

1 **Revision 2:**  
2 **Correlations between cathodoluminescence intensity**  
3 **and aluminum concentration in low-temperature**  
4 **hydrothermal quartz**

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18 **ABSTRACT:**

19 Quartz cathodoluminescence (CL) images are commonly combined with trace  
20 element concentrations to decipher complex histories of hydrothermal systems.  
21 However, the correlations between aluminum content and CL zoning of low-  
22 temperature hydrothermal quartz and their genesis remain controversial. In this

23 contribution, a multiparametric study was carried out on CL-aluminum zoning of low-  
24 temperature hydrothermal quartz (<350°C) from the Shihu and Rushan quartz-vein  
25 type Au deposits in the North China Craton. The results show that aluminum  
26 concentration correlates negatively with CL intensity in quartz from the Shihu Au  
27 deposit. CL-dark quartz zoning has significant Al concentrations as well as detectable  
28 Al-H bonds. However, in the Rushan Au deposit, the correlation is positive, and  
29 aluminum is enriched in the CL-bright quartz zoning. The Al content is positively  
30 correlated with K content with  $r^2=0.769$ . Combined with the electron backscatter  
31 diffraction (EBSD), X-ray single crystal diffraction (XRD) and transmission electron  
32 microscope (TEM) data, we infer that the genesis of CL zoning in the low-  
33 temperature hydrothermal quartz is closely related to  $\text{Al}^{3+}\text{-H}^+$  and  $\text{Al}^{3+}\text{-K}^+$   
34 concentrations. The  $\text{Al}^{3+}\text{-K}^+$  may act as the CL-activator, while the  $\text{Al}^{3+}\text{-H}^+$  may act  
35 as the CL-dampener. Where  $\text{Al}^{3+}\text{-Si}^{4+}$  substitution is charge balanced by hydrogen,  
36 the intensity of CL response decreases; where  $\text{Al}^{3+}\text{-Si}^{4+}$  substitution is charge  
37 balanced by potassium, the intensity of CL response increases. The correlations  
38 between CL intensity and aluminum concentration in the low-temperature  
39 hydrothermal quartz reflect pH fluctuations of hydrothermal system.

40 **KEY WORDS:** Shihu and Rushan Au deposits; Low-temperature hydrothermal  
41 quartz; Cathodoluminescence;  $\text{Al}^{3+}\text{-H}^+$  and  $\text{Al}^{3+}\text{-K}^+$  concentrations; pH

## 42 **1. Introduction**

43 Cathodoluminescence (CL) images can effectively visualize the micro-textures  
44 that record complex history of quartz precipitation and dissolution ([Rusk and Reed  
45 2002](#); [Götze et al. 2001, 2005](#); [Olivia et al. 2020](#)). Concentrations of trace elements  
46 reflect the growth process and physio-chemical conditions during quartz growth such  
47 as pressure, temperature and pH ([Wark and Watson 2006](#); [Thomas et al. 2010](#); [Acosta  
48 et al. 2020](#)). Therefore, CL images combined with trace element concentrations in

49 quartz are extensively used to obtain insight into quartz vein formation processes,  
50 especially in magmatic systems ([Watt et al. 1997](#); [Penniston-Dorland 2001](#); [Monecke](#)  
51 [et al. 2002](#); [Redmond et al. 2004](#)). [Müller et al. \(2005\)](#) used CL zones of quartz to  
52 reveal multiple magma mixing process in the eastern Erzgebirge volcano-plutonic  
53 complex (Germany, Czech Republic). [Allan and Yardley \(2007\)](#) used scanned CL and  
54 trace elements of quartz to track the meteoric infiltration into a magmatic-  
55 hydrothermal system in the Mt. Leyshon, porphyry deposit, Australia. [Takashi et al.](#)  
56 [\(2020\)](#) evaluated the cooling processes of the Toki granitic pluton by using (CL)  
57 oscillatory-zoned quartz. Such studies have helped to better understand the magmatic  
58 processes, origin of granites and genesis of porphyry deposit ([Watt et al. 1997](#); [Müller](#)  
59 [et al. 2005](#); [Takashi et al. 2020](#)). However, there is lack of the application on the low-  
60 temperature hydrothermal deposits due to the controversy on the genesis of CL zoning.

61 Several studies have explored the coupling relationships between CL intensity  
62 and trace element concentrations in quartz crystals ([Müller et al. 2000, 2002](#);  
63 [Penniston-Dorland 2001](#); [Rusk et al., 2011](#)). The formation process of CL-trace  
64 element zoning in high-temperature quartz (>350°C; [Rusk et al. 2008](#)) shows that CL  
65 intensity variations in the high-temperature quartz are positively correlated with the  
66 trace element concentrations, such as Ti, Al, K, Na, H, Li and P ([Götze et al. 2004](#);  
67 [Landtwing and Pettke 2005](#); [Wang et al. 2022](#)). The genesis of CL zoning of the low-  
68 temperature hydrothermal quartz (<350°C; [Rusk et al. 2008](#); [Qiu et al. 2021](#)) is  
69 different from that of the high-temperature quartz and demonstrably more complex.  
70 Correlations between CL intensity and trace elements in the low-temperature  
71 hydrothermal quartz are varied. For example, in most of low-temperature  
72 hydrothermal quartz (such as quartz in the Magmont, Comstock Lode and Butte Main  
73 Stage deposits; [Rusk et al. 2008](#)), aluminum concentration correlates positively with  
74 CL intensity, but in some low-temperature hydrothermal quartz (such as quartz in the  
75 Red Dog and Jerritt Canyon deposits; [Rusk et al. 2008](#)), aluminum concentrations and  
76 CL intensities are inversely correlated. These variations in correlation greatly affects  
77 our understanding of the genesis and implications of low-temperature hydrothermal

78 quartz and raises several questions such as which factors control the correlations  
79 between CL intensity and aluminum concentration in the low-temperature  
80 hydrothermal quartz, and what are the geological processes. In this contribution, it is  
81 necessary to carry out a detailed mineralogical study on the genesis of low-  
82 temperature hydrothermal quartz cathodeluminescence zoning to reveal the  
83 correlation between CL intensity and trace element concentration in the low-  
84 temperature hydrothermal quartz and its geological implications.

85 In the present study, we have integrated scanning electron microscope  
86 cathodoluminescence (SEM-CL), electron microprobe (EMP), electron backscatter  
87 diffraction (EBSD), X-ray single crystal diffraction (XRD), Fourier transform infrared  
88 spectroscopy (FTIR) and transmission electron microscope (TEM) data of quartz from  
89 the Shihu and Rushan Au deposits in the North China Craton (NCC) to evaluate the  
90 genesis of CL-aluminum zoning in low-temperature hydrothermal quartz and the  
91 implications for the evolution of hydrothermal system. Our results provide a new  
92 understanding of the genetic relation between the CL-aluminum zoning of low-  
93 temperature hydrothermal quartz and hydrothermal pH.

## 94 **2. Sampling and analytical methodology**

95 The Rushan Au deposit, hosted by the Mesozoic granitoids in the middle part of  
96 the Muping-Rushan gold belt in the Jiaodong Peninsula of eastern North China Craton  
97 (NCC), is currently the largest lode gold deposit in terms of a single vein Au resource  
98 (>30t) in China (Fig. 1a, b; Goldfarb and Santosh 2014; Li and Santosh 2017; Deng et  
99 al. 2017, 2020a, 2020b; Qiu et al. 2020a, 2020b; Li et al. 2022). The Shihu Au deposit,  
100 located in the Fuping district of the northern Taihang Mountain, is the largest gold  
101 deposit (>40t) in the central North China Craton (Fig. 1a, d; Li et al. 2013; Zeng et al.  
102 2019). Both deposits show similar mechanisms of ore genesis (Hou et al. 2017; Deng  
103 et al. 2020c; Li et al. 2020; Yu et al. 2022). The auriferous quartz-sulfide veins from  
104 both deposits contain numerous coarse-grained quartz crystals with regular zoning  
105 structure (Li et al. 1994; Zeng 2019; Feng et al. 2022). The zoned quartz is thought to

106 have formed in the early ore-forming stage of mineralization with homogenous  
107 temperatures of 260-310°C (Rushan; pyrite-quartz stage; [Cao 2013](#); [Sai et al. 2020](#))  
108 and 268-331°C (Shihu; pyrite-quartz stage; [Zeng 2019](#)) based on fluid inclusion data,  
109 which are consistent with the low-temperature quartz defined by [Rusk et al. \(2008,](#)  
110 [2011\)](#) (<350°C).

111 Fifteen ore samples containing zoned quartz from the elevation 635 m of No. II  
112 orebody in the Rushan Au deposit ([Fig. 1c](#)) and thirteen ore samples from the  
113 elevation 300 m of No. 101 orebody in the Shihu Au deposit ([Fig. 1e](#)) were collected  
114 for this study. All the zoned quartz samples were polished to make thin sections and  
115 prepared for SEM-CL, EMP and EBSD analysis. Hundreds of SEM-CL images were  
116 taken and merged into a complete picture to reveal quartz CL zoning and to carry out  
117 EMP and EBSD mapping ([Fig. 3, 4](#); [Supplementary material](#)). Small quartz grains  
118 (diameter ~40µm) of different CL zoning were picked out from the thin sections for  
119 XRD and FTIR analyses (analyzed locations are shown in [Fig. 4](#)). Two quartz ultra-  
120 thin sections from the aluminum-rich bands were prepared for TEM analysis  
121 (analyzed locations are shown in [Fig. 5](#)).

122 The scanning electron microscope cathodoluminescence (SEM-CL) images were  
123 acquired at the State Key Laboratory of Biogeology and Environmental Geology,  
124 China University of Geosciences (Beijing) using a Zeiss Supra 55 field emission  
125 scanning electron microscope (FESEM) equipped with a Gatan ChromaCL2  
126 cathodoluminescence (CL) detector. Platinum-coated, polished sections were  
127 analyzed at 10 kV with beam current from 1 to 40 nA. The SEM-EBSD analysis was  
128 carried out by a Zeiss Sigma scanning electron microscope (SEM) coupled with a  
129 HKL Nordlys Nano electron backscattered diffraction (EBSD) detector at the Institute  
130 of Geology, China Earthquake Administration, China ([Yang et al. 2019](#)). Crystal  
131 orientation images covering the quartz grains were obtained with a step size of 1 µm  
132 (Shihu) and 5 µm (Rushan) using the Oxford Instruments HKL AZtec software. A  
133 working distance of 18.4 mm was used for pattern acquisition and automatic indexing  
134 was performed using the AZtec software. The CHANNEL 5 software was used for  
135 noise reduction and for filling the missing data with at least 8 identical neighbors with

136 similar orientation. The resulting EBSD data was optimized using MTEX to remove  
137 points with large angular deviation and to smooth intra-crystalline data (Bachmann et  
138 al. 2010; Henry et al. 2017).

139 The major and trace element compositions of the hydrothermal quartz were  
140 analyzed by a JXA-8230 electron microprobe (EMP) (Beijing GeoAnalysis Co., Ltd.).  
141 Operating conditions included 20kV accelerating voltage, the beam diameter of 5 $\mu$ m  
142 and a beam current of 20nA. Standard substances for analysis refer to GB/T 15,074-  
143 2008 general rules. The EPMA X-ray elemental maps were determined at the Beijing  
144 GeoAnalysis Co., Ltd., using a 4 WDS detector-equipped Camaca SX100 electron  
145 microprobe. Operating conditions were 250-300ms dwell time per spot, 80nA beam  
146 current and 20kV acceleration. Aluminum, Ti, Si, K, Na, Ga and Ge were analyzed by  
147 wavelength dispersive spectroscopy (WDS). Elemental maps are up to 256 $\times$ 256  
148 pixels in size, with 1-2 $\mu$ m steps (distance between analysis spots, or spatial definition).  
149 All EMP spot analyzed results and analyzed locations are listed in the Supplementary  
150 material Table. S1 and attached figures.

151 X-ray single crystal diffraction (XRD) analyses were carried out at the State Key  
152 Laboratory of Geological Processes and Mineral Resources, China University of  
153 Geosciences (Beijing), China using a Rigaku Oxford XtaLAB PRO-007HF  
154 microfocus rotating anode X-ray source (1.2kW, MoK $\alpha$ ,  $\lambda=0.71073\text{\AA}$ ) and a hybrid  
155 pixel array detector single-crystal diffractometer. The X-ray powder diffraction data  
156 were recorded with a diffractometer using MoK $\alpha$  radiation. The scanning speed and  
157 range of each sample are listed in the Supplementary material Table. S2. The cell  
158 volume and  $a_0$  values of quartz are obtained from the CIF documents using Mercury  
159 software, which are listed in the Supplementary material Table. S3.

160 Fourier transform infrared spectroscopy (FTIR) analyses were carried out at the  
161 State Key Laboratory of Geological Processes and Mineral Resources, China  
162 University of Geosciences (Beijing), China, using a TENSOR 37 FTIR equipped with  
163 a Rocksol1D<sup>TM</sup> interferometer and Digi tect<sup>TM</sup> detector, with working voltage of 12  
164 kV, a current of 10 A. The detected spectral range is from 7800cm<sup>-1</sup> to 370cm<sup>-1</sup>. The

165 wavenumber accuracy is better than  $0.01\text{ cm}^{-1}$ . Standard resolution is  $0.6\text{cm}^{-1}$  with an  
166 option for  $0.3\text{cm}^{-1}$ . Small quartz grains (diameter  $\sim 50\mu\text{m}$ ) picked out from aluminum-  
167 rich bands in the thin sections (as shown in Fig. 4) were prepared for FTIR analyses.  
168 These grains were ground to a 200-mesh powder in an agate mortar, and then dried to  
169 eliminate the influence of inclusions in quartz on the test results. One mg quartz  
170 sample was thoroughly mixed with 200 mg KBr (200 mesh, prebaked in an infrared  
171 oven for 24 hours) and a hydraulic press was used to make 0.03mm thin section for  
172 FTIR analysis. The FTIR data were processed by Origin 2017 software.

173 The transmission electron microscope (TEM) and high-resolution transmission  
174 electron microscopy (HRTEM) analyses were carried out by a FEI Tecnai F20  
175 transmission electron microscope (TEM) in the Shiyanjia Lab ([www.shiyanjia.com](http://www.shiyanjia.com)).  
176 A ZEISS Crossbeam 540 (Focused Ion beam-SEM) was used to pick out ultra-thin  
177 sections from the aluminum-rich bands of quartz from the Rushan and Shihu Au  
178 deposits for TEM and HRTEM analyses.

### 179 **3. Results and Discussion**

#### 180 **3.1 Correlations between aluminum and CL intensity**

181 Correlations between CL intensity and aluminum concentration of quartz from  
182 the Shihu and Rushan Au deposits are distinctly different. In the Shihu Au deposit,  
183 aluminum concentration and CL intensities of quartz are negatively correlated,  
184 whereas in the Rushan Au deposit, aluminum concentration correlates positively with  
185 the CL intensity. The EMP spot analyses show that in the zoned quartz from Shihu Au  
186 deposit, aluminum concentrations in the CL-bright zone are mostly lower than that of  
187 the CL-dark zone ( $>0.4\text{ wt}\%$ ) (Fig. 3b), and the aluminum concentration shows lack  
188 of any correlation with potassium content with  $r^2=0.014$  (Fig. 2a). However, in the  
189 zoned quartz from Rushan Au deposit, the CL-bright zone mainly shows higher  
190 aluminum concentration ( $>0.2\text{ wt}\%$ ) than that of the CL-dark zone ( $<0.1\text{ wt}\%$ ), and  
191 the aluminum concentration positively correlates with the potassium concentration

192 with  $r^2=0.769$  (Fig. 2b, 3b).

193 The correlations between aluminum concentration and CL intensity of quartz  
194 from the Shihu and Rushan Au deposits are well demonstrated in the EMP element  
195 maps. A good correspondence between CL-dark band and aluminum-rich band in the  
196 quartz from the Shihu Au deposit is presented (Fig. 3b, 4). Conversely, in the quartz  
197 from the Rushan Au deposit, the aluminum-rich band well corresponds to the CL-  
198 bright band (Fig. 3b, 4). It can be seen that the intensity of CL decreases with  
199 increasing aluminum content in the quartz of the Shihu Au deposit, while that of the  
200 Rushan Au deposit is on the contrary. The distribution of potassium in quartz from the  
201 Rushan Au deposit shows obvious zonal structure, which has good correspondence  
202 with the CL zoning and aluminum zoning (Fig. 4). However, in the Shihu Au deposit,  
203 no potassium zoning was detected in quartz. Instead, considerable Al-H bonds are  
204 detected in the aluminum-rich quartz zoning (CL-dark quartz zoning), which is  
205 obviously more enriched than that of quartz in the Rushan Au deposit (Fig. 4).

### 206 3.2 Crystallographic properties of different quartz CL zoning

207 Previous studies have demonstrated that  $Al^{3+}$  substitutes  $Si^{4+}$  in the quartz  
208 structure with charge compensation by additional ions (such as  $H^+$ ,  $Li^+$ ,  $Na^+$  and  $K^+$ )  
209 in interstitial sites related to structural channels (Götze et al. 2001; Larsen et al. 2004;  
210 Landtwing and Pettke 2005). The correlations between CL intensity vs. Al  
211 concentration and Al vs. K, as well as FTIR characteristics of quartz suggest the  
212 substitution of  $Al^{3+}$ ,  $K^+$  and  $H^+$  for  $Si^{4+}$  in the quartz structure of the Shihu and Rushan  
213 Au deposits (Fig. 2, 3b, 4; Landtwing and Pettke 2005; Naoya et al. 2005; Rusk et al.  
214 2008). Due to the larger ionic radius of  $K^+$  than that of  $H^+$  and  $Li^+$ , the effect of  $K^+$   
215 incorporation on lattice structure (cell volume and  $a_0$ ) of quartz should be much  
216 greater than that of  $H^+$  and  $Li^+$  incorporation (Allan and Yardley 2007). The X-ray  
217 single crystal diffraction analyses shows the variations of crystal cell parameters  
218 between CL-dark zone and CL-bright zone in the Shihu and Rushan Au deposits (Fig.  
219 4). In the Shihu Au deposit, the cell volume and  $a_0$  values of the CL-bright zone and



220 CL-dark zone (Al and H rich band) are almost same, which are within  $112 \text{ \AA}^3$  to  $112.5$   
221  $\text{ \AA}^3$  and  $4.9 \text{ \AA}$  to  $4.91 \text{ \AA}$  (Fig. 4). However, in the quartz from the Rushan Au deposit,  
222 the cell volume and  $a_0$  values of the CL-bright zone are obviously larger than those of  
223 the CL-dark zone (Fig. 4). The  $a_0$  value increases from  $4.89 \text{ \AA}$  in the CL-dark zone to  
224  $4.95 \text{ \AA}$  in the CL-bright zone (Al- and K-rich quartz zone). Meanwhile, the cell  
225 volume increases from  $112.1 \text{ \AA}^3$  in the CL-dark zoning to  $115.4 \text{ \AA}^3$  in the CL-bright  
226 zoning (Fig. 4). Comparing the samples from two deposits, the cell volume and  $a_0$   
227 values of the Al-rich quartz zone from Rushan Au deposit are obviously larger than  
228 those of Shihu Au deposit. These results confirm that  $(\text{Al}^{3+}\text{-K}^+)\text{-Si}^{4+}$  substitution in  
229 the quartz from Rushan Au deposit shows more obvious lattice distortion than that of  
230  $(\text{Al}^{3+}\text{-H}^+)\text{-Si}^{4+}$  substitution in quartz from the Shihu Au deposit. Systematically, these  
231 distortions of lattice structure may cause significant crystal deformation in quartz  
232 from the Rushan Au deposit (Passchier and Trouw 2005). In this contribution, two  
233 quartz grains containing both CL-dark and CL-bright zones from the Shihu and  
234 Rushan Au deposits were selected to carry out EBSD analysis to visualize the  
235 deformation distribution (Fig. 3a, b). The band contrast (BC) images show that all  
236 captured pattern images of quartz have high image quality (Fig. 3c). Thus, the results  
237 of deformation distribution of analyzed quartz are reliable. The inverse pole figure  
238 (IPF) colouring images show that the quartz from the Shihu Au preferred crystal  
239 orientation of “a” faceprojected along the Y direction, while the quartz from the  
240 Rushan Au deposit has two crystal orientations, termed as “r” and “m” faces (Fig. 3d),  
241 indicating different degrees of deformation of the quartz crystals from two deposits  
242 (Ghosh et al. 2017). The uniform distribution of kernel average misorientation (KAM)  
243 and grain reference orientation deviation (GROD) angle in the quartz from the Shihu  
244 Au deposit indicate that the crystal is almost undeformed and its crystal structure may  
245 not have changed (Fig. 3e, f; Ghosh et al. 2017; Kleber et al. 2021). However, in the  
246 Rushan Au deposit, the CL-bright band of quartz obviously presents deformation  
247 characteristics, seen in different colors in the KAM and GROD angle maps (Fig. 3e, f),  
248 and suggesting that CL-bright quartz from the Rushan Au deposit may have deformed  
249 (Ghosh et al. 2017; Sai et al. 2020; Kleber et al. 2021). The transmission electron

250 microscopy (TEM) results confirm that both bright and dark zones in quartz from the  
251 Shihu Au deposit display regular lattice stripes and no obvious lattice defects are  
252 found (Fig. 5a, b). However, in the Rushan Au deposit, the aluminum-rich CL-bright  
253 quartz zoning obviously shows considerable lattice defects (Fig. 5c, d), which might  
254 be related to the  $\text{Al}^{3+}\text{-K}^+$  substitution for  $\text{Si}^{4+}$  (Fig. 2b, 5e, f) (Hochella et al. 2008; Yu  
255 et al. 2021). Combing the correlations between Al concentration vs. CL intensity and  
256 Al vs. K, as well as FTIR, XRD, EBSD, TEM analysis results, we consider that the  
257 genesis of CL zoning in the low-temperature hydrothermal quartz is closely related to  
258  $(\text{Al}^{3+}\text{-K}^+)\text{-Si}^{4+}$  and  $(\text{Al}^{3+}\text{-H}^+)\text{-Si}^{4+}$  substitutions in quartz lattice. The negative  
259 correlation between CL intensity and aluminum concentration in quartz of the Shihu  
260 Au deposit are closely related to the  $\text{Al}^{3+}\text{-H}^+$  substitution for  $\text{Si}^{4+}$  (Fig. 5g), while the  
261 positive correlation in quartz of the Rushan Au deposit is more likely to depend on the  
262  $\text{Al}^{3+}\text{-K}^+$  substitution for  $\text{Si}^{4+}$  (Fig. 5h). The  $\text{Al}^{3+}\text{-K}^+$  may act as the CL-activator, while  
263 the  $\text{Al}^{3+}\text{-H}^+$  may act as the CL-dampener. Where aluminum substitution is charge  
264 balanced by hydrogen, the intensity of CL response decreases; where aluminum  
265 substitution is charge balanced by potassium, the intensity of CL response increases.

#### 266 4. Forming conditions of CL-aluminum zoning

267 Variability in quartz CL intensity as observed relates to trace-element  
268 concentrations that could result from changes in fluid chemistry, pressure and  
269 temperature during quartz crystallization (Landtwing and Pettke 2005; Naoya et al.  
270 2005; Wang et al. 2021). The quartz CL zoning reported in this study is thought to  
271 have formed in a narrow range of formation temperature with homogenous  
272 temperatures of 260-310°C (Rushan; Li et al. 1992; Li et al. 1994; Cao 2013; Sai and  
273 Qiu 2020) and 268-331°C (Shihu; Ao 2009; Zeng 2019). Different quartz CL zoning  
274 forms within similar temperature ranges (Cao 2013; Zeng 2019). Sharp contrasts in  
275 aluminum concentration from zone to zone in single quartz grain, where no evidence  
276 of temperature change exists, suggest that CL-aluminum zoning in quartz does not  
277 develop due to temperature fluctuations (Rusk et al. 2008). Pressure fluctuation has a

278 strong influence on quartz solubility and thus the reaction affinity of quartz  
279 precipitation. [Hu et al. \(2005\)](#) pointed out that the mineralization pressure of Rushan  
280 Au deposit fluctuated from 92 MPa to 269 MPa ([Hu et al., 2005](#)). [Cao \(2013\)](#)  
281 indicated that the mineralization pressure of Shihu Au deposit ranges from 55MPa to  
282 95MPa ([Cao, 2013](#)). The wide ranges of mineralization pressure in the two deposits  
283 reflect the process of fluid pulsation during the formation of quartz veins, which  
284 might result in the formation of quartz zones related to the frequency of fluid  
285 pulsation ([Li et al., 1994](#); [Cao, 2013](#)). On these basics, the fluctuation of  
286 mineralization pressure seems to have a certain contribution to the formation of quartz  
287 CL zoning. However, this conclusion cannot be drawn from the experimental data  
288 presented in this paper unless detailed fluid inclusion studies are performed on  
289 different quartz CL zoning.

290 Previous studies noted that although aluminum concentration in hydrothermal  
291 quartz is incapable of reflecting the temperature of quartz precipitation, it is a good  
292 indicator of aluminum solubility, which is strongly dependent on pH ([Rusk et al.](#)  
293 [2008](#)). Generally, with decreasing pH, aluminum solubility increases ([Luo et al. 2001](#)).  
294 Thus, both the CL-dark (Al-rich) quartz in the Shihu Au deposit and CL-bright (Al-  
295 rich) quartz in the Rushan Au deposit are considered to have formed under an acidic  
296 environment. The higher aluminum concentration in CL-bright quartz in the Shihu Au  
297 deposit ([Fig. 2 and Fig. 4e, f](#)) suggests that the crystallization conditions for CL-dark  
298 quartz from the Shihu deposit were more acidic than those of CL-bright quartz from  
299 the Rushan Au deposit ([Luo et al. 2001](#); [Rusk et al. 2008](#)), which is also confirmed by  
300 considerable Al-H bonds detected in the quartz zoning. The widespread distribution of  
301 K-feldspar alteration zones in the Rushan Au deposit ([Li et al. 2013, 2015](#); [Deng et al.](#)  
302 [2020c](#)) indicates that potassium in the ore-forming hydrothermal fluids was consumed  
303 in large quantities. Potassium concentration in the solution might be mostly affected  
304 by the solubility of potassium feldspar, which is strongly dependent on pH ([Luo et al.](#)  
305 [2001](#)). Generally, with the decrease of pH value, potassium feldspar solubility  
306 increases significantly ([Luo et al. 2001](#)). However, in strong acidic environment,  $H^+$   
307 may replace  $K^+$  to compensate the charge of  $Al^{3+}$ - $Si^{4+}$  substitution ([Landtwing and](#)

308 [Pettke 2005](#); [Naoya et al. 2005](#)). Zhu (1991) obtained the fluid pH values of different  
309 mineralization stages in the Rushan Au deposit (4.50 ~ 5.76). [Shen et al. \(2000\)](#)  
310 indicated that pH of ore-forming fluid of the Rushan Au deposit ranged from 5 to 6.  
311 Combining with our study, we consider that the formation of CL-bright quartz from the  
312 Rushan Au deposit is more likely related to an intermediate-acidic environment.  
313 Therefore, aluminum-rich quartz zoning in the Shihu and Rushan Au deposits were  
314 formed in acidic and intermediate-acidic environments, respectively, which are  
315 obviously different from the neutral environment of aluminum deficient quartz zoning.  
316 The formation of CL-aluminum zoning in quartz from the Shihu and Rushan Au  
317 deposits might have been caused by pH fluctuation of the hydrothermal fluid.

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506  
507  
508 **Figure captions**

- 509 Fig. 1 Geological map of the Muping-Rushan gold belt **(a)** ([Deng et al. 2020a](#)) and the  
510 Fuping district of the northern Taihang Mountain **(c)**; Cross-section of the  
511 orebody No. II in the Rushan gold deposit **(b)** ([Li et al. 2015](#)), and No. 101 in the

512 Shihu gold deposit **(d)** ([Li and Santosh 2017](#); [Li et al. 2013](#))

513 Fig. 2 Correlations between Al and K in quartz from Shihu **(a)** and Rushan **(b)** Au  
514 deposits

515 Fig. 3 Scanning electron microscope-cathodoluminescence (SEM-CL) images **(a)**,  
516 electron probe micro-analysis (EPMA) trace element maps **(b)**, and electron  
517 backscatter diffraction (EBSD) maps of quartz from the Shihu and Rushan Au  
518 deposit, North China Craton **(c-f)**. IPF: inverse pole figure; KAM: kernel average  
519 misorientation; GROD: grain reference orientation deviation

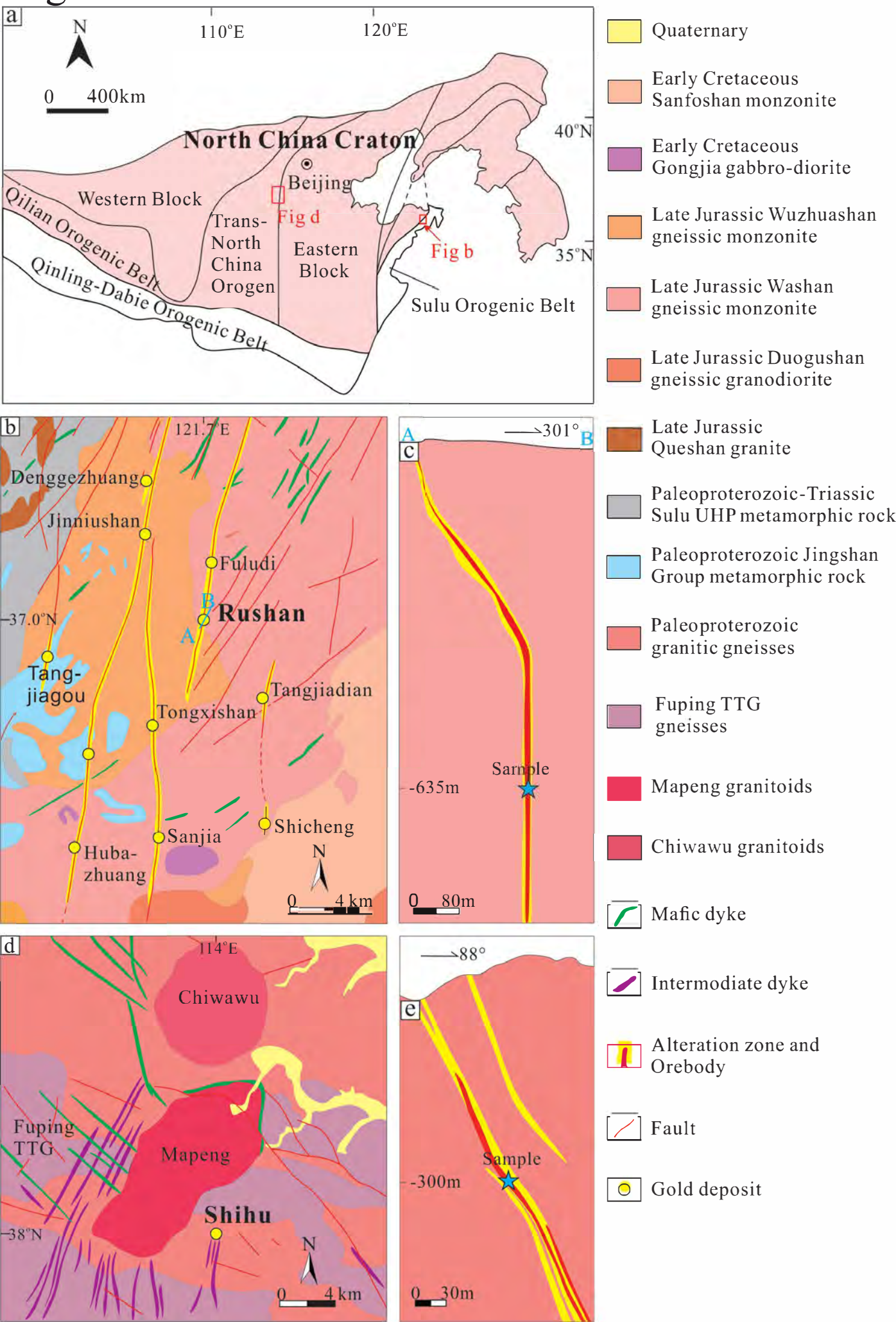
520 Fig. 4 X-ray diffraction and Fourier transform infrared spectroscopy analyses of  
521 different CL zoning in hydrothermal quartz from the Shihu and Rushan Au  
522 deposits, projected on the scanning electron microscope-cathodoluminescence  
523 images and electron probe micro-analysis trace element mappings; The cell  
524 volume and  $a_0$  values of quartz were obtained from the CIF documents using  
525 Mercury software

526 Fig. 5 **(a-d)** TEM and HRTEM images of aluminum-rich bands of quartz from the  
527 Shihu and Rushan Au deposits; **(e)(f)** Line scanning of aluminum, potassium,  
528 sodium and titanium; **(g)(h)** Structural configuration of trace elements in the  
529 quartz atomic lattice (modified after [Larsen et al. 2004](#)); **(g)** Coupled substitution  
530 of  $\text{Al}^{3+}$  and  $\text{H}^+$  ions for  $\text{Si}^{4+}$  i.e.  $(\text{Al}^{3+}-\text{H}^+)-\text{Si}^{4+}$  substitution; **(h)** Coupled  
531 substitution of  $\text{Al}^{3+}$  and  $\text{K}^+$  ions for  $\text{Si}^{4+}$  i.e.  $(\text{Al}^{3+}-\text{K}^+)-\text{Si}^{4+}$  substitution (the  
532 incorporation of Al, H and K is mainly based on the conclusion of this study,  
533 [Larsen et al. 2004](#) and [Götze, 2009](#))

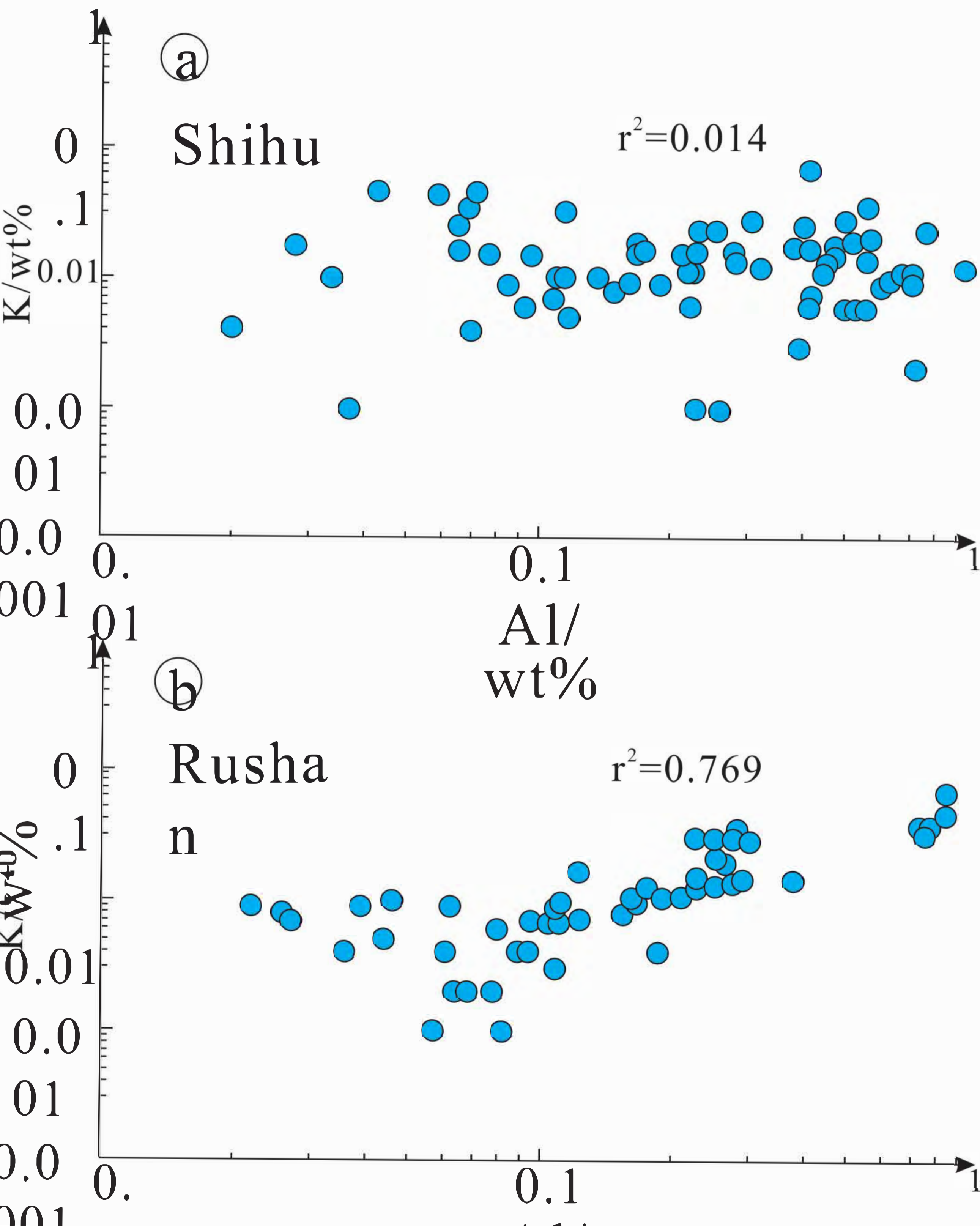
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535

# Figure. 1

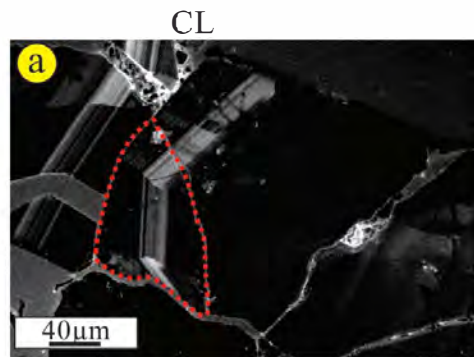


# Figure 2

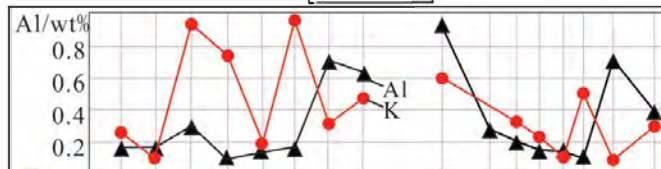


# Figure.3

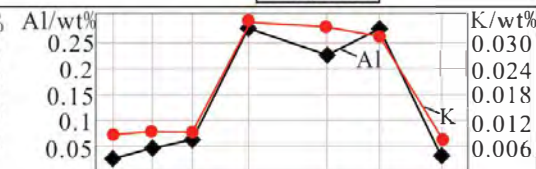
Shihu



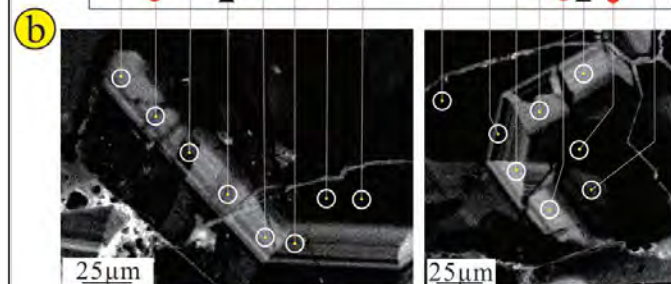
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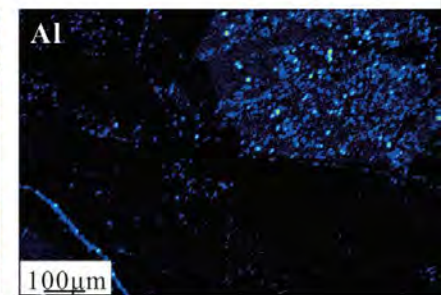
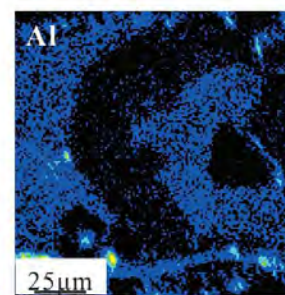
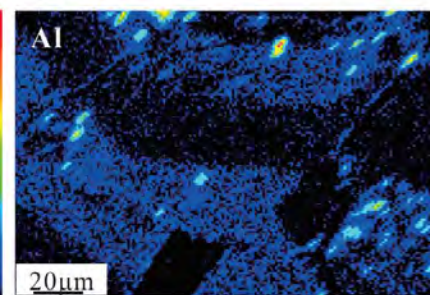
Rushan



Rushan

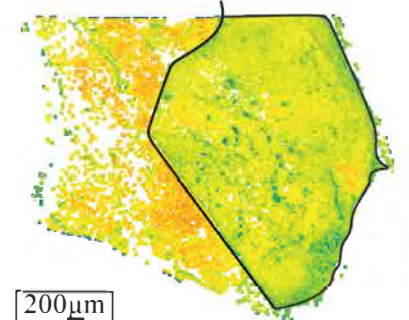
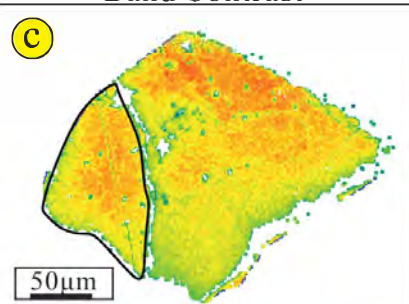


High  
Low



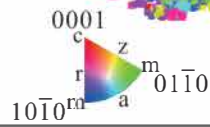
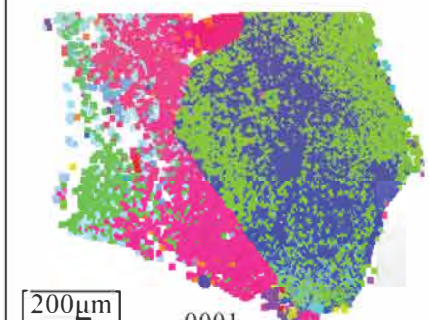
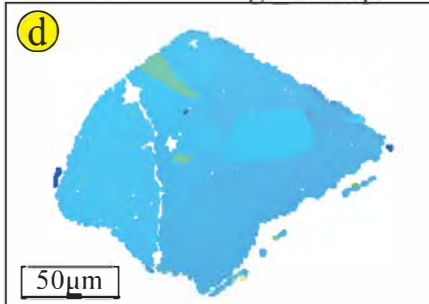
Shihu

Band Contrast

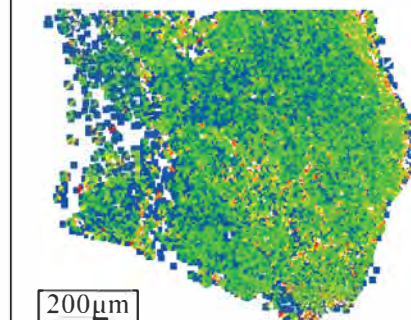
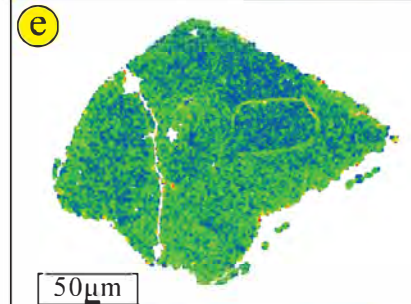


0 180

IPF colouring Y map

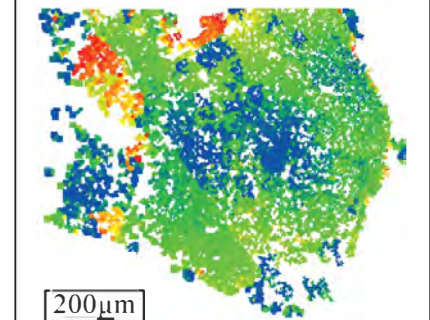
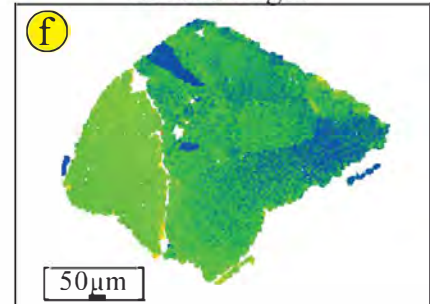


KAM



0° 2°

GROD angle



0 5

# Figure.4

