope fractionation generally decreases in magnitude with increasing temperature, the shift from negative or near-zero values at lower temperatures to positive values at 80°C is unlikely to reflect temperature dependence alone. Our talc experiments provide new sights to reconcile such discrepancies. We observe that $\Delta^{26}\text{Mg}_{\text{talc-soln}}$ increases systematically as talc ripens and its crystallinity improves: early-formed, poorly ordered talc shows smaller heavy-Mg enrichment, whereas more ordered talc at later times exhibits larger Mg isotope fractionation. In this framework, higher synthesis temperature may produce a structurally more mature brucite, with shorter and/or stiffer effective Mg-O bonding relative to low-temperature brucite. This interpretation does not exclude normal temperature dependence once structurally mature brucite has formed, as suggested by the smaller brucite $\pm$ iowaite-serpentine offset in natural serpentinites compared with the 80°C brucite-solution fractionation of Wimpenny et al. (2014) (Nielsen et al., 2024).

Smectite shows a similar sensitivity to crystallinity. Hindshaw et al. (2020) reported light-Mg enrichment in smectite formed at low temperature, whereas Voigt et al. (2020) observed positive smectite-solution Mg isotope fractionation at 250 and 290°C. Furthermore, Hindshaw et al. (2020) and Baldermann et al. (2025) observed that, in both smectites and sepiolite, better-crystallized phases have higher $\Delta^{26}\text{Mg}$ values, directly linking isotopic behavior to crystal order. In summary, these observations suggest that Mg isotope fractionation in Mg-phylosilicates and brucite is governed not only by ideal crystal structure, but also by the degree of structural maturation, and that time- and temperature-dependent ordering can reconcile much of the apparent contradictories in published Mg isotope fractionation factors for Mg-bearing layered minerals.

4.5. Geological implications

4.5.1. Mg isotope heterogeneity in altered oceanic crust

Our experiments demonstrate that Mg-bearing layered minerals do not form in instantaneous Mg isotope equilibrium with aqueous solution. Instead, the $\Delta^{26}\text{Mg}$ is strongly dependent on reaction time and mineral crystallinity. This finding provides a new insight for interpreting $\delta^{26}\text{Mg}$ variations in altered oceanic crust (AOC). Previous studies have reported the $\delta^{26}\text{Mg}$ values of AOC ranged from -2.76‰ to $+0.50\text{‰}$ (Huang et al., 2018; Santiago Ramos et al., 2020), with the most negative values ($\delta^{26}\text{Mg} < -1\text{‰}$) generally attributed to the formation of carbonate minerals, which preferentially incorporate light Mg isotopes (Chen et al., 2020a; Li et al., 2015; Li et al., 2012; Liu and Li, 2023; Liu et al., 2023; Mavromatis et al., 2013; Wang et al., 2013).

However, after excluding these carbonate-influenced low $\delta^{26}\text{Mg}$ outliers, AOC still displays a wide range of $\delta^{26}\text{Mg}$ from -0.73‰ to

$+0.50\text{‰}$. This variability is significant because bulk AOC generally contains both primary igneous Mg and secondary alteration Mg. The inherited igneous component is expected to remain close to mantle-like $\delta^{26}\text{Mg}$ values of $\sim -0.25\text{‰}$ (Teng et al., 2010), whereas the alteration component may be hosted by chlorite, smectite, talc, serpentine, kerolite, sepiolite, or other Mg-bearing secondary phases. Bulk-rock $\delta^{26}\text{Mg}$ values therefore represent mixtures between these reservoirs, and the isotopic variability of the alteration component may be larger than that expressed by the bulk rock, especially where primary igneous Mg remains abundant. The wide non-carbonate $\delta^{26}\text{Mg}$ range in AOC therefore points to heterogeneous Mg isotope fractionation during alteration. Part of this heterogeneity may reflect differences in alteration mineralogy and fluid chemistry, but our results suggest that reaction progress and structural maturity of Mg-bearing phyllosilicates can also be important. Progressive growth and ripening of talc-like or other Mg-bearing layered minerals may shift mineral-fluid Mg isotope fractionation toward heavier solid compositions as local Mg-O bonding environments become more ordered. Thus, $\delta^{26}\text{Mg}$ variations in AOC may record not only fluid composition and carbonate-silicate mixing, but also the mineralogical and structural evolution of non-carbonate Mg alteration phases during hydrothermal alteration.

4.5.2. The oceanic Mg cycle

The temporal evolution of the mass and isotopic composition of Mg in the ocean is mainly controlled by riverine inputs of weathering products and groundwater-derived solute fluxes (Demichio et al., 2005; Mayfield et al., 2021; Tipper et al., 2006), balanced by removal through dolomitization, high-temperature alteration at mid-ocean ridges, low-temperature alteration of the oceanic crust and authigenic clay formation (Elderfield and Schultz, 1996; Hardie, 1996; Higgins and Schrag, 2015; Holland, 2005; Hu et al., 2022; Shalev et al., 2019; Shalev and Ramos, 2025). Among these output fluxes, low-temperature alteration and clay formation remove Mg into a diverse assemblage of Mg-bearing phyllosilicates and related Mg-silicates. In seafloor hydrothermal and sedimentary environments, these phases may include talc, chlorite, saponite/smectite, kerolite, sepiolite and poorly crystalline Mg-silicate phases, depending on temperature, silica activity, fluid chemistry and reaction progress (e.g., Allen and Seyfried, 2003; Dekov et al., 2008; Marumo and Hattori, 1999; Oyanagi et al., 2020; Schroeder et al., 2015).

Our experiments show that talc preferentially incorporate heavy Mg isotopes relative to coexisting fluid, and that the magnitude of this fractionation increases with reaction progress and structural maturation. This behavior indicates that the Mg isotopic composition of the phyllosilicate sink is unlikely to be controlled solely by mineral stoichiometry or by a single equilibrium fractionation factor. Instead, mineral type, crystallinity, hydration state, layer ordering and Mg-O bonding environment may all influence the effective $\Delta^{26}\text{Mg}$ between marine Mg-phylosilicates and seawater-derived fluids. Therefore, the experimentally observed fractionation provides an end-member reference for relatively crystalline Mg-phylosilicates.

Previous studies have shown that the assumed isotope composition of the phyllosilicate sink strongly affects the relative roles assigned to low-temperature alteration and dolomitization in maintaining the seawater Mg isotope budget (e.g., Shalev and Ramos, 2025). Our results reinforce this sensitivity, but also indicate that the bulk $\Delta^{26}\text{Mg}_{\text{phy-sw}}$ is likely to vary with the mineralogical and structural state of the authigenic Mg-silicate assemblage. Quantitative constraints on this sink therefore require mineral-specific fractionation factors for natural marine Mg-phylosilicates with well-characterized crystallinity and Mg coordination environments. Until such constraints are available, Mg isotope mass-balance calculations involving the marine phyllosilicate sink should be regarded as sensitive to this mineralogical and structural uncertainty.

4.5.3. Mg isotope signatures of talc-bearing subduction-interface lithologies

Talc-bearing lithologies are common products of fluid-rock

interaction in ultramafic and mafic–ultramafic systems, including altered oceanic lithosphere and serpentinite-associated metasomatic rocks at the slab-mantle interface (Bucher et al., 2005; Peacock and Hyndman, 1999; Poli and Schmidt, 2002; Spandler et al., 2007). These rocks commonly form where silica-bearing fluids react with serpentinized peridotites, or where mafic, ultramafic and sedimentary lithologies interact along the slab-mantle interface. Our experiments provide a mechanistic basis for interpreting high $\delta^{26}\text{Mg}$ values in such talc-bearing metasomatic rocks: talc preferentially incorporate heavy Mg isotopes relative to coexisting hydrothermal fluids. This interpretation is consistent with field-based Mg isotope studies showing that talc-rich rocks can be isotopically heavier than associated serpentinites or ultramafic protoliths (Beinlich et al., 2014; Easthouse et al., 2025), and that high- $\delta^{26}\text{Mg}$ Mg-rich metasomatic rocks may record interaction with fluids involving talc- or serpentinite-bearing lithologies in subduction channels (Chen et al., 2020b; Chen et al., 2016; Qu et al., 2025).

As subduction proceeds, talc-bearing metasomatic rocks will undergo further reaction with increasing pressure, temperature and fluid flux. Although phase-equilibrium constraints indicate that talc is generally consumed before typical sub-arc magma-generation conditions are reached (Poli and Schmidt, 2002), the heavy-Mg signature acquired during talc formation may be retained within the slab-mantle interface system and redistributed during talc breakdown, fluid-rock interaction, or metamorphic replacement into subsequent fluids or newly formed hydrous assemblages. Similar heavy-Mg signatures may also arise from other Mg-rich reactions in serpentinites, including the breakdown of brucite and iowaite (Nielsen et al., 2024).

5. Conclusion

This study provides the first experimental determination of Mg isotope fractionation between talc and hydrothermal fluids. Closed-system experiments at 130–188°C demonstrate that Mg isotope fractionation evolves systematically with reaction progress, where $\Delta^{26}\text{Mg}_{\text{talc-soln}}$ increases from values near zero during early stage to $\sim +0.80\text{‰}$ as the mineral structure ripens and crystallinity improves. This time-dependent behavior offers a framework to reconcile the apparently conflicting Mg isotope fractionation factors reported for other Mg-bearing layer minerals (e.g., brucite, smectite). Our results suggest that the reported positive and negative fractionations largely reflect differences in structural order and Mg–O bond strength. Therefore, structural maturation plays an important role on the isotopic signatures of Mg-bearing phyllosilicates.

These findings have important implications for the Mg cycle in the oceanic lithosphere. Authigenic Mg-phyllosilicates such as talc, chlorite and smectite represent a significant sink for seawater Mg that preferentially sequesters heavy isotopes, but their effective Mg isotope fractionation is likely to depend on mineral identity and structural state. The broad range of $\delta^{26}\text{Mg}$ values observed in altered oceanic crust may therefore partly reflect variable mineralogical and structural maturation within dynamic hydrothermal systems. By providing an experimental reference for relatively crystalline talc-dominated products, our results highlight the sensitivity of oceanic Mg-budget reconstructions to the assumed isotope fractionation of the marine Mg-silicate sink, rather than providing a single bulk fractionation factor for all marine clay minerals.

Finally, talc-forming reactions may generate heavy-Mg signatures in talc-bearing metasomatic lithologies at the slab-mantle interface. Talc-rich rocks may therefore represent localized heavy-Mg reservoirs in altered oceanic lithosphere and subduction-interface environments. During continued subduction, these signatures may be retained and redistributed during talc breakdown, fluid-rock interaction or metamorphic replacement into subsequent fluids or newly formed hydrous assemblages. This interpretation highlights the potential of Mg isotopes for tracing fluid-rock interaction and Mg redistribution in talc-bearing subduction-interface lithologies, without requiring talc to remain stable to sub-arc magma-generation depths.

Data availability

Data are available through Mendeley Data at <https://doi.org/10.17632/s39hxcmpz8.2>.

CRediT authorship contribution statement

Chuan Liu: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Zhiguang Xia:** Validation, Methodology, Investigation. **Shichao An:** Resources, Methodology. **Weiqiang Li:** Writing – review & editing, Validation, Supervision, Resources, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

The [Supplementary Material](#) provides additional experimental and data documentation. [Fig. S1](#) shows photograph of the fused-glass reaction tube used in the hydrothermal experiments. [Figs. S2 and S3](#) are EDS spectra and SEM images of the Mg-phyllosilicate (talc) formed in the 150°C and 188°C experiments at early reaction time, respectively. [Fig. S4](#) is the evolution of solution composition with reaction time for experiments at 130°C and 188°C. [Fig. S5](#) is the Mg isotope mass balances for each of the experiments. [Fig. S6](#) is comparison between Mg isotopic composition of “Residual Mg” before and after the correction from “HCl-leachable Mg” for experiments at 188°C. [Table S1](#) reports Mg isotopic compositions of test solutions passed through ion-exchange chromatography, showing the accuracy and reproducibility of our analytical method. [Table S2](#) compiles Mg–O bond lengths and mineral-fluid Mg isotope fractionation factors for Mg-bearing layered minerals from experimental calibrations in the literature.

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