

1 Dolomite recrystallization revealed by $\Delta_{47}/U-Pb$
2 thermochronometry in the Upper Jurassic Arab Formation,
3 United Arab Emirates

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14

15 **ABSTRACT**

16 The process of recrystallization affecting dolomitic successions remains a longstanding
17 enigma in carbonate research. Recrystallization influences the accuracy of genetic dolomitization
18 models as well as the prediction of porosity and permeability distribution within dolomitic
19 reservoirs. Here, we investigate early-formed dolomites of the Upper Jurassic Arab Formation
20 reservoir (Arabian Platform), where recrystallization is not easily ascertained based on
21 petrographic and O-C-Sr isotope analyses. Conversely, the application of $\Delta_{47}/U-Pb$ thermo-

22 chronometry revealed the occurrence of burial recrystallization over a temperature/time interval
23 of about 45 °C/45 Ma during Early and Late Cretaceous. The process was initially driven by Late
24 Jurassic mixed marine/meteoric waters, which evolved during burial in a closed hydrologic
25 system and remained in thermal equilibrium with the host-rocks. Recrystallization was a
26 stepwise process affecting the succession heterogeneously, so that samples only few meters apart
27 presently record different temperature-time stages of the process and stopped when hydrocarbons
28 migrated into the reservoir. The results illustrate how Δ_{47} /U-Pb thermo-chronometry may
29 provide a novel approach to unravel dolomite recrystallization and to precisely determine timing
30 and physicochemical conditions (temperature, $\delta^{18}\text{O}$) of the process. This study therefore paves
31 the ground for a better appraisal of recrystallization in dolomitic reservoirs.

32

33 INTRODUCTION

34 Dolomites that originate early at near-surface conditions are metastable and tend to
35 transform and stabilize via recrystallization. This widely documented diagenetic process does not
36 change the overall rock lithology, though it may cause chemical, crystallographic and textural
37 modifications of early-formed dolomites (e.g. Montañez and Read, 1992; Kupecz and Land,
38 1994; Machel, 1997). Recrystallization can commence either immediately after dolomitization or
39 during later diagenesis due to interaction with fluids of marine, meteoric or basinal origin (Gregg
40 et al., 1992; Montañez and Read, 1992; Kaczmarek and Sibley, 2014; Ryan et al., 2022).
41 However, preservation of pristine features can be recorded in early dolomites despite the deep
42 burial conditions (Al-Aasm and Packard, 2000). The expression of the dolomite recrystallization
43 process thus remains enigmatic and needs further understanding since recrystallization impacts

44 the accuracy of reconstructed diagenetic models aiming to establish timing, conditions, and
45 mechanism of dolomitization processes.

46 Despite petrographic and mineralogic clues (e.g. crystal coarsening, increase in non-
47 planar crystal boundaries, stoichiometry and cation ordering, response under
48 cathodoluminescence (CL) or UV-light microscopy, nanotopography type; Kaczmarek and
49 Sibley, 2014; Manche and Kaczmarek, 2021), recrystallization of early dolomites is commonly
50 inferred using isotopic (O, C, Sr) analyses which have been recently complemented by clumped
51 isotope Δ_{47} thermometry (Veillard et al., 2019; Ryan et al. 2022). In parallel, LA-ICPMS U-Pb
52 geochronology has successfully dated early dolomites (Elisha et al., 2020) and opened the
53 possibility to disclose recrystallized dolomites (Montano et al., 2022). However, to our
54 knowledge, no study has combined Δ_{47} thermometry and U-Pb dating (i.e. Δ_{47} /U-Pb thermo-
55 chronometry *sensu* Mangenot et al., 2018) on early-formed dolomites to investigate
56 recrystallization.

57 The Upper Jurassic Arab Formation (Fm.) of Kimmeridgian to Tithonian age (154-150
58 Ma) consists of interbedded shallow marine carbonates and evaporites formed on a ramp system
59 under an arid climate and includes a dolomitic succession hosting among the largest hydrocarbon
60 (HC) reserves in the world. Dolomitization is interpreted to occur in the Late Jurassic by syn-
61 depositional to early diagenetic processes, attributable respectively to the evaporative-sabkha and
62 the seepage-reflux systems (e.g. Grötsch et al., 2003; Cantrell et al. 2004; Al Suwaidi et al.,
63 2005; Morad et al., 2012; Marchionda et al. 2018). Here, this succession was investigated using
64 Δ_{47} /U-Pb thermo-chronometry to: (1) demonstrate the occurrence of dolomite recrystallization
65 and place it in a thermochronological framework; (2) shed new light on the spatial/temporal

66 distribution and physicochemical conditions of recrystallization; (3) relate these findings to HC
67 exploration of other dolomitic reservoirs.

68

69 **MATERIALS AND METHODS**

70 In this study, we investigated Arab Fm. samples from an anticline HC field (Abu Dhabi,
71 UAE) within an intracratonic sedimentary basin of the Arabian Platform (Fig. 1 A-B). Core
72 samples comes from the Arab A-B-C lithostratigraphic members (Fig. 1 C): they formed in
73 supratidal to subtidal environments and consist of high-frequency dolomite-anhydrite
74 shallowing-upward cycles capped by massive anhydrites (Grötsch et al., 2003; Al Suwaidi et al.,
75 2005; Marchionda et al., 2018). The samples come from well A (2834-2850 m) and well C
76 (3303-3366 m), drilled 10 km apart along the anticline crest (Fig. 1 D), i.e. the HC zone.

77 Aiming to reveal dolomite recrystallization, samples were screened with optical, CL and
78 UV-light petrography (n = 79) and analyzed for O-C isotope compositions (n= 37) and $^{87}\text{Sr}/^{86}\text{Sr}$
79 ratios (n = 15). Based on petrographic and geochemical results (Data Repository DR1), a subset
80 of 15 samples was selected for Δ_{47} analyses, including 11 samples that were jointly dated by U-
81 Pb geochronology.

82 Dolomite Δ_{47} compositions were determined following the protocols summarized in Data
83 Repository DR2. Δ_{47} compositions were converted into temperatures ($T\Delta_{47}$) using the calibration
84 from Bonifacie et al. (2017). The oxygen isotope composition of water ($\delta^{18}\text{O}_w$) was calculated
85 from $T\Delta_{47}$ and $\delta^{18}\text{O}_{\text{dol}}$ simultaneously acquired, by using the oxygen isotope fractionation
86 between dolomite and water from Horita (2014). $\delta^{18}\text{O}_{\text{dol}}$ and $\delta^{18}\text{O}_w$ values are expressed in per
87 mil (‰) relative to the VPDB and the VSMOW standards, respectively. Dolomite U-Pb dating

88 was performed by LA-ICPMS following the method of Ring and Gerdes (2016) and analytical
89 protocols are summarized in Data Repository DR3.

90

91 **RESULTS**

92 The investigated samples include two non-ferroan and fabric-preserving dolomite types:

93 (1) Dol1 occurs in supratidal to intertidal facies and consists mostly of dolomicrite with fine
94 crystals (< 10 to 30 μm), closely associated to syn-depositional anhydrite beds/nodules; (2) Dol2
95 occurs in subtidal lagoon facies, and is mainly composed of fine to medium (<10 to 50 μm) and,
96 less commonly, coarse (>50 μm) dolomite crystals, display dolomicrite, planar-s to planar-e
97 textures and is associated to early anhydrite cements. No later dolomite cements were observed.
98 No significant variation in CL response was observed among samples or within single dolomite
99 crystals, whereas UV-light revealed zoned crystals in only a few samples (see Data Repository
100 DR1, Table DR1, Fig. DR1, Table DR4). The $\delta^{18}\text{O}_{\text{dol}}$ and $\delta^{13}\text{C}_{\text{dol}}$ values (whole dataset) vary
101 from -5.7 to -1.3 ‰ and from -1.3 to +2.9 ‰, respectively. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios range from
102 0.7070 to 0.7072. The Δ_{47} values indicate temperatures ranging from 42 to 87 °C, corresponding
103 to $\delta^{18}\text{O}_{\text{w}}$ between -1.4 and +4.9 ‰ (Fig. 2A, B, Table DR4).

104 The $^{238}\text{U}/^{206}\text{Pb}$ and $^{207}\text{Pb}/^{206}\text{Pb}$ ratios measured on most dolomite samples display
105 concordant Tera-Wasserburg linear regressions (isochrons) with mean squared weighted deviates
106 (MSWD) between 0.59 and 1.90 (Fig. 2C, Fig. DR4_A, Table DR4). Overall, the calculated
107 Concordia lower-intercept U-Pb ages range between 136.5 ± 1.7 and 91.6 ± 7.6 Ma. Single
108 dolomite U-Pb ages are reported in Fig. 2C, Fig. DR4_A and Table DR4.

109

110 **DOLOMITE RECRYSTALLIZATION REVEALED BY Δ_{47} /U-Pb THERMO-**
111 **CHRONOMETRY**

112 The studied succession underwent progressive burial from deposition in the Late Jurassic
113 until the onset of a compressional event in the Late Cretaceous which induced faulting and
114 folding together with minor uplift (Johnson et al., 2005). This event was followed by further
115 subsidence during the Paleogene until maximum burial of ~3 km was reached in the Miocene
116 (Morad et al., 2019). Corresponding thermal histories were modelled based on different
117 geothermal gradients between 35 °C/km and 45 °C/km, with the present-day gradient being 38
118 °C/km (Fig. 3A; see Data Repository DR4). To appraise whether thermal resetting may have
119 modified Δ_{47} compositions, the computed thermal histories were integrated into the Δ_{47}
120 reordering models from Lloyd et al. (2018) and Hemingway and Henkes (2021). The models do
121 not predict any significant Δ_{47} reordering over geological time. Only the first model predicts that
122 dolomite Δ_{47} starts to reorder when assuming a geothermal gradient of 45 °C/km, which is
123 unlikely for the intracratonic basin investigated. Consequently, the measured $T\Delta_{47}$ are expected
124 to reflect the temperatures of initial dolomitization or recrystallization (see Data Repository
125 DR4).

126 The temperatures/ages determined for the studied dolomites are consistently
127 higher/younger than those expected for Upper Jurassic dolomites formed in near surface
128 environments. These temperatures/ages do not correlate with sample depth, and samples located
129 only a few meters apart may record difference of both several tens of °C and several tens of Ma
130 (Fig. 2, Table DR4). These Δ_{47} /U-Pb data demonstrate that the original dolomites underwent
131 recrystallization and allow constraining timing and physicochemical conditions (temperature,
132 $\delta^{18}\text{O}_w$) of the process.

133 Conventional petrographic approaches to evaluate recrystallization were not
134 straightforward because: (1) no direct relationship was found between dolomite crystal size and
135 temperatures/ages; (2) non-planar textures were not recorded; (3) UV-light response only
136 showed evidence of dissolution/precipitation from a few samples (see DR1). This
137 demonstrates that the Δ_{47} /U-Pb thermo-chronometer may be used to complement conventional
138 petrographic indicators for dolomite recrystallization and to provide further insights to interpret
139 this process.

140 The observed positive covariance between $\delta^{18}\text{O}_w$ and $T\Delta_{47}$ (Fig. 2A) indicates that the
141 diagenetic fluid became enriched in ^{18}O with progressive burial. By extrapolating the $\delta^{18}\text{O}_w$ - $T\Delta_{47}$
142 regression line towards near-surface temperatures (20-35 °C), at which the original dolomites
143 likely precipitated, it can be inferred that the initial fluids had $\delta^{18}\text{O}_w$ between -2 and -4 ‰. These
144 compositions, depleted in ^{18}O compared to the mean seawater, suggest that the original marine
145 evaporative fluids responsible for dolomitization were flushed by meteoric fluids, as observed in
146 other ancient and present sabkha/reflux systems (Spötl and Burns 1991; Sanford and Wood,
147 2001). These mixed marine/meteoric fluids became progressively enriched in ^{18}O during burial
148 (Fig. 2), due to continuous interaction with the dolomitic host-rocks in a closed hydrologic
149 system with low water renewal (Banner and Hanson, 1990). This scenario agrees with the
150 $^{87}\text{Sr}/^{86}\text{Sr}$ compositions of the recrystallized dolomites (Fig. 2B) that reflect those of Upper
151 Jurassic seawater carbonates (Veizer et al., 1999; Morad et al., 2018, 2019; see Data Repository
152 DR1).

153 The Δ_{47} temperatures (42 to 87 °C) and U-Pb ages (136.5 to 91.6 Ma) jointly measured
154 on dolomite samples define eleven temperature-time pairs that overlap, within uncertainties, with
155 the thermal curves of the studied succession that were reconstructed from burial curves and

156 employing paleogeothermal gradients between 35 and 45 °C/km (Fig. 3A; see Data Repository
157 DR4). This finding suggests that the fluids driving the recrystallization process were in thermal
158 equilibrium with the host-rocks. The temperature-time pairs bracket an interval of about 45 °C
159 and 45 Ma, indicating that recrystallization was a stepwise process from Early to Late
160 Cretaceous times, and was not due to one unique fluid-rock interaction event. The results also
161 reveal that recrystallization affected the dolomitic succession heterogeneously such that different
162 portions of the reservoir record different temperature-time stages of the recrystallization process
163 (Fig. 3B). This recrystallization heterogeneity is recorded on the scale of the hand specimens
164 investigated whereas the maximum scale remains uncertain.

165

166 **CONCLUSIONS AND IMPLICATIONS FOR DOLOMITE RESERVOIR STUDIES**

167 The joint application of Δ_{47} thermometry and U-Pb dating to early-formed dolomites
168 revealed that recrystallization affected the succession heterogeneously and determined timing
169 and some of the physicochemical conditions (temperature, $\delta^{18}\text{O}_w$) of the fluids driving the
170 process. Temperature-age datapoints covary and plot on independent thermal history curves
171 suggesting fluids in equilibrium with host-rocks in a closed system. These conclusions would not
172 have been achieved without the application of Δ_{47} /U-Pb thermo-chronometry. Several
173 implications can be derived for the study of dolomitic successions and their potential economic
174 significance:

175 (1) Dolomite research is widely devoted to the establishment of conceptual genetic models trying
176 to reconstruct timings, diagenetic environments, and fluid delivery mechanisms for
177 dolomitization. However, the common petrographic and geochemical tools used to achieve this
178 aim require that dolomites have not been altered since their initial precipitation (Machel, 1997).

179 Being able to reveal the occurrence of recrystallization via absolute temperature-time data
180 represents therefore a significant advance in this field.

181 (2) Recrystallization of early dolomites upon burial is known to affect the textural evolution of
182 dolomites and consequently their porosity and permeability, therefore having an impact on the
183 storage capacities of these worldwide reservoirs (Saller and Handerson, 1998; Nader et al.,
184 2013). The $\delta^{18}\text{O}_w$ -temperature data from this study agree with those from the Arab Fm. dolomites
185 of the Ghawar field (Swart et al., 2016; Fig. 1 and 2) suggesting that recrystallization occurred in
186 a similar manner (i.e. by mixed marine-meteoric waters in a closed hydraulic system) in
187 reservoirs located hundreds of km apart. It is therefore conceivable to extrapolate the
188 recrystallization scenario proposed here across other reservoirs of the Arabian Platform. Future
189 studies should refine dolomitic reservoir models by also considering the effects of
190 recrystallization on the rock porosity and permeability and its heterogeneous distribution.

191 (3) One of the concerns in sedimentary basin exploration is to constrain the timing of HC
192 migration. In the study area the onset of the first HC migration started in the Late Cretaceous
193 across several reservoirs (Al Suwaidi et al., 2005; Hollis et al. 2017; Morad et al., 2018) and
194 likely occurred along re-activated strike-slip faults during a compressional tectonic phase
195 (Johnson et al., 2005). Our $\Delta_{47}/\text{U-Pb}$ data indicate that the recrystallization of dolomites pre-
196 dated this event and was possibly halted by the Late Cretaceous HC migration (Fig. 3). These
197 data support previous hypotheses stating that HC emplacement may inhibit diagenetic processes
198 in carbonate reservoirs (Nielsen et al., 1998; Cox et al., 2010). The assessment of absolute
199 temperatures-ages for the latest stage of dolomite recrystallization could thus potentially be used,
200 in poorly characterized reservoirs, to indirectly constrain HC migration timing.

201

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208

209 **FIGURE CAPTIONS**

210 **Figure 1. A.** Arabian Peninsula map with location of the UAE (red area) and of the Ghawar field
211 in Saudi Arabia (grey area) investigated by Swart et al. (2016). **B.** Main hydrocarbon fields (in
212 green) located onshore and offshore Abu Dhabi. The frame includes the studied anticline field.
213 **C.** Field stratigraphy with the Arab A-B-C members located between the Arab D member and
214 the Hith evaporites (modified after Morad et al., 2018). **D.** Location of the sampled wells (A and
215 C) in the anticline field. Isolines indicate relative depth: orange and blue colors represent
216 respectively the highest (crest) and lowest (flanks) elevation of the anticline.

217
218 **Figure 2.** Thermo-chronometry ($\Delta_{47}/\text{U-Pb}$) and $^{87}\text{Sr}/^{86}\text{Sr}$ data of the Arab Fm. dolomites. **A:**
219 $T\Delta_{47}$ versus $\delta^{18}\text{O}_w$ cross-plot. The square colors refer to the U-Pb ages. Grey squares refer to
220 samples (n=4) not analyzed (NA) with U-Pb geochronology. Data from Arab Fm. dolomites of
221 Ghawar field (Saudi Arabia; Swart et al., 2016) are reported with grey dots. **B:** $T\Delta_{47}$ versus
222 $^{87}\text{Sr}/^{86}\text{Sr}$ cross-plot. The light blue area shows the $^{87}\text{Sr}/^{86}\text{Sr}$ of carbonates precipitated from Late
223 Jurassic seawater (Veizer et. al., 1999). **C and D:** $^{238}\text{U}/^{206}\text{Pb}$ versus $^{207}\text{Pb}/^{206}\text{Pb}$ Tera-Wasserburg
224 Concordia diagrams and corresponding lower intercept ages for 2 dolomite samples. Data point
225 error ellipses indicate 2σ internal uncertainty of the isotope ratios on “n” analyses. Red lines are
226 the envelopes of the isochrons.

227
228 **Figure 3.** Conceptual scenario of dolomite recrystallization in the Arab Fm. based on $\Delta_{47}/\text{U-Pb}$
229 thermo-chronometry. **A.** Thermal histories of Upper Jurassic rocks from the studied field
230 reconstructed by considering different geothermal gradients. The colored squares represent
231 temperature-time pairs measured on dolomites via $\Delta_{47}/\text{U-Pb}$ thermo-chronometry. Three steps of

232 the reservoir evolution are illustrated: (1) dolomite formation in Late Jurassic; (2) dolomite
233 recrystallization during Early and Late Cretaceous; (3) begin of HC emplacement in Late
234 Cretaceous. **B.** Schematic well cores illustrate the stepwise recrystallization process that affected
235 heterogeneously the original dolomites and pre-dated hydrocarbon (HC) charging.

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238 ¹GSA Data Repository includes: dolomite petrography and O-C-Sr isotope geochemistry (Table
239 DR1, Figure DR1_A, Figure DR1_B, Figure DR1_C), Δ_{47} and U-Pb methods, further Δ_{47} /U-Pb
240 data and discussion (Table DR4, Figure DR4_A, Figure DR4_B), U-Pb raw data (Table DR5). It
241 is available online or on request from editing@geosociety.org

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