Geochemical Perspectives Letters

Published by the European Association of Geochemistry

Mixed paragenesis diamond inclusions form during metasomatic fluid evolution

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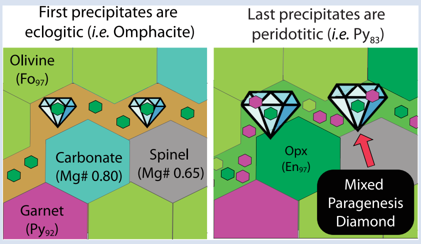
https://doi.org/10.7185/geochemlet.2625

Abstract



Mineral inclusions in diamond provide the context for diamond formation, yet some diamonds contain disequilibrium 'mixed paragenesis' mineral inclusions. Mixed paragenesis diamonds are inferred to somehow record growth of a single diamond in both mafic eclogitic and peridotitic substrates. Here we use thermodynamic modelling with the Extended Deep Earth Water (DEW) model to show that mixed paragenesis mineral inclusions can form during a single episode of diamond growth *via* fluid-rock metasomatism in the lithospheric mantle. Simulations of fluid infiltration and reactions predict sequential mineral precipitation, with early eclogitic minerals (omphacite and garnet) followed by more peridotitic minerals (*e.g.*, olivine and Mg-rich garnet) during continued diamond growth. Concurrently, modelled fluids evolve from silicic toward carbonatitic compositions and reproduce the compositional range of high density fluids observed in fibrous diamonds. These results demonstrate that mixed paragenesis inclusions and the diversity of diamond-forming fluids can arise from progressive metasomatic reactions during a single diamond-forming event, rather than requiring a diamond to grow first in eclogite and then in peridotite.

Received 2 April 2026 | Accepted 5 June 2026 | Published 1 July 2026

Introduction

Mineral inclusions in diamonds provide constraints on the geological environment of diamond formation. However, the relative abundance of minerals encapsulated in diamonds does not reflect the modal mineralogy of Earth's mantle (Stachel and Harris, 2008). This mismatch may reflect [1] preferential inclusion of minerals associated with lower surface energies (Meyer and Boyd, 1972), [2] a syngenetic process where sulphides, garnet, and Mg chromite may precipitate or recrystallise more readily during diamond growth driven by fluid (Mikhail *et al.*, 2019; 2021; Rinaldi *et al.*, 2023) and/or melt metasomatism (Pintér *et al.*, 2022), or [3] a combination of both.

Silicate inclusions in diamond are typically classified into three paragenetic groups: peridotitic (*e.g.*, Cr-rich pyrope, diopside, enstatite, olivine), eclogitic (*e.g.*, Cr-poor pyrope-almandine and omphacite), and websteritic (intermediate compositions; Gurney *et al.*, 1984). Rarely, diamonds contain disequilibrium assemblages termed mixed paragenesis, in which minerals from different paragenetic groups occur within a single crystal (Prinz *et al.*, 1975; Mikhail *et al.*, 2019; Lai *et al.*, 2022). For example, eclogitic minerals may occur in the core of a diamond (*e.g.*, omphacite, eclogitic garnet, or coesite), whereas peridotitic minerals occur near the rim (*e.g.*, forsteritic olivine or Mg chromite) (Lai *et al.*, 2022). Mixed paragenesis diamonds have been suggested to record multiple episodes of diamond growth in different host rocks which requires physical movement of the diamond between different mantle environments (*e.g.*, Wang, 1998; Lai *et al.*, 2022). In contrast, our results show that, despite comments to the contrary (Lai *et al.*, 2022), the mixed parageneses in a single diamond may form from the progressive trapping of minerals formed during metasomatic reactions (*e.g.*, Mikhail *et al.*, 2021). We use predictive reaction path models of irreversible reactions between rocks and diamond forming aqueous fluids (Sverjensky and Huang, 2015; Huang and Sverjensky, 2020; Mikhail *et al.*, 2021; Rinaldi *et al.*, 2023) with thermodynamic data from the DEW model (Huang and Sverjensky, 2019; Sverjensky, 2019) as a process based framework to explore the possibility that mixed paragenesis inclusions can form during a single episode of diamond growth *via* fluid-rock metasomatism. In addition to examining the mineral assemblages produced during metasomatism, we evaluate whether such reactions reproduce the compositional range of diamond forming fluids observed in fibrous diamonds.

Methods

Modelling approach. We used the Extended Deep Earth Water (DEW) model to simulate fluid-rock metasomatism in the lithospheric mantle. DEW extends the EQ3/EQ6 geochemical modelling framework for aqueous speciation and irreversible mass transfer reactions to mantle pressures and temperatures (Huang and Sverjensky, 2019). EQ3 was used to calculate equilibrium fluid compositions, and EQ6 was used to simulate irreversible fluid-rock reactions during metasomatism. The thermodynamic data set is calibrated using experimentally

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determined solubilities at upper mantle pressures and temperatures (*e.g.*, Kessel *et al.*, 2015) and has previously been applied to model diamond forming fluids and metasomatic reactions in the lithospheric mantle (Sverjensky and Huang, 2015; Huang and Sverjensky, 2020; Mikhail *et al.*, 2021; Rinaldi *et al.*, 2023).

Model parameterisation. Simulations were performed at 5 GPa, 1000 °C and logfO_2 = -2 ΔFMQ, conditions consistent with lithospheric diamond formation (Stachel and Harris, 2008). The initial fluid compositions were calculated from equilibrium between water and eclogite or water and carbonated dunite assemblages following Huang and Sverjensky (2020). Mineral solid solutions were represented using ideal site mixing between pyrope, almandine, and grossular in garnet, and non-ideal mixing between diopside, hedenbergite, and clinoenstatite in clinopyroxene following Huang and Sverjensky (2020). The chosen temperature of 1000 °C lies at the lower end of the estimated diamond inclusion entrapment temperature range (1155 ± 105 °C; Stachel and Luth, 2015). Two model scenarios were investigated, termed Model I and Model II, as described below.

Results

Model I. Infiltration of eclogitic fluid into dry carbonated peridotite. This simulation models the infiltration of an externally derived fluid into a dry mantle rock, following an approach similar to that of Rinaldi *et al.* (2023). An eclogitic fluid was generated by equilibrating water with an eclogite assemblage (clinopyroxene + garnet + coesite + diamond) using EQ3. The resulting fluid was then reacted with an anhydrous carbonated peridotite using EQ6 to simulate irreversible fluid-rock interaction. Because the infiltrating fluid and host rock are far from equilibrium, progressive reaction results in sequential mineral precipitation (Fig. 1a). At early reaction progress, the precipitating phases have an eclogitic character, dominated by omphacite (Jd_{67}) and garnet (Py_{46}). Diamond precipitation begins during these early stages and may encapsulate these minerals as inclusions in the cores of growing crystals. With increasing reaction progress, the mineral assemblage evolves toward more peridotitic compositions, including olivine (Fo_{95}) and a more magnesian garnet (Py_{83}). Continued diamond growth during these later stages allows these minerals to be preserved as inclusions near diamond rims. This sequence predicts that a single diamond growth episode may first encapsulate eclogitic and then later peridotitic inclusions as metasomatism progresses.

Model II. Infiltration of carbonatitic fluid into eclogite with mixing of eclogitic pore fluid. Model II simulates a more complex scenario involving simultaneous fluid mixing and reaction with the host rock, similar to the model of Huang and Sverjensky (2020) for fluid-rich diamonds from the Panda kimberlite. In the present model, a carbonatitic fluid infiltrates eclogite containing an eclogitic pore fluid. The assumption of localised pore or grain boundary fluids in eclogitic substrates is motivated by the broader evidence for fluid mediated metasomatism in mantle lithologies, including the transport of incompatible elements, carbonate species, and volatile-rich metasomatic agents through the lithospheric mantle (see Steele-MacInnis, 2025 for a review). As infiltration proceeds, the invading fluid progressively mixes with the resident pore fluid while reacting with the surrounding rock. The carbonatitic fluid was generated by reacting water with olivine + diamond and minor orthopyroxene and garnet using EQ6, producing a fluid in equilibrium with olivine (Fo_{97}) and garnet (Py_{92}). This fluid was then allowed to react with eclogite containing clinopyroxene, garnet and minor coesite while simultaneously mixing with the resident pore fluid. During early reaction progress, the composition of precipitating minerals is dominated by the eclogitic pore fluid, resulting in the formation of omphacite (Jd_{67}) and garnet (Py_{45}) (Fig. 1b). Diamond precipitation begins during these early stages and may encapsulate these minerals within the cores of growing diamonds. As the proportion of carbonatitic fluid increases with continued reaction, the mineral assemblage evolves toward more peridotitic compositions, including a more magnesian garnet (Py_{83}). These later minerals may become trapped as inclusions near diamond rims.

Discussion

The formation of mixed paragenesis diamond inclusions. To place the model results in a natural context, we compare them with

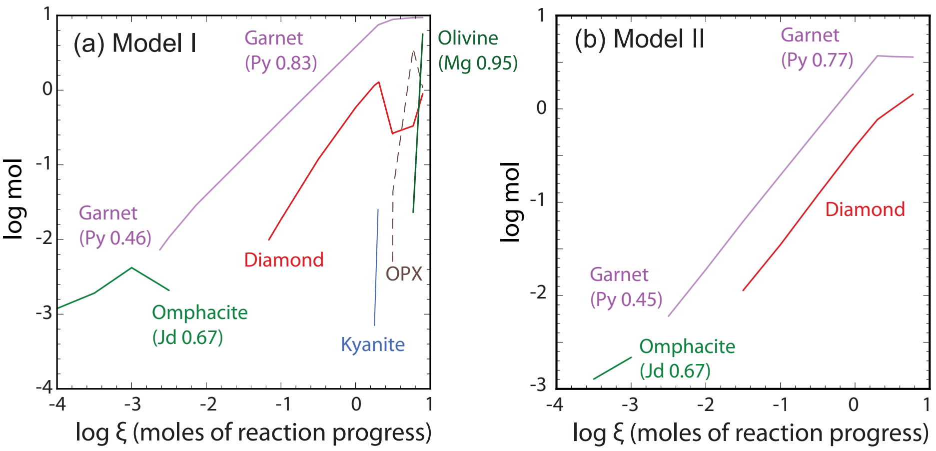

Figure 1 Predicted mineral precipitation during reaction progress in two DEW fluid-rock interaction models. Log mineral abundance (mol) is plotted against log reaction progress (ξ), where ξ represents the extent of fluid-rock interaction simulated by irreversible mass transfer in EQ6. **(a)** Model I predicts progressive garnet growth (Py 0.46–0.83), followed by omphacite (Jd ~0.67), diamond precipitation, and late olivine (Mg# ~0.95), with minor orthopyroxene and kyanite at advanced reaction progress. **(b)** Model II shows a simpler sequence dominated by garnet and omphacite formation followed by diamond precipitation. Values in parentheses indicate representative mineral compositions predicted by the models. The simulations illustrate mineral assemblages produced during mantle fluid-rock interaction capable of forming diamond bearing eclogitic-peridotitic assemblages.

mixed paragenesis diamonds from the Koidu kimberlite complex, Sierra Leone (Lai *et al.*, 2022). The predicted mineral sequences closely reproduce the most common inclusion relationships documented in these diamonds. For example, their diamond #138-7 contains an eclogitic garnet inclusion in the core and olivine near the rim, a sequence consistent with Model I (Fig. 1a). Similarly, their diamond #133-6 contains omphacite in the core and Mg chromite near the rim, consistent with a broader evolution toward increasingly magnesian mineral assemblages during metasomatism. Our models did not produce coesite. However, variants of Model II involving higher initial coesite abundances could plausibly permit preservation and encapsulation of coesite during diamond growth, consistent with previous studies (Rinaldi *et al.*, 2023).

Chromium is not included in our DEW models, and therefore Mg chromite is not predicted by these models. However, the Mg chromite reported by Lai *et al.* (2022) is an Mg-Fe-Al chromite ($(Mg,Fe)O(Cr,Al,Fe)_2O_3$), although the present simulations do not explicitly reproduce Mg chromite because chromium is not included within the current modelling framework. The appearance of these minerals during later stages of reaction reflects a shift toward more peridotitic mineral assemblages during progressive metasomatism (as predicted here; Fig. 1).

Taken together, the models demonstrate that disequilibrium mineral assemblages can be produced and preserved during a single episode of diamond growth under constant pressure-temperature conditions. Mixed paragenesis inclusions may therefore form as direct products of diamond forming fluid-rock interaction, without requiring a single diamond to grow first in one mantle lithology and then again in a second mantle lithology.

Relationship between diamond inclusions and the diamond forming substrate. The Koidu kimberlite complex is unusual in that mantle xenoliths recovered from the pipes are exclusively eclogitic (Hills and Haggerty, 1989). Nevertheless, diamonds from Koidu contain inclusions with clearly peridotitic compositions, including olivine and Mg-Fe-Al chromite (Lai *et al.*, 2022). This apparent mismatch between xenolith mineralogy and diamond inclusions has important implications for the nature of the diamond forming substrate. Our models predict that olivine precipitates from the fluid only when the reacting host rock is highly Mg-rich. This suggests that olivine bearing, mixed paragenesis diamonds likely formed within depleted peridotitic lithologies rather than within eclogite itself. Although such lithologies are not represented among xenoliths from the Koidu kimberlites, indicator mineral concentrates include both eclogitic and peridotitic compositions, with garnets characterised by high Cr and low Ca contents (Harder *et al.*, 2013). These garnet compositions indicate the presence of highly depleted harzburgitic or dunitic mantle beneath the Koidu kimberlite field. Mixed paragenesis diamonds may therefore provide direct evidence of the lithologies involved in diamond formation in the absence of xenoliths. Where spatial relationships between inclusions are preserved, the sequence of inclusion entrapment may record metasomatic reaction pathways operating during diamond growth (*i.e.* progressive geochemical evolution; Figs. 1, 2).

These data support a growing body of evidence that diamond and diamond inclusion formation is intimately linked to metasomatism of mantle rocks (illustrated in Fig. 2). For example, fluid-rock interaction can partially dissolve existing mantle minerals and precipitate new phases during diamond growth (*e.g.*, Walter *et al.*, 2011; Aulbach *et al.*, 2013; Thomson *et al.*, 2014; Huang and Sverjensky, 2020; Mikhail *et al.*, 2021; Pasqualetto *et al.*, 2022; Rinaldi *et al.*, 2023; Bruno *et al.*, 2024; Howarth *et al.*, 2025). The dissolution and precipitation of phases explains why the modal abundance of silicate and oxide inclusions in diamond do not reflect the mineralogy of the mantle (Stachel and Harris, 2008; Stachel and Luth, 2015), and why the major element geochemistry of many garnet and clinopyroxene diamond inclusions

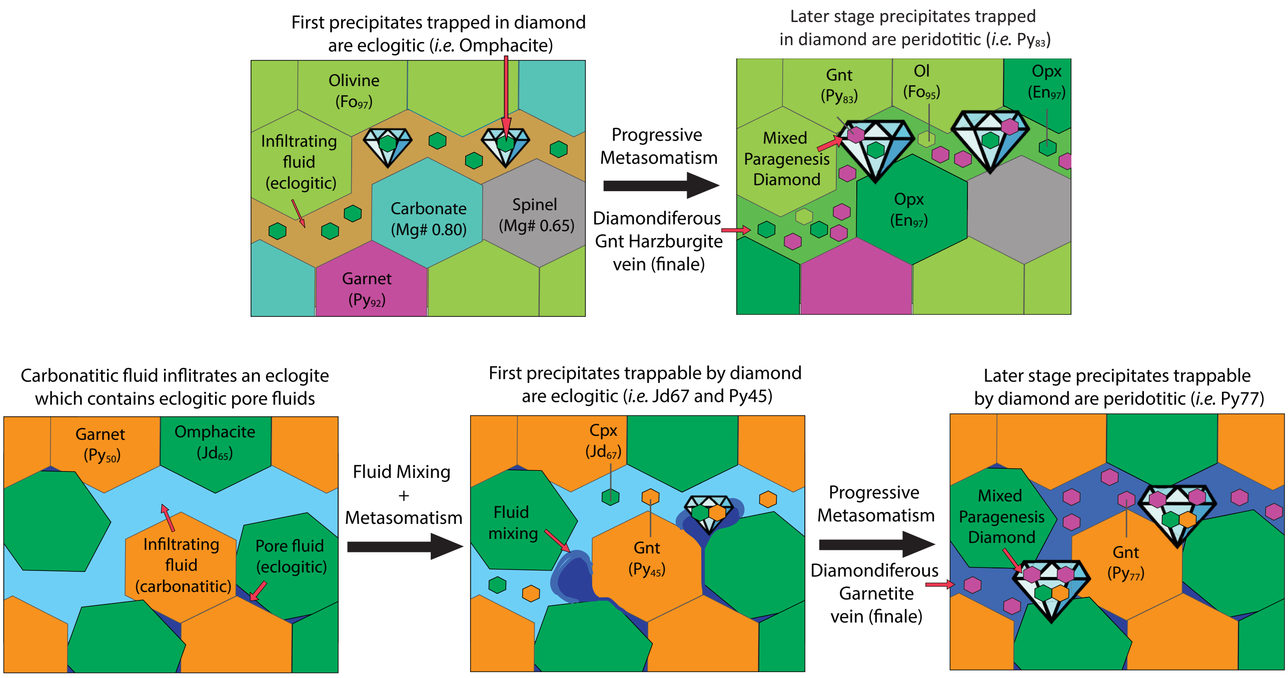

Figure 2 Conceptual illustrations of metasomatic fluid-rock interaction represented by Models I and II (top and bottom). Fluids infiltrate mantle rocks along veins and react with the surrounding lithologies, precipitating minerals and diamond along the reaction path. Although the DEW models have no spatial dimension, the cartoons illustrate processes observed in diamondiferous mantle xenoliths (Howarth *et al.*, 2025). Progressive diffusive equilibration may remove the original metasomatic veins, leaving inclusion-bearing and inclusion-free diamonds preserved within the mantle matrix.

do not match the major element geochemistry of the garnets or clinopyroxenes of their host rocks (Ireland *et al.*, 1994; Taylor *et al.*, 1996; Anand *et al.*, 2004; Misra *et al.*, 2004; Stepanov *et al.*, 2008; Liu *et al.*, 2009; Smart *et al.*, 2012; Aulbach and Stachel, 2022).

Evolution of diamond forming fluids. Irreversible reaction path models using the DEW model thermodynamic data predict that metasomatism should result in the precipitation of silicates which mirror those observed as diamond inclusions (Mikhail *et al.*, 2021; Rinaldi *et al.*, 2023; this study). Here we investigate whether our simulations reproduce natural diamond forming fluids. We compare modelled fluid compositions with the compositional range of high density fluids (HDFs) observed in fibrous diamonds (Fig. 3). In both Model I and Model II, progressive fluid-rock interaction drives fluid compositions away from silicic compositions toward increasingly carbonatitic compositions. The comparison also shows that different metasomatic scenarios generate different fluid evolution paths. In Model I, infiltration of an eclogitic fluid into peridotite produces fluid compositions that evolve along the silicic-carbonatitic transition. In Model II, mixing between carbonatitic and eclogitic fluids produces trajectories that intersect a similar region of HDF compositional space but follow a different path. The simulations of Rinaldi *et al.* (2023) represent reactions between fluids of different initial compositions (eclogitic, silicic, transitional silicic-carbonatitic, and carbonatitic) and mantle lithologies including carbonated peridotite (Models 151 and 152 of Rinaldi *et al.*, 2023) and eclogite or websterite (Models 154 and 158 of Rinaldi *et al.*, 2023), and the model of Huang and Sverjensky (2020) describes the evolution of saline fluids as they mix and react with highly Mg-rich carbonatitic compositions (Fig. 3; Table S-1). The DEW simulations reproduce the direction and extent of fluid evolution observed in natural systems, where the model fluid trajectories overlap the compositional arrays defined by natural HDF samples, including the silicic, silicic low-Mg carbonatitic, low-Mg carbonatitic, and saline fields (Fig. 3). These results suggest that multiple metasomatic pathways can generate fluids within the observed range of diamond forming compositions. Taken together, these trajectories follow the same general compositional gradients defined by natural HDF arrays (Weiss *et al.*, 2022), thus indicating that the diversity of diamond forming fluids can arise from progressive metasomatic reactions akin to those predicted by DEW simulations (Figs. 1–3).

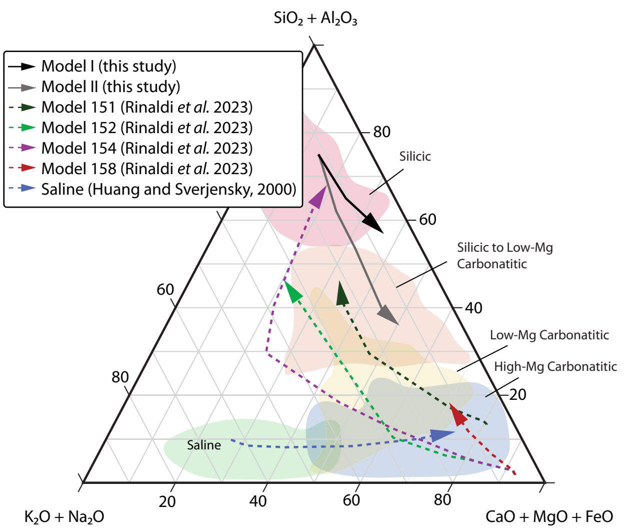

Figure 3 Ternary diagram comparing predicted model fluid compositions (curves and arrows) with high density fluid (HDF) compositions in fibrous diamonds (coloured fields from Weiss *et al.*, 2022). Solid curves and arrows show fluid evolution paths predicted by DEW models from this study (Models I and II). Dashed curves and arrows show paths from Rinaldi *et al.* (2023), and Huang and Sverjensky (2020). Models I and II trajectories reproduce the transition from silicic to carbonatitic compositions and overlap natural HDF arrays. The simulations of Rinaldi *et al.* (2023) represent reactions between fluids of different initial compositions (eclogitic, silicic, transitional silicic-carbonatitic, and carbonatitic) and mantle lithologies including carbonated peridotite (Models 151 and 152) and eclogite or websterite (Models 154 and 158) and overlap numerous natural fluid inclusion trends. The model of Huang and Sverjensky (2020) describes the evolution of saline fluids as they mix and react with highly Mg-rich carbonatitic compositions.

Conclusions

Thermodynamic models of fluid-rock metasomatism in the lithospheric mantle reproduce general metasomatic pathways consistent with observations of mixed paragenesis inclusions observed in diamonds from the Koidu kimberlite complex which evolve from Mg-poor to Mg-rich. The simulations demonstrate that disequilibrium inclusion assemblages can form during a single episode of diamond growth under constant pressure-temperature conditions as infiltrating fluids react with mantle rocks with and without pore fluids. Reaction pathways involving dunitic host rocks generate olivine during late stages of metasomatism, indicating that olivine-bearing mixed paragenesis diamonds formed within Mg-rich lithologies in the lithospheric mantle. Modelled fluid compositions evolve from silicic to carbonatitic and span the total range observed in fibrous diamond inclusions. These results show that both mixed paragenesis inclusions and the diversity of diamond forming fluids can arise from progressive metasomatic reactions, and that diamond inclusions do not necessarily represent samples of pristine mantle minerals.

Acknowledgements

This work was supported by grants from the UKRI NERC (OPP341 and NE/V011383/1) to SM, the UK Space Agency (ST/T001763/1) to SM, and builds on the results of previous published work funded by the US Department of Energy and the National Science Foundation from 2019 to 2025 to DAS. The codes and thermodynamic data used were first presented at a workshop in Milan in 2025 hosted by Simone Tumiati at the Universita degli Studi di Milano and can be found on the Deep Earth Water Community site at http://www.dewcommunity.org/. DAS greatly appreciates the help in developing the EQ3/6 code applications for deep Earth conditions from Simon Matthews and Mark Ghiorso. This work benefited from thoughtful reviews by Thomas Stachel, one anonymous reviewer, and editorial handling of Ambre Luguet.

Editor: Ambre Luguet

Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2625.

Cite this letter as: Mikhail, S., Rinaldi, M., Sverjensky, D.A. (2026) Mixed paragenesis diamond inclusions form during metasomatic fluid evolution. *Geochem. Persp. Let.* 41, 7–11. https://doi.org/10.7185/geochemlet.2625

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