

Supplementary file

1. Evidences of the initial growth of natrolite include our observations of the sample's euhedral crystal morphology by naked eyes (Figure S1 A and B), under an SEM image (Figure S1 C), as well as the exterior appearance dissolved and replaced by pectolite based on polarizing microscopy (Figure S2).

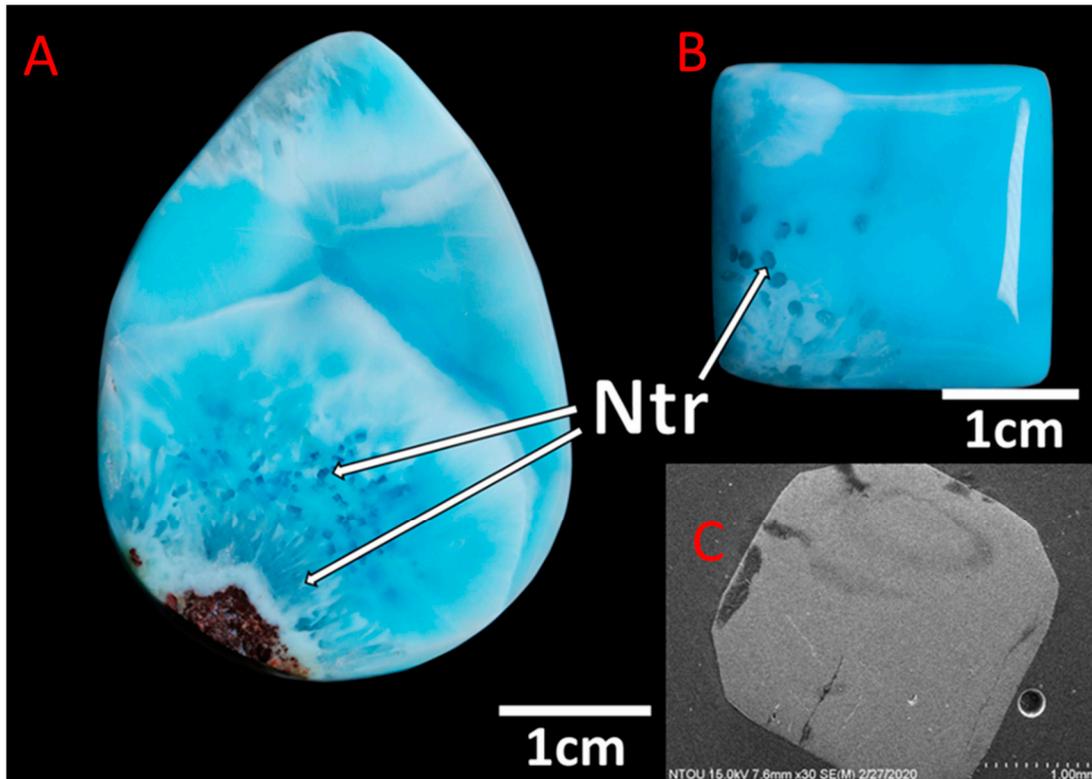


Figure S1. (A) Natrolite initiates its growth as columnar crystals along the periphery of the host rock, displaying transparent and well-formed crystal structures. Later on, blue pectolite fills the entire cavity during the later stages. (B) Hexagonal prismatic crystals of natrolite are enclosed within the interior of blue pectolite. (C) SEM image of a vertical c-axis section of natrolite.

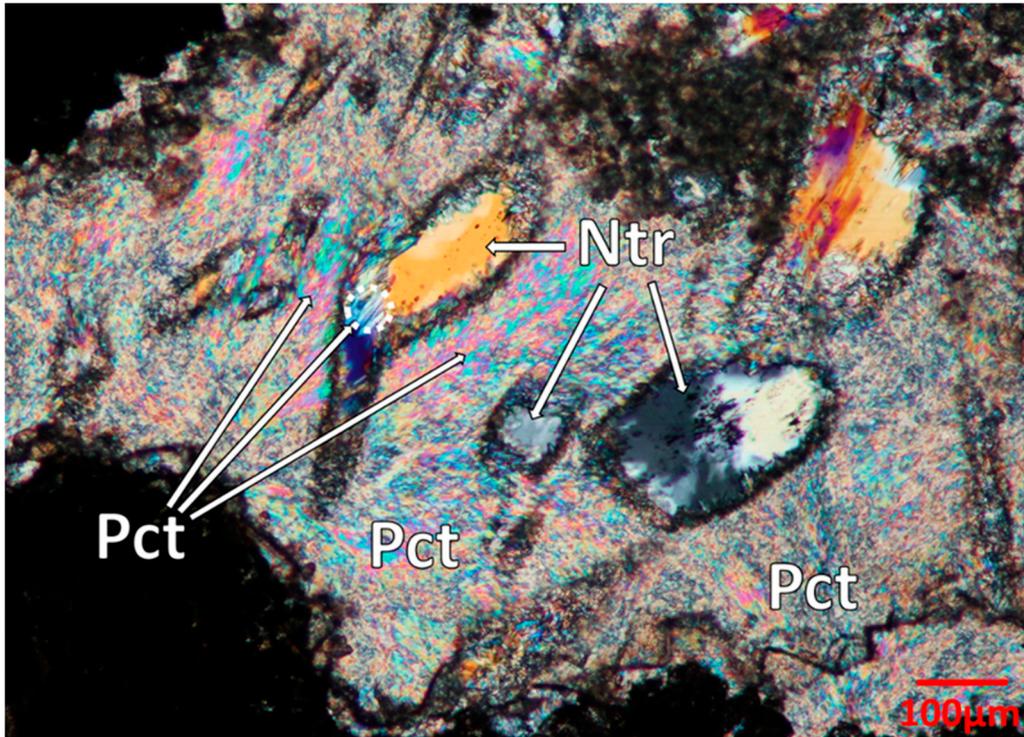


Figure S2. Portions of the natrolite crystals have undergone surface dissolution, with some sections being replaced by later-stage pectolite, preserving pseudomorphic features.

2. Raman analysis results of samples N1 to N9.

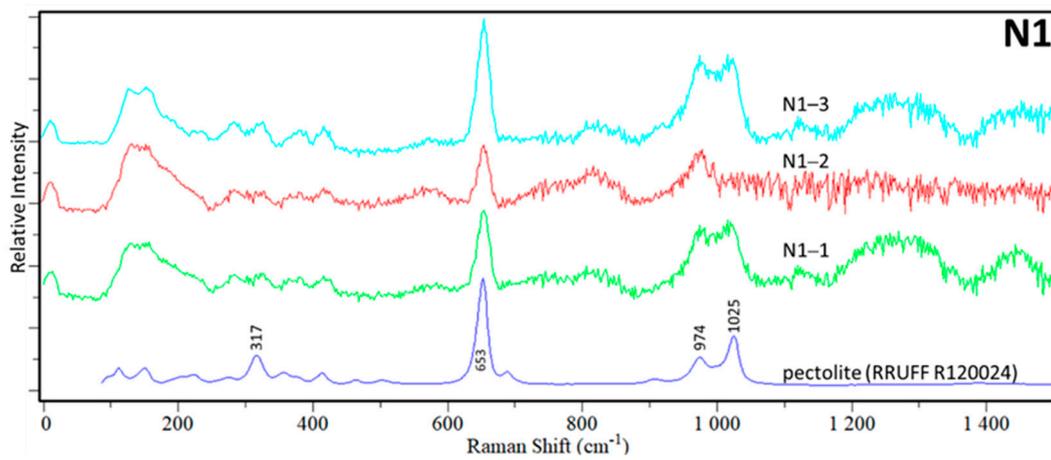


Figure S3. Raman analysis results of sample N1, but the results indicated the presence of fluorescence interference.

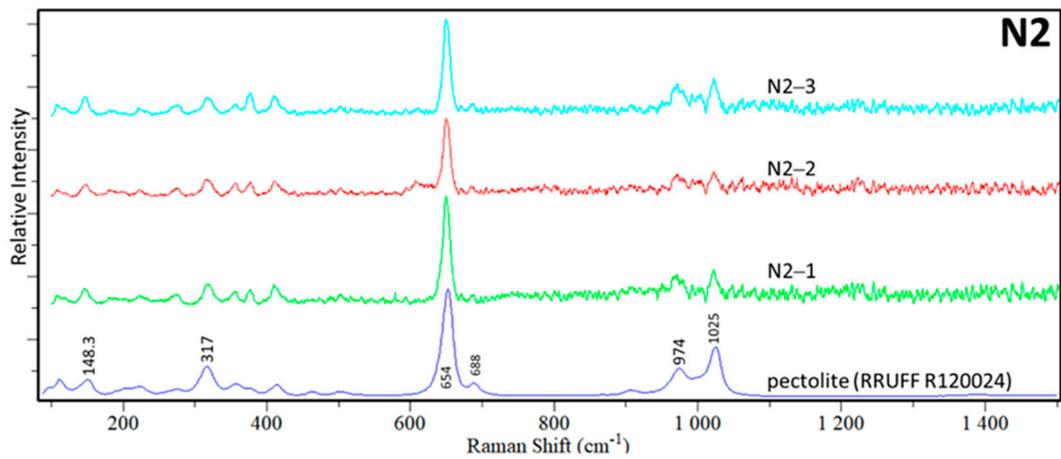


Figure S4. Raman analysis results of sample N2.

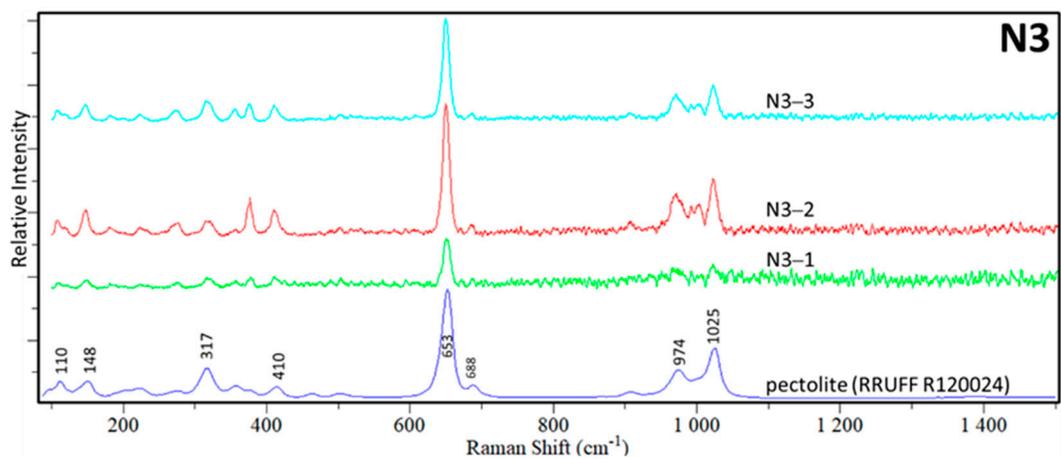


Figure S5. Raman analysis results of sample N3.

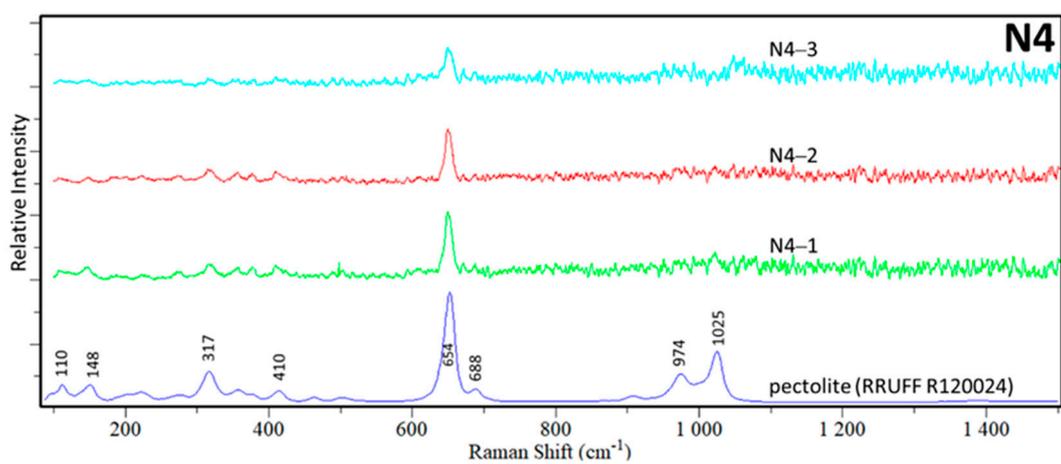


Figure S6. Raman analysis results of sample N4.

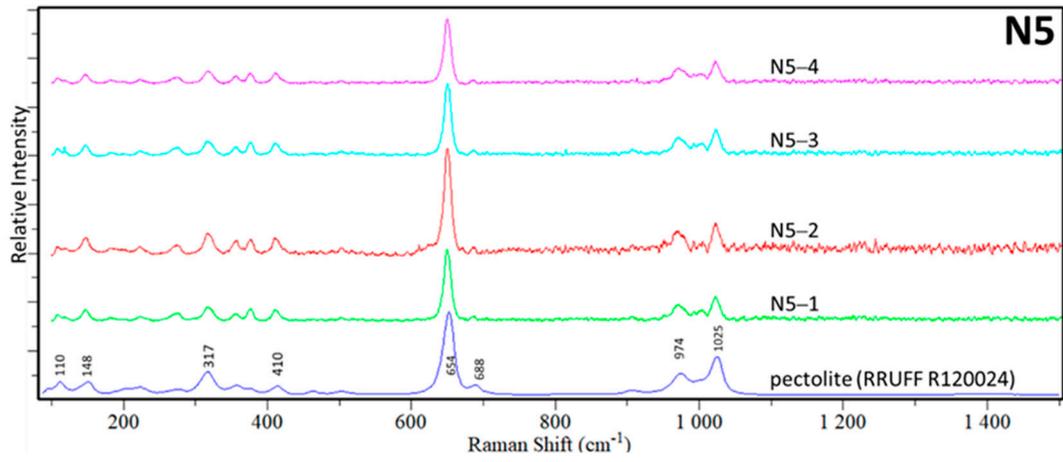


Figure S7. Raman analysis results of sample N5.

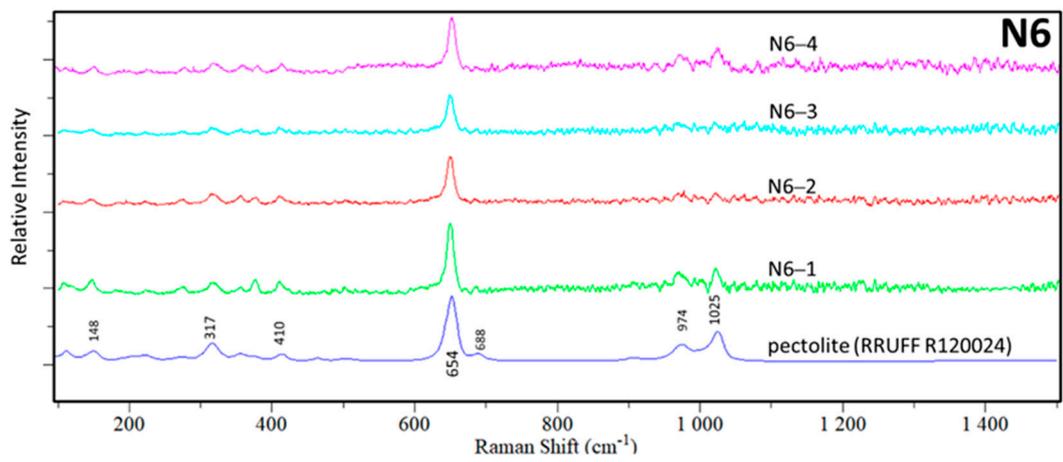


Figure S8. Raman analysis results of sample N6.

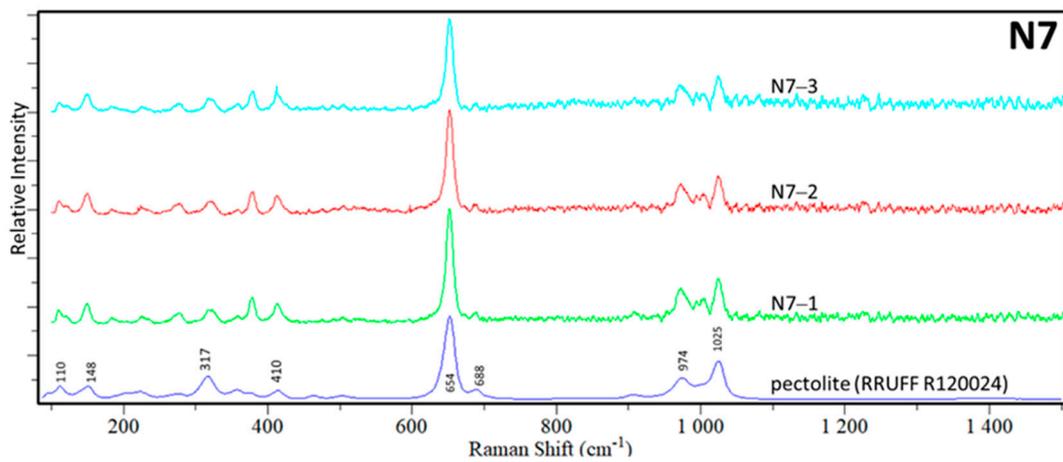


Figure S9. Raman analysis results of sample N7.

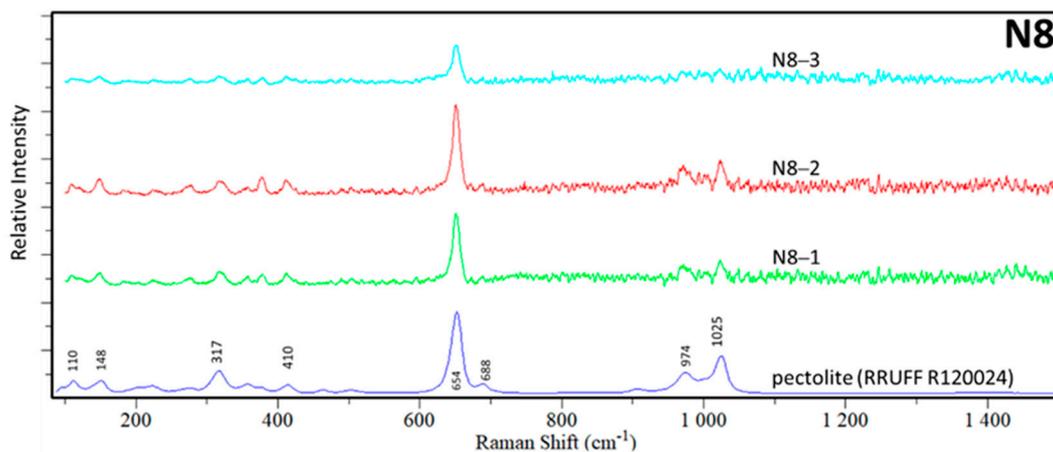


Figure S10. Raman analysis results of samples N8.

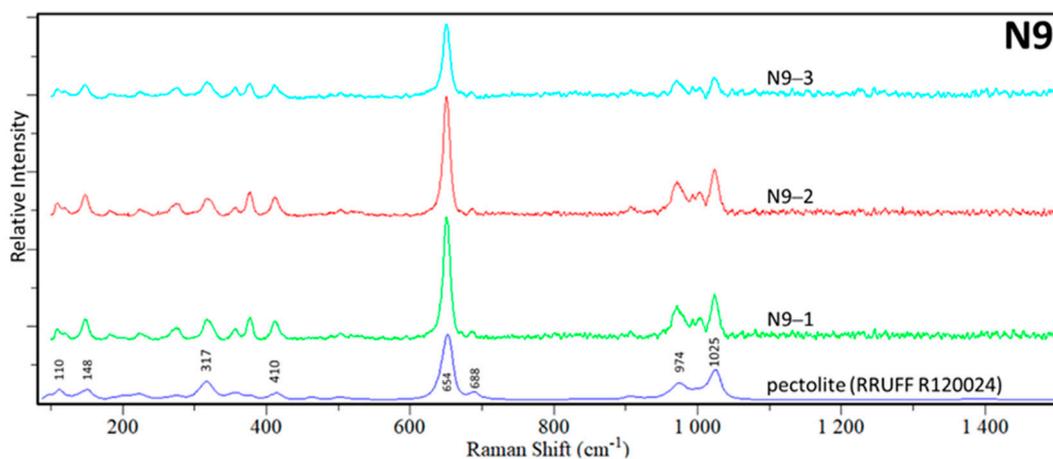


Figure S11. Raman analysis results of sample N9.

3. The UV-Vis experimental results for samples N1 to N5 were presented in terms of transmittance, where higher transmittance indicated a more pronounced color appearance. The analysis outcomes for the blue component in the mineral were depicted using blue lines, the green component with green lines, and the white component with white lines.

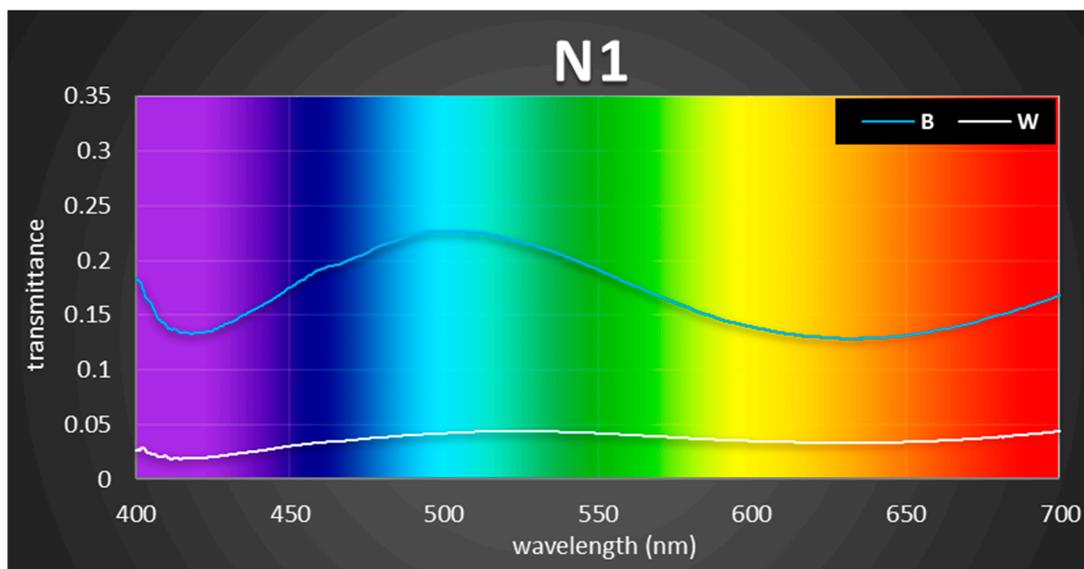


Figure S12. UV-Vis result of sample N1.

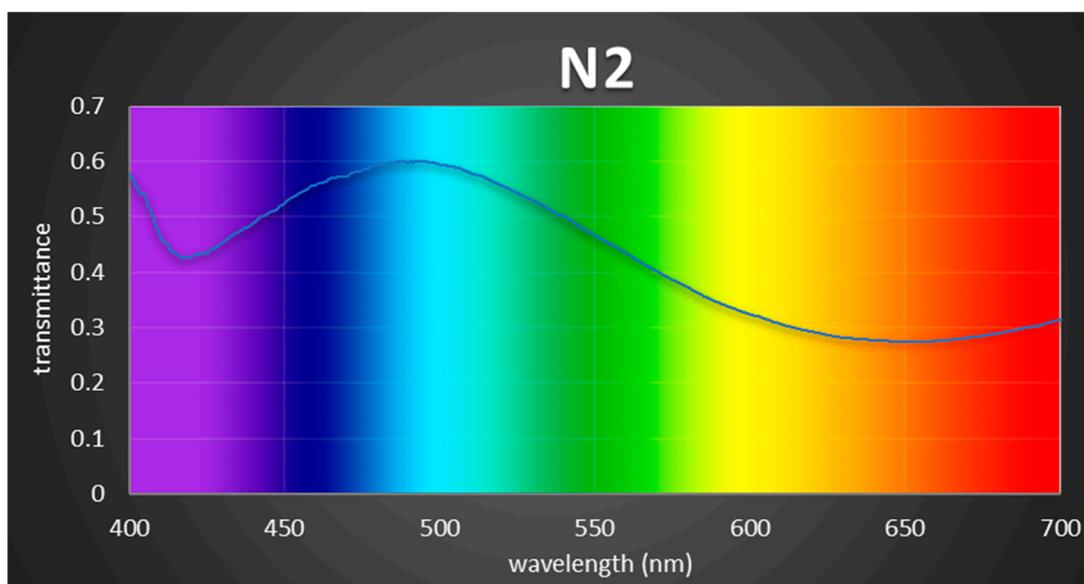


Figure S13. UV-Vis result of sample N2.

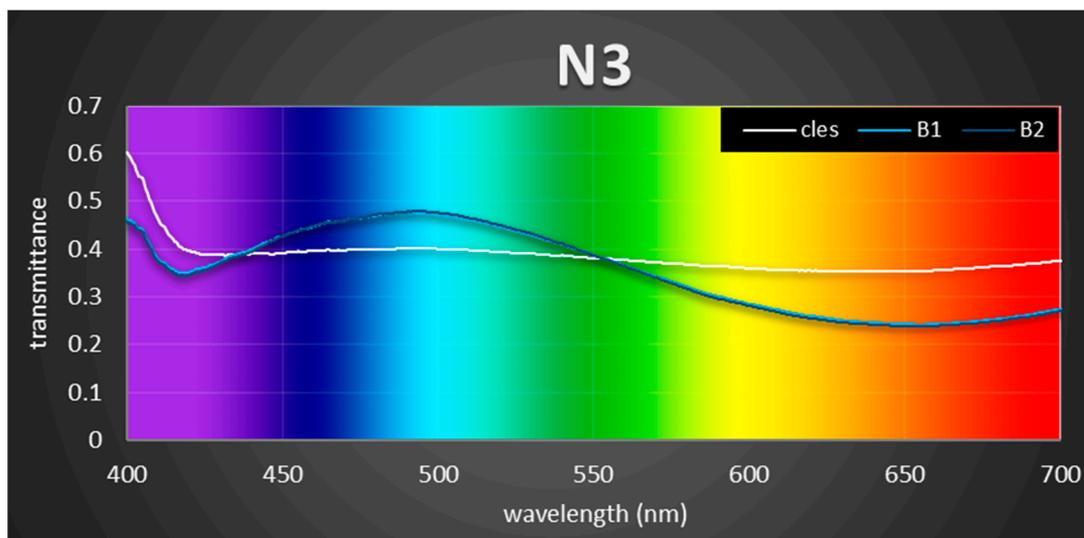


Figure S14. UV-Vis result of sample N3.

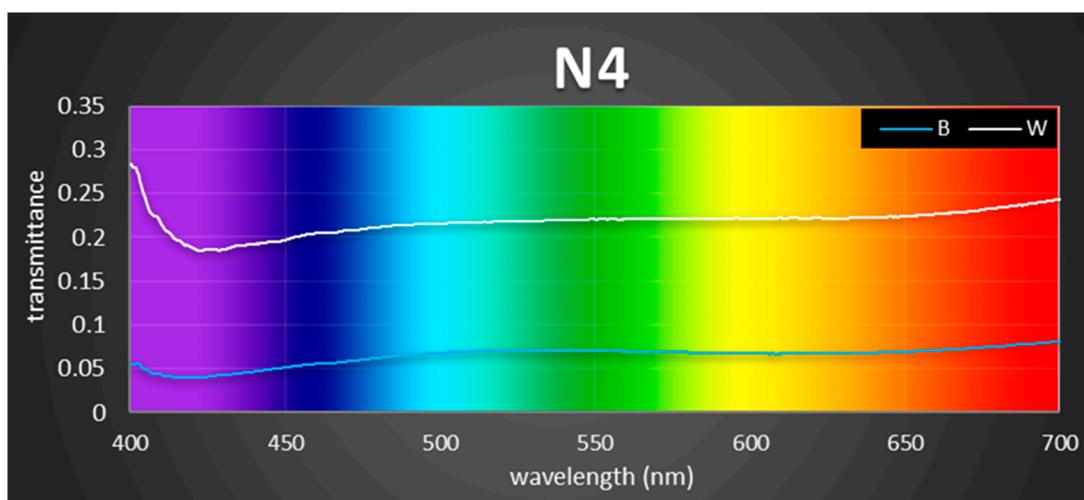


Figure S15. UV-Vis result of sample N4.

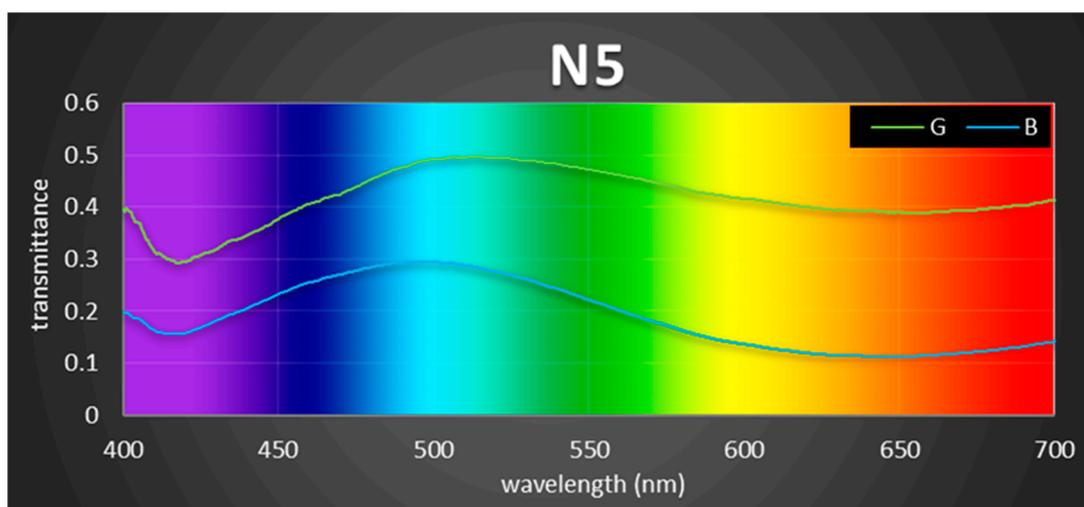


Figure S16. UV-Vis result of sample N5.

4. The heating experiments of Larimar under 250°C

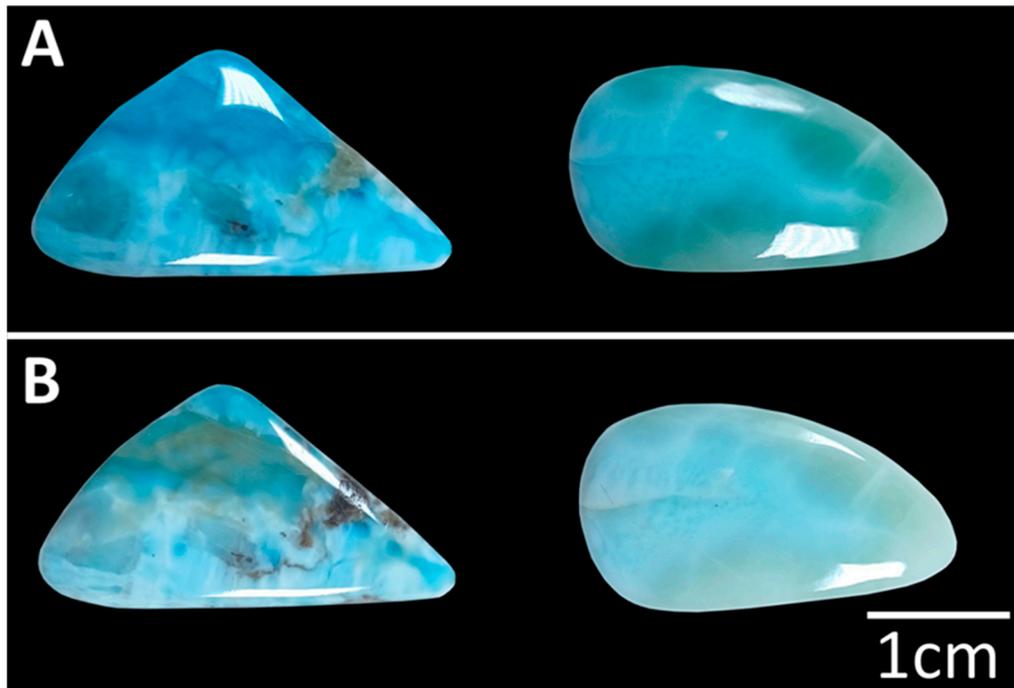


Figure S17. The results of heating Larimar to 250°C display a gradual change in color. Among these changes, the fading of the blue color is most prominent, ultimately leaving a gloomy green color. (A) the initial color of two samples at 0 hour, (B) fading of blue color and thickening of green and black color after heating 216 hours.

5. Comment minerals which coloration is due to V and Fe element.

Table S1. Mineral compositions and coloration by vanadium and iron ions.

| Mineral | Chemical composition | Color | Ion |
|-----------------------------------|--|--|------------------------------------|
| Contain V ⁴⁺ | | | |
| Cavansite | Ca(VO)Si ₄ O ₁₀ · 4H ₂ O | Greenish blue | V ⁴⁺ |
| Pentagonite | Ca(VO)Si ₄ O ₁₀ · 4H ₂ O | Greenish-blue to blue | V ⁴⁺ |
| Cloncurryite | Cu _{0.5} (VO) _{0.5} Al ₂ (PO ₄) ₂ F ₂ · 5H ₂ O | Sky-blue to colourless | V ⁴⁺ , Cu ²⁺ |
| Minasragrite | (V ⁴⁺ O)(SO ₄) · 5H ₂ O | Vivid cobalt-blue to pale aquamarine-blue | V ⁴⁺ |
| Orthominasragrite | (V ⁴⁺ O)(SO ₄) · 5H ₂ O | blue | V ⁴⁺ |
| Anorthominasragrite | (V ⁴⁺ O)(SO ₄) · 5H ₂ O | Bluish green | V ⁴⁺ |
| Phosphovanadylite-Ca | Ca[V ⁴⁺ ₄ P ₂ O ₁₂ (OH) ₄] · 12H ₂ O | bright greenish blue | V ⁴⁺ |
| Bobjonesite | (V ⁴⁺ O)(SO ₄) · 3H ₂ O | pale blue, blue-green | V ⁴⁺ |
| Davidbrowneite-(NH ₄) | (NH ₄) ₅ (V ⁴⁺ O) ₂ (C ₂ O ₄)[PO _{2.75} (OH) _{1.25}] ₄ · 3H ₂ O | Light green-blue | V ⁴⁺ |
| Karpovite | Tl ₂ VO(SO ₄) ₂ (H ₂ O) | White, light blue | V ⁴⁺ |
| Prehnite | Ca ₂ Al ₂ Si ₃ O ₁₀ (OH) ₂ | blue-gray | V ⁴⁺ |
| Zoisite var. Tanzanite | {Ca ₂ }{Al ₃ }(Si ₂ O ₇)(SiO ₄)O(OH) | blue to blue-violet | V ⁴⁺ |
| Contain V ³⁺ | | | |
| Goldmanite | Ca ₃ V ³⁺ ₂ (SiO ₄) ₃ | Green, brownish-green | V ³⁺ |
| Vanadium-bearing Tremolite | {Ca ₂ }{(Mg,Cr,V) ₅ }(Si ₈ O ₂₂)(OH) ₂ | Green | V ³⁺ , Cr ³⁺ |
| Vanadium-bearing Diopside | Ca(Mg,Cr,V)Si ₂ O ₆ | Green | V ³⁺ , Cr ³⁺ |
| Grossular var. Tsavorite | Ca ₃ (Al,Cr,V) ₂ (SiO ₄) ₃ | Green | V ³⁺ , Cr ³⁺ |
| Contain Fe ²⁺ | | | |
| Actinolite-Tremolite Series | Ca ₂ (Mg _{4.5-2.5} Fe _{0.5-2.5})Si ₈ O ₂₂ (OH) ₂ to Ca ₂ Mg ₅ (Si ₈ O ₂₂)(OH) ₂ | Colorless to dark green | Fe ²⁺ |
| Diopside-Hedenbergite Series | CaMgSi ₂ O ₆ to CaFe ²⁺ Si ₂ O ₆ | Colorless to dark green | Fe ²⁺ |
| Fayalite-Forsterite Series | Fe ²⁺ SiO ₄ to Mg ₂ SiO ₄ | Colorless to green | Fe ²⁺ |
| Ferro-hornblende | Ca ₂ (Fe ²⁺ ₄ Al)(Si ₇ Al)O ₂₂ (OH) ₂ | Dark green to black, greenish-brown, more rarely lighter green | Fe ²⁺ |
| Clinozoisite-Epidote Series | (CaCa)(AlAlAl)O[Si ₂ O ₇][SiO ₄](OH) to (CaCa)(AlAlFe ³⁺)O[Si ₂ O ₇][SiO ₄](OH) | Colourless to dark green | Fe ²⁺ |
| Ludlamite | Fe ²⁺ ₃ (PO ₄) ₂ · 4H ₂ O | Apple-green to bright green | Fe ²⁺ |
| Chamosite | (Fe ²⁺) ₅ Al(Si,Al) ₄ O ₁₀ (OH,O) ₈ | Green, grey-green, brown-green, dark | Fe ²⁺ |

| | | | |
|----------------------------------|---|--|------------------|
| Melanterite | $\text{Fe}^{2+}(\text{H}_2\text{O})_6\text{SO}_4 \cdot \text{H}_2\text{O}$ | green, black Colorless to white or green | Fe^{2+} |
| Pumpellyite-(Fe^{2+}) | $\text{Ca}_2\text{Fe}^{2+}\text{Al}_2[\text{Si}_2\text{O}_6\text{OH}][\text{SiO}_4](\text{OH})_2(\text{OH})$ | Green | Fe^{2+} |
| Julgoldite-(Fe^{2+}) | $\text{Ca}_2\text{Fe}^{2+}\text{Fe}^{3+}_2[\text{Si}_2\text{O}_6\text{OH}][\text{SiO}_4](\text{OH})_2(\text{OH})$ | Greenish black | Fe^{2+} |
| Ferro-pargasite | $\text{NaCa}_2(\text{Fe}^{2+}_4\text{Al})(\text{Si}_6\text{Al}_2)\text{O}_{22}(\text{OH})_2$ | Greenish-brown to dark green, black | Fe^{2+} |
| <hr/> | | | |
| Contain Fe^{3+} | | | |
| Andradite | $\text{Ca}_3\text{Fe}^{3+}_2(\text{SiO}_4)_3$ | Yellow, brown, brownish red, brownish yellow | Fe^{3+} |
| Cacoxenite | $\text{Fe}^{3+}_{24}\text{AlO}_6(\text{PO}_4)_{17}(\text{OH})_{12} \cdot 75\text{H}_2\text{O}$ | Yellow to brownish yellow | Fe^{3+} |
| Jarosite | $\text{KFe}^{3+}_3(\text{SO}_4)_2(\text{OH})_6$ | Amber-yellow, yellow-brown, to brown or light yellow. | Fe^{3+} |
| Amarantite | $\text{Fe}^{3+}_2(\text{SO}_4)_2\text{O} \cdot 7\text{H}_2\text{O}$ | Amaranth-red to brownish red and red-orange | Fe^{3+} |
| Yukonite | $\text{Ca}_3\text{Fe}^{3+}(\text{AsO}_4)_2(\text{OH})_3 \cdot 5\text{H}_2\text{O}$ | Red-brown, dark brown, nearly black | Fe^{3+} |
| Phosphosiderite | $\text{FePO}_4 \cdot 2\text{H}_2\text{O}$ | Purple to reddish violet, rose-red, peachy-pink, brownish yellow | Fe^{3+} |
| Arseniosiderite | $\text{Ca}_2\text{Fe}^{3+}_3(\text{AsO}_4)_3\text{O}_2 \cdot 3\text{H}_2\text{O}$ | Golden-yellow to yellow-brown, reddish-brown, brown, black | Fe^{3+} |
| Pyroaurite | $\text{Mg}_6\text{Fe}^{3+}_2(\text{OH})_{16}[\text{CO}_3] \cdot 4\text{H}_2\text{O}$ | Yellow to brownish-white, white, gray | Fe^{3+} |