1   **Fault-controlled hydration of the upper mantle during continental rifting**

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21

22   **Water and carbon are transferred from the ocean to the mantle in a process that**

23   **alters mantle peridotite to create serpentinite and supports diverse**

24   **ecosystems[1]. Serpentinised mantle rocks are found beneath the seafloor at slow- to**

25   **ultraslow-spreading mid-ocean ridges[1] and are thought to be present at about half**

26   **the world's rifted margins[2,3]. Serpentinite is also inferred to exist in the downgoing**

plate at subduction zones[4], where it may trigger arc magmatism or hydrate the deep Earth. Water is thought to reach the mantle via active faults[3,4]. Here we show that serpentinisation at the rifted continental margin offshore from western Spain was probably initiated when the whole crust cooled to become brittle and deformation was focused along large normal faults. We use seismic tomography to image the three-dimensional distribution of serpentinisation in the mantle and find that the local volume of serpentinite beneath thinned, brittle crust is related to the amount of displacement along each fault. This implies that seawater reaches the mantle only when the faults are active. We estimate the fluid flux along the faults and find it is comparable to that inferred for mid-ocean ridge hydrothermal systems. We conclude that brittle processes in the crust may ultimately control the global flux of seawater into the Earth.

The formation of serpentinite requires a supply of fluids to the mantle, but the bulk continental crust typically has low permeabilities (10^{-14} to 10^{-18} m^2)[5]. Active faults have been shown to transmit fluids during earthquakes and fault damage zones form potential fluid pathways[6], but low-permeability fault gouge can be a barrier to fluid flow[7]. In short, the complex distribution of low and high permeability features within fault zones leads to extreme permeability heterogeneity and anisotropy[7], so that the importance of faults as fluid pathways over geological timescales is unclear.

During rifting at magma-poor continental margins, crustal thinning leads to a reduction in overburden pressure at depth, which together with cooling moves the originally viscous lower crust into the brittle field[3]. Numerical models that track the thermal and rheological evolution of rifted margins have suggested that, for a wide range of strain rates and starting rheologies, the entire crust should become brittle at thicknesses of

between 14 and 4 km (Fig. 1a). Sufficient coincident deep seismic reflection and refraction profiles across magma-poor rifted margins now exist to enable a rigorous comparison between the observed maximum crustal thickness above or juxtaposed against serpentinised mantle, and the thickness at which complete crustal embrittlement is expected to occur[8] (Fig. 1 and Supplementary Fig. S1; Methods). Our compilation shows that serpentinite is found only where the crust is thin enough to have become entirely brittle during rifting and for many margins, such as Porcupine Basin[9] and Southeast Flemish Cap[10], conjugate to Galicia margin the predictions exactly match the observations (Fig. 1a-c and Supplementary Fig. S1). At some margins (e.g., Nova Scotia[11]; Fig. 1e and Supplementary Fig. S1) serpentintite only occurs beneath crust both thinner and some distance oceanward than predicted. However, in each case thick synrift sediments and/or salt, both known to reduce permeability[12], are present. It is clear that a brittle crust is not the only criterion for the serpentinisation; suitable fluid pathways must also exist. Perhaps most intriguingly, at other margins, the misfit is small, with serpentinite only occurring under crust that is thinner than predicted but only about a fault block spacing (5-10 km) further oceanward than predicted (Figs. 1a and d, Supplementary Fig. S1). This observation suggests that the serpentinisation process may be controlled by faults cutting down from the seafloor to the mantle. Thus at all serpentinite margins, where block bounding normal faults are well imaged, there is a strong link between the presence and the spacing of the brittle faulting in the thin crust and the serpentinisation of the underlying mantle.

The Deep Galicia segment of the magma-poor west Iberia rifted margin (Figs 1 and 2d) is one such margin. Here, rifting led to the formation of hyperextended continental crust with serpentinisation, detachment faulting and subsequent mantle exhumation[13]. Serpentinite has a lower density and coefficient of friction than fresh mantle peridotite

79	and causes weakening of the mantle[14]. The most recent normal faults soled out into the
80	weaker serpentinite layer, forming a detachment-like surface at the boundary between
81	the fault-bounded tilted crustal blocks and serpentinised mantle, a boundary described
82	from seismic reflection profiles as the "S reflector"[13].
83	
84	We compare the 3D compressional (P-) wave velocity structure of the Deep Galicia
85	Margin obtained from first-arrival travel-time tomography (Methods) with tectonic
86	structure from representative pre-stack depth migrated multi-channel seismic reflection
87	profiles, migrated using independent velocity models (Fig. 2). We obtained velocities
88	similar to those previously obtained from a two-dimensional transect through the same
89	area[15], but our denser sampling and 3D survey geometry (Supplementary Figs. S2 and
90	S3) yield much improved resolution. The smooth velocity model closely matches the 5-
91	10 km wide tilted basement blocks imaged on the seismic profiles. From the interpreted
92	top of the crystalline basement down to the S reflector, velocities vary between 5.5 and
93	6.5 km/s; the 6.5 km/s iso-velocity contour follows closely the S reflector where it
94	underlies the tilted basement blocks (root-mean-square difference 0.4 km), illustrating
95	the resolution of the model, confirmed using standard resolution tests (Supplementary
96	Information, Supplementary Figs. S4 - S7) for velocities both above and below S.
97	Velocities beneath the S reflector increase to 7.5 km/s over a mean depth interval of 1.2
98	km. If these velocity variations are attributed solely to variations in the degree of
99	serpentinisation, they would correspond to a decrease from ~45% serpentinisation (6%
100	by weight of water) immediately below the S reflector to ~15% at depths of ~1.2 km
101	below the S reflector. The inversion yields a smooth, isotropic model. Anisotropy is
102	likely to be present in the uppermost mantle but limited to a few per cent when
103	velocities are reduced by serpentinisation to 6.5-7.5 km/s[16].
104	

105   Intriguingly, at the intersections between the block-bounding normal faults and the S

106   reflector, this velocity increase occurs over a larger depth interval (~2 km), suggesting a

107   locally thicker zone of highly serpentinised mantle (Figs. 2 and 3a). This observation

108   implies that faults in the hanging-wall of the S reflector have had a role in the transport

109   of seawater to the mantle. Alternatively, velocities could be reduced by the presence

110   beneath S of lower crustal material or of frozen melt. However, lower crustal material is

111   unlikely because none is imaged on coincident seismic reflection profiles, and mafic

112   intrusions are also unlikely as no magmatism is known at this margin, and we would not

113   expect its location to be controlled by faulting processes above S.

114

115   The volume of seawater supplied to the mantle has been estimated from the volume of

116   the serpentinised region and the degree of serpentinisation (Methods). Each block-

117   bounding fault is associated with a maximum in water uptake, implying not only that the

118   water uptake was controlled by the deformation associated with those faults but also that

119   the serpentinised mantle has subsequently moved with the overlying fault blocks,

120   requiring the S detachment progressively to have become inactive, as expected for

121   brittle sequential faulting[17] such as in rolling hinge model[18]. Hydrothermal systems

122   commonly involve focused upflow along faults and diffuse downflow away from

123   faults[19]. Our data only tell us the water volume required for the observed degree of

124   serpentinisation, from which we infer the total net downflow during serpentinisation

125   (Methods). The inferred net downflow (minimum time-integrated flux) for individual

126   faults varies between 1.2×10^4 and 5.9×10^4 m^3 per square metre of fault cross-sectional

127   area. There are very few published estimates for comparison, but our estimated time

128   integrated fluxes are comparable to values of $\sim 10^5$ m^3/m^2 inferred for mid-ocean ridge

129   faults in the Oman Ophiolite[20].

130

131   Along the southernmost profile the fault that intersects the S reflector beneath the basin

132   oceanward of the last continental fault block (Supplementary Fig. S8c, F6, profile ISE1),

133   is not used for the estimation of time integrated fluxes. At that location the observed

134   high degree of serpentinisation may be due to the direct connection at breakup time

135   between the ocean and the exhumed foot-wall mantle. This environment may have been

136   similar to off-axis, low-temperature, ultra-mafic hosted foot-wall hydrothermal systems

137   observed at mid-ocean ridges[21].

138

139   The strong correlation between the water volumes and the fault displacements

140   (correlation coefficient: 0.91, probability of no correlation: 0.006; Fig. 3c and

141   Supplementary Figs. S9 - S10; Methods) suggests that a physical process links these

142   parameters, perhaps associated with increased connectivity of fluid pathways when

143   faults move[22]. If we assume that the faults above the S reflector moved sequentially to

144   accommodate a constant extension rate[17], the duration of their activity, and therefore the

145   corresponding flow rates, may be inferred from their displacement (Methods). Inferred

146   flow rates vary between 6.2×10^{-8} and 3.8×10^{-7} $m^3 s^{-1}$ per metre along the rift. These rates

147   are around one order of magnitude less than those associated with high-temperature

148   fluid fluxes at the TAG hydrothermal field, where a flux of ~100 kg/s[23] is inferred to

149   be sourced from 35-61 km of rift[24], corresponding to a flow rate of $2\text{-}3 \times 10^{-6}$ $m^3 s^{-1}$ per

150   metre.

151

152   Water circulation in the crust may be driven by episodic changes in tectonic stress and

153   in fault zone permeability[6]. A combination of coupled fluid pressure and mean stress,

154   suction pumping action and fault-valve action leads to the cyclic accumulation of water,

155   followed by fault reactivation and release of fluids[25]. Rather than using the fault

156   damage zone, the downflow may occur through antithetic faults created at the tips of

157 normal faults when they penetrate into regions of reduced yield stress[6], with any upflow

158 through the fault damage zone. At the Deep Galicia Margin such antithetic faults might

159 have started to form where the normal faults sole out onto the S reflector detachment

160 surface. As the opening of such antithetic faults is driven by the slip cycle on the main

161 block-bounding fault, fluid flow will also be proportional to the slip on that fault,

162 explaining the fit in Figure 3c. This latter mechanism may explain why serpentinisation

163 appears to be focused beneath the hanging-walls of the normal faults detaching onto S

164 (Fig. 2).

165

166 We do not infer 100% serpentinisation anywhere below the S reflector. The production

167 of new serpentinite depends on the temperature of the medium and the access of the

168 water to the unreacted peridotite, which is controlled by the water supply and the

169 porosity and the permeability of the medium[26,27]. Serpentinite has a lower permeability

170 (10^{-23}-10^{-22} m^2) than crustal rocks and faults and serpentinisation is a volume-increasing

171 process (up to 40% of volume expansion[3,27]) that reduces the initial rock porosity to

172 near zero. However, due to the volume expansion, cracking occurs and locally increases

173 the permeability. Within the serpentinite the water flow uses these cracks and is driven

174 by the pressure gradient created by the serpentinisation[27]. The incomplete

175 serpentinisation may suggest an insufficient water supply, in which case all of the

176 available water may have been consumed.

177

178 Alternatively, high temperatures and/or the limited access of the water to fresh mantle

179 peridotite may have resulted in a low serpentinisation rate. In that case, the water may

180 have been recycled by upflow along the faults. At the onset of serpentinisation during

181 continental breakup, the crust is less than 14 km thick (Fig. 1a), and the mantle is at

182 around 400-500 °C, i.e. the upper limit of the serpentinite stability[26]. The geothermal

183 gradient is high and remains high due to heat released during serpentinisation, providing

184 suitable conditions for focused upflow along the fault damage zones. If such

185 hydrothermal circulation is initiated, it will continue until the pore-filling reactions

186 between the hydrothermal fluids and the cold crustal rocks and ambient fluids lower the

187 damage zone permeability again[22].

188

189 Here we quantified the fluid fluxes in a rifted margin setting, but our approach could

190 also be at mid-ocean ridges and subduction zones given seismic data of sufficient

191 resolution. Our results show that when the entire crust becomes brittle during extension,

192 bulk serpentinisation in the upper 2 km of the mantle is enhanced close to crustal faults,

193 and thus that hydration of the upper mantle is fault-controlled. Therefore brittle

194 processes in the crust ultimately may control the global flux of seawater into the solid

195 Earth.

196

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280   **Author Contributions**

281   D.S.S., T.J.R., T.A.M., D.K., D.J.S., C.R., J.B. and J.K.M. designed the seismic

282   experiment. D.S.S. led the survey on *R/V Marcus Langseth* and D.K. and C.P. led the

283   deployment and recovery of seafloor instruments aboard *F/S Poseidon*. G.B. conducted

284   the seismic data analysis, with some assistance from R.G.D. T. J. R. compiled the

285   North Atlantic seismic profiles and M.P.G. carried out the numerical modelling. G.B.

286   and T.A.M. wrote the first draft of the paper and all authors contributed to subsequent

287   revisions.

288

289   **Additional Information**

290   Supplementary information is available in the online version of the paper. Reprints and

291   permissions   information   is   available   online   at   www.nature   .com/reprints.

292   Correspondence and requests for materials should be addressed to T.A.M

293

294   **Competing financial interests**

295   The authors have no competing financial interests.

296

Figure Legends

298   **Figure 1: a)** Comparison between the crustal thickness at which complete crustal

299   embrittlement is predicted to occur[3] (grey region covering three different modelled

300   rheologies) and the maximum crustal thickness observed above or juxtaposed against

301   serpentinised mantle at various North Atlantic magma-poor margins[8]. NS: Nova Scotia,

302   PB: Porcupine Basin, FC: Flemish Cap, IAP: Iberian Abyssal Plain, ARM: Armorican

303   Margin, NB: Newfoundland Basin, TAP: Tagus Abyssal Plain. The prefixes W, E, N, S

304   are West, East, North and South respectively. Cross sections of **b)** Porcupine Basin[9], **c)**

305   Southeast Flemish Cap[10], **d)** Galicia[15] and **e)** Nova Scotia[11] margins.

306

307   **Figure 2:** Compressional (P-) wave velocities superimposed on coincident seismic

308   reflection profiles illustrate the concentration of serpentinisation beneath the hanging-

309   wall of normal faults (expansion of 6.5 – 7.5 km/s iso-velocity interval). Yellow circles

310   are seabed instrument locations. Iso-velocity contours are marked by thin black lines.

311   Dashed lines mark the seabed (pale blue), interpreted base of post-rift sediments (green),

312   top of the pre-rift sediments (blue), top of the crystalline basement (red), S reflector

313   (black). Thick black lines indicate faults. **a)** ISE4 profile[13]; **b)** IAM 11 profile[17]; **c)** ISE1

314   profile[28]; **d)** location of the Galicia 3D survey with colour-coded bathymetry. **e)**

315   Schematic illustrating serpentinization associated with a single normal fault "F".

316

317   **Figure 3:** Water volume and amount of serpentinisation associated with faults on the

318   seismic reflection profile shown in Fig. 2b, assuming a two-dimensional structure. **a)**

319   Degree of serpentinisation (white contours) and water content (black contours). Black

320   and green boxes show the vertical and horizontal integration domains, respectively.

321   Bold black lines are the faults and dashed black line is the S reflector. **b)** Vertically

322   integrated water content (black) and horizontal extent associated with faults (green). **c)**

323   Correlation between water volume within hydrated mantle, representing integrated net

324   fluid flux through the fault, and fault displacement at the top of crystalline basement.

325   Data derived from seismic profiles (Fig. 2a – c) are shown by the magenta, green and

326   blue colors respectively. Faults F4 – F6 are shown by the triangle, square and circle

327   symbols respectively.

328

329

Methods

330

331 We calculated the stretching factor at which the entire crust becomes brittle using

332 hundreds of runs of a one-dimensional numerical model, in which a 125-km lithosphere

333 with a 32 km crust undergoes extension by a uniform pure strain rate[3]. The initial basal

334 and Moho temperatures were 1300° of 550° C, respectively[3]. We used a wet quartz

335 rheology for the upper crust, anorthosite, dry quartz and aggregate rheologies for the

336 lower crust, and a dry olivine rheology for the mantle[3]. For various Atlantic rifted

337 margins, we used published profiles showing the depth to the top of crystalline crust

338 from seismic reflection data and the depth to the Moho and distribution of mantle

339 serpentinisation from wide-angle seismic data (Supplementary Fig. S1). From these

340 profiles, we determined the maximum crustal thickness either beneath which detectable

341 serpentinisation occurs (e.g. Galicia), or where crust and serpentinite are juxtaposed (e.g.

342 S Iberia Abyssal Plain).

343

344 Seismic data on the Deep Galicia Margin were collected between June and September

345 2013. The primary aim of the survey was to acquire a three-dimensional seismic

346 reflection volume. Airgun shots were fired from *R/V Marcus Langseth* along fifty

347 parallel profiles with 400 m spacing between them (Supplementary Fig. S2), alternating

348 between two 3000 cu. in. airgun arrays to give a shot interval of 37.5 m (~16 s). A grid

349 of 72 ocean bottom instruments, comprising 44 four-component ocean bottom

350 seismometers (OBSs) and 28 ocean bottom hydrophones (OBHs) was deployed on the

351 seabed for three months to record these shots, with sample rates of 250 Hz and 200 Hz,

352 respectively. OBS/H data were corrected for internal clock drifts and instruments were

353 relocated using a 2D iterative inversion (Supplementary Information). In our analysis

354 we used first arrival travel-times from 48 OBS/H spread across the survey area

355 (Supplementary Fig. S2). The pick uncertainty was estimated visually as a function of

356   signal noise ratio and varied from 50 to 230 ms.

357

358   The P-wave velocity image was constructed using a non-linear iterative tomographic

359   technique (FAST[29]). A total of 155,924 first arrival times was used, corresponding

360   mostly to crustal refractions, since there are few first arrivals from shallower parts of the

361   structure (Supplementary Fig. S3). We used a forward grid of 0.5 km node interval and

362   an inverse grid of 1 km cell size. The starting models used in this study and the

363   resolution and uncertainty of the inversion results are discussed in the Supplementary

364   Information (Supplementary Figs. S4 – S7).

365

366   P-wave velocities were converted to percentage of serpentinization below the depth of

367   the S reflector and the water content by weight was estimated using an empirical

368   relationship[30] (Fig. 3 and Supplementary Fig. S8). The water content may be

369   overestimated because of model smoothing across a velocity discontinuity. The weight

370   of serpentinite was calculated using the estimated density of the serpentinite at each grid

371   node and the water weight percentages were used to calculate the water volume. The

372   water volume associated with a given block-bounding normal fault was estimated by

373   integrating the water volume over a vertical interval of 2.5 km beneath the S reflector

374   and a horizontal interval of 7 km (Fig. 3 and Supplementary Fig. S8). The bottom of the

375   depth interval was chosen to be close to the limit of ray coverage (Supplementary Fig.

376   S5). The horizontal integration interval approximates the width of the region of reduced

377   velocities around an isolated fault (Figs 2b and 3a, F5, profile IAM 11). The values

378   resulting from the vertical and horizontal integration (Fig. 3c) represent the water

379   volume per unit length along the margin that is associated with a given fault. Some of

380   this fluid will have been fed by the S detachment itself, perhaps represented by the

381   intercept on the vertical axis of the regression line shown in Fig. 3c. The minimum time-

382   integrated fluxes are estimated by dividing the water content associated with the faults

383   by the thickness of the faults. A thickness of 50 m was inferred for the S reflector by the

384   full waveform inversion along profile ISE1[31], and for this approximate calculation we

385   assume that the block-bounding faults have comparable thickness.

386

387   Fault displacements at the top of crystalline basement were estimated from foot-wall

388   and hanging-wall cut-off[32] measured for each fault block, on pre-stack depth migrated

389   seismic profiles (Fig. 2a-c and Supplementary Fig. S9). Due to the topography of the

390   crystalline basement, a minimum and a maximum displacement were calculated for

391   each fault and the mean displacement was used in the discussion. Water content

392   estimation errors were calculated using the standard deviations of the P-wave velocity at

393   the nodes (Fig. 3c and Supplementary Fig. S7). We estimated the duration of activity of

394   the faults by dividing the heaves by the half extension rate of 0.8 ± 0.36 cm/yr[33]. We then

395   estimated the flow rates by dividing the water volume related with one fault by the

396   duration of activity of that fault.

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398   **Code Availability:** The tomographic FAST[29] code used for this study is available at

399   http://terra.rice.edu/department/faculty/zelt/fast.html

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