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Liquid - nitrogen quenching induced pore evolution and quantitative structure– activity modeling for high-purity quartz purification

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Abstract

Chemical purification is the key route to produce high-purity quartz sand (≥ 4N), yet the relationship between micro-pore evolution and purification efficiency remains unclear. In this study, natural quartz sand was subjected to liquid-nitrogen quenching and hydrothermal acid leaching, and the effects of calcination temperature, calcination time, quenching frequency, and mechanical grinding on pore distribution were systematically investigated. A quantitative predictive model describing the multi-factor interactions between pore structure, impurity removal rate, and quartz purity was established. The results showed that thermal expansion and phase transformation are the dominant factors for pore evolution, and connectivity microcracks can significantly improve the purity and impurity removal rate of quartz sand. Thermal expansion causes the specific surface area of quartz sand to decrease to 45%–55% of the raw sample, while high temperature also promotes the segregation of impurity elements. Phase transformation has a significant impact on pore connectivity, and the highest purity of acid-washed quartz sand can reach 99.93%, with an average removal rate of aluminum elements being approximately 80%. Partial correlation analysis confirmed the strong correlation between different pore types. The regression model follows a quadratic polynomial framework, with R^2 values ranging from 0.896 to 0.994, indicating its high reliability. This study elucidated the quantitative structure-activity relationship between pore structure and purification performance, providing a theoretical basis for the optimization of the high-purity quartz sand production process.

1 Introduction

High-purity quartz sand serves as a strategic raw material extensively utilised in sectors such as semiconductors, photovoltaics, optical fibres, advanced manufacturing, and aerospace. The total impurity content must remain below 20 ppm, while the silicon content should exceed 99.998% [1]. Currently, the preparation technology for high-purity quartz sand primarily relies on the purification of natural quartz ore and artificial synthesis. Natural quartz mineral resources are widely distributed and readily accessible, making them the principal raw materials for producing high-purity quartz sand. However, the metallogenic causes of quartz in nature are extremely complicated, leading to significant variations in associated minerals, lattice impurities, gas–liquid inclusions, and other contaminants [2]. Hence, the quality of quartz ore is pivotal in assessing high-purity quartz sand quality, as exemplified by the Unimin Company in the U.S. with its unparalleled pegmatite mine, underpinning its dominance in the global high-purity quartz sand market [3]. Due to the scarcity of high-quality quartz ore resources, the development based on the purification technology of 2N grade natural quartz sand is still lagging behind. To date, a reliable and universal technical solution for 2N grade quartz purification has not yet been established. In the existing technical solutions, conventional separation methods such as magnetic separation, gravity separation, flotation, and colour separation can only remove impurities like feldspar, magnetic iron oxide, and coloured inclusions [4, 5].

For lattice impurities and gas–liquid inclusions, pretreatment methods such as water quenching [6, 7] and microwave treatment [8, 9] are considered to be very effective. Nevertheless, the theories behind these physical separation techniques are still not mature, and the effective removal of lattice impurities and gas–liquid inclusions remains one of the technical bottlenecks in this field. In general, Al^{3+}, Fe^{3+}, Ge^{4+}, Ti^{4+}, B^{3+} and P^{5+} can act as lattice impurities, substituting the Si^{4+} site in the lattice, with aluminium being the most common impurity element [10]. Due to its abundance in natural quartz and similar ionic radius to silicon ions, aluminium is frequently utilised as a crucial indicator for assessing quartz sand purity. Monovalent cations, including Li^+, Na^+ and K^+, occupy the lattice gaps as compensating ions [11, 12]. Additionally, titanium, iron, sodium, and potassium may be present as mineral inclusions, such as rutile or iron oxide, or as fluid inclusions.

Currently, water quenching is considered the most effective and versatile technology for addressing this complex issue. This process typically involves heating quartz sand to temperatures exceeding 600°C, followed by rapid cooling in a subcooled medium. The thermal stress generated during this process leads to the formation of micro-cracks and new

grain boundaries, which disrupt gas–liquid inclusions and create diffusion channels and reaction interfaces for reactive molecules. Meanwhile, high-temperature calcination can activate high-energy chemical bonds, such as Al–O and Ti–O, facilitating the migration of impurity elements to the interface and enhancing the dissolution of trace impurity elements [13, 14]. Many similar studies have shown that calcination and quenching can effectively improve the removal rate of impurities and the purity of quartz sand. Ren et al. [6] showed that water quenching can produce micro-cracks, which makes quartz sand have larger specific surface area, pore volume, and pore diameter and enhances the adsorption capacity of quartz sand to acidic solution. Khalifa et al. [15] calcined natural sand in oxygen at 1000°C and quickly quenched it. The result of the acid leaching experiment shows that the content of metal impurities in the natural sand is greatly reduced, and its purity is increased from 99.72% to 99.92%. An experimental study also showed that after the quartz was roasted at 900°C for 4 h, the removal rate of iron impurities and total impurities after acid leaching reached 98.7% and 88.2%, respectively [16]. Yang et al. [17] used calcination pretreatment combined with ultrasonic-assisted leaching to remove iron impurities in quartz sand, and the quartz content increased from 99.6828% to 99.9047%. Lin et al. [18] mixed quartz raw materials with 2 wt.‰ of KCl, calcined at 900°C for 4–5 h, and leached them to lower the Al content in the sample to 11.7 ppm. The aforementioned studies have validated the efficacy of calcination and quenching in enhancing the purity of quartz sand. However, these studies have not clearly defined the structure–activity relationship and quantitative model between pore structure of quartz sand and the purification effect [6, 19]. Additionally, the purification effect is closely related to the source of raw materials and purification technology, and the complexity of the types and distribution of impurities limits their reference value and application value significantly [20].

From the perspective of mass transfer within the pores of quartz sand, the presence of a sufficient number of open pore structures in quartz sand particles is essential for assessing the quality of chemical purification. This condition ensures that the liquid-phase reaction medium can penetrate the pores and participate in the chemical reaction, ultimately facilitating the dissolution and diffusion of impurity elements. Currently, there are no research reports addressing the impact of quartz sand's pore structure on purification efficacy and the structure–activity relationship between the two. Based on our previous research on the ultra-low temperature cracking of quartz sand [20], this paper delves into the evolutionary relationship of pore structure between quenched sand and acid-impregnated sand, revealing the influence of pore structure changes on the removal rate of impurities and purity, and establishing a corresponding structure–activity prediction model that can be used for quantitative prediction.

2. Experimental section

2.1 Raw material and reagents

The samples of natural quartz sand used in this study were obtained from Fengyang, Anhui Province. The grain size of raw sand is 80–140 mesh, and the specific surface area is 0.0728 m^2/g. The chemical compositions and pore distribution analysis of natural quartz sand are reported in Table 1. Liquid nitrogen is a commercial product; deionised and distilled water was used throughout the work. The reagents used were oxalic acid (AR) and hydrofluoric acid (AR) purchased from Sinopharm Chemical Reagent Co., Ltd.

Table 1
Chemical composition and pore volume distribution of the raw silica sands

Component	Amount (%)	Pore size	Pore volume distribution (%)
SiO_2	99.60	Ma (130 nm ≥ PS > 50 nm)	40.63
Al	0.078	Me50 (50 nm ≥ PS > 10 nm)	26.53
Ti	0.004	Me10 (10 nm ≥ PS > 2 nm)	32.84
Na	0.0052	Mi (2 nm ≥ PS ≥ 0.35 nm)	0
K	0.019		
Mg	0.0064		
Ca	0.02		
Fe	0.0078		

2.2 Methods

Figure 1 illustrates the flowchart of the liquid-nitrogen quenching and purification process for natural quartz sand. Initially, selected natural quartz sand (designated as NQS) is placed in a muffle furnace, where it is heated and maintained at a specific temperature for a designated duration. Following this, the sand is immersed directly in liquid nitrogen for quenching. The quenched samples are then dried for 2 h at 100°C in a drying oven to yield cracked quartz sands (referred to as CQS). Subsequently, 30 g of cracked sand is combined with a 1 M oxalic acid solution containing 5 wt% HF in a hydrothermal reactor, where it undergoes hydrothermal treatment for 10 h at 120°C. The liquid–solid volume ratio is maintained at 4:1 to ensure complete immersion of the quartz sand in the solution. After hydrothermal treatment, the resulting purified sand is washed multiple times with deionised water and dried to produce acid-leached quartz sands (designated as AQS).

To investigate the effects of extreme temperature variations and acid leaching on purification, two sets of experiments were designed. The first set aimed to assess the influence of calcination temperature on pore structure. Cracked sand samples calcined for 1 h were designated as CQS-500, CQS-600, CQS-700, CQS-800, CQS-900, and CQS-1000, corresponding to their respective calcination temperatures. The purified sands derived from these samples were labelled as AQS-500, AQS-600, AQS-700, AQS-800, AQS-900, and AQS-1000. The second set involved quartz sands calcined at 900°C for varying durations to examine the impact of phase transitions on pore structure. Samples with different calcination times were designated as CQS-1, CQS-2, CQS-4, CQS-6 and CQS-8, with the corresponding purified sands labelled as AQS-1, AQS-2, AQS-4, AQS-6, and AQS-8, respectively.

2.3 Characterisations and evaluation methods

The specific surface area and pore distribution analysis of quartz particles were analysed by a specific surface area analyser (BSD-660S A6). The impurity content in quartz sand was determined by ICP-MS and measured by the national standard GB/T 384–2015 “Analytical method of the chemical composition in the quartz glass”. The efficiency of impurity removal can be calculated by the following Eq. (1).

$$\mathrm{E}\ (\%) = \left[1 - \frac{\text{impurity content in AQS}}{\text{impurity content in CQS}}\right] \times 100 \tag{1}$$

To assess the impact of changes in pore structure on the purity and impurity removal rate of quartz sand, this study utilises the difference in pore volume distribution, denoted as $\Delta PD = PD_A - PD_C$, where PD_A and PD_C represent the pore volume distributions of quenched sand and purified sand, respectively.

3. Results and discussion

3.1 Influence of calcination temperature on cracked structure and purity of quartz sand

The impact of various calcination temperatures on the pore volume distribution of purified sand elucidates the evolution of quartz sand's pore structure under different thermal stresses. The alterations in these laws are depicted in Fig. 2. Figure 2 illustrates contrasting trends in pore changes between purified sand and quenched sand, with the variations in Ma and Me50 being more pronounced compared to Mi and Me10. The discrepancy is caused by corrosion of Mi and Me50 during the purification process, which results in the formation of corresponding Me10 and Ma. As a result, the porosity of Mi and Me50 decreases, while the porosity of Ma and Me10 increases. With the escalation of calcination temperature, the thermal expansion-induced cracking of the quartz lattice promotes the generation of numerous micro-cracks, thereby augmenting pore connectivity. The rise in the proportion of Mi and Me50 in samples subjected to calcination after corrosion, coupled with the decline in Me10 pore ratio in samples treated at 900°C and 1000°C, substantiates the impact of micro-cracks and through cracks on their structures. The elevation in the proportion of Mi and Me50 in the sample treated at 1000°C, along with a 13.42% reduction in Me10, effectively corroborates this assertion.

Furthermore, phase transformation significantly affects the alteration of pore structure in quartz sand. Due to the effects of impurities, the corresponding temperatures for the α–β phase transformation and the α–α phase transformation are 600°C and 800°C, respectively. As illustrated in Fig. 2, the AQS-600 and AQS-800 samples exhibit marked differences compared to other samples. The increase rates of Me10 and Ma in the AQS-800 sample reached + 4.01% and + 25.3%, while the corresponding proportions of Mi and Me50 decreased by 5.83% and 23.47%, respectively. A notable characteristic of the CQS-600 sample is the 17.46% reduction in the proportion of Me10, which accounts for only 0.9%. The growth rates of Mi and Ma were + 4.54% and + 11.76%, respectively, while the proportion of Me50 increased by only 1.17%. The significant difference is that the microcracks produced by the α–β phase transition are more conducive to the corrosion of Me10 pores and the formation of Ma pores, while the sharp increase in the Ma pore ratio during the α–α phase transition mainly comes from the contribution of the corrosion of Me50 pores. Clearly, the phase transformation expansion process is a significant mechanism for generating micro-cracks and through cracks. The micro-cracks produced by the α–β phase transition facilitate the pore corrosion of Me10 and the formation of Ma, whereas the substantial increase in the proportion of Ma pores during the α–α phase transition primarily arises from the pore corrosion of Me50. This distinction may be attributed to the varying phase transition characteristics of quartz, with the α–β transition occurring at approximately 573°C and the transformation from α-quartz to α-squamous quartz at around 870°C [21–23]. The α–β transition is a rapid low-temperature transformation process involving simultaneous reversible displacement on both the surface and interior of quartz sand, resulting in a minimal volume expansion of only 0.82–1%. In contrast, the transformation from α-quartz to α-squamous quartz represents a slow, high-temperature phase transition process, with a theoretical volume expansion rate of up to 16% [20, 24]. The significant volume expansion during the α–α phase transition leads to increased micro-crack formation, thereby enhancing connectivity between microporous and mesoporous pores.

3.2 Influence of calcination time on cracked structure and purity of quartz sand

To investigate the relationship between phase transformation and the pore structure of quartz sand, a calcination temperature of 900°C, which is close to the theoretical temperature for α–α phase transformation, was selected. This study aims to examine how the phase transformation process, characterised by a high volume expansion rate, influences the pore structure of quartz sand. Figure 3 illustrates the effects of varying phase transformation durations on the pore structure of quartz sand. As shown in Fig. 3, different calcination times result in notable variations in the extent of phase transformation, with the degree of phase transformation cracking emerging as a critical factor influencing the pore structure of purified sand. During the initial phase of transformation (~ 2 h), the impact on porosity is most pronounced. The pore ratios of Me50 and Ma exhibit the greatest increases, at − 13.88% and + 16.52%, respectively. Additionally, the ratio of Mi to Me10 has decreased, suggesting improved pore connectivity within the structure.

When the calcination time reaches 4 h, the phase transformation cracking effect is maximised, leading to a significant increase in the number of micro-cracks and connected cracks. This results in a marked rise in the proportion of Mi and Me10 pores formed by corrosion within the pore structure. As calcination time extends, thermal expansion notably intensifies pore bridging, primarily evidenced by the dissolution of Me10 pores into Me50 and a reduction in the proportion of Ma. Furthermore, an excessively prolonged calcination period causes the accumulation of thermal energy in the quartz sand, resulting in lattice thermal cracking that disrupts both lattice and chemical bonds. Consequently, the proportions of Ma and Mi in the AQS-8 sample increased to 29.25% and 10.49%, respectively. Obviously, lattice thermal cracking under overheated conditions can effectively enhance the formation of micro-cracks and connected cracks, thereby further corroborating the analytical results of the AQS-1000 sample.

3.3 The influence of porous structure on specific surface area under different conditions

Table 2 illustrates the relationship between the specific surface area of quartz sand and various calcination conditions. As indicated in Table 2, the specific surface area of cracked sand has significantly decreased, and the specific surface area of most cracked sand is less than 50% of that of the original sand. And the specific surface area of all purified sands is only 50%–60% of the raw sand. Furthermore, the specific surface areas of the CQS-600, CQS-1000, and CQS-4 demonstrate notable differences. Specifically, the CQS-600 and CQS-1000 achieve specific surface areas of 87.09% and 89.70% of the original sand, significantly exceeding those of the other samples. Comparison of pore structure changes reveals that the larger specific surface area of the CQS-600 and CQS-1000 samples primarily stems from increased micropores, with their pore volume distributions rising by 4.54% and 4.65%, respectively. The specific surface areas of CQS-4 and AQS-4 samples were 0.0453 m^2/g and 0.043 m^2/g, respectively, exhibiting minimal change. Correspondingly, the micropore ratios of CQS-4 and AQS-4 were 9.99% and 8.53%, respectively, decreasing by only 1.46%, indicating the least alteration in micropores. Evidently, the notable decline in the specific surface area of quartz sand results from pore bridging intensification during high-temperature calcination, where even the extreme temperature difference stress induced by nitrogen quenching fails to promote significant pore cracking. Both higher temperature and phase transition significantly facilitate micropore formation, thereby augmenting the specific surface area of quartz sand.

It is worth noting that the specific surface area of quenched sand ranges from 0.0278 to 0.0653 m^2/g, whereas that of purified sand varies from 0.033 to 0.043 m^2/g. The decrease in the specific surface area of purified sand indicates good connectivity of the pore structure, which can provide a mass transfer channel for reaction substances. The increase in the proportion of large-sized pores in Me50 and Ma also precisely confirms this. Except for CQS-700, CQS-1 (CQS-900), and CQS-4, the increment of the specific surface area of other samples is significant, ranging from 11.81% to 41.76%. Among them, CQS-700 and CQS-4 samples mainly benefit from pore closure or the formation of stable pore structures during phase transformation, while the CQS-1 sample was mainly due to the shorter calcination time resulting in a smaller degree of pore expansion. Based on these analyses, we propose that high temperature and α–β phase transition contribute to micropore cracking, thereby enhancing the connectivity among pores [25, 26].

Table 2
The specific surface areas of cracked sand and purified sand under different calcination conditions

Sample	$S_{BET-CQS}$ (m^2/g)	$\frac{S_{BET-CQS}}{S_{BET-Raw}} \times 100\%$	$S_{BET-AQS}$ (m^2/g)	$\frac{S_{BET-AQS}}{S_{BET-Raw}} \times 100\%$	$\frac{S_{BET-CQS}}{S_{BET-Raw}} \times 100\% - \frac{S_{BET-AQS}}{S_{BET-Raw}} \times 100\%$
CQS-500	0.0278	38.19%	0.0402	55.22%	17.03% ↑
CQS-600	0.0361	49.59%	0.0375	51.51%	1.92% ↑
CQS-700	0.0306	42.03%	0.0409	56.18%	14.15% ↑
CQS-800	0.0352	48.35%	0.0397	54.53%	6.18% ↑
CQS-900	0.0352	48.35%	0.0397	54.53%	6.18% ↑
CQS-1000	0.0653	89.70%	0.0395	54.26%	35.44% ↓
CQS-2	0.0303	41.62%	0.0403	55.36%	13.74% ↑
CQS-4	0.0453	62.23%	0.0430	59.07%	3.16% ↓
CQS-6	0.0331	45.47%	0.0421	57.83%	12.36% ↑
CQS-8	0.0343	47.12%	0.0429	58.93%	11.81% ↑
Average		45.09%		54.80%	

3.4 Correlation analysis of pore structure and purification effect of quartz particles

The critical step in producing high-purity quartz sand involves disrupting the crystal lattice and inclusion structure of quartz to create micro-cracks within the crystal. This process facilitates the exposure of impurity ions on the pore surface, enabling their effective removal. The widespread adoption of the water quenching process in current high-purity quartz sand processing is attributed to this mechanism. Therefore, the multi-scale pore structure emerges as the primary determinant of quartz sand purification quality. The primary role of micropores and pores is to enhance the specific surface area and provide numerous adsorption sites. Mesoporous structures function as channels, facilitating the diffusion and transport of the reaction medium within the particles. In contrast, macropores act as temporary storage spaces for adsorbates, thereby reducing diffusion resistance. The purification process encompasses various physical and chemical mechanisms, including reactions, diffusion, and mass transfer, all occurring within a complex pore system comprised of micropores, mesopores, and macropores.

Since the pore volume remains almost constant during the purification process, the change in pore volume distribution directly reflects the mutual conversion relationship between different pores. An increase in pore ΔPD corresponds to a decrease in ΔPD of other pores, as evidenced by experimental results. Moreover, the change in pore volume distribution also proves that pore corrosion is the fundamental cause of its pore structure changes. According to the law of pore corrosion, the change in pore ratio mainly depends on the relative proportion of corrosion-induced transformation of small-sized pores and self-corrosion transformation. For example, variations in the Me10 ratio in purified sand stem from Me10 formation via micro-pore corrosion and Me10 conversion to Me50 through pore corrosion. Therefore, a positive ΔPD suggests a prevalence of small-sized pores formed by extensive corrosion over self-corrosion transformation. Conversely, a negative ΔPD indicates a relatively closed pore structure with limited pore corrosion transformation. The pore corrosion can not only directly reflect the mass transfer properties of the reaction medium within the pores but also signifies enhanced connectivity of pores.

(1) Correlation between pore structure and purity

Figure 4 shows the relationship between ΔPD and the purity of quartz sand. From Fig. 4a and 4b, it can be seen that the ΔPD_{Mi} and ΔPD_{Me50} values of quartz sand are mostly negative, while the ΔPD_{Ma} values are all positive. This indicates that small-sized pores can be corroded during the purification process, forming macropores. As the calcination temperature increases, ΔPD_{Mi} and ΔPD_{Me50} show an increasing trend, while ΔPD_{Ma} and ΔPD_{Me10} show a decreasing trend. Particularly at post temperatures exceeding 700°C, the variations in ΔPDMe10 and ΔPD_{Mi} become increasingly pronounced. Notably, the ΔPD_{Me50} and ΔPD_{Mi} of the CQS-1000 sample surged to 4.65% and 8.33%, respectively. The primary cause of this phenomenon is the combined impact of thermal expansion and thermal cracking of quartz crystals. Throughout the entire high-temperature calcination process, thermal expansion exacerbates pore bridging and pore closure, leading to a notable decrease in specific surface area. The pores in CQS-600 and CQS-700 samples underwent significant shrinkage, accompanied by a reduction in the interconnected pores. Although the α–β phase transformation expansion is conducive to the formation of micropores, the decreasing trend of purity indicates that the pores show more obvious pore closure under the combined effect of α–β phase transformation expansion and thermal expansion. Especially for the AQS-700 sample, the specific surface area increased by only 1.92%. This largely inhibited the mass transfer and diffusion of reaction media within the pore channels, and was also the key reason for the decrease in purity of AQS-600 and AQS-700 samples.

When the calcination temperature exceeds 700°C, the α–α phase transformation is induced, resulting in the formation of numerous immeasurable microcracks and interconnected pores, which further evolved into detectable micro-pores through corrosion. Simultaneously, higher calcination temperatures can induce α–α phase transitions and facilitate the displacement, exchange, and diffusion of atoms within the lattice. This is also the reason for the significant increase in ΔPD_{Mi}, ΔPD_{Me50} and SiO_2 purity of AQS-900 and AQS-1000 samples. The difference is that the proportion of macropores in the AQS-800 sample increased by 25.3% compared to the CQS-800 sample, with the purity of silica reaching 99.87%. Therefore, we infer that both α–α phase transformation cracking and thermal cracking have generated more interconnected pores, which effectively enhance the mass transfer and reaction of the acid etching solution within the pore channels. Additionally, the decreasing trend of ΔPD_{Ma} further substantiates this perspective regarding pore structure.

To further elucidate the beneficial effects of phase transformation on the cracking behaviour of quartz sand, we conducted a comprehensive analysis of the relationship between ΔPD and purity during the α–α phase transformation process. As illustrated in Figs. 4c and 4d, the purity of quartz sand exhibits an upward trend, yet there is no significant correlation with ΔPD. This observation is intrinsically linked to the completeness of the phase transition. Based on varying degrees of phase transformation, the α–α phase transformation process of quartz generally goes through three distinct stages. During the initial phase of transformation (~ 2 h), quartz sand begins to absorb substantial amounts of heat energy, primarily for the phase transformation and thermal expansion of silicon oxide crystals. The expansion associated with phase transformation results in the development of numerous unmeasurable cracks and micropore cracks. New pores promote the connection of the original large-sized pores, facilitating reactions and mass transfer within the pores, and resulting in a significant increase in ΔPD_{Ma} and the purity of quartz sand.

As the phase transformation progresses towards completion, expansion cracking from the phase transformation becomes the primary factor influencing the pore structure change. The ΔPD_{Mi} and ΔPD_{Me10} of the AQS-4 sample decreased by − 1.46% and − 2.99%, respectively, while ΔPD_{Me50} and ΔPD_{Ma} increased by + 1.32% and + 3.14%. The rise in the relative ratio of Ma and Me50 suggests effective pore corrosion, diffusion and reactions in the cracking structure, attributed to the formation of numerous unmeasurable pores, microporous pores, and connected pores during the α–α phase transformation process with a substantial volume expansion effect. Beyond 4 h of calcination time, the cracking structure of pores stabilises, following a trend similar to the temperature's influence on purity and pore structure. Due to

thermal expansion effects, Mi and Me50 corrode and transform into more Me10 and Ma, leading to an increasing trend in ΔPD_{Ma} and ΔPD_{Me10}.

It is worth noting that the purity of quartz sand did not change with the variation of the pore structure. This observation indicates that the pore structure is not the sole determinant of quartz sand purity. The more critical reason is the increase in temperature, which enhances lattice vibration and increases crystal defects. This elevation in energy facilitates the release of impurity ions and accelerates their migration. In addition, prolonged exposure to high temperatures may result in a greater degree of ion migration. Consequently, we can deduce that the purification process of quartz sand primarily relies on the synergistic interaction between crystal structure and pore structure, with significant structural alterations in both being influenced by temperature. A notable example is the AQS-800 sample. The high-temperature phase transition not only increased the number of micropores and interconnected pores but also induces M–O fractures in the crystal structure, thereby promoting the migration of lattice impurities and significantly enhancing purity.

(2) Correlation between pore structure and ion removal rate

In addition to silicon dioxide content, the presence of impurity elements serves as a critical indicator of quartz sand quality. Generally speaking, natural quartz ores primarily consist of common minerals, including feldspar, mica, clay, and magnetite. Impurity elements are mostly present in the lattice in the form of substituting atoms or interstitial atoms. Therefore, this study selects the removal rates of high bond energy elements (Ti and Al), alkali metal elements (Na and K), alkaline earth metal elements (Ca and Mg), and transition metal elements (Fe) as characteristic indicators. Figure 5 illustrates the relationship between calcination temperature and time on the removal rate of impurities in AQS samples.

As can be seen from Fig. 5, the removal rates of impurity elements exhibit a distinct pattern of initial decrease followed by an increase, with potassium and calcium being exceptions. Notably, the AQS-700 sample demonstrates the lowest ion removal rate. The behaviour of impurity removal rates at temperatures equal to or below 700°C aligns with ΔPD_{Ma} and ΔPD_{Me10}, whereas at temperatures exceeding 700°C, it corresponds to ΔPD_{Mi} and ΔPD_{Me50}. Different from non-phase change samples, CQS-600 and CQS-800 samples correspond to the α–β phase transformation process and α–α phase transformation process, respectively. The variations in impurity element removal rates in the AQS-600 sample mirror those in ΔPD_{Ma} and ΔPD_{Me10}, suggesting that impurity elimination primarily stems from the transformation of Mi and Me50 pores during the purification process.

Surprisingly, the removal rate of iron element during the α–α phase transformation shows a significant decrease, and it exhibits a trend consistent with ΔPD_{Mi} and ΔPD_{Me50}. Moreover, the trend of the removal rate of impurity elements during the α–α phase transformation also shows significant differences. The removal rate of sodium and iron elements in the AQS-6 sample also shows a similar change pattern to that of iron element in the AQS-800 sample, and both removal rates reach the peak. The trends of removal rates of magnesium and calcium elements are consistent with ΔPD_{Ma} and ΔPD_{Me10} changes, while the trends of removal rates of titanium, aluminium, and potassium elements are consistent with ΔPD_{Mi} and ΔPD_{Me50} changes. This diversified change pattern indicates the correlation between pore structure and the substitution structure of impurities in the crystal.

Generally speaking, Ti^{4+} substitution of Si^{4+} forms the isovalent isomorphism structure, while Al^{3+} and Fe^{3+} substitution of Si^{4+} can form the neutral balanced charges of $[M^{3+}O_4/M^{+}]^0$ [27, 28]. In the incipient stage of phase transformation (~ 2 h), the quartz lattice expands and generates a large number of cracking pores, and almost all impurity elements begin to be exposed on the pore surface, and the removal rate significantly increases. When the calcination time is 4 h, the phase transformation cracking structure reaches a stable state. The removal rates of Al, K and Ti reach their maximum values, which are 82.05%, 75.26% and 37.5%, respectively. This phenomenon

indicates that the cracking of the α–α phase transformation is conducive to destroying the Ti–O bonds and $[Al^{3+}O_4/K^+]^0$ in the quartz crystal. During this process, the removal rates of Ca and Mg significantly decrease, and the Fe removal rate of sample CQS-4 drops to 76.92%. Therefore, we infer that relatively stable $[Fe^{2+}O_4/Ca^{2+}]^0$ and $[Fe^{2+}O_4/Mg^{2+}]^0$ structures may have formed in the quartz crystal. When the quartz sand is calcined for 6 h, the removal rates of Ca and Mg increased significantly, reaching 97.1% and 85.16%, respectively. However, the removal rate of iron continued to decrease and reached its minimum value of 61.54%. This change is attributed to the oxidation reaction of Fe^{2+} to Fe^{3+}, which causes $[Fe^{2+}O_4/M^{2+}]^0$ to transform into $[Fe^{3+}O_4/M^+]^0$. Another important piece of evidence is that the trend of Na^+ changes over the calcination time is opposite to that of iron ions. Obviously, this might be the result of the mutual transformation among $[Fe^{3+}O_4/Na^+]^0$, $[Fe^{2+}O_4/2Na^+]^0$ and $[Fe^{2+}O_4/M^{2+}]^0$ during the phase transition process. As the calcination time increases, the removal rate of Fe rises while the removal rate of Na decreases. The process of its structural changes is as follows [10, 13, 29].

2 h: $[Fe^{3+}O_4/Na^+]^0 + [Al^{3+}O_4/K^+]^0 \rightarrow Al^{3+} + K^+ + Fe^{3+} + Na^+$

4 h: $[Fe^{2+}O_4/2Na^+]^0 + [Al^{3+}O_4/K^+]^0 \rightarrow [Fe^{3+}O_4/Na^+]^0 + Na^+ + Fe^{3+} + Al^{3+} + K^+$

6 h: $[Fe^{2+}O_4/M^{2+}]^0 + 2Na^+$ (interstitial atom) $\rightarrow [Fe^{3+}O_4/Na^+]^0 + Ca^{2+} + Mg^{2+} + Na^+$

8 h: $[Fe^{3+}O_4/Na^+]^0 \rightarrow Fe^{3+} + Na^+$ (interstitial atom)

Moreover, the heat absorbed by the quartz particles will significantly facilitate the migration of ions such as calcium, magnesium, and iron. In particular, for the calcium element, the removal rate of Ca in the AQS-8 sample reached 99.45%. Since the calcium element in quartz crystals mainly exists in the form of gas–liquid inclusions or independent minerals such as fluorite, we speculate that the phase transformation has a very good effect on the gas–liquid inclusions. By comparing the removal rates of other elements, it can be known that the crystal structure of the AQS-8 sample mainly consists of $[Al^{3+}O_4/M^+]^0$ and $[Fe^{3+}O_4/M^+]^0$, as well as some magnesian analogous structures.

3.5 Construction of multi-factor interactive regression model

To further establish a quantitative model linking the pore structure to the purity and impurity removal rate of quartz sand, we performed three quenching and three quenching–grinding treatments on the CQS-2 and CQS-4 samples, respectively. This approach allowed us to thoroughly investigate the effects of repeated thermal stress and mechanical force on the phase change pore structure, resulting in a larger dataset for statistical analysis. Table 3 presents the pore structure, purity, and removal rate data for the CQS-2-X and CQS-4-X samples. The experimental results indicate that the ΔPD values for the CQS-2-2 and CQS-4-2 samples exhibit significant changes. Both repeated quenching and mechanical grinding effectively modify the pore structure.

The purity of quartz sand did not exhibit a significant increase with the number of quenching and quenching–grinding cycles. In comparison to the CQS-2 and CQS-4 samples, the purities of CQS-2-X and CQS-4-X increased slightly. The purities of CQS-2 and CQS-4 rose to 99.90% and 99.93%, respectively, reflecting increases of only 0.02% and 0.04%. Furthermore, the removal rates of impurities generally displayed a decreasing trend and appeared to stabilise. This relatively stable trend may result from the impurities in the quartz crystal reaching an equilibrium state between crystal and purification solution. Consequently, to achieve high-purity quartz sand, it is crucial to reduce the concentration of impurity ions in the purification solution.

Table 3
The specific surface areas of cracked sand and purified sand under different calcination conditions.

sample	CQS-2-1	CQS-2-2	CQS-2-3	CQS-4	CQS-4-1	CQS-4-2	CQS-4-3
ΔPD_{Mi}	-4.52	1.13	-8.16	-1.46	-12.1	-5.41	-3.47
ΔPD_{Me10}	1.87	-10	14.04	-2.99	14.79	8.21	5.49
ΔPD_{Me50}	-13.88	7.68	-20.05	1.32	-13.88	-12.97	-7.12
ΔPD_{Ma}	16.52	1.19	14.17	3.14	11.19	10.17	5.09
Purity	99.88	99.80	99.90	99.89	99.86	99.88	99.93
ΔP	0.28	0.2	0.3	0.29	0.26	0.28	0.33
E(Al)	80.77	79.49	76.92	82.05	75.64	76.92	76.92
E(Ti)	30.00	35.00	25.00	37.50	25.00	10.00	30.00
E(Ca)	96.10	90.50	92.50	86.00	90.00	88.00	89.00
E(Na)	57.69	53.85	48.08	61.54	53.85	46.15	46.15
E(Fe)	80.77	79.49	79.49	76.92	79.49	76.92	79.49
E(K)	74.74	71.58	68.42	75.26	66.32	68.42	71.58
E(Mg)	85.31	23.44	84.38	78.13	84.38	84.38	84.38

The complexity of pore structure and the interrelated transformations during pore cracking and dissolution indicate that the relationships among pores are not unidirectional. To accurately assess the correlation coefficient of ΔPD as a reflection of linear correlation, we performed a partial correlation analysis of ΔPD across micropores, mesopores, and macropores in 14 groups of phase change samples (Table 4). As shown in Table 4, there is a significant correlation between different pores, indicating that there is a transformation relationship between pores of similar sizes. Thus, an increase or decrease in ΔPD_{Mi} and ΔPD_{Me50} corresponds to a decrease or increase in ΔPD_{Me10}, ΔPD_{Me50}, and ΔPD_{Ma}, respectively. Additionally, there is a significant positive correlation between ΔPD_{Me10} and ΔPD_{Ma}, and between ΔPD_{Mi} and ΔPD_{Me50}. That is, an increase in Mi and Me10 will correspondingly lead to an increase in Me50 and Ma. This indicates that the pore change relationship between discontinuous-scale pores may be influenced by the connected pores. These analysis results align closely with the results of our analyses presented in Sects. 3.1 to 3.4.

Table 4
Partial correlation analysis of ΔPD

ΔPD_{Mi}	average value	standard deviation	ΔPD_{Mi}	ΔPD_{Me10}	ΔPD_{Me50}	ΔPD_{Ma}
	-2.818	**4.517**	**1**			
ΔPD_{Me10}	0.230	9.515	-0.818**	1		
ΔPD_{Me50}	-7.086	9.575	0.851**	-0.979**	1	
ΔPD_{Ma}	9.675	6.464	-0.931**	0.930**	-0.976**	1
*$p < 0.05$ ** $p < 0.01$						

Given the interaction and significant correlation among pores, we selected purity and impurity removal rate as dependent variables. We used ΔPD and its interaction terms as independent variables to analyse the impact of multi-factor interactions on sample purity and impurity removal rate, constructing a linear regression model to represent this relationship. Table 5 displays the statistical analysis results of the model. As can be seen from Table 5, the regression model prediction equation indicates that significant two-factor interactions influencing the purity of quartz sand and the impurity removal rate primarily include $\Delta PD_{Mi} \times \Delta PD_{Ma}$, $\Delta PD_{Me10} \times \Delta PD_{Me50}$, and $\Delta PD_{Me10} \times \Delta PD_{Ma}$. Notably, only $\Delta PD_{Me10} \times \Delta PD_{Ma}$ exerts a significant or relatively significant effect on the removal rates of magnesium, calcium, titanium, and iron. This significant interaction suggests that the impacts of Ma and Me50 on purity and removal rates are contingent upon variations in Mi and Me10. This phenomenon can be attributed to the calcination and quenching processes, during which smaller fissures of Mi and Me10, as well as unmeasurable fissures, are more readily formed. The significant interactions between these pores may be attributed to the fact that Mi and Me10 have enhanced the connectivity of large-sized pores, thereby strengthening the dissolution and diffusion of impurities throughout the purification process.

The statistical analysis results presented in Table 5 also show that the F values of all prediction equations are statistically significant ($p < 0.05$). However, the F values vary considerably, ranging from 13.09 to 227.90, which highlights the differing intensities of various indicators influenced by pore structure. The Na removal rate exhibited the most pronounced inter-group differentiation, with an F value of 227.90. Next were Al and K, with F values of 117.38 and 114.81, respectively. The values of ΔPD, Fe, Mg, Ca, and Ti decreased successively. From the R^2 value, it can be seen that the variables with high F values have both high statistical significance and practical significance. The high F value indicators such as E(Na), E(Al), and E(K) serve as key markers for distinguishing high-purity quartz sand, and they effectively explain the quantitative relationship between the pore structure of quartz sand and the removal rate of impurities. This provides a more direct theoretical basis for evaluating the quality of high-purity quartz sand. Nonetheless, the low F value for E(Ti) does not diminish its practical significance, as evidenced by an R^2 value of 0.8962. This importance is linked to the substitution form of titanium within quartz crystals and its high bond energy, which complicates the separation process of titanium in the pore structure.

The scatter plot depicting the residuals against the predicted values for purity and impurity removal rates is presented in Fig. 6. As illustrated, the purity residuals predominantly fall within the range of [− 0.04, 0.04]. The distribution of model residuals and predicted values generally oscillates around the zero line, aligning with the anticipated performance of the linear regression model, thereby indicating reliable prediction results. With the exception of the titanium impurity removal rate, the residuals for the impurity removal rates of other elements also fluctuate around the zero line, consistent with the expected behaviour of the linear regression model and suggesting a certain degree of reliability in the predictions. Overall, the linear regression model serves as a crucial foundation and research avenue for further investigations into structure–activity relationships and the quantitative evaluation of pore structures, offering significant reference value. The prediction equations of the quadratic regression model are as follows:

$$Y(\Delta P) = -10.22\,\Delta PD_{Mi} - 10.07\,\Delta PD_{Me10} - 9.93\,\Delta PD_{Me50} - 9.91\,\Delta PD_{Ma} - 0.00102\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 0.027\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 0.053\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 0.012\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 0.018\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.0038\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Al) = -1751.62\,\Delta PD_{Mi} - 1723.94\,\Delta PD_{Me10} - 1691.34\,\Delta PD_{Me50} - 1684.63\,\Delta PD_{Ma} - 0.075\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 5.67\Delta PD_{Mi} \times \Delta PD_{Me50} + 10.66\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 2.54\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 4.02\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.56\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Ti) = -891.34\,\Delta PD_{Mi} - 881.01\,\Delta PD_{Me10} - 866.49\,\Delta PD_{Me50} - 863.67\,\Delta PD_{Ma} - 0.36\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 1.57\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 4.13\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 1.05\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 1.86\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.30\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Fe) = -2036.12\,\Delta PD_{Mi} - 2014.74\,\Delta PD_{Me10} - 1987.56\,\Delta PD_{Me50} - 1979.86\,\Delta PD_{Ma} - 0.35\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 3.85\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 8.40\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 1.90\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 3.40\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.41\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Na) = -1103.92\,\Delta PD_{Mi} - 1084.19\,\Delta PD_{Me10} - 1059.90\,\Delta PD_{Me50} - 1055.10\,\Delta PD_{Ma} - 0.36\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 3.69_{\Delta PDMi} \times \Delta PD_{Me50} + 7.73\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 1.93\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 3.09\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.48\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(K) = -1753.27\,\Delta PD_{Mi} - 1727.09\,\Delta PD_{Me10} - 1695.94\,\Delta PD_{Me50} - 1689.85\,\Delta PD_{Ma} - 0.081\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 5.53\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 10.30\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 2.49\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 3.87\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.55\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Mg) = -1190.39\,\Delta PD_{Mi} - 1163.87\,\Delta PD_{Me10} - 1129.10\,\Delta PD_{Me50} - 1119.30\,\Delta PD_{Ma} + 0.53\,\Delta PD_{Mi} \times \Delta PD_{Me10} + 9.47\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 13.64\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 3.81\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 4.99\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.35\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

$$Y(Ca) = -731.12\,\Delta PD_{Mi} - 705.24\,\Delta PD_{Me10} - 668.48\,\Delta PD_{Me50} - 655.76\,\Delta PD_{Ma} + 0.65\,\Delta PD_{Mi} \times \Delta PDMe10 + 10.96\,\Delta PD_{Mi} \times \Delta PD_{Me50} + 15.14\,\Delta PD_{Mi} \times \Delta PD_{Ma} + 4.41\,\Delta PD_{Me10} \times \Delta PD_{Me50} + 5.56\,\Delta PD_{Me10} \times \Delta PD_{Ma} - 0.27\,\Delta PD_{Me50} \times \Delta PD_{Ma}$$

Table 5
Statistical Analysis of the Quadratic Regression Model.

		p							
		ΔP	**Al**	**Ti**	**Ca**	**Na**	**Fe**	**K**	**Mg**
ΔPD_{Mi}		0.012*	0.046*	0.26	0.60	0.022*	0.11	0.036*	0.32
ΔPD_{Me10}		0.013*	0.048*	0.26	0.61	0.023*	0.11	0.037*	0.33
ΔPD_{Me50}		0.013*	0.050	0.27	0.63	0.025*	0.11	0.039*	0.34
ΔPD_{Ma}		0.013*	0.051	0.27	0.63	0.025*	0.11	0.039*	0.34
$\Delta PD_{Mi} \times \Delta PD_{Me10}$		0.58	0.87	0.51	0.52	0.18	0.65	0.85	0.52
$\Delta PD_{Mi} \times \Delta PD_{Me50}$		0.014*	0.029*	0.45	0.036*	0.012*	0.23	0.024*	0.031*
$\Delta PD_{Mi} \times \Delta PD_{Ma}$		0.0019**	0.0049**	0.12	0.018*	0.0012**	0.051	0.0041**	0.013*
$\Delta PD_{Me10} \times \Delta PD_{Me50}$		0.0033**	0.008**	0.14	0.015*	0.0017**	0.086	0.0064**	0.012*
$\Delta PD_{Me10} \times \Delta PD_{Ma}$		8.83×10^{-4}**	0.0017**	0.036*	0.0077**	3.19×10^{-4}**	0.017*	0.0015**	0.0056**
$\Delta PD_{Me50} \times \Delta PD_{Ma}$		0.0033**	0.024*	0.16	0.46	0.0034**	0.18	0.019*	0.27
Modle	Adj. R-Square	0.9833	0.9939	0.9879	0.9654	0.9587	0.9881	0.8962	0.9664
	F Value	83.41	227.9	114.81	40.01	33.45	117.38	13.09	41.23
	p	3.37×10^{-4}**	4.58×10^{-5}**	1.79×10^{-4}**	0.0014**	0.002**	1.71×10^{-4}**	0.012*	0.0014**

*$p < 0.05$; ** $p < 0.01$

4. Conclusion

Based on the microscopic pore structure characteristics, a systematic purification strategy for natural quartz sand was established. The synergistic effects of liquid nitrogen quenching, hydrothermal acid leaching and heat treatment processes on the pore structure of quartz sand and the removal of impurities were studied. The following were the primary conclusions:

(1) The evolution of pore structure is one of the important factors affecting the purification of quartz sand. It mainly manifests as the formation and expansion of micropores and mesopores in the pore network. After optimizing the calcination temperature, calcination time and quenching frequency, quartz sand with a SiO_2 content of 99.93% was finally obtained. The removal rates of characteristic impurities such as aluminum and titanium reached 82.05% and 37.5% respectively, and the removal rate of calcium was as high as 99.45%. This sand can be used as the raw material for high-

end silicate products such as photovoltaic quartz crucibles, and can lay the foundation for further deep-bed chemical purification to produce semiconductor-grade quartz sand.

(2) The calcination temperature and time have the most significant impact on the specific surface area and pore structure of quartz sand. The specific surface area of the phase-change quartz sand can exceed that of the original sand by 87%, while the average specific surface area of non-phase-change quartz sand and refined sand is between 45% and 55% of that of the original sand. As the temperature increases, ΔPD_{Ma} and ΔPD_{Me10} show a downward trend, while ΔPD_{Mi} and ΔPD_{Me50} show an upward trend. After calcination at 900°C for 4 hours, the pore structure of the quartz sand becomes stable, the phase change process ends, and ΔPD_{Mi} and ΔPD_{Me50} reach peak values of -1.46% and + 1.32% respectively. The α–β phase change is conducive to the connectivity between mesopores, while the α–α phase change promotes the formation of micropores and enhances the connectivity between mesopores. The phase transformation cracking process is the most critical mechanism for enhancing quartz sand purity. The α–α phase transition generates substantially more interconnected microcracks than the α–β transition, which is the principal reason why AQS-800 and AQS-900 samples achieve the highest purity levels (up to 99.93%). By regulating the phase change path, it is possible to effectively control and optimize the pore structure, providing a theoretical basis for structure-oriented purification design.

(3) The correlation analysis between the pore structure and the purity and impurity removal rate of quartz sand indicates that the inflection point temperature for the purity and impurity removal rate of quartz sand is 700℃, which is consistent with the trend of ΔPD decreasing first and then increasing. In contrast, a longer phase transformation time is more conducive to the improvement of the purity of quartz sand, with the purity of the AQS-8 sample reaching 99.92%. The trend of ΔPD value changes shows a multi-regional variation characteristic with the differences in the degree of phase transformation. This trend is mainly attributed to the dynamic equilibrium established between the impurity elements in the lattice and those in the purification solution. It is worth noting that non-structural factors such as the breakage of M-O bonds and the migration of lattice impurities are also one of the main reasons for the monotonic increase in the purity of quartz sand.

(4) The experimental results show that repeated quenching and grinding can alter the pore structure, but they cannot significantly improve the purity of quartz sand. On the contrary, the removal rate of impurity elements in quartz sand shows a downward trend, indicating that the final purification efficiency is mainly influenced by the migration of lattice impurities and the breakage of M–O bonds, rather than simply the opening of physical pores. Based on the data of 14 samples, this paper constructs a multi-factor interaction regression model between ΔPD and purity/impurity removal rate, revealing the quantitative relationships between ΔPD_{Mi}, ΔPD_{Me10}, ΔPD_{Me50}, ΔPD_{Ma} and their interaction quantities and purity/impurity removal rate. Partial correlation analysis confirms that there is a strong interdependence between different pore types. The R^2 value of the quadratic polynomial regression model ranges from 0.896 to 0.994, indicating that the model has good reliability. The significant interaction terms $\Delta PD_{Mi} \times \Delta PD_{Ma}$ and $\Delta PD_{Me10} \times \Delta PD_{Ma}$ indicate that the connectivity between pores is the main structural factor determining the impurity removal efficiency. The structural-activity quantitative model established in this study provides a theoretical basis for the selective regulation of pore structure and offers guidance for the industrial production of high-purity quartz sand.

Declarations

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Author Contribution

Conceptualisation, methodology, investigation, data curation, writing — original draft: Y.G.; experimental validation, data curation: N.C. and R.D.; methodology, resources, investigation, data curation: G.S.; experimental validation, analysis: W.X. and X.X.; conceptualisation, validation, investigation: J.C. All authors reviewed and approved the final manuscript.

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Data availability

No datasets were generated or analysed during the current study.

References

1. Yu ZX, Luo DW, Zhao K, Rong K, Deng JB, Gao ZJ (2026) High-purity quartz sand from mineral purification: a review on process, mechanism, and environmental strategies. Miner Eng 235:109801. https://doi.org/10.1016/j.mineng.2025.109801
2. Wang T, Zhang YM, Zhang YS, Gu FH, Peng YW, Yao SY (2024) Quartz fluid inclusions and trace elements in the Lailisigaoer porphyry Mo deposit, Western Tianshan, Xinjiang: fluid evolution and metallogenic mechanism in magmatic-hydrothermal system. Ore Geol Rev 169:106081. https://doi.org/10.1016/j.oregeorev.2024.106081
3. Zhang RY, Tang CH, Ni W, Yuan J, Zhou Y, Liu XL (2023) Research status and challenges of high-purity quartz processing technology from a mineralogical perspective in China. Minerals 13(12):1505. https://doi.org/10.3390/min13121505
4. Li SH, Liang ZY, Zhou WT, Zhou W, Chen J (2024) Review of upscale quartz sand purification process: from raw material properties to efficient technologies. Mineral Process Extractive Metall 133(2):171–190. https://doi.org/10.1177/25726641241301583
5. Ibrahim SS, Selim AQ, Hagrass AA (2013) Gravity separation of silica sands for value addition. Part Sci Technol 31(5):590–595. https://doi.org/10.1080/02726351.2013.800930
6. Ren ZJ, Liu Z, Liu YH, Gao HM, Wu FD, Song YH (2023) The effects of calcination-water quenching on quartz purification and its mechanism. Min Metall Explor 40(6):2519–2527. https://doi.org/10.1007/s42461-023-00786-2
7. Jeyaprabha B, Elangovan G, Prakash P (2016) Effects of elevated temperature and water quenching on strength and microstructure of mortars with river sand substitutes. Constr Build Mater 114:688–698. https://doi.org/10.1016/j.conbuildmat.2016.03.189
8. Li FF, Jiang XS, Zuo QX, Li JW, Ban BY, Chen J (2021) Purification mechanism of quartz sand by combination of microwave heating and ultrasound-assisted acid leaching treatment. Silicon 13(2):531–541. https://doi.org/10.1007/s12633-020-00457-7
9. Song WF, Jiang XS, Chen C, Ban BY, Wan SM, Chen J (2023) Purification of quartz via low-temperature microwave chlorinated calcination combined with acid leaching and its mechanism. Silicon 15(2):971–981. https://doi.org/10.1007/s12633-022-01749-w
10. Wei ZL, Li YB, Li PY, Pan L, Hu XL, Gu YX, Tian Y (2023) Migration mechanisms of Al^{3+}/Li^{+} lattice impurities during phase transition from α-quartz to β-quartz: an implication for purification of high-purity quartz. Minerals 13(10):1280. https://doi.org/10.3390/min13101280

11. Xie YP, Li SQ, Pan XD, Li YK, Zhang A (2023) Recent advances in the marketing, impurity characterization and purification of quartz. Minerals Mineral Mater 2(4):16. https://doi.org/10.20517/mmm.2023.17
12. Long HL, Zhu DQ, Pan J, Li SW, Yang CC, Guo ZQ (2024) Advanced processing techniques and impurity management for high-purity quartz in diverse industrial applications. Minerals 14(6):571. https://doi.org/10.3390/min14060571
13. Wei ZL, Li YB, Li PY, Pan L, Li WQ, Hu XL, Gu YX (2024) New insight into the plastic deformation mechanisms during the SiO_2 phase transition process. RSC Adv 14(9):6262–6269. https://doi.org/10.1039/d3ra07633d
14. Guo WD, Lu HL, Zhang ZY, Jiang L, Wu HJ, Liu DF, Chi R (2024) Crystal structure transformation and lattice impurities migration of quartz during chlorine roasting. Int J Min Sci Technol 34(6):1465–1474. https://doi.org/10.1016/j.ijmst.2024.09.004
15. Khalifa M, Ouertani R, Hajji M, Ezzaouia H (2019) Innovative technology for the production of high-purity sand silica by thermal treatment and acid leaching process. Hydrometallurgy 185:204–209. https://doi.org/10.1016/j.hydromet.2019.02.010
16. Zhang HW, Guo SJ, Wu JJ, Wu DD, Wei KX, Ma WH (2021) Effect of quartz crystal structure transformations on the removal of iron impurities. Hydrometallurgy 204:105715. https://doi.org/10.1016/j.hydromet.2021.105715
17. Yang CQ, Li SQ, Bai JX, Han SS (2018) Advanced purification of industrial quartz using calcination pretreatment combined with ultrasound-assisted leaching. Acta Geodynamica et Geomater 15(2):187–195. https://doi.org/10.13168/AGG.2018.0014
18. Lin M, Pei ZY, Li YB, Liu YY, Wei ZL, Lei SM (2018) Separation mechanism of lattice-bound trace elements from quartz by KCl-doping calcination and pressure leaching. Miner Eng 125:42–49. https://doi.org/10.1016/j.mineng.2018.05.029
19. Wang SJ, Yu DS, Ma C, Wei FS, Zhang HQ (2024) A new insight into the influence of fluid inclusions in high-purity quartz sand on the bubble defects in quartz glass: a case study from vein quartz in the Dabie Mountain. Minerals 14(8):794. https://doi.org/10.3390/min14080794
20. Guo Y, Cheng NS, Ding R, Chen JH, Shu LX, Xu W, Shi GL (2025) The impact of ultra-low temperature quenching treatment on the pore structure of natural quartz sand. Minerals 15(1):52. https://doi.org/10.3390/min15010052
21. Johnson SE, Song WJ, Cook AC, Vel SS, Gerbi CC (2021) The quartz α↔β phase transition: does it drive damage and reaction in continental crust? Earth Planet Sci Lett 553:116622. https://doi.org/10.1016/j.epsl.2020.116622
22. Klimenko NN, Mikhailenko NY, Delitsin LM, Sigaev VN (2021) Effect of particle size of quartz sand filler on microstructure and strength of alkali-activated slag-based materials. Arab J Sci Eng 46(5):4337–4352. https://doi.org/10.1007/s13369-020-04935-3
23. Xia M, Yang XY, Hou ZH (2024) Preparation of high-purity quartz sand by vein quartz purification and characteristics: a case study of Pakistan vein quartz. Minerals 14(7):727. https://doi.org/10.3390/min14070727
24. Wang SR, Huang Y (2022) Experimental study on the shear characteristics of quartz sand exposed to high temperatures. Acta Geotech 17(11):5031–5041. https://doi.org/10.1007/s11440-022-01603-8
25. Zhang C, Wang FZ, Hu LB, Jiang F, Huang GQ (2024) Experimental investigation on the mechanical behavior and fracture mechanism of sandstone after heat treatment. Bull Eng Geol Environ 83(6):220. https://doi.org/10.1007/s10064-024-03717-7
26. Zhao Y, Du C, Bi J, Li JS, Wang CL (2022) Influence of multiple porous structure evolution on physical-mechanical properties and damage mechanism of sandstone heated continuously/periodically with the identical heating time. Bull Eng Geol Environ 81(12):507. https://doi.org/10.1007/s10064-022-03020-3
27. Götze J, Pan YM, Müller A (2021) Mineralogy and mineral chemistry of quartz: a review. Mineral Mag 85(5):639–664. https://doi.org/10.1180/mgm.2021.72
28. Zaichuk AV, Amelina AA, Hordieiev YS (2024) Cobalt-containing diopside pigments based on granulated blast furnace slag. Pigm Resin Technol 53(5):624–630. https://doi.org/10.1108/PRT-10-2022-0112

29. Gui YY, Chen XT, Zhu XX, Fu H, Jiang XS, Sun JF, Wang L, Ban BY, Chen J (2025) Quartz lattice Al impurity removal: a simple annealing-leaching process and analysis. Mater Lett 391:138478. https://doi.org/10.1016/j.matlet.2025.138478