

1 U-Pb geochronology of calcite-mineralized faults: Absolute
2 timing of rift-related fault events on the NE Atlantic margin

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8 **ABSTRACT**

9 Constraining the timing of brittle fault events is critical in understanding crustal
10 deformation and fluid flow, but a number of regional-scale fault systems lack readily
11 available techniques to provide absolute chronological information. Calcite
12 mineralization occurs in crustal faults in many geological settings, and can be suitable for
13 U-Pb geochronology. This application has remained under-utilized because traditional
14 bulk dissolution techniques require uncommonly high U concentration. As U and Pb are
15 distributed heterogeneously throughout calcite crystals, high spatial-resolution sampling
16 techniques can target domains with high U and variable U/Pb ratios. Here we present a
17 novel application of in situ laser ablation inductively coupled mass spectrometry (LA-
18 ICPMS) to basaltic fault rock geochronology in the Faroe Islands, NE Atlantic margin.
19 Faults that are kinematically linked to deformation associated with continental break-up
20 were targeted. Acquired ages for fault events range from Mid-Eocene to Mid-Miocene,
21 and are therefore consistently younger than the regional Early Eocene onset of ocean
22 spreading. These new absolute ages highlight a previously unrecognized protracted brittle

deformation within the newly developed continental margin. LA-ICPMS U-Pb calcite geochronology represents an important and novel method to constrain the absolute timing of fault and fluid-flow events.

Introduction

Constraining the timing of brittle faulting is critical in understanding crustal deformation and fluid flow in the upper crust, but for many settings there is a lack of readily available techniques to provide absolute chronological information. Calcite is a common fault-hosted mineral that has the potential to be dated by U-Pb geochronology. Calcite growth associated with slip (such as slickenfibres) or inter-slip periods can therefore be used to constrain the timing of slip events along the host fault. U-Pb geochronology of calcite has been applied to various geological systems including the depositional, diagenetic, and formation ages of sediments, fossils, and ore deposits (Rasbury and Cole, 2009). Bulk dissolution techniques have been used traditionally, targeting material with high U (>1 ppm) contents. Precise age determinations also require low initial Pb contents, and hydrothermal settings typically have unfavorable initial U/Pb ratios (Rasbury and Cole, 2009). Recently, U-series dating has been applied successfully to the dating of precipitates and striations on fault structures (Uysal et al., 2011; Nuriel et al., 2012), but this can be applied to only relatively young faults (i.e., < 0.6 My). Before now, calcite U-Pb geochronology has not been applied successfully to the absolute dating of faulting. Laser Ablation Inductively Coupled Mass Spectrometry (LA-ICPMS) U-Pb geochronology has recently been applied to dating diagenetic calcite in fossils (Li et al., 2014), and to hydrothermal veins in oceanic crust (Coogan et al., 2016). These studies highlight the utility of the LA-ICPMS method, whereby a large spread in U-Pb ratios can

be determined from a single sample, thereby potentially increasing the precision of a determined age compared to that of the averaging effect of dissolution-based methods. To our knowledge, the technique has not been presented for the successful characterization and dating of calcite-bearing fault rocks.

Here we use LA-ICPMS on calcite hosted within continental basaltic fault zones from the Faroe Islands, NE Atlantic margin, to present the first absolute ages for fault sets associated with rifting synchronous with continental break-up. Volcanic passive margins provide a crucial record of rift processes and continental break-up. The Faroe Islands lava sequence represents an onshore expression of the North Atlantic Igneous Province (NAIP). Fault and dike sets in the Faroe Islands show cross-cutting relationships, which have been fit to a relative chronology of deformation events associated with rifting, leading to continental break-up and formation of the NE Atlantic (see Walker et al., 2011a). Current age constraints for faults along the margin use offset marker horizons in the host stratigraphy, but there are a number of significant limitations with that approach. Faults in the Faroe Islands cut all of the Paleocene sequence (Moy and Imber 2009; Walker et al. 2011a) and thus these markers constrain only maximum ages of faulting. Additionally, NAIP ages for onshore samples, acquired through K-Ar and Ar-Ar techniques, range from ~60.5–54.5 Ma (Jolley and Bell, 2002), predating magnetochron ages for break-up (i.e., 55–53 Ma). The ages for volcanism therefore do not provide any lower age bracket for phases of continental break-up, but more importantly, volcanism is not unique to deformation stages during break-up. Direct dating of faults is therefore important in constraining the history of continental break-up. Faults

in the Faroe Islands host abundant zeolite and calcite mineralization, lending the area to U-Pb calcite geochronology.

Relative Chronology

Faults in the Faroe Islands, suitable for dating using U-Pb in calcite, were identified during detailed onshore mapping (Fig. 1; see Supplementary Files; Walker et al., 2011a). The following fault sets are recognized based on their orientation and kinematics (Walker et al., 2011a): (1) N-S and NW-SE striking normal faults that accommodated E-W to NE-SW extension; (2) ENE-WSW to ESE-WNW conjugate strike-slip and normal faults that accommodated N-S extension; and (3) NE-SW and NNE-SSW-striking oblique-slip faults that accommodated NW-SE extension. Some Set 1 faults can be relative-age-constrained to syn-emplacement of the Faroes lavas (57–54 Ma), by stratigraphic thickness variations across faults. Sets 2 and 3 cut the entire onshore sequence, with no clear evidence of thickness variations, hence are inferred to post-date the entire lava sequence (54 Ma and younger). Where local cross-cutting relationships are observed, Set 1 is cut by Set 2, which is in turn cut by Set 3, leading to the interpretation that faults in the Faroe Islands represent a progressive rotation in the extension direction prior to, during, and following break-up (Walker et al., 2011a).

Absolute Dating Method

Fault rocks were characterized to constrain deformation textures, and in particular to identify *crack seal type* veins (Fig. 2A,B). Crack seal texture represents vein-widening as a function of repeat fracture events (Petit et al., 1999); individual veins are inferred to seal rapidly, limiting the potential for long-lived open cavities. The calcite within crack-seal veins is inferred to represent instantaneous mineralization, recording the age of the

fracture within the resolution of the dating technique. For comparison, vein material was selected from faults that do not display crack seal texture, such as implosion breccia mineralization associated with the sudden creation of an open cavity (Sibson, 1986), and dilational jog zones. Walker et al. (2012) and Walker et al. (2013) showed that the faults used in the present study accumulated displacements through repeat fault episodes, involving fracture growth, linkage, and slip, with several stages of mineralization. It is therefore important to note that a successful age for a crack seal vein represents the age of the sampled vein, but does not represent the full age range of faulting associated with a given fault zone. Here we aim to use the range of ages for a given fault population, to constrain the duration of the associated stress state (Fig. 1B), and the potential persistence of open cavities along faults.

Calcite samples were collected from each of the three fault sets. Calcite chips were extracted from the fault rock samples and mounted in epoxy. After optical examination, elemental mapping using LA-ICPMS (Fig. 2) was conducted to identify primary growth and secondary alteration zones. Suitable domains containing high U and low Pb were targeted with spot analyses using LA-ICPMS to provide the best achievable precision and accuracy. See supplementary file for a full description of the method. Results are displayed as Tera-Wasserburg plots shown in Figure 3.

Absolute Dating Results

Elemental mapping shows that U and Pb contents of the samples are highly variable. Average U and Pb contents across the nine successful samples range from 12–161 ppb and 0.2–13 ppb, respectively; some chips are homogeneously low, whereas others feature zoning in uranium (Fig. 2; see also supplementary file). Uranium content is

distributed similarly to most trace metals (see Fig. 2C-F), indicating the preserved elemental pattern represents a primary (crystal growth) distribution.

Seventeen samples were analyzed for U-Pb, and age determinations were obtained from nine samples (Fig. 3), taken from eight different faults. Set 2 samples (n=7) provide a range of ages between 44.8 ± 2.0 Ma to 11.2 ± 1.1 Ma (Fig. 3). Set 3 samples (n=2) provide ages of 41.7 ± 1.9 Ma and 16.3 ± 1.2 Ma (Fig. 3). Analyses were unsuccessful on the Set 1 N-S to NW-SE normal faults, due to very low U contents. Unsuccessful analyses fall into two categories: (1) those that are dominated by high common lead; and (2) samples with analytical uncertainties that preclude a regression. Obtained ages that are deemed to be successful have variable precision owing to the combination of low U abundance and variable proportions of radiogenic to common Pb (Fig. 3). Of the successful results, seven show mean squared weighted deviates (MSWD) values outside of the expected range for a single population, with scatter in these cases consistent with variable common Pb isotope composition. The quoted uncertainties take account of this scatter, but absolute uncertainties should be viewed with caution. In all cases, the obtained ages, including uncertainties, are younger than the host basaltic lavas (57–54 Ma; Jolley & Bell, 2002).

Discussion

Set 2 samples taken from crack seal veins on the slipped portions of faults cluster within an age bracket of 44.8 ± 2.0 Ma to 40.1 ± 4.8 Ma, and samples from along a single fault (MOL-1–1 and MOL-1–2; Fig. 3) have overlapping ages within uncertainty. Calcite in these cases must precipitate between slip events (Petit et al., 1999), hence we interpret these dates as recording the age of slip within uncertainty. The Set 2 sample from an

implosion breccia (TJN-1-3: 40.9 ± 8.1 Ma) looks to fall within this age range, but for poor uncertainty, as anticipated for a near-instantaneous mineralization (see e.g., Sibson, 1986). Samples taken from dilational oversteps on Set 2 faults (TJN-6-1: 37.7 ± 1.9 Ma), and with potentially incomplete crack seal texture (LEY-2-1: 11.2 ± 1.1 Ma) provide ages that are younger than demonstrated crack seal veins. We infer that these young ages record the maintenance of open cavities within the mechanically strong basalt lavas (Walker et al., 2011b), rather than representing a record of slip events along the fault.

A crack seal vein sample from Set 3 (TJN-2-1: 16.3 ± 1.2 Ma) fits with the Walker et al. (2011a) stepwise rotation in extension direction through time, though it is noted the age is considerably younger than their predicted Eocene age. However, TJN-5-2 (Set 3: 41.7 ± 1.9 Ma) falls within error of the main grouping of Set 2 samples, which is not easily reconciled with this stepwise deformation history. Both of the Set 3 faults benefit from good relative age constraints, as the structures cut and offset faults (and dikes) associated with Set 2 (Fig. 4). It should be noted that deformation histories based on observed cross-cutting relationships are vulnerable to the impact of unobserved relationships, and it is possible that structures in the Faroe Islands represent a more gradual change in extension directions, potentially with overlap between kinematic fault sets. In any case, we are presenting a single age, and clearly further age-dating is required to constrain this and elucidate a full geodynamic history.

Faults in the Faroe Islands are geometrically and kinematically linked to stages of continental rifting and break-up to form the NE Atlantic (Walker et al., 2011a), which is generally constrained to Magnetochron 24R (~ 55 – 53 Ma). Initial spreading began on a segmented ridge system involving a NE-propagating Reykjanes segment, and SE-

propagating Aegir, and Mohns ridge segments (Lundin, 2002; see Fig. 1A, 4), but the age of continental break-up in the sense of a through-going oceanic crust, is difficult to define. The Aegir and Reykjanes ridges were separated by a continental relay zone between Kangerlussuaq (East Greenland) and the Faroe Islands (Gernigon et al., 2012; Ellis and Stoker, 2014). Extension on these ridges is thought to result in an anticlockwise stress-field rotation in the continental relay zone, which is consistent with the progressive rotation documented for structures in the Faroe Islands (Walker et al., 2011a). Detailed characterization of sea floor magnetic anomalies suggests break-up of the relay zone, and formation of a through-going oceanic crust, by the Early to Mid Eocene (~49.7–47.9 Ma; Gernigon et al., 2012). Alternatively, regional tectonostratigraphic correlation on the East Greenland and European margins (Ellis and Stoker, 2014) suggests the continental relay zone remained intact until the Early Oligocene (~33 Ma), and possibly as late as the Late Oligocene (~25 Ma). Our fault-slip related calcite ages are Mid Eocene (44.8–40.1 Ma), and we tentatively suggest that this represents stages of dismemberment of the continental relay zone consistent with the Ellis and Stoker (2014) model. The range in U-Pb calcite ages, including Miocene age crack seal veins (LEY-2-1) suggests faulting persisted on the continental margin, potentially for a period of time following formation of a through-going oceanic spreading centre. Further detailed fault rock dating across the region, and ideally to include the conjugate Greenland margin, has the potential to constrain this complex rift and break-up history.

LA-ICPMS U-Pb geochronology of calcite mineralization presents a novel approach in obtaining absolute ages of fault episodes, as well as the potential to constrain the timing of fluid flow in the subsurface. Given the abundance of fault-hosted calcite in

various settings, and provided that structural characterization can constrain the calcite mineralization to discrete slip events, this technique has wide application in determining the age of upper crustal faults and fractures.

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FIGURE CAPTIONS

Figure 1. **Simplified structural elements map for the Faroe Islands, NE Atlantic**

margin. A: The East Greenland and European conjugate margin, showing sea floor

magnetochrons, main rift basin ages, and major lineaments. Map was compiled using:

basin ages from Lundin and Doré (1997); oceanic magnetic anomalies from Gaina et al.

(2009); Iceland stratigraphic ages from Doré et al. (2008). **B**: Hillshaded topographic and bathymetric map of the northern Faroe Islands. Lower hemisphere stereographic projections for idealized fault orientations and paleostress axis calculations for Sets 1, 2, and 3 (summarized from Walker et al., 2011a; see text for explanation).

Figure 2. Calcite geochronology sample method and analysis. **A**: Example of a Set 2 crack seal vein. **(B)**: Calcite sample showing area for elemental maps (crosses for reference in C-F). **C-F**: Elemental maps for U, Pb, Mn, and V respectively, for the dated region.

Figure 3. Tera-Wasserburg Concordia plots showing $^{238}\text{U} / ^{206}\text{Pb}$ versus $^{207}\text{Pb} / ^{206}\text{Pb}$. Samples are ordered top-left to bottom-right from oldest to youngest, excluding uncertainty ranges.

Figure 4. A: Summary of U-Pb calcite ages in the Faroe Islands, including relative timings of oceanic spreading (after Doré et al., 2008; see text for details). Set 3 sample names are shown in italics. **B-C**: Maps showing the distribution of ages with respect to mapped structures in the Faroe Islands.

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