

Article

Cavity-Filling Calcite in the Deccan Traps as a Natural Analogue for Mineral Carbon Sequestration and Low-Temperature Deformation

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ABSTRACT

Cavity-filling calcite is among the most widespread secondary phases in the Deccan Traps, and its position late in the amygdale paragenesis, after zeolites and silica phases, is long established. Two questions that remain open are addressed here: whether this calcite retains a mechanical record of deformation imposed after the cavities were sealed, and what a naturally carbonated flood-basalt pile can tell us about the durability of engineered mineral carbon storage. Calcite from three closely spaced amygdaloidal basalt exposures in the district of Maharashtra, Ane Ghat in Junnar taluka, Pune, Shindewadi 1 and Shindewadi 2 in Parner taluka, Ahilyanagar was examined by transmitted-light petrography, scanning electron microscopy with energy-dispersive X-ray spectrometry (SEM-EDS) and micro-Raman spectroscopy, with growth features deliberately separated from post-growth strain features. All three occurrences comprise coarse, optically continuous calcite grown into open voids: rhombohedral cleavage is perfect, birefringence is extreme, planar crystal faces survive in secondary-electron images, and no replacement front is developed against the basalt wall. Superimposed on this growth fabric are closely spaced polysynthetic e-twin lamellae, undulose extinction, cleavage-controlled intracrystalline microfractures, and healed fractures carrying trails of secondary inclusions. Because these features truncate cleavage and crystal boundaries that are continuous elsewhere in the same individual, they post-date crystal growth. Twin morphology is dominated by thin, straight lamellae, which on the Ferrill classification indicates deformation below roughly 170–200 °C. EDS spectra confirm a Ca-carbonate composition; because light-element quantification in a coated carbonate is not reliable, measured Ca/C atomic ratios of 1.31–1.61 depart substantially from the ideal value of unity and the analyses are treated as semi-quantitative throughout. Micro-Raman spectra are those of well-ordered calcite, with lattice modes near 156 and 279–281 cm⁻¹ and internal modes at 711–712 cm⁻¹ (ν_4) and 1085–1087 cm⁻¹ (ν_1); differences between sites do not exceed instrument calibration uncertainty. The results are placed in the framework of *in situ* mineral carbon sequestration. Deccan amygdale calcite is a completed natural experiment in basalt carbonation: divalent cations liberated by fluid–rock interaction was fixed as a thermodynamically stable carbonate that has survived approximately 66 million years in the same pore volume now targeted for engineered CO₂ injection. The microstructural record modifies that otherwise reassuring picture in a specific way. Mineralized amygdals were subsequently strained, brittle microfractures propagated through the carbonate cement, and those fractures were later healed by carbonate-bearing fluids. Permanence of mineral trapping in a tectonically quiescent plateau interior is therefore neither unconditional nor catastrophically compromised: it is a balance between fracture generation and carbonate self-sealing that can be read directly from the cement.

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Research Highlights

- Mechanical deformation of calcite and implications for engineered carbon storage, post growth deformation.
- Vibrational spectroscopy of calcite amygdules and relevance to carbon storage implication as a natural analogue.
- The twin morphology of calcite is consistent with low temperature deformation, broadly below $\sim 170\text{--}200\text{ }^{\circ}\text{C}$ according to the Ferrill framework.

1. Introduction

The Deccan Traps blanket close to 500,000 km² of western and central India in their present eroded state, and an original extent of order 1.5 million km² is generally inferred. Eruption was rapid and straddled the Cretaceous–Palaeogene boundary; high-precision geochronology confines the bulk of the erupted volume to a few hundred thousand years around 66 Ma [1–4]. The origin and distribution of secondary minerals in the Deccan Traps have been debated for several decades. Early studies recognized recurring associations of zeolites, silica, calcite and other alteration minerals within vesicles, cavities and fractures of the basaltic flows, with subsequent studies providing broader perspectives on the mineralogical and geochemical characteristics of volcanic materials in the Indian context [5]. Walker [6] proposed a broad vertical zonation of zeolite minerals, with laumontite, scolecite and heulandite defining successive zones with increasing elevation, and interpreted this distribution primarily in terms of burial depth and the associated geothermal gradient. Sukheshwala et al. [7] subsequently recognized these three zones in the Western Deccan and suggested that their distribution reflected increasing depth together with the action of circulating fluids. They also noted that structural disturbances and intrusive activity could enhance fluid circulation. Subsequent investigations, however, demonstrated that the distribution of secondary minerals is more complex than a simple regional burial depth model. Jeffery [8] based on the distribution and mineral chemistry of zeolites over a large part of the Western Deccan, found no consistent support for the proposed regional zonation. Common zeolite species, including heulandite, stilbite, mordenite and apophyllite, occur at different stratigraphic levels and in different formations, indicating that their distribution cannot be explained solely by present by altitude or burial depth. The study instead emphasized the importance of temperature and post emplacement fluid circulation in controlling secondary mineral formation. More recent studies have further developed this interpretation by demonstrating the secondary mineralization in the Deccan Volcanic Province was multistage and involved changing physiochemical conditions during post volcanic fluid circulation. Ottens et al. [9] showed that commonly occurring secondary minerals are not restricted to a simple vertical sequence and documented multiple stages of cavity

mineralization. Their observations indicate that temperature, fluid composition and the availability of suitable cavities and fluid pathways played important roles in determining mineral assemblages. Accordingly, sequence of clay minerals, zeolites silica and calcite observed in the Deccan basalt cavities should not be regarded as a universally fixed paragenetic sequence applicable to the entire province. Rather, secondary mineralization considered to reflect the interaction between evolving post emplacement fluids and the basaltic host rocks under changing temperature, fluid chemistry and permeability conditions. Pre-existing vesicles, cavities, fractures and flow related discontinuities could have provided localized pathways and sites for fluid rock interaction and mineral precipitation. The relative abundance and sequence of individual secondary minerals may therefore vary between localities and stratigraphic units. In the present study, the observed mineral assemblages are consequently interpreted in terms their local petrographic relationships and mineralogical characteristics rather than being assigned solely to a regional depth-controlled zeolite zone.

That paragenesis is not in dispute, and this paper is not offered as a rediscovery of it. Integrating cathodoluminescence imaging, fluid-inclusion work, carbon and oxygen isotope analysis and radiometric dating across the province, Ottens and co-workers resolved at least three separate mineralization stages and more than one calcite generation, with the youngest carbonate and apophyllite returning ages tens of millions of years younger than the volcanism [9, 10]. Their synthesis changes the question that a newly described Deccan calcite has to answer. It is no longer whether the phase is late, almost invariably it is, but which generation it represents, and what happened to it after it crystallized.

Two lines of inquiry follow from the second half of that question, and both are pursued here.

The first is structural. Calcite deforms by mechanical twinning one {0118} at differential stresses of only a few megapascals and at temperatures far below the threshold of any other common crystal-plastic mechanism, which is why calcite twins have become a routine instrument for reconstructing low-temperature stress and strain in fold–thrust belts and forelands [11–15]. Amygdale calcite in a flood-basalt pile is an unconventional but promising host for that instrument. It is coarse, chemically simple and monomineralic; it grew into a void at a known

point in a well-constrained paragenetic sequence; and it is distributed through hundreds of metres of stratigraphy across the plateau. Any twin it carries records strain that necessarily post-dates cavity sealing. Whether the plateau interior has in fact experienced significant post-eruptive deformation is contested: geomorphometric analysis of drainage basins across the Western Ghats found no statistically meaningful signal of active tectonics and assigned the western Deccan to a low-activity class [16], while structural, geophysical and Quaternary-sediment studies have argued for reactivation of sub-trappean basement discontinuities and differential block uplift. A microstructural strain marker independent of landscape morphology is therefore worth developing.

The second line of inquiry is applied, and it is the one that gives the microstructural result its wider significance. The increasing environmental pressures associated with climate change and resource depletion have strengthened the need for sustainable approaches to carbon management and long-term environmental protection, and the transition toward cleaner energy systems [17, 18]. Basaltic rock is now among the most actively investigated hosts for permanent geological disposal of carbon dioxide, because divalent cations released by dissolution of plagioclase, clinopyroxene and glass react with dissolved CO_2 to precipitate carbonate minerals, converting a mobile fluid into an immobile solid [17, 19, 20]. Field injection has demonstrated that the reaction is fast: the majority of CO_2 injected into basaltic lavas at the Hellisheidi site in Iceland was mineralized within two years, far more rapidly than pre-injection models predicted [21, 22]. Flood basalts specifically have been identified as high-capacity candidate reservoirs [23], and the Deccan Volcanic Province has been repeatedly nominated as a strategic component of India's storage portfolio [24–27]. Modelling studies of CO_2 storage in the Eastern Deccan Basalt further demonstrates the potential for rapid carbonate mineralization within the basaltic reservoir [28]. Recent reviews of basalt– CO_2 –water interactions likewise emphasize the importance of basalt dissolution, cation release and carbonate precipitation for long term carbon storage, including the Deccan Volcanic Province [29, 30]. Laboratory carbonation experiments on Deccan lithologies confirm the reactivity directly, with secondary carbonate growing on grain surfaces and in intergranular space under supercritical conditions [31, 32]. Experimentation investigation of altered Deccan basalt has further demonstrated the basalt dissolution can provide the reactive cations required for subsequent carbonate mineralization during CO_2 injection [33].

The connection between these experimental and modelling studies of basalt– CO_2 mineralization and the natural carbonate record preserved within the Deccan Volcanic Province forms the basis of the present study. Every filled amygdale in the Deccan is a completed natural experiment in exactly the reaction that engineered carbonation seeks to induce, run at ambient conditions and preserved for approximately 66 million years. Amygdale calcite therefore constitutes a direct empirical constraint on

the permanence of mineral trapping over geological time and, because the same crystals also record whatever deformation the pile subsequently underwent, on whether that permanence was mechanically disturbed. This is a question about reservoir integrity that no short-duration injection pilot can address, and it is answerable with ordinary petrographic tools.

Three exposures within a single district and a comparable stratigraphic interval were selected so that lithological and altitudinal variables are minimized. The objectives are stated narrowly. They are: to establish texturally that the calcite grew in open space and was subsequently strained; to characterize the twin population and extract a first-order deformation temperature; to determine whether SEM–EDS and Raman data resolve inter-locality differences at all, and to state plainly where they do not; and to develop the implications of a naturally carbonated, subsequently deformed basalt pile for engineered carbon storage in the same province.

2. Geological Setting and Sampling

2.1. Study Area

The study area lies in the Pune and Ahilyanagar districts of Maharashtra within the Deccan basalt outcrop. The present location which belongs to Upper Ratangarh Formation (Figure 1) constitutes an important basaltic unit of the Sahyadri Group of the Deccan Volcanic Province and is predominantly characterized by compound lava flows and is restricted to the north-western parts of Ghod valley and in the central part in the Bhima valley [34]. In the Pune-Ahilyanagar region, the formation is mainly represented by thick sequences of compound-pahoehoe flows composed of dark grey, fine to medium grained basalt that is generally massive, hard and compact [35]. The basalts range from non-porphyrific to sparsely or moderately porphyritic varieties, with plagioclase and locally olivine phenocrysts occurring within a fine-grained basaltic groundmass [36]. Individual pahoehoe flows commonly comprise a relatively massive central portion bounded by vesicular zones, with pipe amygdaloids, spherical vesicles and local cavity and crack filling features. The compound nature of the formation is expressed by the occurrence of multiple overlapping lava lobes and flow units. The lava pile locally contains megacrystic plagioclase bearing horizons that serve as regional stratigraphic marker units and are important for correlation within the Deccan lava succession [37]. In the broader Pune-Ahilyanagar sector, the Upper Ratangarh Formation is associated with a thick pahoehoe dominated lava sequence, and its stratigraphic relationships have been refined using Giant Plagioclase Basalt (GPB) horizons [35, 38]. An important component of the Deccan basalt succession is the occurrence of interflow red bole horizons, which commonly occur between successive lava flows. These reddish to reddish-brown, fine-grained horizons are generally associated with weathering of volcanic material and development of paleosol-like or altered interflow horizons during intervals of volcanic quiescence.

Red bole beds have been documented extensively within the Deccan Volcanic Province, including the Pune region, and may occur between individual basaltic flow units [39]. The presence of such interflow horizons introduces additional heterogeneity into the fluid flow architecture of the Deccan basalt pile. Depending on their thickness, continuity, degree of induration and alteration, red bole horizons may act as relatively low permeability horizontal barriers that impede vertical fluid movement, while in other circumstances, their altered or fractured character may provide preferential lateral pathways for groundwater or hydrothermal fluids. Consequently, fluid circulation within the Deccan basalt sequence should not be considered as a simple system in which massive It represents an interflow stratigraphic horizon that may have influenced the direction and degree of fluid circulation through the host basaltic pile. The formation of the secondary Calcite is consequently interpreted in the context of a heterogeneous basaltic reservoir in which fluid pathways were controlled by the spatial relationships among vesicles, fractures, flow contacts and interflow horizons.

The vesicular and amygdaloidal portions of these basaltic flows are particularly significant for secondary

mineralization because they provide cavities and fracture networks that can be subsequently occupied by hydrothermal or low temperature secondary minerals. Such secondary mineral assemblages in the Deccan basalts commonly include zeolites, calcite, silica minerals and other cavity filling phases [9]. Therefore, the vesicular, fractured and amygdaloidal character of the Upper Ratangarh basalt provides an appropriate geological host for the occurrence of secondary calcite investigated in the present study.

2.2. Field Sampling

Hand specimens were collected from fresh quarry and road-cut faces (Figure 2a,c,e), weathered surfaces and soil-covered zones being avoided. Calcite was sampled together with a rim of adhering host basalt wherever possible so that the wall-rock contact could be examined in thin section. Because the present dataset comprises a limited number of specimens, all comparative statements between localities are treated with corresponding caution throughout, and the compositional and spectroscopic results are interpreted principally as a pooled description of a single texturally defined calcite generation rather than as evidence of inter-site variation.

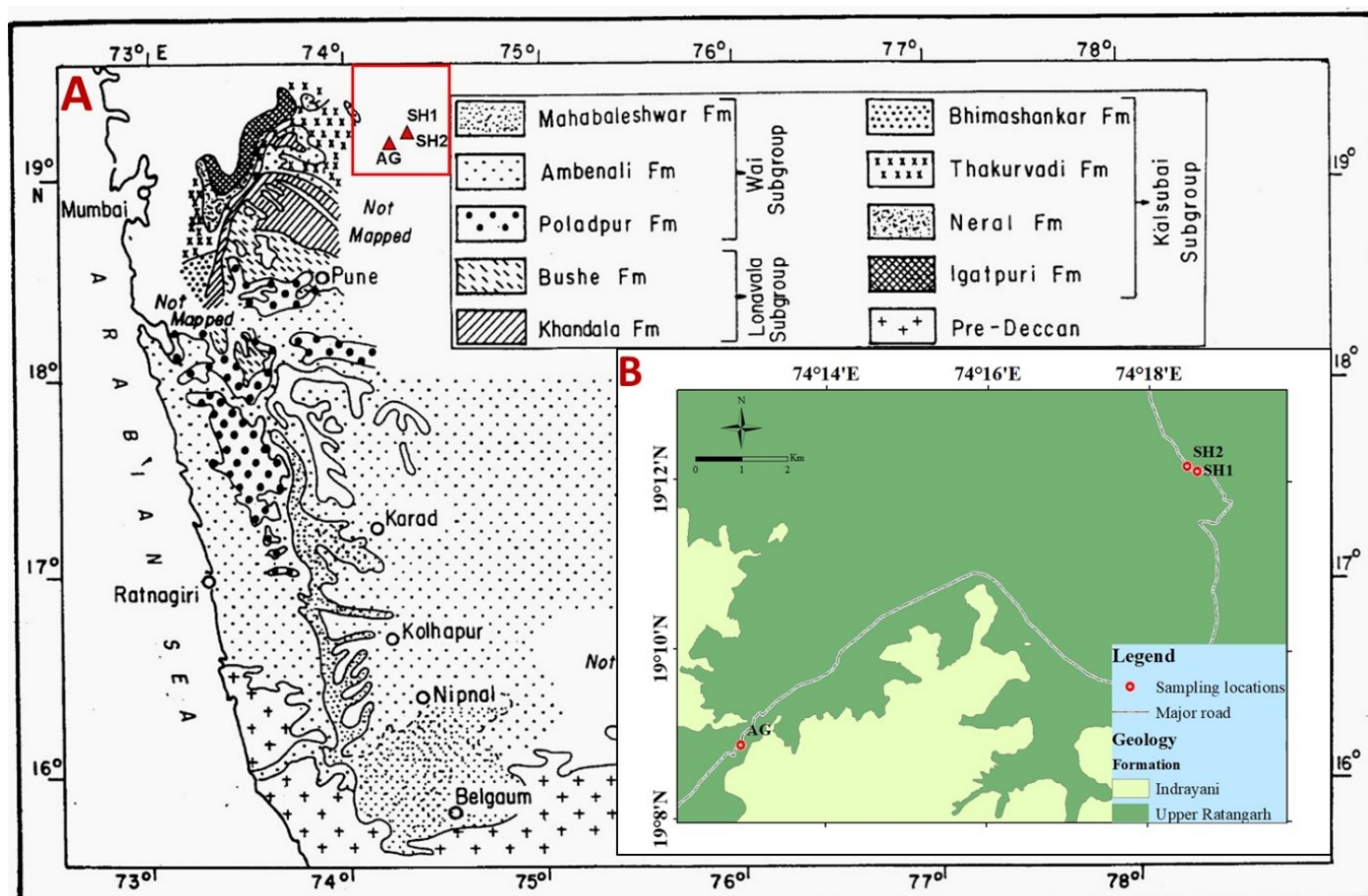


Figure 1. Geological map of Deccan Volcanic Province, after Parthasarathy 2006 [40]. (A) Location of study area marked in red box. (B) Geology of the study area [Junnar, Parner Taluka in Pune & Ahilyanagar].



Figure 2. (a,c,e) Outcrops of Ane ghat, Shindewadi 1, Shindewadi 2; (b,d,f) Secondary minerals from locations—Ane ghat, Shindewadi 1, Shindewadi 2.

2.3. Cavities in the DVP

The Deccan Volcanic Province (DVP) contains various types of open spaces within basaltic lava flows including vesicles, cavities, vugs and fractures. These features differ in their morphology, origin and relationship with secondary mineralization. The term cavity is used here as a general morphological term for an open space with a roughly rounded to irregular boundary, ranging in size from millimetres to centimetres. Cavities may develop through vesiculation, dissolution cooling related processes, or structural modification of the host basalt. Vesicles are primary cavities produced by the exsolution and entrapment of volatile rich gas bubbles during the cooling and solidification of lava. When such vesicles are subsequently filled partly or completely by secondary minerals, they are referred to as amygdaloids (amygdules). In the study area, amygdaloids range from a few millimetres to several centimetres in size and are commonly filled by secondary minerals such as Stilbite, Scolecite, apophyllite, Calcite, Quartz, and/or Chalcedony.

A vug refers to an open cavity, commonly within the rock or a mineralized zone, whose walls are lined or coated by crystals. In contrast, a fracture is a linear or planar mechanical discontinuity produced by brittle failure of the rock. Fractures may develop as a consequence of tectonic stresses, contraction during cooling of the lava, or unloading associated with erosion and removal of overlying material. Where such fractures become mineral filled, they may form mineralized veins or fracture filled assemblages.

The investigated localities show distinct morphological and mineralogical characteristics of these open

spaces. At Ane Ghat, the secondary mineralization occurs within a cavity that is filling predominantly by Calcite and is associated with Stilbite. At Shindewadi 1, Calcite occurs within an irregularly shaped cavity, representing cavity fill mineralization, where Calcite occurs in association with Scolecite within pre-existing vesicular spaces (Figure 2b,d,f). Thus, the investigated Calcite occurrences represent predominantly cavity fill and amygdaloidal mineralization, rather than unequivocal fracture or vein fill mineralization. The morphological distinction is important for interpreting the mode of emplacement of Calcite and its paragenetic relationship with the associated zeolite minerals.

3. Analytical Methods

3.1. Optical Petrographic

Petrographic examination of the thin section was carried out using a polarizing optical microscope at the Department of Geology, Savitribai Phule Pune University, Pune, India.

Petrographic examination of the Calcite bearing samples reveals variations in crystal morphology, deformation intensity, fracture development and the relationship between Calcite and the surrounding basaltic host. The Calcite generally occurs as coarse to medium grained secondary crystals occupying vesicular/cavity spaces and fractured domains within the basalt. Individual Calcite crystals are predominantly anhedral to subhedral, although, locally well-developed crystal boundaries can be observed. The crystals are generally optically homogeneous and do not show conspicuous, concentric or sectorial growth zoning in the examined thin sections.

The Ane Ghat Calcite (Figure 3a,b) is characterized by relatively coarse crystals displaying abundant intersecting planar features. These features form a prominent grid like pattern within the crystals and are interpreted primarily as Calcite twinning and deformation related fractures rather than compositional growth zoning. Several fractures transect the Calcite crystals indicating that fracturing occurred after at least part of the Calcite had crystallized. The Calcite-host contact is locally irregular, and dark, fine grained material occurs along portions of the crystal margin. However, a systematic replacement front or dissolution-replacement texture is not sufficiently evident in the available thin-section images to establish replacement of an earlier mineral.

The Shindewadi 1 sample (Figure 3c,d) contains comparatively homogeneous Calcite with relatively subdued internal optical variation. Several straight to slightly irregular fractures intersect the Calcite crystals. Unlike the Ane Ghat sample, the Calcite does not display a pronounced

network of intersecting Twin/deformation features. No obvious concentric growth zoning is observed. The relatively uniform optical character suggests crystallization of a comparatively homogeneous Calcite generation, followed subsequently by brittle fracturing.

The Shindewadi 2 sample (Figure 3e,f) exhibits a more complex textural association between Calcite and the surrounding altered basaltic material. Calcite occurs as irregular to locally fragmented domains adjacent to the darker host material with numerous fractures developed through the carbonate. Some Calcite boundaries are irregular rather than euhedral, indicating growth into an irregular open space or fractured domain. The available image therefore supports an interpretation of Calcite filling an existing void or fracture network rather than permitting a definitive identification of a replacement texture. Where Calcite occupies an open cavity, the contact with the basaltic wall is irregular and locally associated with fine grained alteration products.

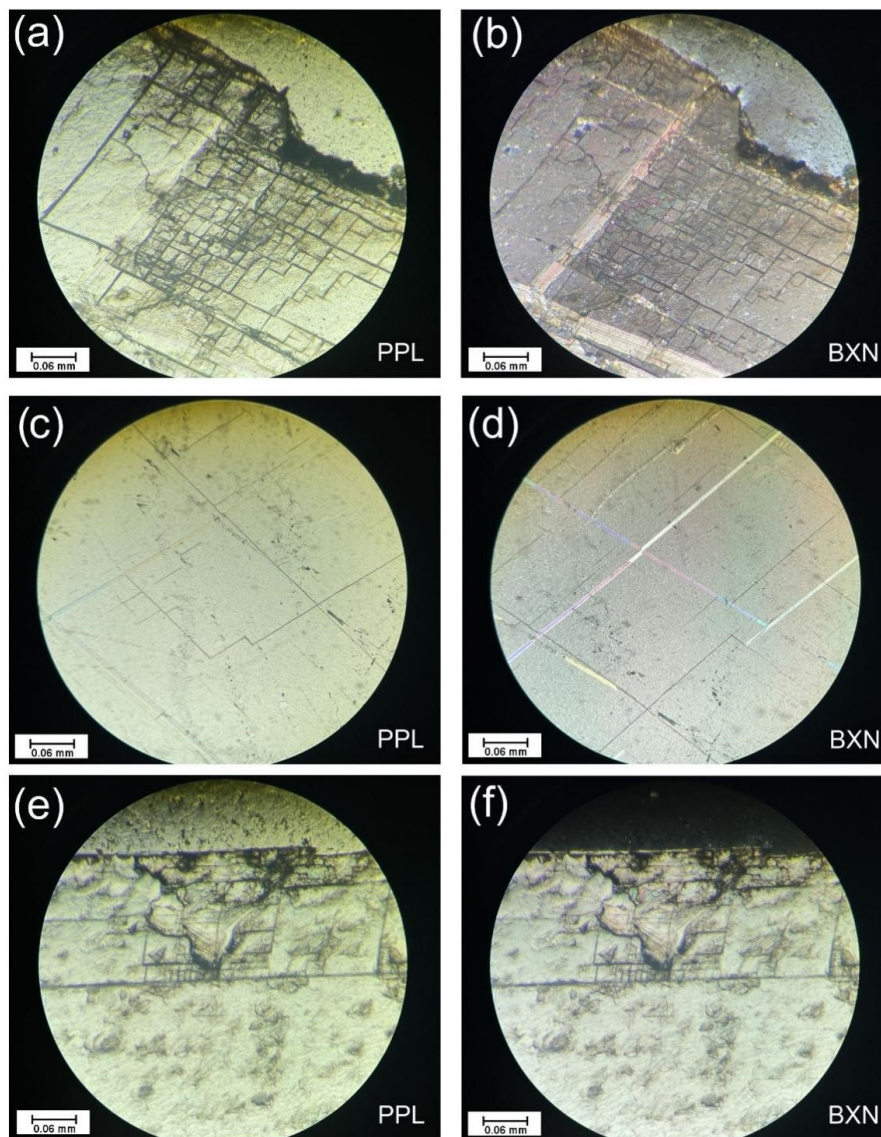


Figure 3. Photomicrographs of calcite (a,b) Ane Ghat. (c,d) Shindewadi 1. (e,f) Shindewadi 2.

Across the three samples, the most conspicuous post-crystallization feature is fracturing and deformation of Calcite, particularly in the Ane Ghat sample. The fractures commonly cross the individual Calcite crystals, demonstrating that at least part of the deformation post-dates Calcite crystallization. However, the available thin sections do not provide unequivocal evidence for repeated crack-seal growth, because discrete successive generations of carbonate separated by healed fracture bands are not clearly resolved. Similarly, although Calcite occurs in association with altered basalt and secondary mineral assemblages, the present petrographic images do not permit a definitive identification of Calcite replacing an earlier zeolite or other secondary mineral.

3.2. Scanning Electron Microscope & EDS

Imaging and spot analyses were carried out at the Central Instrumentation Facility, Savitribai Phule Pune University. Secondary-electron and backscattered-electron images were used to assess crystal face preservation, surface texture and evidence for dissolution or overgrowth; energy-dispersive spectra were collected from flat regions of individual crystals.

A limitation is stated at the outset rather than buried. Energy-dispersive analysis is not a quantitative technique for carbon and oxygen in a carbonate. Low-energy characteristic X-rays from light elements are strongly self-absorbed, fluorescence yields are low, and surface contamination together with any conductive coating contributes directly to the measured carbon signal [41]. The EDS data reported below are used to confirm phase identity and to place upper limits on minor elements, and are used for nothing else.

3.3. Micro-Raman Spectroscopy

Raman spectra were acquired in the Department of Earth Sciences, Indian Institute of Technology Bombay, on the same polished sections used for petrography, with the analysed spot located optically within a single crystallographic domain. Peak positions were obtained by fitting individual bands after background subtraction.

Band width and band position, not absolute intensity, are used as structural indicators. Absolute Raman intensity in a micro-Raman experiment depends on laser power, focal quality, objective, integration time and in an optically anisotropic mineral such as calcite, on the orientation of the crystal relative to the incident polarization. The E_g lattice modes of calcite are strongly orientation-sensitive, so a favourably oriented grain returns a stronger band irrespective of its defect density. Intensity comparisons between separately mounted grains are not physically meaningful, and none are made here. The full width at half maximum of the ν_1 mode, which broadens systematically with lattice disorder and with magnesium substitution, is the appropriate quantitative crystallinity metric and is identified below as a target for future measurement [42–46].

4. Result

4.1. Growth Textures

In every section the calcite occupies vesicles and short fracture segments and is bounded by the basalt wall along a sharp, non-reactive contact. No embayment of the wall rock, no corroded plagioclase or pyroxene relics enclosed within the calcite, and no reaction rim were observed. Crystals are coarse and commonly fill an entire void as one or a few optically continuous individuals, displaying the perfect rhombohedral cleavage and extreme birefringence characteristic of the phase, with interference colours in the high-order pale range under crossed polars (Figure 3a–f).

In backscattered and secondary-electron images the crystals are euhedral to subhedral, with planar faces, sharp edges and stepped growth surfaces. Etch pits, rounded corners, dissolution embayment and overgrowths of a second carbonate generation were not observed at the scale examined. The sharp wall contact, the absence of replacement textures and the preservation of crystal faces together indicate growth into a fluid-filled void rather than metasomatic replacement of the basalt (Figure 4a–c).

4.2. Deformation Features

A consistent set of strain features is superimposed on the growth fabric. Most conspicuous are closely spaced, straight, parallel lamellae that remain optically continuous through stage rotation and extinguish at an angle to the host, identifying them as mechanical e-twins rather than mineral-filled partings or exsolution lamellae. Two intersecting twin sets are developed in several grains. Twin lamellae terminate at crystal boundaries and do not continue into adjacent grains or into the wall rock (Figure 3a–f).

Grains at Shindewadi 1 show pronounced undulose and patchy extinction during rotation, indicating residual lattice distortion. Intracrystalline microfractures occur at all three sites; they are dominantly cleavage-parallel with irregular linking segments and locally offset cleavage traces by a few tens of micrometres. A subset of fractures is healed and marked by planar trails of small inclusions, a geometry characteristic of secondary fluid-inclusion planes.

The relative timing is unambiguous. Twins, microfractures and healed fractures all cross-cut cleavage planes and growth surfaces that are continuous elsewhere in the same crystal, so they were imposed after the crystal attained its final size. Neither dynamic recrystallization nor sub grain development nor bulging grain boundaries were observed, indicating that strain was accommodated almost entirely by twinning and brittle failure.

Twin morphology is dominated by thin, straight lamellae that appear as fine dark lines at the working magnification, corresponding to Type I of the Ferrill classification [12]. Thick, lensoid and bent twins, and patchy twinning, were not observed. Type I dominance corresponds to deformation at temperatures below approximately 170–200 °C. Table 1 summarizes the microstructural observa-

tions; the qualitative twin-type assignment is reported, and quantitative twin width and density statistics, which would sharpen the temperature bracket, are identified in Section 6.7 as required future measurement.

4.3. SEM-EDS

Spectra from all three localities are dominated by Ca, C and O with minor Fe and Mg, and, at Ane Ghat and Shindewadi 1, sub-1 wt% Si and Al. No other element was detected above background. Normalized results appear in Table 2.

The stoichiometric ratios are the informative rows. Calcite requires Ca:C:O in the atomic proportion 1:1:3. Measured Ca/C ratios of 1.31–1.61 depart from unity by 30 to 60%, and measured Ca/O ratios of 0.62 exceed the ideal value of 0.33 by nearly a factor of two. This is not a property of the samples. It is the expected behaviour of energy-dispersive analysis applied to light elements in a coated carbonate, and the constancy of the Ca/O ratio at 0.62 across all three sites demonstrates that the offset is systematic and instrumental rather than random.

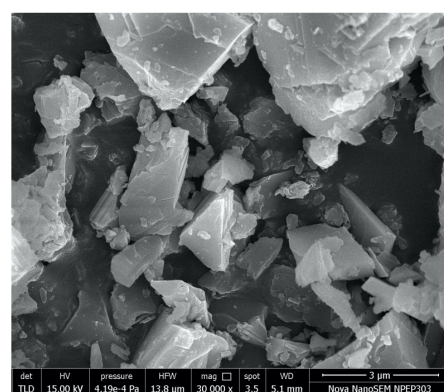
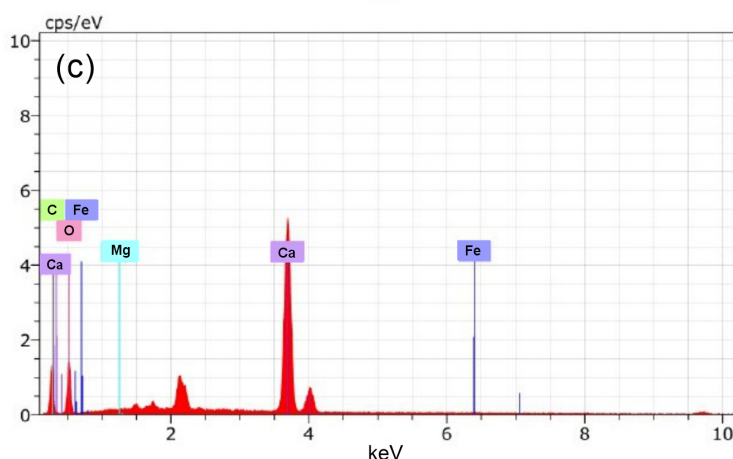
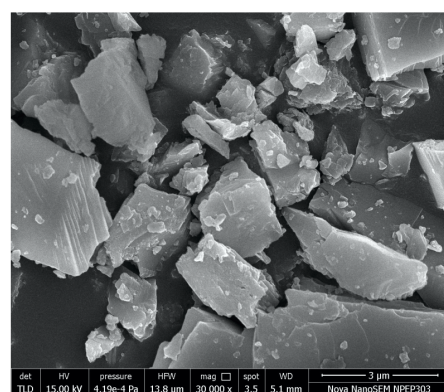
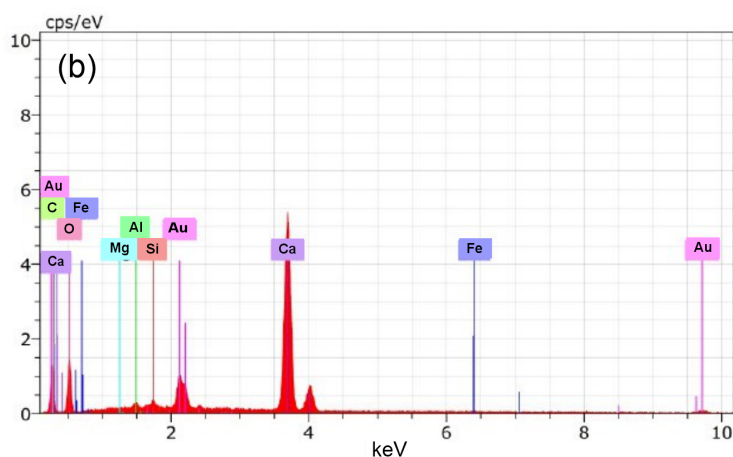
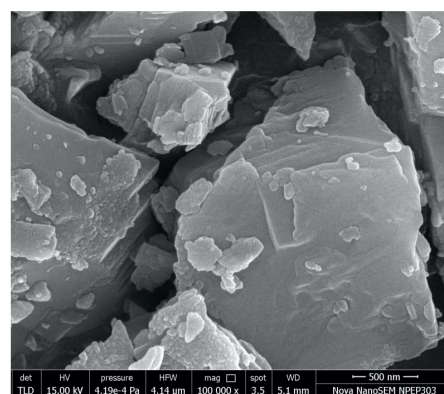
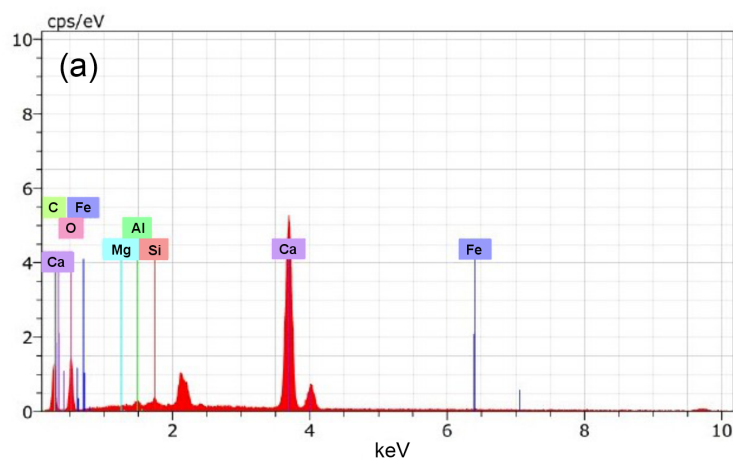


Figure 4. SEM EDS Images; (a) Ane Ghat. (b) Shindewadi 1. (c) Shindewadi 2.

Table 1. Microstructural observations in cavity-filling calcite.

Feature	Ane Ghat	Shindewadi 1	Shindewadi 2
Polysynthetic e-twins	Present	Present	Present
Dominant twin type	I	I	I
Intersecting twin sets	Observed	Observed	Observed
Undulose extinction	Moderate	Pronounced	Moderate
Cleavage-controlled microfractures	Present	Present	Present
Cleavage offsets	Observed	Observed	Observed
Healed fracture planes	Present	Present	Present
Dynamic recrystallization	Absent	Absent	Absent
Inferred T of deformation	<170–200 °C	<170–200 °C	<170–200 °C

Table 2. Semi-quantitative SEM-EDS analyses (weight per cent, normalized; atomic per cent in parentheses).

Locations	Ane Ghat		Shindewadi 1		Shindewadi 2	
Elements	Wt%	At.%	Wt%	At.%	Wt%	At.%
Ca	52.58	29.33	53.75	30.54	53.81	30.13
O	33.63	46.99	34.53	49.15	34.39	48.24
C	11.99	22.32	10.01	18.98	11.49	21.48
Si	0.89	0.71	0.87	0.71	b.d	b.d.
Al	0.62	0.51	0.61	0.52	b.d	b.d.
Fe	0.62	0.1	0.19	0.08	0.25	0.1
Mg	0.05	0.04	0.04	0.04	0.05	0.05
Ca/C (atomic)	1.31		1.61		1.4	
Ca/O (atomic)	0.62		0.62		0.62	

b.d. = below detection. Ideal calcite has Ca/C = 1.00 and Ca/O = 0.33. Values are not stoichiometric and must not be read as quantitative mineral chemistry; see Section 4.3.

Two consequences follow and are stated rather than worked around. No claim about carbonate stoichiometry, purity or crystal chemistry can be supported by these data. And the differences between localities 1.2 wt% in Ca, 0.06 wt% in Fe, are far smaller than the systematic error just demonstrated and cannot be read as real compositional differences.

Twin-type assignment follows the four-type morphological classification of [12]. The temperature bracket derives from twin morphology alone and is a first-order constraint; quantitative twin width and density statistics are identified as required future measurement in Section 6.7.

4.4. Raman Spectroscopy

All spectra are those of calcite in the R $\bar{3}c$ structure (Figure 5a–c). The two external lattice modes lie near 156 cm^{−1} (translational, E_g) and 279–281 cm^{−1} (vibrational, E_g), and the internal carbonate modes at 711–

712 cm^{−1} (ν_4 in-plane bend, E_g) and 1085–1087 cm^{−1} (ν_1 symmetric stretch, A_{1g}). Weak features near 1435 cm^{−1} and 1749 cm^{−1} were recorded at all sites. The former corresponds to the ν_3 antisymmetric stretch, formally Raman-inactive in ideal calcite but weakly present in most natural spectra; the latter is a well-known combination band. Neither requires explanation in terms of chemical substitution [47].

No aragonite, dolomite, magnesite or amorphous calcium carbonate signature was detected. Peak positions agree between the three localities—Ane Ghat (Figure 5a), Shindewadi 1 (Figure 5b), Shindewadi 2 (Figure 5c) to within 4 cm^{−1}, which is at or below the wavenumber accuracy of a routinely calibrated dispersive system; the published range for the vibrational mode of natural calcite spans this interval, and the difference is not interpreted. Band positions and reference values are compared in Table 3.

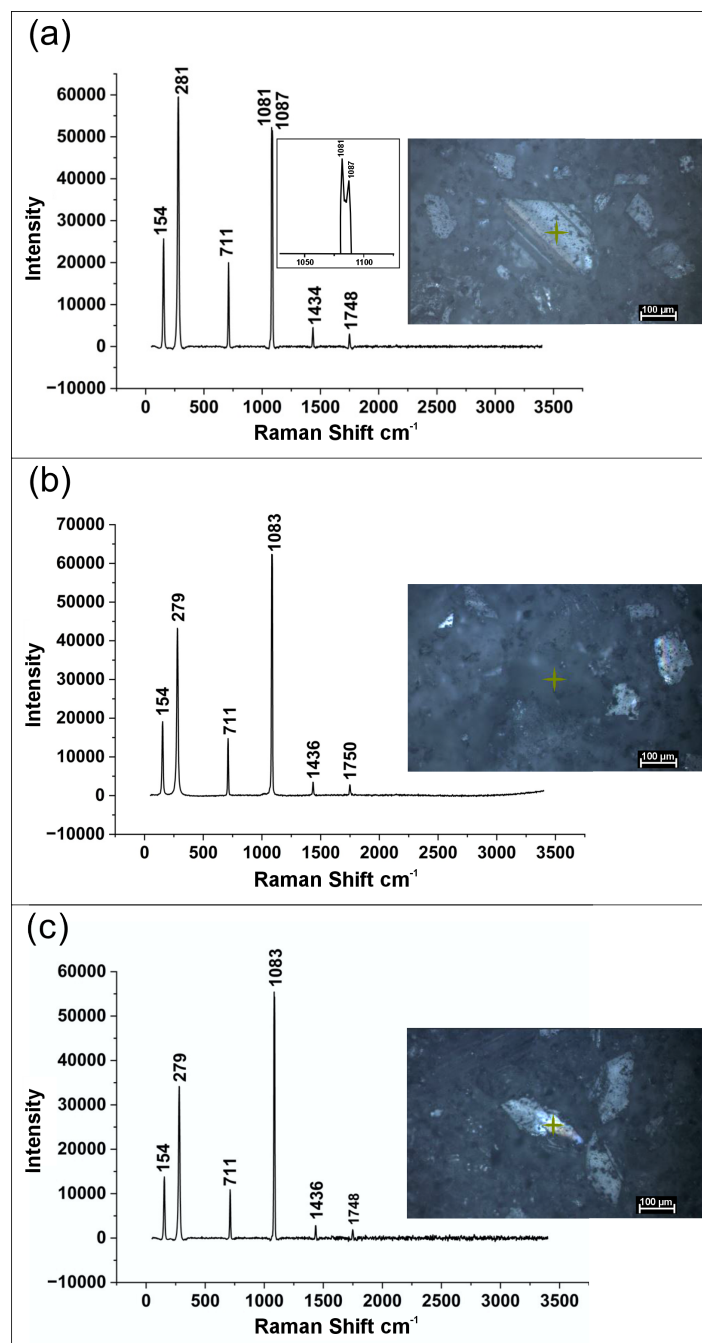


Figure 5. Raman Shifts cm^{-1} of calcite, (a) Ane ghat. (b) Shindewadi 1. (c) Shindewadi 2.

Table 3. Micro-Raman band (cm^{-1}) positions of calcite.

Band (Assignment)	Ane Ghat	Shindewadi 1	Shindewadi 2	Reference
Lattice T (E_g)	156	154	154	~156
Lattice L (E_g)	281	279	279	~282
ν_4 (E_g)	711	711	711	~712
ν_1 (A_{1g})	1081, 1087	1083	1083	~1086
ν_3 (weak)	1434	1436	1436	~1435
Combination band	1748	1750	1748	~1749

Reference values after [46, 48]. Inter-site differences do not exceed the wavenumber accuracy of a routinely calibrated dispersive system and are not interpreted. The ν_1 mode is singly degenerate (A_{1g}) and is not split in calcite. However, the Ane ghat Raman peak showing splitting at 1081 and 1087 cm^{-1} (Figure 5a) which could reflect the possible cause of lattice strain, structural disorder.

5. Discussion

5.1. What the Growth Textures Establish

The textural evidence for open-space growth is solid and internally consistent: sharp non-reactive wall contacts, absence of replacement fronts, coarse optically continuous crystals filling voids, and euhedral faces preserved in secondary-electron images. Growth from a supersaturated aqueous fluid moving through connected vesicles and fractures is the only reasonable reading. The association with zeolite and silica phases in the same cavities, with calcite occupying the residual central volume, places this calcite late in the local filling sequence, in agreement with the paragenesis long established for the province [7, 9, 49, 50].

The limit of that inference deserves to be explicit. Late in the filling of a given cavity is a relative statement about one void. It does not establish that the calcite is late in the history of the province, and it does not identify which of the calcite generations recognized province-wide is represented. Calcite associated with a burial-related stage and a considerably younger carbonate generation with mineralization ages in the Eocene–Oligocene range have both been documented [9], and texturally similar, chemically indistinguishable calcite may belong to either. Any genetic model resting on textures alone therefore carries an irreducible ambiguity, which is left visible here rather than closed by assumption.

5.2. The Deformation Record

The strain features are the substantive new observation. Their timing is fixed by geometry: e-twins, microfractures and healed fracture planes cut across cleavage and growth surfaces that are continuous elsewhere in the same crystal, so the strain post-dates completion of crystal growth, which in turn post-dates sealing of the vesicle. The calcite therefore brackets a deformation event younger than hydrothermal cavity filling.

Mechanical twinning is the dominant crystal-plastic mechanism in calcite below roughly 400 °C and operates at critical resolved shear stresses of only a few megapascals, so its presence alone does not require a large stress. Morphology is what carries diagnostic weight. Thin straight Type I twins form below about 170–200 °C; thick, lensoid and bent twins appear above that threshold, and patchy twinning above roughly 250 °C [11–13, 51]. Experimental investigation further demonstrates the utility of calcite deformation twins as indicators of stress and strain under low temperature condition [52]. The observed dominance of Type I therefore confines deformation to the low-

temperature field, and the complete absence of recrystallization is consistent.

Post-emplacement deformation of the lava pile is the most economical explanation, but three alternatives must be excluded before that interpretation is secure, and they are flagged rather than passed over. Twinning can be induced during rock sawing and section preparation. It can be produced by differential compaction and load transfer within the pile with no tectonic driver. And in a cavity mineral it can arise from local stress concentration at the amygdale wall, generated by the elastic contrast between calcite and basalt, which would not scale to a regional strain field. The first is testable by examining twins in grains fully enclosed by basalt and remote from cut surfaces. The second and third require orientation data: twin-set orientations measured relative to a geographic frame from oriented samples and inverted for a stress tensor by an established method [13, 14]. Compression axes consistent between grains, cavities and localities would support a regional driver; randomly distributed axes would not.

Against the regional literature the result occupies a useful point of tension. Geomorphometric indices across thirty basins of the western Deccan returned no signal of active tectonics [16], whereas structural and geophysical work has argued for reactivation of sub-trappean basement structures. The two need not agree. Evidence from Koyna-Warna region documents fault zone deformation and calcite bearing veins within the Deccan basalt succession, demonstrating the brittle deformation and fluid assisted mineralization continued after emplacement [53]. Landscape indices integrate only the most recent deformation, whereas a twinning strain integrates everything that has occurred since the cavity was sealed, and the microstructural record may register events that the topography no longer preserves. The twin data are offered as evidence bearing on that debate rather than as confirmation of either side.

5.3. Constraints on the Compositional and Spectroscopic Data

Three interpretations that recur in the earlier literature on Deccan cavity calcite do not survive scrutiny and are set aside.

The first treats EDS-detected Si and Al as elements incorporated into the calcite lattice from the mineralizing fluid. Neither ion has a plausible site in the calcite structure. Ca^{2+} occupies a six-fold coordinated site and is substituted by divalent cations of comparable radius Mg^{2+} , Fe^{2+} , Mn^{2+} , Sr^{2+} whereas Si^{4+} is four-coordinated in silicates and Al^{3+} cannot be accommodated without a charge-balancing mechanism for which no evidence exists [54]. Sub-1 wt% Si and Al in a spot analysis of calcite are far more simply explained by micrometre-scale silicate or zeolite inclusions within the interaction volume, or by beam overlap onto an adjacent phase. Their absence at Shindewadi 2 indicates that the particular spot analysed was cleaner. It does not demonstrate a more chemically evolved fluid.

The second uses absolute Raman peak intensity as a crystallinity index. For the reasons set out in Section 3.3, intensity in a micro-Raman measurement is governed by instrumental and geometric factors and, in an anisotropic crystal, by orientation relative to the incident polarization. Band width and position are the correct metrics, and the ν_1 full width at half maximum is the appropriate one here [42, 45]. No ranking of the three localities by lattice order is defensible on the present data.

The third reads the 2 cm^{-1} difference in the vibrational lattice mode as a compositional or crystallization signal. This lies at or below the wavenumber accuracy of a routinely calibrated dispersive Raman system, and the published range for natural calcite spans the interval. Interpreting it would be over-reading the data.

What the Raman data do establish is worth retaining, stated modestly. All three samples are calcite; they are structurally ordered; no other carbonate polymorph and no magnesium-rich carbonate is present; and the carbonate framework was not measurably disturbed by the twinning strain, an observation itself consistent with the low deformation temperature inferred from twin morphology, and one with direct bearing on the durability argument developed below.

6. Implications for Mineral Carbon Sequestration in the Deccan Traps

6.1. Why Basalt, and Why Carbonate

Geological disposal of carbon dioxide in sedimentary reservoirs relies principally on structural and residual trapping, in which a buoyant supercritical phase is retained beneath a low-permeability seal. Retention in that configuration is contingent on the seal remaining intact, so the storage site requires monitoring on a timescale set by the residence of the buoyant phase [55]. Mineral trapping in reactive silicate rock operates on an entirely different principle. Dissolution of CO_2 in formation water produces carbonic acid, which attacks primary silicates and liberates Ca^{2+} , Mg^{2+} and Fe^{2+} ; when carbonate saturation is reached, those cations are fixed as calcite, dolomite, ankerite, siderite or magnesite [19, 20]. The carbon is then held in a thermodynamically stable solid of essentially the same density as the host rock. Buoyancy is eliminated, leakage pathways become irrelevant, and the storage becomes permanent in a sense that structural trapping cannot match.

Basalt is the preferred host because it is unusually rich in the necessary divalent cations. The controlling doubt was never whether the reaction occurs but whether it occurs fast enough to be useful, and field injection has settled that question decisively: at the Hellisheidi site in southwest Iceland the great majority of injected CO_2 was mineralized within approximately two years, an outcome substantially faster than pre-injection modelling anticipated [21]. However, experimental and field observations from the CarbFix project at Hellisheidi demonstrate that basalt–fluid reactions and carbonate mineralization can proceed rapidly

when reactive basalt interacts with CO_2 -bearing water under favourable geothermal and hydrological conditions. However, this system should not be considered a direct kinetic analogue for the Deccan Volcanic Province. The Deccan basalts represent an ancient continental flood-basalt sequence that underwent prolonged cooling, burial, alteration and multiple episodes of secondary mineralization. Thus, the CarbFix results are used here only to demonstrate the potential for relatively rapid basalt–fluid interaction, rather than to assign a specific mineralization rate to the Deccan system. The timing and duration of Deccan secondary mineralization must instead be constrained from its mineral assemblages, paragenetic relationships, fluid characteristics and available geochronological constraints.

Comparable reactivity has been demonstrated in continental flood basalt [23] and in deep-sea basaltic crust [56], and the approach is now treated as a mature component of the storage portfolio [22].

6.2. The Deccan Traps as a Candidate Reservoir

The Deccan Volcanic Province has been identified as a strategic CO_2 storage domain for India for nearly two decades [24], and the case has since been developed on several independent lines. Laboratory carbonation of Deccan lithologies confirms reactivity directly: picrites from the Igatpuri Formation reacted with water and supercritical CO_2 at 100°C and 60 bar for five months developed secondary carbonate on grain surfaces and in intergranular space, identified by combined SEM–EDS, infrared and Raman spectroscopy [31]. Reactive-transport modelling of the Mandla lobe indicates that more than half of injected carbon mineralizes within roughly 140 days, with ankerite, siderite and calcite as the dominant product phases [32]. Longer-duration capture experiments on Deccan basalt have tracked crystallographic evolution of the calcite product itself [32]. Structural and residual trapping behaviour has been modelled for the Saurashtra sector [27], and Deccan dyke swarms have been evaluated separately as potential sinks [26].

The most quantitatively constrained assessment to date integrates 12,072 line-km of two-dimensional seismic data with borehole and petrophysical control from the Koyna–Warna and Killari regions, and reports a gross rock volume of $88,937\text{ km}^3$ at depths exceeding 750 m, the minimum burial required to maintain supercritical CO_2 conditions [25]. That study identifies the reservoir architecture explicitly: vesicular and amygdaloidal flow tops constitute the principal storage intervals, while massive flow interiors act as laterally continuous natural seals, giving the province an intrinsic layered trapping geometry.

It is this last point that connects the engineering literature to the present observations. The amygdaloidal flow tops nominated as reservoir intervals are the very horizons sampled at Ane Ghat, Shindewadi 1 and Shindewadi 2, and the vesicles nominated as pore volume are the voids whose calcite fill is described in Section 4.

The calcite filled amygdalites investigated in this study provide a natural geological example of carbonate mineral formation and long-term carbon immobilization; however, they should not be regarded as a direct or exact analogue of modern engineered carbon capture and storage (CCS) operations. The geological system developed over substantially longer timescales and under fluid flow conditions that differ fundamentally from those associated with industrial CO₂ injection. In the studied basaltic system, the observed paragenetic sequence indicates that calcite precipitation occurred during a late stage of hydrothermal burial evolution after the formation of zeolite minerals and progressive reduction in the available pore space and permeability. Consequently, Calcite formation probably occurred from relatively evolved and residual fluids within a progressively sealed vesicular network, rather than from a rapidly injected CO₂ rich fluid through an actively permeable reservoir. This distinction is important when considering the relevance of the studied system to CCS. Modern geological CO₂ storage involves deliberate injection of CO₂, commonly under elevated pressure followed by multiphase fluid migration, dissolution and potential mineral reactions within an engineered reservoir. The natural system examined here does not reproduce these injection rates, pressure conditions, fluid flow regimes, or reservoir management strategies. Therefore, the present study does not claim that the observed amygdalites constitute a direct experimental simulation of engineered CCS. Instead, their significance lies in demonstrating a natural end member of long-term mineral trapping. The sequential development of zeolite and carbonate minerals illustrates how basaltic mineral assemblages can evolve through fluid rock interaction toward progressive sealing and carbonate precipitation. The occurrence of calcite at the late stage of the paragenetic sequence provides geological evidence that carbonate minerals can persist as stable as mineral products within basaltic cavities over geological timescales. Thus, the studied amygdalites are better considered a natural geological analogue for the long-term mineralogical consequences of carbon bearing fluid rock interaction rather than a direct analogue for the kinetics or hydrodynamics of industrial CCS. Accordingly, the observations provide a geological perspective on the potential for basalt hosted systems to retain carbon through carbonate mineralization, while recognizing that the efficiency and rate of retained mineral trapping in engineered CCS depend on factors such as CO₂ injection conditions, fluid pressure, permeability, reactive surface area, temperature, fluid composition, and reservoir scale hydrology. The natural example therefore informs the long-term mineralogical end point of carbon sequestration, but does not by itself establish the performance or efficiency of an engineered CCS system.

6.3. Amygdale Calcite as a Completed Natural Experiment

Sections 4.1 and 5.1 establish a sequence of events that reproduces, step for step, the reaction pathway that engineered carbonation is designed to induce. Aqueous fluid circulated through a connected void network in tholei-

itic basalt. Fluid–rock interaction altered calcic plagioclase, clinopyroxene and glass, releasing Ca and other cations into solution. Silica and alumina were progressively removed from that fluid by precipitation of zeolite and silica phases. When the residual fluid attained saturation with respect to CaCO₃, calcite grew into the remaining open volume and sealed it.

Two features of the resulting cement carry direct evidential weight for storage permanence, and both are established above.

The first is the absence of dissolution. No etch pits, rounded corners or dissolution embayment were observed in secondary-electron imaging, and no replacement fronts or corroded relics occur at the wall contact. Whatever fluids traversed the pile after cavity sealing did not appreciably redissolve the carbonate. Given the interval concerned of order 6.6×10^7 years, this is a stronger empirical statement about long-term mineral-trap stability than any laboratory or pilot programme can furnish, because no experiment can be run for the timescale that actually matters.

The second is structural integrity at the lattice scale. Micro-Raman spectra are those of well-ordered calcite, with no aragonite, no magnesian carbonate, and no amorphous calcium carbonate signature. The carbonate framework has neither inverted to a metastable polymorph nor accumulated detectable disorder, and critically it was not measurably disturbed by the twinning strain documented in Section 4.2. Since crystallographic modification of calcite is one of the properties tracked in long-duration Deccan CO₂-capture experiments [32], the demonstration that a 66-million-year-old natural carbonate in the same province retains a well-ordered structure through a subsequent deformation episode is a directly relevant benchmark.

Quantitative assessment of the cavity-filling secondary mineral assemblage indicates that calcite constitutes approximately 0.8–16 molar volume% of the local secondary precipitate volume in the studied samples. The Ca concentration was also converted to CaCO₃ equivalent using the stoichiometric factor $\text{CaCO}_3/\text{Ca} = 100.09/40.08 \approx 2.497$; accordingly, 29.33 wt% Ca corresponds to approximately 73.25 wt% CaCO₃ equivalent, assuming that the measured Ca is entirely hosted by calcite. These values demonstrate the measurable contribution of carbonate mineral precipitation within the cavity-filling assemblage and provide quantitative support for the occurrence of mineral carbon trapping.

Taken together, the mineralogical evidence supports permanence. The microstructural evidence complicates it, and that complication is the substantive contribution of this study to the storage question.

6.4. The Carbon Source Problem

A natural analogue is quantitatively useful only to the extent that the source of its carbon is known, and the present dataset does not constrain that source. Stable carbon and oxygen isotope analysis would resolve it directly. Carbon derived from magmatic or mantle CO₂ occupies a $\delta^{13}\text{C}$ range near -5 to -8‰ relative to VPDB; carbon from

soil-zone or organic sources is markedly lighter; carbon from dissolution of marine carbonate is heavier. Oxygen isotopes, combined with an independent temperature estimate, yield the $\delta^{18}\text{O}$ of the parent fluid and therefore discriminate meteoric-dominated from evolved or burial fluids.

A regional endpoint for that comparison already exists, and it is informative. Carbonates separated from crystalline basement recovered from the KLR-1 borehole in the Killari region, beneath the Deccan cover, from samples KIL-13 at 440.5 m and KIL-20 at 499.6 m, return $\delta^{13}\text{C}$ values of -6.23 and -6.22% versus PDB and $\delta^{18}\text{O}$ values of 7.94 and 8.11% versus SMOW. These plot within the primary igneous carbonatite field and were interpreted as carbonate of mantle origin, produced by metasomatism drawing on the deep mantle carbon reservoir; the same basement rocks carry 2.00 – 2.28 wt% CO_2 [57, 58].

Three points follow. First, and most importantly for the present purpose, the values are a comparison framework and not a measurement on the material described in this paper. They derive from 2.5 Ga amphibolite–granulite facies basement at 440 – 500 m depth in a different district, not from amygdale calcite in the lava pile, and they are used here strictly as an independently published endpoint. Second, they document a substantial deep carbon reservoir directly beneath the province, together with a demonstrated mechanism for transferring that carbon upward and fixing it as stable mineral carbonate, a natural precedent for the process that engineered injection seeks to reproduce. Third, they convert a vague expectation into a sharp and falsifiable prediction. If Deccan amygdale carbon is dominantly deep-sourced, $\delta^{13}\text{C}$ in the calcite described here should cluster near -6% ; values substantially lighter would indicate a soil-zone or organic contribution, and values substantially heavier would indicate dissolution of pre-existing carbonate. The distinction matters for the analogue, because a natural system that mineralized mantle CO_2 at depth is a closer structural parallel to engineered injection than one that mineralized shallow soil-derived carbon.

The relatively low $\delta^{18}\text{O}$ of the Killari basement carbonates, near 8% SMOW, is itself diagnostic of a high-temperature, deep-sourced fluid rather than shallow meteoric water, and provides a second axis of comparison once amygdale values are available.

6.5. Post-Mineralization Deformation and Reservoir Integrity

The microstructural results bear on storage security in a way that the mineralogical results do not, and the argument is worth stating carefully because it cuts against the prevailing emphasis.

Basalt carbonation is generally presented as permanent, and at the level of the mineral phase that presentation is correct: the observations in Section 6.3 support it. But permanence of the carbonate phase is not the same thing as permanence of the reservoir. The amygdale calcite described here was mechanically strained after it sealed its host cavity. Twins formed, cleavage-controlled microfractures propagated through the carbonate cement,

and cleavage traces were offset by tens of micrometres. Mechanical twinning in amygdaloidal calcite records post-crystallization, intracrystalline strain accommodation and provides evidence for the stress deformation history of the reservoir. However, Calcite Twinning alone does not demonstrate a loss of reservoir containment. Any inference regarding compromised integrity or enhanced fluid leakage requires independent evidence of interconnected brittle fractures, faulting, cataclasis, or other macroscale permeability pathways [59].

The plateau interior is conventionally treated as tectonically quiescent, and geomorphometric analysis across thirty western Deccan basins found no signal of active tectonics [16]. Even so, the cement records brittle failure.

Three consequences follow for storage assessment in this province.

A quiescent surface expression does not warrant an assumption of mechanical stability at reservoir depth. The strain documented here is invisible in landscape morphology yet plainly recorded in the cement, precisely because a twinning and micro fracturing strain integrates the entire interval since cavity sealing whereas geomorphic indices integrate only the most recent increment. Site screening that relies on geomorphic quiescence alone will systematically underestimate the cumulative deformation a candidate reservoir has experienced. Comprehensive monitoring approaches, including spatial information-based techniques, are therefore important for evaluating storage behaviour and identifying potential anomalies during geological CO_2 sequestration [59, 60].

Second, carbonate cement is the mechanically weak link, not the seal. The massive flow interiors that constitute the natural seals in the Deccan architecture [25, 61] are unfractured basalt, whereas the carbonate filling the vesicular flow tops fails brittly at low differential stress. Fracturing of the cement does not by itself breach containment, since the seal is a separate lithology, but it does create secondary permeability within the storage interval, with implications for fluid redistribution, for the effective porosity available to subsequent injection, and for the mechanical response of an already carbonated horizon to renewed pore-pressure increase. Experimental work has established that carbonation measurably alters the strength of Deccan basalt [58]; the present observations show that the natural product of the same reaction, once emplaced, is itself susceptible to brittle failure.

Third, the temperature bracket sets the conditions of failure. Type I twin dominance places the deformation below approximately 170 – 200 °C in the range of shallow crustal, near-ambient conditions rather than any thermal excursion. Carbonate cement in a flood-basalt reservoir can therefore be strained under precisely the conditions in which an engineered store would operate. This is not an exotic failure mode confined to orogenic settings.

6.6. Fracture Healing and Self-Sealing

The picture is not one-sided, and the same microstructural record supplies the counterweight. A subset

of the microfractures described in Section 4.2 is healed, marked by planar trails of small inclusions with the geometry characteristic of secondary fluid-inclusion planes. Fractures that opened in the carbonate cement were subsequently sealed by carbonate-bearing fluid moving through them.

This is self-sealing, and for storage assessment it is a favourable observation. A reactive reservoir in which fractures are progressively re-cemented by the same reaction that filled the pore space originally is fundamentally more robust than an inert one, in which a fracture once opened remains a permanent conduit. The secondary inclusion planes are direct textural evidence that the Deccan system possesses this capacity, and they indicate that carbonate precipitation continued after the deformation episode.

The security of mineral trapping in a naturally carbonated flood basalt is therefore best framed as a competition rather than an absolute. Fracture generation degrades the cement; continued carbonate precipitation repairs it. Which term dominates over a given interval determines whether secondary permeability accumulates or is annealed, and that balance is legible in the cement of any amygdale in the province. Reading it does not require a drilling programme. It requires thin sections.

6.7. Measurements Required to Close the Argument

Four measurements would convert the framework above into a quantitative result, and all are routine.

Quantitative twin analysis on oriented samples is the most valuable extension available without new laboratory access. Mean twin width and twin density measured over a statistically adequate population would sharpen the temperature bracket and yield a differential stress estimate [12, 15]. Twin-set orientations measured in a geographic frame, by universal stage or electron backscatter diffraction, and inverted for a stress tensor [13, 14, 62, 63] would establish whether the strain reflects a regional field or purely local stress concentration at the amygdale wall. Consistency of compression axes across cavities and localities is the discriminant. A control observation should accompany this work: twins must be confirmed in grains fully enclosed by basalt and remote from saw cuts, to exclude preparation-induced twinning.

Carbon and oxygen isotope analysis across a suite of samples spanning the three localities and, if possible, more than one stratigraphic level would identify the carbon source and test the prediction set out in Section 6.4 against the Killari basement endpoint [58]. Isotopic spread within a single cavity would additionally reveal whether more than one calcite generation is present, which cannot be detected optically.

Fluid-inclusion micro thermometry on primary inclusions would supply the trapping temperature that oxygen isotope thermometry requires, together with salinity from final ice-melting temperatures. Secondary inclusions along the healed planes are an equally valuable and separate target, because they trap fluid present during or after the deformation; comparison of the two populations would

bracket the deformation event between two measured fluid states.

Wavelength-dispersive electron probe microanalysis using carbonate standards, with CO₂ calculated by stoichiometry, or laser-ablation ICP-MS trace-element analysis, would replace the semi-quantitative EDS data of Table 2 and permit a defensible discussion of minor-element chemistry. Together with ν_1 full-width-at-half-maximum measurement, this would allow the crystallinity and substitution arguments deferred in Section 5.3 to be made properly.

7. Paragenetic Model

The secondary mineralisation investigated in the Volcanic Province (DVP) is interpreted as a multiple process that developed following emplacement and cooling of the tholeiitic basaltic lava flows at approximately 66 Ma. Cooling and thermal contraction of individual lava flows promoted the development of vesicles, amygdaloidal zones, fractures, and joints, which subsequently provided pathways and open spaces for fluid circulation and mineral precipitation. During the early stages of cooling and incipient alteration, interaction between the basalt and circulating fluids promoted alteration of volcanic glass and primary mafic minerals and the development of clay and Fe–Mg-bearing alteration phases [64, 65]. Such early alteration is consistent with previously proposed models for secondary mineralisation in the DVP.

With progressive burial and increasing temperature, continued fluid–rock interaction promoted alteration of basaltic minerals, particularly glass and plagioclase [66, 67] and resulted in the precipitation of secondary zeolites and silica phases. Heulandite, Stilbite, and Scolecite are common secondary minerals in the DVP and have been interpreted to form under low-temperature to low-grade hydrothermal/ burial conditions. Previous studies indicate that the mineral assemblages and their sequence vary between localities, reflecting differences in temperature, fluid composition, permeability, and the extent of fluid–rock interaction [9, 10, 68, 69].

Silica minerals such as Chalcedony and Quartz occur as important components of the secondary mineral assemblage and locally formed in association with zeolites. In some parts of the DVP, later mineralisation was associated with more evolved hydrothermal fluids capable of precipitating minerals such as apophyllite. The occurrence of apophyllite and other late-stage minerals has previously been interpreted as evidence for a later hydrothermal stage in the multistage mineralisation history of the DVP.

Calcite represents an additional stage of carbonate mineralisation in the investigated samples. The occurrence of coarse euhedral to subhedral calcite crystals, well-developed rhombohedral forms, planar crystal faces, and sharp contacts with the basaltic cavity walls is consistent with precipitation from a fluid into pre-existing open cavities and fractures rather than direct replacements of the basalt. The association of calcite with Stilbite at Ane Ghat

and with Stilbite and Scolecite at Shindewadi indicates that carbonate precipitation occurred within the broader sequence of secondary mineralization. However, because the present study does not include fluid-inclusion or stable-isotope measurements, the precise temperature and relative timing of Calcite precipitation cannot be independently constrained.

The chemical and spectroscopic characteristics further support the identification of the investigated material as well-crystallized Calcite. SEM-EDS analysis show Ca, C, and O as the dominant constituents, with minor Si, Al, Fe, and Mg. The Raman spectra are characterised by the diagnostic carbonate bands of Calcite, including the strong band near 1085 cm^{-1} . These observations indicate a well-developed Calcite structure and support its interpretation as a secondary mineral precipitated within the basaltic cavities and fractures.

Following Calcite crystallization, some crystals exhibit mechanical twins, intracrystalline microfractures, off-set along cleavage planes, and locally healed fractures. These features cross-cut or affect previously formed crystal growth features and are therefore interpreted as evidence of deformation after Calcite crystallization. The deformation may reflect post-mineralisation structural reactivation within the basaltic succession. However, because Calcite-filled vesicles can also concentrate local stresses owing to the mechanical contrast between Calcite and Basalt, the observed deformation should not be considered by itself as definitive evidence for a province-wide tectonic stress field.

Thus, the investigated Calcite record at least two major geological episodes: (1) precipitation during secondary mineralisation of cavities and fractures within the Deccan basalts and (2) subsequent deformation after Calcite crystallization. The integrated petrographic, SEM-EDS, and Raman observations therefore indicate that the calcite-bearing cavities preserve a record of both post-volcanic fluid-mediated mineralisation and subsequent structural modification.

8. Genetic Model

The genetic model proposed for the investigated secondary mineralization involves progressive interaction between basaltic rocks and circulating aqueous fluids under changing temperature, fluid-composition, and pressure conditions.

Following emplacement and cooling of the Deccan basalt, thermal contraction generated vesicles, fractures, and joints that acted as sites of fluid circulation and secondary mineral growth. Early interaction between basalt and circulating fluids promoted alteration of volcanic glass and primary minerals and released elements such as Ca, Si, Al, Fe, Mg, and Na into the fluid. These components subsequently precipitated in the formation of clay minerals, zeolites, silica phases, and carbonates.

During burial and continued fluid circulation, increasing temperature promoted progressive alteration and zeolite formation. Previous investigations of the DVP have

demonstrated that zeolite and associated secondary-mineral assemblages developed over a range of relatively low-temperature conditions. At Savda, Ottens documented a multistage sequence involving early alteration, Calcite and Silica mineralisation, followed by zeolite and later hydrothermal mineralisation, with fluid-inclusion temperatures locally extending to approximately $250\text{ }^{\circ}\text{C}$ for Calcite III. In Salsette [10] similarly documented several mineralisation stages, with early high-temperature mineralisation followed by decreasing temperatures and formation of low-temperature zeolites including Scolecite, Heulandite, and Stilbite.

The Stilbite-Scolecite Calcite associated with assemblages in the present study is therefore consistent with relatively low-temperature secondary mineralization. Literature values indicate that these zeolites can form over approximately $80\text{--}160\text{ }^{\circ}\text{C}$ [10, 70, 71] although their actual formation temperatures depend on fluid composition, pressure, host-rock alteration, and mineral stability. Consequently, the occurrence of these zeolites provides a reasonable basis for interpreting the associated Calcite as a relatively low-temperature secondary mineral, but it does not provide a direct temperature measurement for Calcite.

For the investigated samples, Calcite occurs in association with Stilbite at Ane Ghat and Shindewadi 1, with Scolecite at Shindewadi 2. On the basis of these mineral associations and published temperature ranges for these mineral associations and published temperature ranges for capable DVP secondary-mineral assemblages, Calcite precipitation is tentatively interpreted to have occurred under approximately $100\text{--}130\text{ }^{\circ}\text{C}$ conditions. This temperature should be regarded as an inferred range rather than a directly measured crystallization temperature, because fluid-inclusion and stable-isotope thermometry were not performed on the present Calcite samples.

The Ca required for Calcite precipitation could plausibly have been supplied largely through alteration of Ca-bearing minerals, particularly Plagioclase, during prolonged fluid–rock interaction. This interpretation is consistent with the ability of circulating fluids to extract Ca and other major elements from basalt during secondary mineralisation. The carbon component, however, cannot be uniquely established from the present mineralogical and chemical data. Published isotope work on Savda [9] Calcites indicates that their carbon was derived from dissolved inorganic carbon (DIC), involving mixed organic/inorganic carbon sources. Accordingly, a DIC-bearing circulating fluid is considered a plausible carbon source for the present Calcites, although confirmation would require stable carbon- and oxygen-isotope analyses of the investigated samples. The fluid may have contained both meteoric and hydrothermal components. This interpretation is consistent with previous Deccan Volcanic Province studies in which secondary mineralisation has been attributed to evolving fluid systems involving burial-related and hydrothermal processes. At Savda, for example, isotope and fluid-inclusion data indicate that the mineralizing fluids changed through time and that different Calcite gener-

ations formed under different physicochemical conditions. Similarly, the Salsette study demonstrates that the DVP secondary-mineral system experienced changing temperatures and fluid composition through multiple stages.

The final stage of the proposed genetic model involves post-mineralisation deformation. After Calcite crystallization, subsequent structural reactivation and stress within the basaltic succession may have produced mechanical twinning, micro fracturing, cleavage offsets, and partial healing of fractures within the Calcite. These features indicate that deformation post-dated crystal growth. However, the deformation is interpreted conservatively as post-crystallization structural modification, rather than being used alone to infer a regional tectonic stress field, because local stress concentration around vesicle walls may also contribute to deformation.

Overall, the genetic model suggests that the investigated Calcites formed through fluid-mediated secondary mineralisation of the Deccan basalts, involving basalt-fluid interaction, mobilization of Ca, and precipitation from DIC-bearing aqueous fluids under relatively low-temperature conditions, followed by later post-crystallization deformation. The model is therefore consistent with the broader multistage mineralisation history recognised in the DVP while remaining appropriately constrained by the evidence available from the present study.

9. Conclusions

Calcite filling amygdales in Deccan basalts of the Junnar and Parner sectors grew into open voids after zeolite and silica phases, and was subsequently strained. The strain is expressed as thin straight e-twins, cleavage-controlled microfractures and healed fracture planes, all of which cut growth features and therefore post-date cavity sealing; twin morphology places the deformation below approximately 170–200 °C. This microstructural record, rather than the mineralogy, is the substantive result. It demonstrates that cavity calcite functions as a distributed, low-temperature strain marker in a flood-basalt province where the extent of post-eruptive deformation remains debated.

The compositional and spectroscopic data support phase identification and structural ordering. Given the well-known limitations of energy-dispersive analysis for light elements and the orientation dependence of Raman intensities, they do not resolve differences between the three localities, and no such claim is made. Assigning this calcite to a specific generation within the province-wide paragenesis requires carbon and oxygen isotope data and fluid-inclusion micro thermometry on a larger sample suite.

For mineral carbon sequestration, the material examined here is a completed natural experiment in basalt carbonation, preserved in the amygdaloidal flow tops that constitute the nominated reservoir intervals of the province. The absence of dissolution textures and the retention of a well-ordered carbonate structure over approximately 66 million years constitute empirical support for

the permanence of mineral trapping on a timescale no experimental programme can reach. That support is qualified in a specific and useful way. The mineralized cement was mechanically strained after emplacement, at near-ambient temperature, in a region conventionally regarded as tectonically quiescent and where geomorphic indices detect no active tectonics. Brittle failure of carbonate cement within a storage interval is therefore a realistic process in this province, and screening based on geomorphic quiescence alone will underestimate it. The healed fractures supply the counterweight, showing that the system re-cements its own damage. Long-term security of mineral trapping in the Deccan is accordingly a balance between fracture generation and carbonate self-sealing, and that balance is directly readable in thin section from any filled amygdale. Petrographic examination of natural carbonate cement deserves a place alongside seismic volumetrics and laboratory carbonation experiments in the assessment of flood-basalt storage sites.

Author Contributions

A.M.: Geological Field work, Conceptualization, Visualization, Formal analysis, Writing—original draft. S.H.: PhD scholar, Geological Field work, Sample Characterization, Data curation. G.P.: Spectral analysis. CO₂ Writing—review and editing.

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All data used in the work are presented in the paper.

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Conflict of 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.

Use of AI and AI-Assisted Technologies

No AI assisted tools were used during the preparation of this paper.

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