

Article

Multimodal Analysis Unveils the Correlation Between Graphite Anode Characteristics and Operational Longevity in Pr/Nd Rare Earth Metals Electrolysis

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Abstract

The service life of graphite anodes—key consumables in the Pr/Nd fluoride molten salt electrolysis process—directly governs production continuity, cost-efficiency, and supply chain stability. This study systematically evaluated five industrial-grade anodes produced from different raw materials and processes. Multimodal characterization—combining macroscopic and microscopic morphology, SEM/EDS, XRD, Raman, and physical property analysis—was employed to correlate initial anode properties with corrosion-induced morphological and mass changes during electrolysis. The results show that the raw material quality and preparation methods synergistically regulate both the crystal structure and microstructure, thereby governing the corrosion behaviour and mass loss. Anodes #2 and #3, which were fabricated from high-quality petroleum coke and subjected to full densification and graphitization, exhibited high graphitization (93.7–94.5%), large crystallites (59.6–64.5 nm), minimal defects (low I_D/I_G), and suppressed microporosity, leading to the lowest mass loss (10.2 ± 0.8 kg and 10.6 ± 0.9 kg). In contrast, anodes #1, #4, and #5, made from recycled graphite without graphitization, contained abundant structural defects and large pores and led to greater morphological changes and quality losses. Moreover, for recycled graphite anodes, the presence of large pores and cracks is one of the important reasons for their failure. This work clarifies the “process–microstructure–mass loss” relationship in graphite anodes for Pr/Nd electrolysis, offering key insights for designing high-performance anodes and advancing sustainable rare earth production.

Keywords: graphite anode; fluoride molten salt; Pr/Nd metal; corrosion

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1. Introduction

Praseodymium/Neodymium (Pr/Nd), a rare earth element, serves as a critical strategic resource for the advancement of new energy, new materials, and high end manufacturing sectors worldwide. It is essential for producing permanent magnets, alloys, catalysts, and glass colorants. As “dual carbon” goals are pursued and fields such as new energy vehicles, wind power generation, industrial automation, robotics, and consumer electronics expand, the demand for Pr/Nd increases steadily. The global demand for Pr/Nd oxide is projected to reach 120,000 tons by 2026 [1]. Currently, oxide-fluoride molten salt electrolysis

is the predominant technology for the industrial production of Pr/Nd metals and alloys, and the stability and cost-effectiveness of this process directly influence the safety assurance capabilities of the rare earth supply chain [2]. In this process, the electrolytic cell features a steel shell lined with refractory materials, a graphite crucible, and the anode consists of graphite, whereas the cathode is made from a high temperature resistant tungsten rod. The molten salt system serves as a reaction medium for the migration and discharge of Pr/Nd ions; however, it imposes stringent requirements on the performance of electrolytic cell components because of its high fluorine content and strong oxidizing properties at elevated temperatures [3,4].

Graphite materials exhibit excellent electrical conductivity and chemical stability, making them the primary electron transport channels and essential carriers for electrochemical reactions in the Pr/Nd electrolysis process [5]. The service life of graphite anodes is directly linked to production continuity and cost management. Current industrial practices indicate that the average service life of graphite anodes is approximately 48 h. Abnormal wear and tear, such as fractures, slag shedding, and excessive corrosion, results in consumption costs comprising 25% to 30% of the total expenses of Pr/Nd electrolysis. The graphite anodes used in Pr/Nd electrolytes experience a threefold deterioration effect: “electrochemical consumption, high temperature oxidation, and fluoride salt erosion.” Electrochemical consumption arises from both anode and side reactions, which contribute 60% to 65% of the overall consumption. High temperature oxidation primarily occurs in the “cavitation zone,” where atmospheric air and electrolytic products such as CO and CO₂ synergistically accelerate the oxidation of carbon within the graphite pores, contributing 30% to 35% of the total consumption. Fluoride salt etching exacerbates structural deterioration by facilitating the penetration of molten salt and reacting with the carbon at the edges of graphite. The cumulative effects of these deterioration processes make the performance of the graphite anode performance a critical bottleneck limiting service life [6,7]. Moreover, molten salt electrolysis has proven to be a direct and versatile route for synthesizing functional graphitic electrodes. For example, this technique can generate a MnO_x–graphitic carbon composite from CO₂ in molten Li₂CO₃ for battery applications, and produce a sustainable nanoscale CO₂-derived carbon support for a highly active Pt catalyst (Pt/CO₂-C) [8,9]. These studies highlight the versatility of molten salt electrolysis in tailoring carbon materials, which underscores the importance of understanding anode behaviour in specific systems such as Pr/Nd electrolysis.

Currently, research has been conducted on the behaviour of graphite anodes in Pr/Nd electrolytes. For example, Lin et al. reported through industrial investigations that the service life of graphite anodes is closely related to their installation position and whether it is standardized [10]. By adding Pr₆O₁₁ to catalyse the graphite electrode, Wang et al. reduced the graphite layer spacing, increased the graphitization degree to 95.06%, and reduced the resistivity to 5.0 μΩ m [11]. Xue et al. [12] conducted onsite research on the consumption of graphite anodes during the electrolysis of rare earth molten salts to analyse their failure mechanism and experimentally studied the protective characteristics of impregnated borates. Chemical erosion and oxidation promote each other. Both damage mechanisms involve active reactions within the pores, causing the shedding of graphite particles and leading to contamination by molten salts and rare earth metals. Borate impregnation can fill the pores of graphite and cover the anode surface, but the protective effect is lost over time. Studies have carried out surface coating treatment on graphite anodes and simulation studies, indicating that coating treatment is a relatively effective extension method [13–15].

Most studies have primarily examined how electrolytic parameters, such as temperature and current density, affect the extraction of Pr/Nd. Existing research predominantly

employs specific types of graphite but has not systematically investigated how initial performance variations in graphite anodes affect the Pr/Nd electrolysis process. Furthermore, the mechanisms of performance degradation and failure in graphite anodes following the electrolytic degradation reaction remain poorly understood. Significant scientific and technological challenges persist regarding the interactions between the performance regulatory mechanisms of graphite anodes and electrolytes. Consequently, an in depth exploration of the initial performance of graphite anodes, their behavioural evolution during the electrolysis of fluoride molten salts, identification of key factors impacting electrolysis efficiency, and analysis of the performance degradation mechanisms of graphite anodes after electrolysis are crucial for elucidating the effects of surface and interface characteristics, as well as the corrosion behaviour of graphite anodes within molten salt systems. The research results not only help reveal the failure mechanism of graphite anodes and fill the research gap concerning the influence of the performance differences in graphite anodes in rare earth electrolysis but also provide a scientific basis for the development of high performance anode materials and the optimization of rare earth electrolysis processes, further promoting the upgrading of the rare earth metallurgy industry towards low consumption, high quality, and environmental protection.

2. Materials and Methods

2.1. Materials

Five industrial graphite anodes, each comprising ten pieces, were selected based on varying processes and raw material compositions; detailed information is shown in Table 1. Each component has an arc shaped structure, featuring an inner ring radius of approximately 125 mm, an outer ring radius of approximately 215 mm, a height of approximately 500 mm, and a mass of approximately 20 kg.

Table 1. Information on raw materials, processes, and sources of graphite anodes.

Anodes	Raw Materials	Processes	Sources
#1	Recycled graphite anodes: Graphite chips, and coal tar pitch as the binder.	After pretreatment such as crushing, screening and purification, kneading, Extrusion molding, baking, and mechanical processing; No impregnation, re-roasting, graphitization treatment.	Fangda Carbon New Material Co., Ltd., Lanzhou, China.
#2	Common petroleum coke, the aggregate particle size is approximately 2–4 mm, 4–8 mm.	Crushing, screening, kneading, extrusion molding, high-temperature calcination, impregnation, re-roasting, graphitization, mechanical processing.	
#3	High-quality petroleum coke, the aggregate particle size is approximately 0.5–2 mm.	Crushing, screening, kneading, Isostatic pressing molding, high-temperature calcination, impregnation, re-roasting, graphitization, mechanical processing.	
#4	Recycled graphite anodes:	After pretreatment such as crushing, screening and purification, kneading, extrusion molding, baking, and mechanical processing; No impregnation, re-roasting, graphitization treatment.	Kangda New Materials Co., Ltd., Jiexiu, China.
#5	High-power graphite powder, coal tar pitch is used as the binder.		Aohua New Materials Co., Ltd., Xiangcheng, China.

The entire experiment was conducted in a continuously operating industrial-grade 8 kA open-type electrolytic cell with a cathode inserted. The core components of this electrolytic cell include: a cylindrical graphite crucible, an anode assembly composed of four arc-shaped graphite sheets, a tungsten cathode rod placed on the central axis of the crucible, and a metal receiver installed at the bottom of the crucible. The single effective usage time of each graphite anode is approximately 49 h. The electrolysis process uses a Pr/Nd–lithium fluoride molten salt system, with a ratio of Pr/Nd to lithium fluoride of 8:1. The electrolytic current is (7000 ± 200) A; the temperature of the molten salt is 1050 ± 30 °C; the cathode current density is $(5\text{--}6)$ A/cm²; and the anode current density is $(1.7\text{--}1.9)$ A/cm².

2.2. Analysis and Testing Methods

The basic performance of the original graphite anode was tested according to reference standards: GBT 24528–2009 for volume density determination of carbon materials [16], GBT 1431–2019 for compressive strength determination [17], GBT 3074.4–2016 for the coefficient of thermal expansion (CTE) [18], GBT 24525–2009 for resistivity determination [19], GBT 24529–2009 for apparent porosity determination [20], and GBT 22588–2008 for measuring thermal diffusivity or conductivity via flash emission [21].

Macroscopic and microscopic characterization: Before and after the experiment, a macroscopic appearance comparison of the entire graphite anode was conducted (photographed with a high-definition digital camera, surface corrosion, cracks, and wear conditions were recorded), and the samples were weighed via an electronic balance. Parallel weighing was carried out three times to eliminate random errors. The anode was subsequently cut into blocks 10 mm × 10 mm × 5 mm in size. The microscopic morphology was observed via a digital microscope and a scanning electron microscope (TESCAN MIRA 3SEM). The SEM acceleration voltage was 15 kV, the working distance was 8 mm, and the secondary electron imaging mode was adopted. The elemental composition and distribution were analysed in combination with an attached energy spectrometer (BRUKER XFlash 6130), and more than three different regions were selected for the test.

XRD analysis: Graphite anode samples were ground in an agate mortar to achieve a particle size of less than 75 µm. The XRD patterns of the powdered graphite anodes were obtained via a D8 ADVANCE powder X-ray diffractometer with a step size of 0.02°, a scanning rate of 10°/min, and a scan range from 20° to 80°. The XRD patterns were segmented and fitted via MDI Jade 6.5 software. Following background noise removal, the interplanar spacing (d_{002}) was calculated via the Bragg equation. The degree of graphitization (g) was determined via the Franklin equation: $g = (0.3440 - d_{002}) / (0.3440 - 0.3354) \times 100\%$, where 0.3440 nm corresponds to the d_{002} value of amorphous carbon and 0.3354 nm represents the d_{002} value of ideal graphite. The grain size (L_c) was calculated via the Scherrer equation ($L_c = K\lambda / (\beta \cos\theta)$, $K = 0.89$, where β is the (d_{002}) peak half-height width, in radians) [22–24].

Raman Spectroscopy Analysis: Raman spectra of the powdered graphite anodes were obtained via a Renishaw In Via Reflex Raman spectrometer (UK). The test parameters were as follows: 532 nm semiconductor laser, spectral scanning range of 1000–3500 cm^{-1} , and resolution of 1 cm^{-1} . The Raman spectra were baseline-corrected via Origin 2021 software to identify the D peak ($\sim 1350 \text{ cm}^{-1}$) and the G peak ($\sim 1580 \text{ cm}^{-1}$). The intensity ratio (I_D/I_G) between the D and G peaks were then calculated. This ratio exhibited a positive correlation with the density of graphite defects and was employed to quantitatively assess the changes in defects within the electrolytic graphite anodes [25,26].

2.3. Statistical Analysis

All the data are presented as the mean $\pm$ standard deviation (SD) of more than three experiments. All the statistical results presented were analysed via one-way analysis of variance via Origin 2021 software.

3. Results

3.1. Morphological Changes in the Corroded Graphite Anode

A schematic diagram of the production of rare earth metals by fluoride molten salt electrolysis is shown in Figure 1. The degradation of graphite anodes in fluoride molten salts primarily occurs through electrochemical processes and oxidation, resulting in significant alterations in the anode shape and eventual failure. During the electrolysis process, graphite anodes are affected by temperature and mainly undergo two types of chemical reactions [27,28].

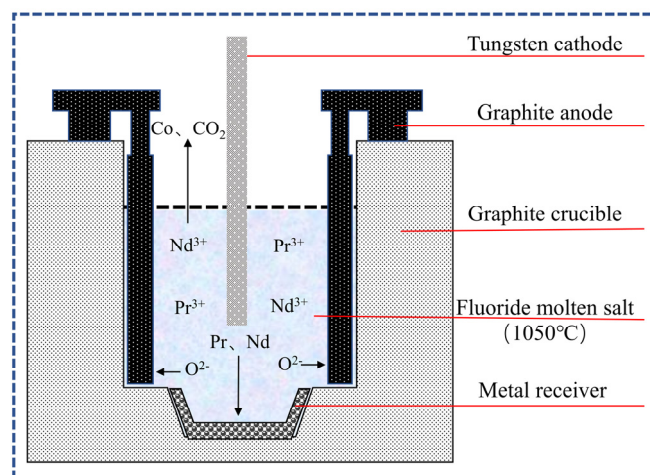


Figure 1. Schematic diagram of the production of rare earth metals by fluoride molten salt electrolysis.

(1) Primary electrochemical reactions:



(2) Secondary chemical reactions:



Changes in the morphology of graphite anodes after corrosion can be assessed at both the macroscopic and microscopic levels, revealing variability among different anodes. This variation reflects the complexity of the corrosion processes and the diverse factors influencing them. Figure 2 shows that the surface of the original graphite sample is flat, characterized by a uniform shape with distinct edges and corners. After operation, significant corrosion marks become evident on the surface compared with those on the original graphite. The edges and corners completely eroded away, resulting in a rough surface that exposed the internal aggregates of the electrode. In some areas, material shedding and morphological changes were still observed. Overall, the anode was larger, wider, and thicker at the top, tapering to a smaller, narrower, and thinner base. These observations indicate that fluoride molten salts exert a significant corrosive effect on graphite electrodes. Furthermore, significant differences exist in the macroscopic damage levels of the graphite anodes identified by their respective post corrosion values. Specifically, anode #2 has the smallest shape change, whereas anode #5 has the greatest shape damage. This suggests inherent differences among the graphite electrodes that influence the corrosion outcomes.

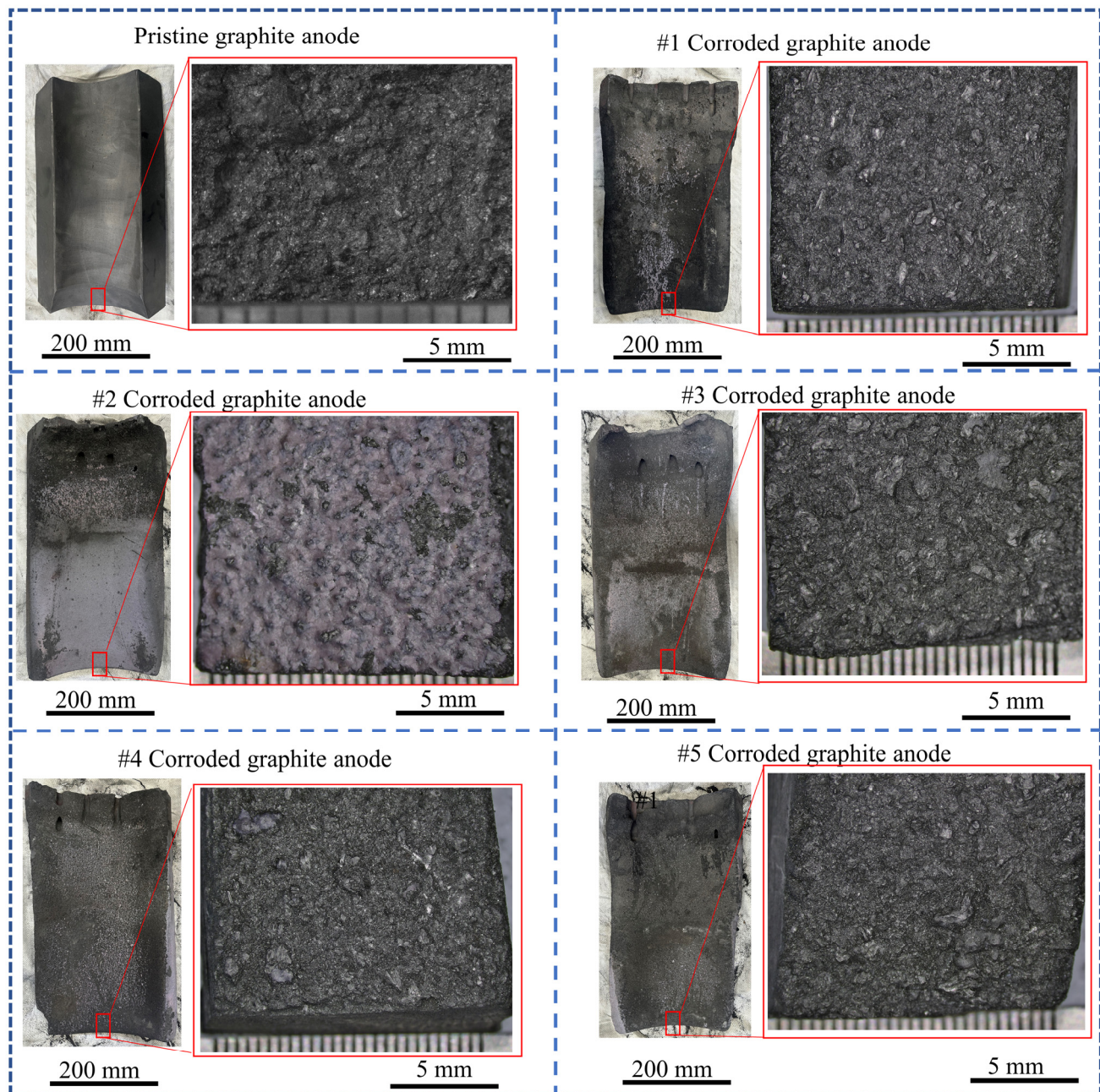


Figure 2. Changes in the morphology of the graphite anode before and after corrosion.

Figure 2 shows a local magnification of the bottom of the graphite anode and the surface morphology observed with a digital microscope, revealing the morphology of the untreated sample after corrosion. The surface of graphite anode #1 exhibited a rough granular morphology characterized by a dispersed particle distribution and low surface flatness. This indicates erosion of the surface material and structural damage to the graphite caused by corrosion. Graphite electrode #2 displays unique characteristics that distinguish it from other electrodes, particularly in colour and morphology. A specific layer of corrosion products forms during the corrosion process, leading to a distinctive texture and colour on the surface. The surface of graphite electrode #3 is another rough granular structure; however, its particle size and distribution differ from those of #1, further demonstrating the uneven corrosion process. Graphite electrodes #4 and #5 also exhibit rough particle morphologies. However, variations in particle fineness and stacking configurations reflect changes in the degree of corrosion and microscopic morphology evolution as corrosion

progresses. Overall, these findings suggest that the corrosion of fluoride molten salts on graphite is significant at the microscopic scale, resulting in damage to the graphite surface microstructure, and that the inherent fine and regular structure of the graphite has been lost.

3.2. Mass Change of the Graphite Anodes

In the Pr/Nd electrolysis process, the electrolysis temperature is typically maintained at 1050 °C. The molten fluoride salt reacts with graphite, producing gaseous by products, including CF₄, CO, and CO₂, resulting in the ongoing consumption of the graphite anode. After corrosion, the surface of the graphite anode retains a cooled and solidified salt layer. As the mechanical removal of this salt layer could damage the existing corrosion structure, this study indirectly assessed the extent of corrosion progress by analysing the changes in mass of the graphite anode. Specifically, the mass of the graphite anode was measured both before and after corrosion. The corrosion induced mass change was then calculated via the following formula: Corrosion mass change = Original mass – Mass after corrosion. The results are presented in Table 2.

Table 2. Corrosion mass changes of the graphite anodes.

Anodes	Mass of Pristine Anodes (kg)	Mass of Corroded Anodes (kg)	Corrosion Mass Change (kg)
#1	19.2 ± 0.2	7.9 ± 0.8	11.3 ± 0.8
#2	20.6 ± 0.3	10.4 ± 0.7	10.2 ± 0.8
#3	20.7 ± 0.2	10.1 ± 0.9	10.6 ± 0.9
#4	19.3 ± 0.3	8.9 ± 0.5	10.4 ± 0.6
#5	19.1 ± 0.3	7.8 ± 0.5	11.3 ± 0.6

The data are presented as the means ± SD (*n* = 10).

Based on the measured values, a consistent trend in mass change was observed. Anodes #1 and #5 presented the largest mass change of 11.3 kg, suggesting more substantial material loss under the given conditions. In contrast, Anodes #2 and #4 showed relatively lower mass changes of 10.2 kg and 10.4 kg, respectively, whereas anode #3 presented an intermediate value of 10.6 kg. It is critical to note that the absolute differences between these samples are within a narrow range (approximately 5–10% of the average mass change). In the absence of a comprehensive statistical analysis of the standard errors, the absolute significance of these differences should be interpreted with caution. Nonetheless, the observed variations indicate potential differences in the corrosion behaviour of the graphite anodes.

The differences in mass change have direct implications for industrial operation. A greater mass change per operating cycle, as seen in Anodes #1 and #5, implies faster consumption of the anode matrix. To project these findings to an industrial context, a common practice is considered where an anode is replaced after losing half of its original mass to maintain operational performance and structural integrity. Under this premise, an anode with a lower mass change per cycle will require more cycles to reach this replacement threshold, thereby extending its service life.

In practical terms, for every 100 anodes of types #1, #5 consumed, only approximately 91 anodes of types #2, #4 would be required to achieve the same cumulative operating time. This 9% reduction in material consumption confers a direct cost advantage. Assuming an average anode mass of 20 kg and a market price of 15,000 RMB per tonne, the graphite material cost for 100 anodes of type #1, #5 anodes would be approximately 30,000 RMB. In contrast, only 91 anodes of type #2, #4, costing approximately 27,300 RMB, would be needed for the same total service output, resulting in a direct saving of 2700 RMB per 100-anode service cycle. In a large-scale facility with hundreds of anodes undergoing repeated cycles, this per-unit savings accumulates into substantial annual cost reductions.

Furthermore, the reduced replacement frequency minimizes operational interruptions and mitigates risks associated with anode handling, such as altered current distributions due to positioning deviations.

In summary, although the absolute differences in mass change measured in this experiment are relatively small, their cumulative effects on material consumption and operational stability in a continuous industrial setting are meaningful. Employing anodes with lower mass changes per cycle can contribute to reduced raw material expenditure, enhanced process predictability, and improved overall process economics for Pr/Nd electrolysis.

3.3. Influence of the Basic Physical Properties of Pristine Graphite Anodes

The failure behaviour of graphite anodes in Pr/Nd fluorinated molten salt electrolysis, namely, changes in morphology and quality, is fundamentally determined by their inherent physical properties. As mentioned in Section 2.1, these characteristics are the direct result of raw material selection and the manufacturing process. Table 3 presents the basic performance parameters of the five types of pristine graphite anodes, including the bulk density, compressive strength, thermal conductivity at room temperature, resistivity, apparent porosity, and coefficient of thermal expansion (CTE). The data from Table 3, when correlated with the corrosion morphology (Section 3.1) and mass loss (Section 3.2), reveal a definitive and causal relationship between the initial anode characteristics and its performance. Specifically, anodes #2 and #3 demonstrated superior longevity with minimal mass loss (10.2 ± 0.8 kg and 10.6 ± 0.9 kg, respectively) and morphological damage, whereas anodes #1 and #5 experienced the greatest mass loss (11.3 ± 0.6 kg) and severe structural deterioration.

Table 3. Basic properties of the pristine graphite anodes.

Anodes	Bulk Density (g/cm ³)	Apparent Porosity (%)	CTE ($\times 10^{-6}/^{\circ}\text{C}$)	Thermal Conductivity (Room Temperature) (W/(m·k))	Electrical Resistivity ($\mu\Omega\cdot\text{m}$)
#1	1.63 ± 0.02	19.07 ± 0.51	2.92 ± 0.10	30.61 ± 1.58	21.66 ± 1.15
#2	1.72 ± 0.02	16.82 ± 0.43	1.56 ± 0.08	120.73 ± 3.54	9.43 ± 0.33
#3	1.74 ± 0.02	13.80 ± 0.38	1.56 ± 0.08	151.37 ± 4.13	7.15 ± 1.08
#4	1.67 ± 0.02	19.22 ± 0.55	2.87 ± 0.09	29.43 ± 1.52	20.28 ± 1.10
#5	1.59 ± 0.02	16.90 ± 0.49	3.91 ± 0.12	35.05 ± 1.67	23.60 ± 1.74

The data are presented as the means $\pm$ SD ($n = 3$).

The dominant role of the bulk density and apparent porosity. Bulk density and apparent porosity directly regulate the penetration depth of corrosive fluoride molten salts—the primary step initiating both electrochemical corrosion and oxidative consumption (Equations (1)–(5), Section 3.1). Anodes #2 and #3, manufactured from high-quality or precisely sized petroleum coke and subjected to multiple cycles of impregnation and graphitization, achieved the highest bulk densities (1.72–1.74 g/cm³) and the lowest apparent porosities (13.80–16.82%). This dense, low-porosity microstructure functions as an effective barrier, largely confining corrosion reactions to the external surface. Their post-corrosion morphology—characterized by minimal damage with only shallow, isolated pores—corroborates this barrier effect and explains their correspondingly low mass loss. In contrast, anodes #1 and #4, produced from common petroleum coke or graphite powder without advanced densification treatments, exhibited significantly greater porosities (19.07–19.22%). In the case of anode #1, this porous network served as a continuous conduit for deep molten salt infiltration, resulting in extensive internal corrosion, a severely eroded granular surface, and the highest mass loss among all samples. Anode #4 presents an

instructive exception: despite its similarly high porosity, it sustained only moderate mass loss and retained a relatively intact microstructure. The underlying reasons for this deviation, related to its distinct surface pore structure, will be further explored in Section 3.6. Anode #5, with its notably low bulk density (1.59 g/cm^3), exhibited inherent structural weakness. Although its porosity was only moderate, this deficiency—combined with other detrimental properties—significantly contributed to its overall poor performance.

The elevated operating temperature of 1050°C renders thermal properties paramount in determining the service life of graphite anodes. Among these, the CTE serves as a key differentiator. Anodes #2 and #3, which underwent graphitization, exhibited an exceptionally low and identical CTEs of $1.56 \times 10^{-6}/^\circ\text{C}$, conferring outstanding dimensional stability and effectively suppressing the formation of thermal stress-induced cracks. In stark contrast, anode #5 displayed the highest CTE ($3.91 \times 10^{-6}/^\circ\text{C}$). Under repeated thermal cycles during operation, this pronounced expansivity induced substantial internal stresses, resulting in the wide surface cracks observed morphologically. These cracks subsequently acted as rapid infiltration pathways for the molten salt, markedly accelerating the degradation process. The thermal conductivity and electrical resistivity further govern the temperature and current distribution across the anode. Anodes #2 and #3, benefitting from graphitization, demonstrated high thermal conductivity ($>120 \text{ W/(m}\cdot\text{K)}$) and low electrical resistivity ($<9.5 \mu\Omega\cdot\text{m}$). These attributes promoted uniform heat dissipation and current flow, thereby preventing localized overheating that would otherwise exacerbate corrosive reactions. Conversely, anodes #1 and #4, which lacked graphitization, exhibited inferior thermal transport and higher resistivity.

The service life of graphite anodes is governed by the synergistic interplay of multiple physical properties, where microstructure and thermomechanical characteristics collectively determine corrosion resistance. Anodes #2 and #3 exemplify high-performance materials, where the combination of high bulk density ($>1.72 \text{ g/cm}^3$), low apparent porosity ($<16.82\%$), low CTE ($1.56 \times 10^{-6}/^\circ\text{C}$), and high thermal conductivity ($>120 \text{ W/(m}\cdot\text{K)}$) creates a robust barrier system. The dense microstructure restricts molten salt penetration, whereas the exceptional thermal stability prevents crack initiation under thermal cycling, resulting in minimal mass loss and morphological damage. In contrast, anodes #1 and #5 demonstrate how the absence of synergistic properties leads to accelerated degradation. Anode #1 suffers from the combined deficiencies of high porosity and poor thermal properties, creating interconnected corrosion pathways. Anode #5 exhibited the most severe degradation due to its extremely high CTE ($3.91 \times 10^{-6}/^\circ\text{C}$) coupled with low bulk density (1.59 g/cm^3), where thermal stress-induced cracking dominated the failure mechanism.

This analysis confirms that optimal anode performance requires a balanced combination of properties, where deficiencies in one characteristic cannot be fully compensated by excellence in another. The most critical parameter combination for prolonged service life involves high density, low porosity, and low thermal expansivity, which are achievable through advanced processing routes including graphitization and impregnation treatments.

3.4. Impact of Elemental Composition

The elemental composition of graphite anodes significantly influences their corrosion resistance by modulating the reaction pathways during corrosion. EDS, as a rapid and quantitative technique for elemental analysis, effectively characterizes carbon purity and impurity profiles in graphite, thereby providing a scientific basis for correlating intrinsic material properties with service life. The EDS results for the five pristine graphite anodes (#1–#5) are presented in Figure 3.

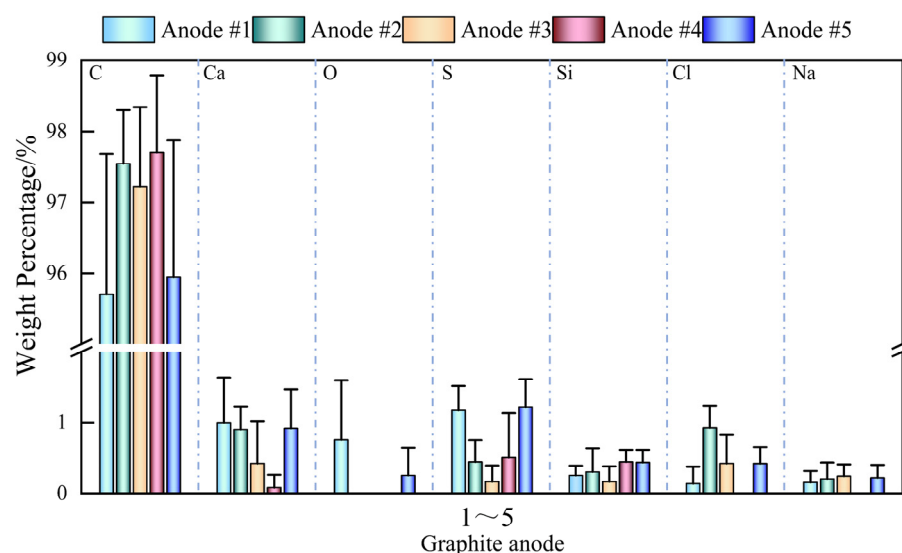


Figure 3. Distribution map of the main elements of the pristine graphite anodes. The data are presented as the means $\pm$ SD ($n = 3$).

Carbon, which serves as the matrix of graphite anodes, directly determines the structural integrity and corrosion resistance of a material. A higher carbon content generally corresponds to a more complete graphitic layered structure and superior corrosion resistance [29]. Anodes #2, #3, and #4, with higher carbon contents (#2 $\approx 97.5\%$, #3 $\approx 97.2\%$, #4 $\approx 97.7\%$), possess a structural foundation that contributes to their excellent corrosion resistance. In contrast, Anodes #1 ($\approx 95.5\%$) and #5 ($\approx 96.0\%$) have relatively lower carbon contents and significantly higher proportions of impurities, resulting in inferior corrosion performance.

Impurity elements (e.g., O, S, Cl, Ca, and Na) adversely affect the corrosion resistance of graphite anodes through mechanisms such as oxidative reactions, catalytic effects, and structural degradation. Higher impurity contents lead to more pronounced deterioration in corrosion resistance. Specifically: O exists primarily in the form of surface functional groups (e.g., -OH, and -COOH). In high-temperature molten salt environments, these groups can undergo oxidation reactions with F^- , accelerating the consumption and spallation of the anode surface. Anode #1, with a significantly higher O content ($\approx 0.8\%$) than the other samples do, is particularly susceptible to this degradation mechanism. Anode #5 contains elevated levels of S ($\approx 1.0\%$) and Cl ($\approx 0.5\%$). Under electrolysis conditions, these elements can form corrosive compounds such as SF_4 and Cl_2 , which induce a “gas erosion” effect within the graphite matrix, exacerbating structural degradation. Compared to anodes #2, #3, and #4, anodes #1 and #5 also have noticeably higher contents of metallic impurities (Ca, Na). These metallic species can catalyse carbon gasification reactions and facilitate the penetration of molten salt into the graphite interior, further compromising the resistance of the material [30,31].

3.5. Impact of the Crystallography and Molecular Structure

The impact of the crystal structure characteristics on the service life. The diffraction peaks of the graphite anodes are essentially identical, all displaying significant peaks at 26.5° , 44.4° , 54.5° , and 77.4° , which correspond to the (002), (101), (004), and (110) crystal planes, respectively. The peak angles align with the 2θ values from the graphite spectrum identified as PDF00-041-1487. Figure 4 shows the variations in the relative intensity of the intensity's peaks among the five anodes. The (002) peak of anode #3 is sharper, suggesting a higher degree of crystallization for this graphite anode. The XRD pattern revealed no additional diffraction peaks from other crystalline phases.

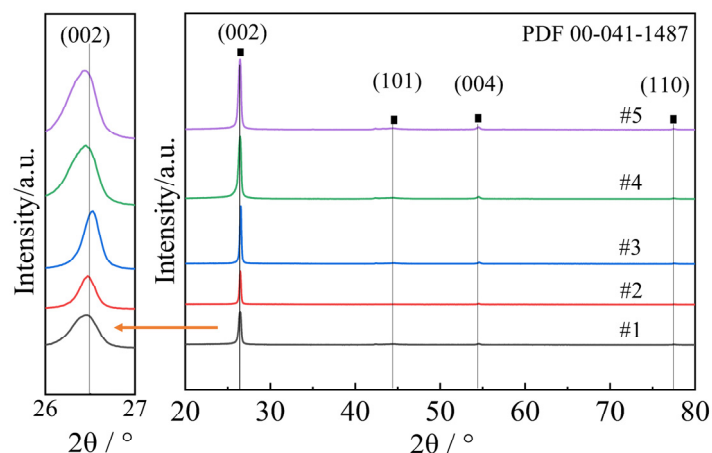


Figure 4. XRD patterns of the pristine graphite anodes.

The crystal plane spacing (d_{002}), degree of graphitization, and grain size of the graphite anode were calculated via the Bragg formula, Franklin equation, and Scherrer formula [32,33]. The results are detailed in Table 4. By comparing the crystal plane spacing (d_{002}) of the graphite anode to that of ideal hexagonal crystal system graphite, the degree of graphitization (g) for the five types of graphite anodes was calculated. A higher degree of graphitization indicates a more complete graphite crystal structure within the anode, characterized by fewer defects and active sites. Consequently, impurities can be removed, leading to increased purity of the anode, reduced specific resistance, and increased electrical conductivity and true specific gravity. The grain size influences the electrical conductivity and mechanical properties of the graphite anode.

Table 4. Lattice constants of the pristine graphite anodes.

Graphite Anodes	Crystal Plane Spacing d_{002} (nm)	Degree of Graphitization (%)	Grain Size (nm)
#1	0.33658 ± 0.00021	86.2 ± 1.5	29.9 ± 3.6
#2	0.33594 ± 0.00015	93.7 ± 0.8	64.5 ± 3.1
#3	0.33588 ± 0.00012	94.5 ± 0.9	59.6 ± 2.8
#4	0.33654 ± 0.00018	86.6 ± 1.8	30.2 ± 2.3
#5	0.33667 ± 0.00023	85.3 ± 1.3	16.8 ± 2.1

The data are presented as the means $\pm$ SD ($n = 3$).

The crystal plane spacing d_{002} is inversely correlated with the degree of graphitization, thereby confirming the impact of crystal integrity on corrosion. The ideal value of d_{002} for the hexagonal crystal system of graphite is 0.3354 nm. In the experiment, the d_{002} value for anode #3 is 0.33588 nm, the smallest among the samples, indicating that its interlayer arrangement is the most compact and closest to the ideal value. A smaller d_{002} indicates a stronger interlayer interaction force, making it more difficult for molten salt ions to penetrate the graphite interlayers and initiate corrosion. Consequently, anodes #2 and #3 display enhanced corrosion resistance. In contrast, the larger d_{002} values for anodes #1, #4 and #5 result in wider interlayer gaps, making them more susceptible to the penetration of molten salt ions, thereby accelerating the degradation of the graphite structure and reducing its lifespan.

The degree of graphitization, a key indicator of graphite crystal integrity, directly influences the chemical stability of the anode. A higher degree of graphitization typically indicates a more complete crystal structure, a more regular layered arrangement, fewer defects (e.g., edge dislocations and vacancies), and an enhanced resistance to fluoride molten salt corrosion [34,35]. The experimental data indicate that anode #3 has the highest degree of graphitization (94.5%) and the sharpest peak (002), suggesting a highly complete

crystal structure. Anode #2 ranks second with a graphitization degree of 93.7%, followed by #4 (86.6%) and #1 (86.2%), whereas #5 has the lowest degree at 85.3%. The manufacturing process of anodes #2 and #3 graphitization, characterized by high degrees of graphitization, feature layered structures that effectively hinder the penetration of molten salt ions and lower the likelihood of reactions between C and F. Consequently, their corrosion mass changes are relatively low. In contrast, anode #5, which has the lowest degree of graphitization, experiences a corrosion mass change of 11.3 kg.

The grain size is indirectly related to the service life, as it influences both the mechanical properties and the reaction interface [36]. According to the Scherrer formula calculations, anode #2 has the largest grain size (64.5 nm), followed by anode #3 (59.6 nm), anode #4 (30.2 nm), anode #1 (29.9 nm), and anode #5, which have the smallest grain size (16.8 nm). Larger grain sizes can improve the mechanical strength and crack resistance of graphite, thereby reducing the occurrence of stress-induced cracks during service [37]. Anode #2, which has the largest grain size, exhibited minimal shape change after corrosion, indicating superior structural integrity. Anode #3 has relatively large grains. Despite the presence of large pores on its surface, the intergranular bonding is robust, preventing severe structural collapse and resulting in a relatively low corrosion rate. Anode #5, with the smallest grain size and weak intergranular bonding, is susceptible to grain shedding under molten salt corrosion and thermal stress. This leads to significant surface roughness and material loss, resulting in severe morphological damage.

The Raman-active vibrational modes of graphite electrodes primarily fall within the wavenumber range of 1000–3000 cm^{-1} . For the detailed attribution of spectral generation, please refer to [38,39]. Notably, the D peak, located at approximately 1350 cm^{-1} , corresponds to structural defect-induced vibrations in graphite crystals associated with defects (e.g., edge defects, vacancies, and impurity doping) and disordered regions. The presence and intensity of the D peak indicate the level of defects in the graphite material. A higher density of defects correlates with greater D peak intensity. The G peak, found at approximately 1580 cm^{-1} , arises from the in-plane stretching vibrations of sp^2 hybridized carbon atoms in the graphite structure. This peak serves as a characteristic indicator of the structural order in the graphite crystal, with its intensity closely linked to the integrity and order of the material. Typically, a higher structural order results in greater G peak intensity. This peak occurs at approximately 2700 cm^{-1} and is a harmonic overtone of the D peak. The shape, position, and intensity of this peak provide insights into the interlayer stacking arrangement and the number of layers in graphite materials. For multi-layered stacked materials such as graphite, the characteristics of the 2D peak can be utilized to assess the orderliness of the layer structure [40,41]. Analysing the Raman spectra of the graphite electrodes, based on their crystal structure characteristics, revealed the results presented in Figure 5.

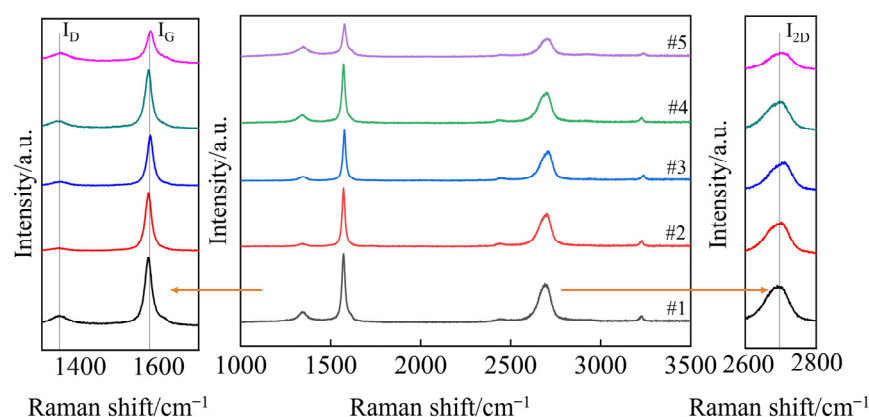


Figure 5. Raman patterns of the pristine graphite anodes.

The local magnification of the G peak on the left shows that the G peak positions of the five graphite anodes are clustered at approximately 1580 cm^{-1} . This observation indicates that all the samples exhibit typical in-plane vibration characteristics of graphitic sp^2 hybrid carbon. However, significant differences exist in the G peak intensity among the samples. Specifically, sample #5 has the weakest G peak intensity, followed by sample #3. In contrast, samples #1, #2, and #4 display significantly stronger G peak intensities. The characteristics of the graphite materials indicate that the strength of the G peak is positively correlated with the degree of order in the graphite crystals. This indicates that the graphite crystals of samples #1, #2, and #4 have a higher degree of order, and the in-plane arrangement of the carbon atoms is more regular. In contrast, the graphite crystals of sample #5 demonstrate relatively poor ordering and may contain more crystal distortions or disordered regions.

The intermediate spectrum diagram reveals that the appearance and intensity variation in peak D are critical factors contributing to graphite defects. The #1 graphite anode exhibited the most pronounced D peak, with a peak intensity significantly greater than that of the other samples. #5 has the second highest D peak intensity. The D peaks of the #2 and #3 graphite anodes and #4 are extremely weak, making them nearly impossible to observe. These results indicate that #1 and #5 exhibit the highest defect density, likely due to the substantial presence of edge defects, vacancies, or other structural imperfections. The #2 graphite anode has a relatively low defect density, indicating a more complete graphite crystal structure.

The magnified view of the 2D peak on the right reveals that the 2D peak positions of the five graphite anodes are approximately 2700 cm^{-1} ; however, their peak shapes and intensities differ significantly. The 2D peak of the #5 graphite anode has a relatively broad shape and weak intensity, which aligns with the expected characteristics of disordered mesoporous graphite structures. This finding further confirms the lack of order in the interlayer stacking of the #5 graphite anode. In contrast, the 2D peaks of the other graphite anodes display sharper and more intense characteristics, indicating more orderly interlayer stacking of graphite within these anodes. This observation corroborates the conclusions drawn from the G peak analysis.

The intensity of the D peak indicates the defect density, whereas the intensity of the G peak indicates the proportion of ordered structures. Consequently, a lower I_D/I_G ratio is correlated with a lower defect density [32,42]. Figure 6 shows the I_D/I_G ratios of the graphite anodes. Among these materials, the #2 graphite anode has the lowest I_D/I_G value, suggesting that its structure is relatively ordered with fewer defects. Owing to its superior structural integrity, the Pr/Nd electrolyte has fewer corrosion effects than the other anodes do. Conversely, graphite anodes with higher I_D/I_G values, such as the #5 anode, exhibit significant disorder and numerous defects. In the Pr/Nd electrolyte, defect areas are more susceptible to reactions with the electrolyte, leading to a greater likelihood and severity of corrosion. The disorder of graphite, as indicated by the I_D/I_G ratio, is a crucial factor affecting its corrosion behaviour in the Pr/Nd electrolyte. Increased disorder heightens susceptibility to corrosion.

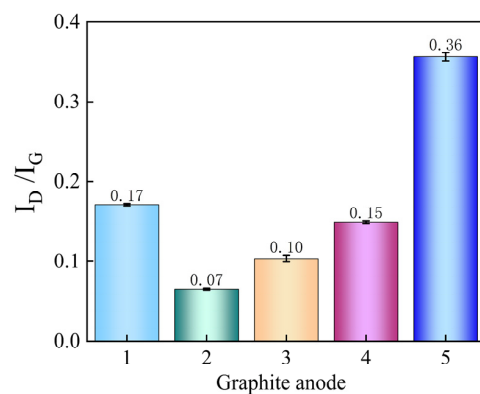


Figure 6. Raman I_D/I_G of the pristine graphite anodes. The data are presented as the means $\pm$ SD ($n = 3$).

3.6. Influence of Surface Macropores on Recycled Graphite Anodes

In a previous study, anodes #2 and #3 exhibited excellent overall performance, which can be attributed to the use of high-quality raw materials. By contrast, anodes #1, #4, and #5 were all produced from recycled graphite. Owing to their cost advantages, such recycled anodes have been widely adopted in the industrial production of rare earth metals via molten salt electrolysis. It is noteworthy that although these three types of recycled anodes had relatively similar basic physical properties—as evaluated in Section 3.3 (e.g., bulk density and apparent porosity)—their corrosion behaviours differed significantly. Specifically, the surface and cross-sectional morphologies of the three regenerated graphite anodes after corrosion are shown in Figure 7. The samples are from the bottom of the graphite anode, that is, the area where corrosion is most obvious for anodes #1 clearly has a loose and porous surface structure, with large voids and fragmented regions, indicating substantial material loss and severe structural degradation during corrosion.

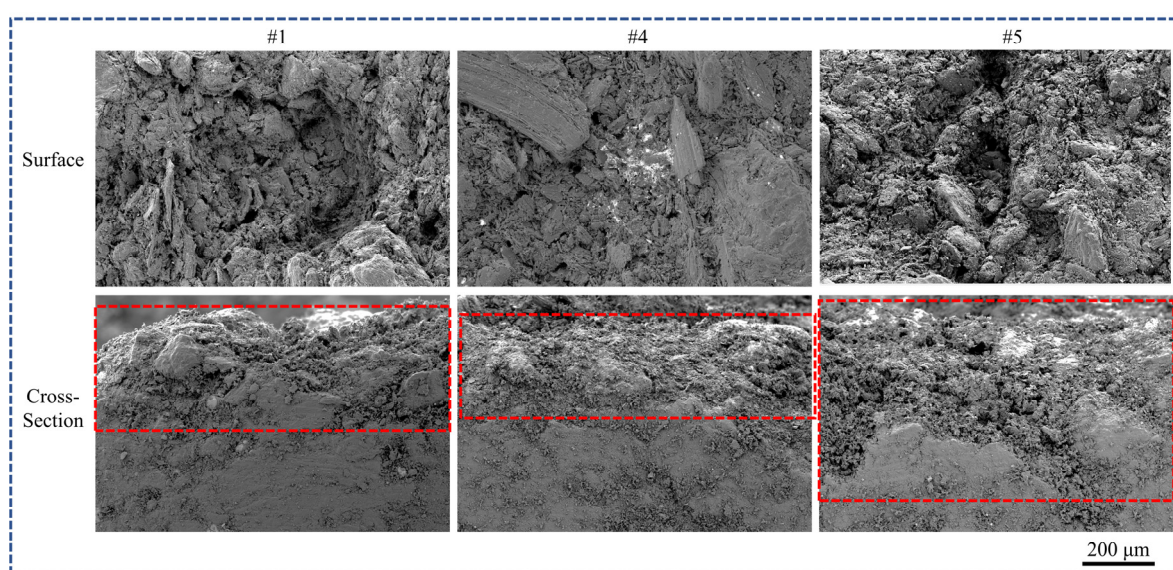


Figure 7. The surface and cross-sectional morphologies of three types of recycled graphite anodes after corrosion. The red dotted box in the figure indicates the thickness of the corrosion layer.

Anodes #4 display a relatively dense surface with visible corrosion traces; however, the extent of voids and fragmentation is significantly less than that of #1, demonstrating better structural integrity. Anodes #5 present an extremely rough and loose surface, characterized by severe fragmentation and high porosity, representing the most pronounced material loss and structural damage among the three samples. In cross-section, anodes #1 has a loose corrosion layer with a thickness ranging from approximately 140–360 μm . The presence of visible pores and an uneven interface with the substrate suggests deep internal structural damage caused by corrosion. Anodes #4 exhibit a relatively dense and uniform corrosion layer, approximately 150–300 μm thick, with a relatively flat interface to the substrate. Anodes #5 possess the most porous and nonuniform corrosion layer among the three samples, with thickness varying between approximately 200 and 500 μm .

To clarify the underlying mechanisms responsible for these behavioural differences, we performed a detailed comparison of the microscopic surface morphologies of the three recycled graphite anodes. Figure 8 displays representative SEM images of these pristine graphite anode samples. To ensure that the microstructural analysis was both representative and statistically meaningful, four distinct regions were examined for each anode. We focused particularly on the distribution of surface macropores (with sizes approximately $>20\ \mu\text{m}$) and cracks present on the pristine anodes. For clarity, selected

macropores and cracks are highlighted with red dashed circles in Figure 8 (only partially illustrated for representative purposes).

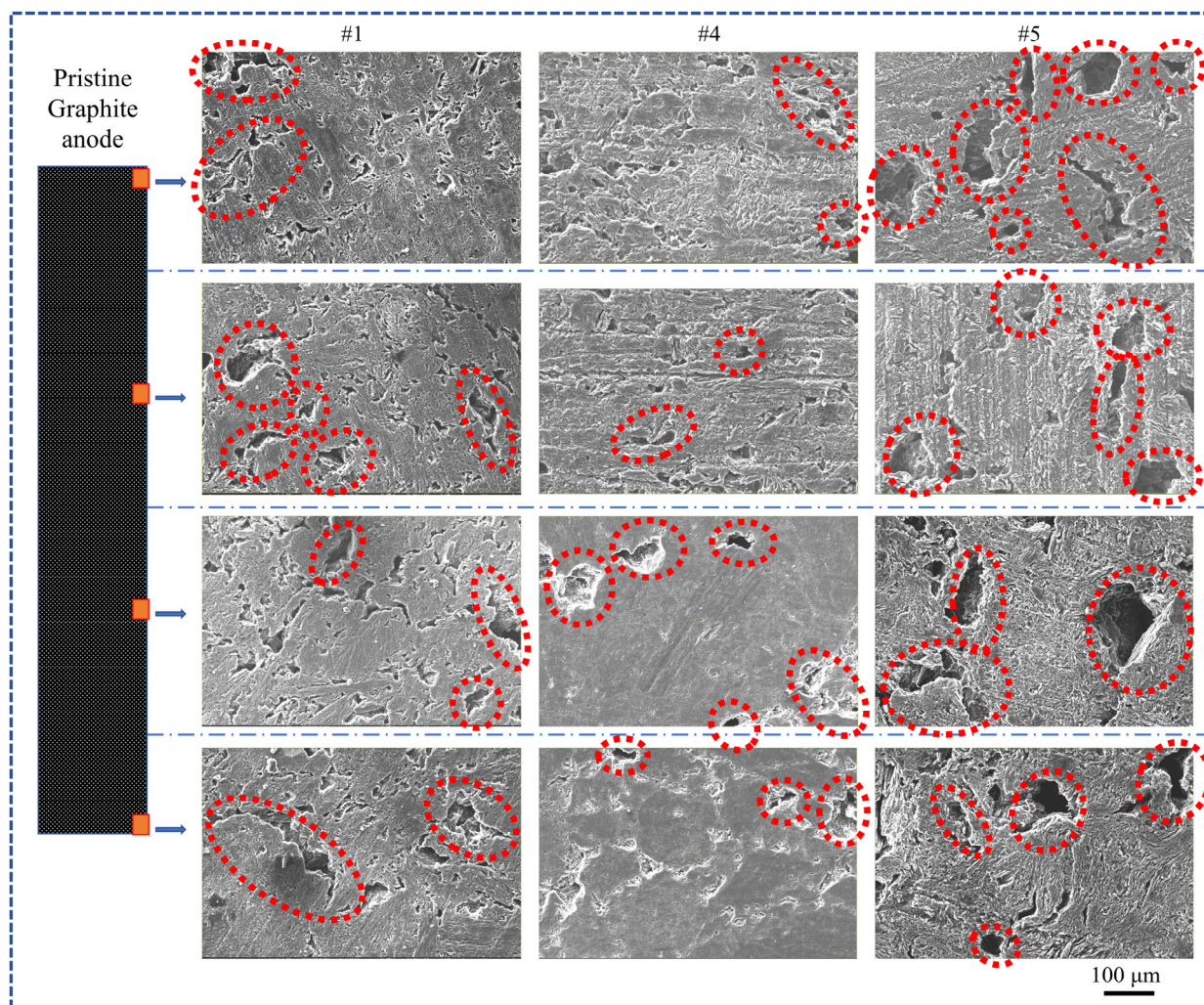


Figure 8. Pristine graphite anode corrosion surface macropores and cracks. The red dotted box highlights the large pores and cracks on the surface of the graphite anode.

A clear distinction should be made between the apparent porosity discussed in Section 3.3 and the surface macropores analysed here. Apparent porosity represents a macroscopic property that reflects the total volume of open pores within a material. In contrast, surface macropores and cracks are localized morphological features that are directly exposed to the molten salt environment. These defects can serve as preferential pathways, facilitating rapid penetration of corrosive media into the anode interior and significantly accelerating corrosion [43]. On the other hand, fine and uniformly distributed pores help restrict both the rate and depth of molten salt infiltration.

A systematic examination of Figure 8 reveals that the surface of anode #5 contains numerous interconnected wide cracks and macropores, which explains its pronounced corrosion failure. Anode #1 also exhibits visible cracks and irregular macropores, although with less connectivity than anode #5. In contrast, the surface of anode #4 is the densest and uniform, containing only isolated macropores. In addition, the colour of these large holes is light, and there is less obvious penetration. This ranking in terms of microstructural integrity is consistent with the macroscopic corrosion severity observed across the three anodes.

In summary, for recycled graphite anodes with similar bulk physical properties, the distribution characteristics of surface macropores and cracks are critical microstructural factors governing their corrosion behaviour. These findings provide valuable theoretical insight and practical guidance for selecting and optimizing recycled graphite anodes in industrial applications.

4. Conclusions

This study systematically elucidates the correlation between the initial characteristics of graphite anodes and their service life in Pr/Nd fluoride molten salt electrolysis through multimodal characterization and industrial-scale testing. The following conclusions are drawn, which align with and substantiate the findings presented in the Results and Abstract sections:

The raw material quality and manufacturing processes synergistically affect and determine anode performance. Anodes #2 and #3, produced from high-quality petroleum coke via complete impregnation, roasting, and graphitization, exhibited optimal properties, including high graphitization (93.7–94.5%), large crystallite sizes (59.6–64.5 nm), minimal structural defects (lowest Raman I_D/I_G ratios), high bulk density ($\geq 1.72 \text{ g/cm}^3$), and low apparent porosity ($\leq 16.82\%$). These characteristics collectively contributed to their superior corrosion resistance, as reflected by the lowest mass losses ($10.2 \pm 0.8 \text{ kg}$ and $10.6 \pm 0.9 \text{ kg}$) and minimal morphological degradation.

The integrity of the crystal structure is the predominant factor governing corrosion resistance. A high degree of graphitization, large grain size, and low defect density effectively inhibit the penetration and electrochemical reactions of molten salts, thereby extending the anode service life. In contrast, anodes #1, #4, and #5, which were fabricated from recycled graphite without graphitization, exhibited poor crystallinity (graphitization $< 87\%$, grain size $\sim 30 \text{ nm}$ or smaller), high defect density, and inferior physical properties, resulting in greater mass losses (up to $11.3 \pm 0.8 \text{ kg}$) and severe structural deterioration.

The elemental composition significantly influences the corrosion process. Anodes #2 and #3, with higher carbon contents ($\geq 97.2\%$) and lower impurity levels, demonstrated more stable electrochemical behaviour. In comparison, the elevated contents of O, S, Cl, and metallic impurities in anodes #1 and #5 catalysed carbon gasification reactions and exacerbated molten salt penetration erosion.

Surface macropores and cracks serve as critical microstructural features that accelerate corrosion, particularly in recycled graphite anodes. For recycled anodes with similar bulk physical properties (e.g., #1 and #4, both with $\sim 19\%$ apparent porosity), the surface macropore distribution emerged as a key differentiator. Anode #1 contained interconnected surface macropores ($> 20 \mu\text{m}$) and microcracks, which acted as “fast channels” for molten salt, resulting in a thick corrosion layer ($140\text{--}360 \mu\text{m}$) and a mass loss of $11.3 \pm 0.8 \text{ kg}$. In contrast, anode #4 featured predominantly isolated surface macropores, which limited molten salt penetration to a thinner corrosion layer ($150\text{--}300 \mu\text{m}$) and reduced mass loss to $10.4 \pm 0.6 \text{ kg}$. This demonstrates that while apparent porosity reflects the overall pore volume, the distribution and connectivity of surface macropores directly govern the penetration pathways of molten salt.

Importantly, this study was conducted in an industrial-scale electrolyser, where slight operational fluctuations are inherent. While this environment provides high practical relevance, the observed corrosion behaviour is a complex interplay of the intrinsic properties of the anode and these external factors. Future work involving well-controlled laboratory-scale experiments could further decouple these effects.

The intrinsic “raw material–process–structure–performance–lifetime” relationship established in this study provides a scientific basis for the rational selection of anode

materials and the optimization of manufacturing protocols. Graphite anodes produced from high-purity raw materials and subjected to advanced densification and graphitization treatments demonstrate significantly enhanced corrosion resistance, contributing to the sustainable development of rare earth electrolysis technology.

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