



J Adv Ceram 2026, **15**: 9221373

<https://doi.org/10.26599/JAC.2026.9221373>

Research Article

Key role of Ti₂SnC surface oxides in tailoring interfacial diffusion behavior and electrical contact properties of Ag-based composites

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Received: June 27, 2026; Revised: August 24, 2026; Accepted: September 4, 2026

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Abstract: To clarify the role of Ti_2SnC surface oxides in the arc erosion resistance of Ag-based electrical contacts, a pre-oxidation strategy is employed to construct a controllable oxide layer at the Ag/ Ti_2SnC interface. Results show that pre-oxidation induces surface decomposition of Ti_2SnC , leading to the in-situ formation of a TiO_2 -dominated oxide layer, and promoting the diffusion of Sn into the Ag matrix. Although such interfacial reconstruction decreases the initial density, tensile strength, and electrical conductivity, it enhances local solid-solution strengthening and increases interfacial hardness. After 10,000 arc cycles, the pre-oxidized Ag/ $\text{Ti}_2\text{SnC}@ \text{TiO}_2$ composite exhibits lower mass loss, average arcing time, and arc energy. Combined with the post-erosion surface morphology, the results suggest that the preformed oxide interface regulates interfacial diffusion behavior, which further affects arc-root attachment and migration, thereby suppressing localized arc action and molten Ag splashing. This study reveals the role of Ti_2SnC surface oxides in regulating interfacial structure and arc erosion behavior, providing insights into the interfacial design of Ag/MAX-phase electrical contact materials.

Keywords: Ag-based electrical contact materials; MAX phase; Pre-oxidation; Interfacial reconstruction; Arc erosion resistance

1 Introduction

Under extreme service conditions, coupled electrical, thermal, magnetic, and mechanical degradation processes can alter the surface phases, morphology, and interfacial structures of materials, ultimately determining their structure, properties, and service life [1,2]. Therefore, upfront design of the composition and microstructure of existing material is crucial for tailoring key performance and controlling service life. As core materials in low-voltage switches, electrical contacts endure prolonged high-frequency arc impact and high-temperature corrosion [3]. As core components in low-voltage switches, silver (Ag)-based electrical contacts endure prolonged high-frequency arc impacts [4,5], yet their intrinsic low hardness and poor welding resistance under strong-voltage and large-current

operations severely limit their service life [6]. While traditional alloying strategies enhance mechanical durability, they inevitably trigger a drastic degradation in electrical conductivity [7].

In the early 2000s, the oxide reinforcing strategy, exemplified by the "universal" Ag/CdO composite, circumvented this trade-off via a unique arc-quenching vaporization mechanism [8]. However, stringent global toxicity bans have restricted Cd usage, compelling the industry to pursue eco-friendly alternatives such as Ag/SnO₂ composites [9,10]. Despite their remarkable arc-dispersing capabilities [11], SnO₂-reinforced contacts suffer from high intrinsic hardness and poor wettability with the Ag matrix, resulting in high contact resistance and inferior processability [12,13]. Consequently, resolving the contradiction between banned toxic traditional materials and underperforming green alternatives remains a critical bottleneck for the switchgear industry.

To overcome the performance limitations dictated by conventional oxides, exploring novel reinforcing phases with diversified compositions, such as graphene, TiB₂, high-entropy alloys and MAX phases has become a paramount research direction [14–17]. Among these, ternary layered MAX phase ceramics have attracted extensive attention, as they uniquely integrate the dual performance advantages of both metals and ceramics [18–20]. Notably, the representative MAX phase Ti₂SnC exhibits exceptionally low electrical resistivity (22 $\mu\Omega\cdot\text{cm}$) and extraordinary metal wettability, effectively eliminating the agglomeration and severe temperature rise issues inherent to oxide reinforcements [21]. Furthermore, the differentiated bonding between Ti-C and Ti-Sn bonds ensures the excellent processability of Ti₂SnC and endows it with structural tunability.

Previous studies have shown that Ag/Ti₂SnC composites can be fabricated by powder metallurgy [22] and exhibit material transfer resistance comparable to that of Ag/CdO, together with good arc erosion resistance [23]. This superior performance is mainly attributed to the good interfacial wettability between Ti₂SnC and Ag, which increases the viscosity of the molten pool under arc action and suppresses the flow and splashing of liquid Ag [24]. However, during fabrication process and high-temperature arc action, Sn atoms from the interlayers of Ti₂SnC may diffuse into the Ag matrix,

leading to Sn–Ag interdiffusion [25]. Such interdiffusion can improve hardness, tensile strength, and welding resistance, but the solid solution of Sn in Ag also markedly decreases the electrical conductivity of the Ag matrix, which is detrimental to the reduction of contact resistance and temperature rise [26]. Predictably, precise regulation of interfacial diffusion behavior is one of the key directions for breakthroughs in the electrical contact performance of Ag/Ti₂SnC. In addition to interfacial diffusion, the oxidation behavior of Ti₂SnC under the high temperature of electrical arcs is another key factor affecting the service performance of Ag/Ti₂SnC composites [27,28]. Staged discharge experiments and microstructural characterizations have revealed that Ti₂SnC undergoes a “slow decomposition–gradual oxidation” process during arc erosion, which plays a beneficial role in absorbing arc energy and mitigating arc-induced damage [29]. Meanwhile, Ti-rich oxides formed during the middle and late stages of arc erosion can fill interfacial gaps in Ag/Ti₂SnC composites and retard the degradation of their overall mechanical properties [30]. However, these oxides are mainly generated randomly by the non-uniform high-temperature field of electrical arcs, making their composition, morphology, content, and spatial distribution difficult to control [31]. Such random oxidation not only makes the effects of oxides on arc behavior and electrical contact performance difficult to predict, but may also introduce new failure factors such as interfacial defects, crack propagation, and reinforcing-phase agglomeration. Therefore, the current understanding of the “oxidation-sacrifice” mechanism of Ti₂SnC remains insufficient, particularly regarding how the composition and structural evolution of oxides regulate arc–material interactions.

Based on these observations, we considered whether an oxide interface that would otherwise form progressively during arc erosion could be introduced in advance through pre-oxidation, allowing the interfacial oxide structure to be established before service. By comparatively evaluating the microstructure, interfacial diffusion, and contact performance of Ag/Ti₂SnC@Oxide composites, we examined the possible role of preformed oxides in modifying arc erosion behavior and protecting the Ag matrix.

2 Experimental procedures

2.1 Preparation of samples

Raw powders of Ti, Sn, and C were uniformly mixed at the molar ratio of 2:1:1. The mixture was then heated to 1450 °C in a tube furnace (GSL-1700X, Kejing, China) under the protection of Ar and held for 2 h to synthesize Ti_2SnC powder (5~10 μm). The detailed synthesis procedure has been described in our previous report [32]. As shown in Fig. 1, the as-synthesized Ti_2SnC powder was first ball-milled for 2 h (QM-3SP2, Nanjing Nanda Instrument, China) at a powder-to-ball mass ratio of 1:20. The ball-milled Ti_2SnC powder and Ag powder were then weighed at a mass ratio of 1:9 and subsequently wetting ball-milled for 0.5 h (mass ratio of powder : ball : ethanol = 1:1.5:1). After drying, the mixed powder was cold-pressed at 550 MPa to obtain green bodies of Ag/10wt.% Ti_2SnC composites (abbreviated to Ag/TSC). Subsequently, the green bodies were subjected to heat treatment via the following processes: (1) a direct heat treatment at 600 °C/2 h in Ar atmosphere was conducted on Ag/TSC, designated as sample S1. (2) Ag/TSC was first subjected to a pre-oxidation at 550 °C for 5 h in O_2 atmosphere, followed by heat treatment at 600 °C for 2 h in Ar atmosphere, yielding the Ag/10wt.% Ti_2SnC @Oxides, designated as sample S2. Finally, samples S1 and S2 were processed into rectangular pieces (15 mm $\times$ 1.5 mm $\times$ 1 mm), dumbbell-shaped pieces (40 mm $\times$ 1.5 mm $\times$ 7.5 mm), and disc-like pieces (Φ = 7 mm, thickness = 1.5 mm) for subsequent tests of electrical resistivity, tensile strength, and electrical contacting, respectively.

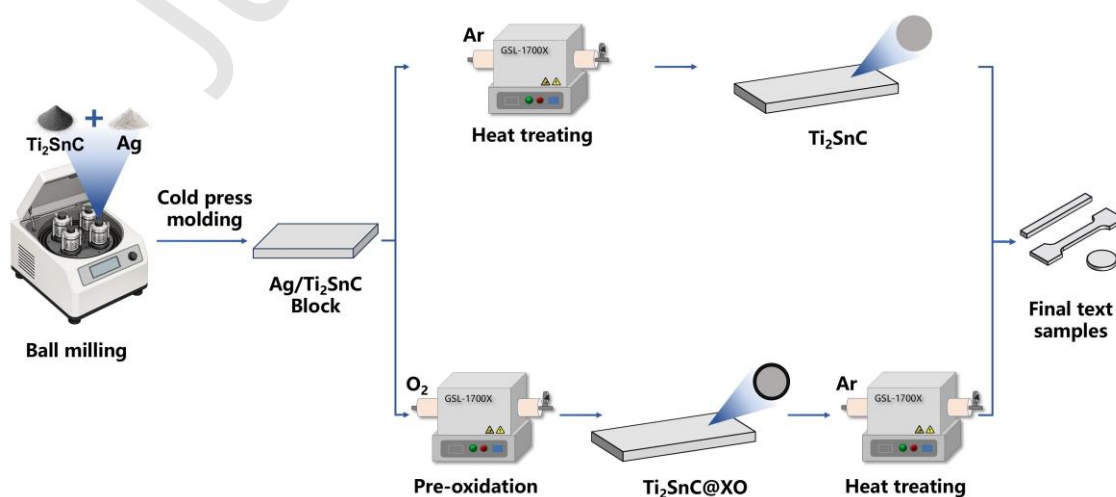


Fig. 1 Schematic illustration for preparing the Ag/TSC and Ag/TSC@XOs composite samples.

2.2 Electrical arc discharging test

The polished electrical contact samples of S1 and S2 were placed into a Relay Life Monitoring System (ADCES-300, Maihang, China). Then, the make-and-break operations 10,000 discharge cycles (2 s on, 2 s off) were performed under a resistive load of 36 V/20 A in an ambient air atmosphere (GB/T 24273-2009), to evaluate the arc erosion resistance of the Ag/TSC and Ag/TSC@XOs composites.

2.3 Characterization

For all test items, more than ten test values for one sample were conducted to ensure the accuracy of data, and then the average value was calculated after removing the highest and lowest values. The bulk density of the Ag/TSC sample (S1) and Ag/TSC@XOs sample (S2) was measured using a densitometer (AR-150PM, Dahong Meituo, China). The metallographic observation was performed using an optical microscope (OM, DM2700P, Leica, Germany). Vickers microhardness were obtained by a microhardness tester (XHVT-1000Z, Shanghai Shangcai, China). The resistivities of rectangular S1 and S2 samples were measured using a high-precision micro-ohmmeter (Metra HIT 27I, Gossen Metrawatt, Germany). The tensile strength of dumbbell-shaped S1 and S2 samples was obtained via a universal testing machine (AGS-X, Shimadzu, Japan) at a speed of 1 mm/min, in accordance with the GB/T 228.3-2019 standard. Nanoindentation tests were performed point by point on the sample surfaces using a nanoindenter (iNano, KLA Instruments, USA) equipped with a Berkovich indenter. The testing protocol utilized a peak load of 1 mN, a loading/unloading rate of 0.1 mN/s, and a dwell time of 10 s. Phases of the bulk specimens S1 and S2 were identified by X-ray diffraction (XRD; Ultima IV, Rigaku, Japan) using Cu *Ka* radiation ($\lambda_{K\alpha 1} = 0.154060$ nm, $\lambda_{K\alpha 2} = 0.154443$ nm) operated at 40 kV and 36 mA. The diffraction patterns were recorded over a 2θ range of 10° to 80° at a scan rate of $3^\circ/\text{min}$. X-ray photoelectron spectroscopy (XPS) was performed using an ESCALAB 250Xi spectrometer (Thermo Fisher Scientific, USA) to analyze the surface chemical states and elemental compositions of the samples. The binding energies were calibrated with reference to the C

1s peak at 284.8 eV. Finally, the microstructures of the samples S1 and S2 before and after arc erosion were observed using a field-emission scanning electron microscope (SEM, Nova Nano 450, FEI Company, USA), and their elemental compositions were analyzed utilizing an attached energy-dispersive X-ray spectrometer (EDS, AZtec X-MaxN80, Oxford Instruments, UK).

3 Results

3.1 Microstructural and compositional characterization

Figs 2a1 and b1 present the SEM micrographs of the as-prepared Ag-based composite samples (S1 and S2). The continuous bright regions correspond to the Ag matrix, and the uniformly distributed dark particles are the Ti_2SnC reinforcing phase. As shown in the SEM images of Figs. 2a2 and b2, the particle-size distributions of the reinforcing phases were further quantified as 2.17 μm of S1 and 2.26 μm of S2, respectively. Further high-magnification SEM observations in Figs. 2a3 and b3 show clear interfaces between the reinforcing phases and the Ag matrix in both samples. Compared with sample S1 (Fig 2a3), a distinct dark interfacial layer with a thickness of approximately 0.5~1.0 μm is observed around the Ti_2SnC reinforcing phase in sample S2 (Fig. 2b3) after pre-oxidation at 550 °C for 5 h. This result indicates that a new unknown phase is likely generated on the surface of the Ti_2SnC reinforcing phase during the pre-oxidation process.

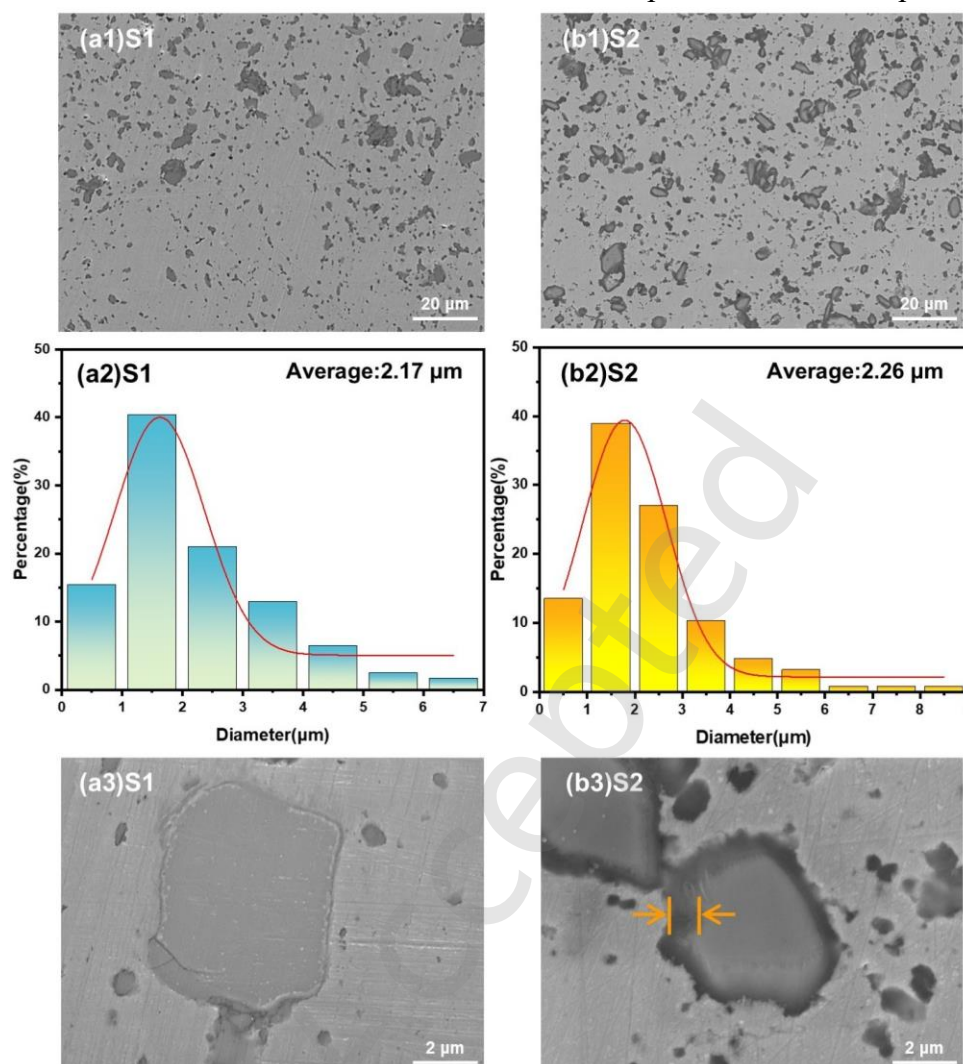


Fig. 2 SEM micrographs, particle-size distributions, and microstructural images of the Ag/TSC composites, (a) sample S1 prepared via direct heat treatment; (b) sample S2 prepared via pre-oxidation followed by heat treatment.

The XRD characterization with a slower scanning speed of 3°/min was applied on these Ag-based composite bulk samples to detect their phase change. For sample S1 merely by heat treatment, as shown in Fig. 3a1, primary diffraction peaks of Ag and Ti₂SnC phase are detected. After the pre-oxidation of 550 °C/5 h followed by heat treatment, in addition to phases of Ag and Ti₂SnC, weak characteristic diffraction peaks of TiO₂ are observed in sample S2. The results of XRD and SEM (Fig. 2) indicate that a small amount of TiO₂ is generated on the Ti₂SnC surface through the pre-oxidation process. Furthermore, as shown in Fig 3a2, the 36~46° window diffraction peaks of both samples S1 and S2 shift towards lower angles in contrast to the standard Ag peaks. This peak shift behavior is

probably attributed to the formation of an Sn-containing solid solution in the Ag matrix [33]. Notably, the offset values of Ag peaks in sample S1 are 0.041° and 0.056° , while those in sample S2 increase to 0.141° and 0.157° . This suggests that the degree of Sn solid solution in the Ag matrix of sample S2 is higher than that in sample S1. In other words, the Sn diffusion of Ti_2SnC into the Ag matrix in sample S2 is more pronounced than in sample S1. Therefore, in addition to generating new phases at the interface, the pre-oxidation process also induces more Sn to diffuse from the Ti_2SnC into the Ag.

The XPS characterization was conducted to further determine the chemical composition of sample S2 by the valence state analysis. In the high-resolution Ti 2p spectrum (Fig. 3b1), the binding energy peaks at 458.48 eV and 464.23 eV correspond to Ti^{4+} . In the Sn 3d spectrum (Fig. 3b2), the binding energy peaks at 486.29 eV and 494.75 eV are assigned to Sn^{4+} . In the O 1s spectrum (Fig. 3b3), the peak at 530.21 eV corresponds to lattice oxygen. This result further confirms the generation of the TiO_2 phase at the interface between Ti_2SnC and Ag matrix during the pre-oxidation process, which is highly consistent with the XRD analysis. It is confusing that the Sn^{4+} and the lattice oxygen were detected via XPS, while the SnO_2 phase was not found in XRD. It is presumed that a portion of Sn diffused into the Ag matrix, whereas only a negligible amount was oxidized and confined at the interface, making it undetectable by XRD. The above results imply that the composition of the oxide layer at the interface of sample S2 is mainly TiO_2 , with only a small amount of SnO_2 present, and thus is abbreviated as Ag/TSC@TO.

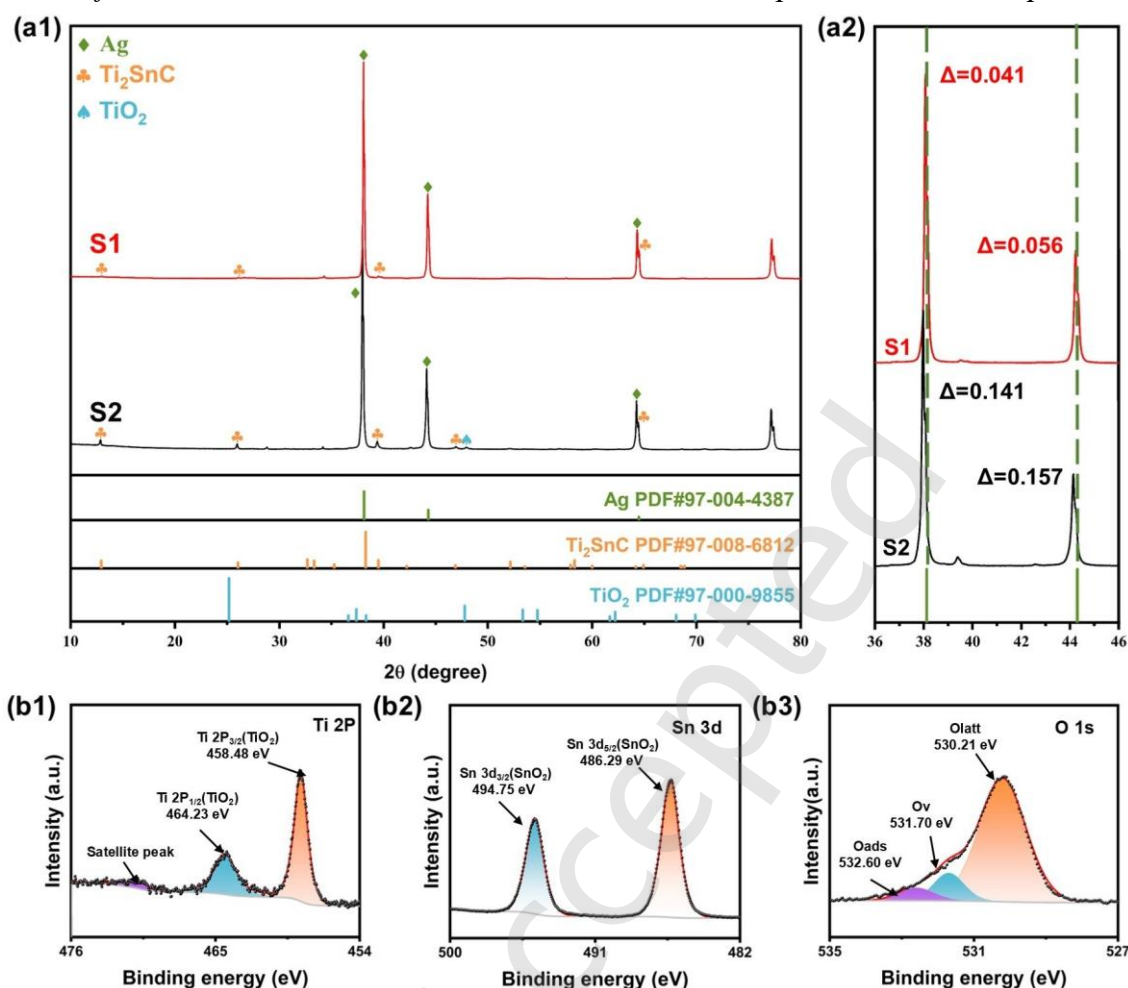


Fig. 3 XRD and XPS characterizations of the Ag/Ti₂SnC composites. (a) XRD patterns of samples S1 and S2 and comparisons with the standard peaks of TiO₂, Ti₂SnC and Ag. (b1), (b2), and (b3) are the high-resolution XPS spectra of sample S2 corresponding to Ti 2p, Sn 3d, and O 1s, respectively.

3.2 Physical and electrical contact properties

Fig 4 presents the bulk density, tensile strength, interfacial Vickers hardness, and electrical resistivity of samples S1 and S2. Compared to sample S1, the bulk density of sample S2 decreases from 9.24 g/cm³ to 8.59 g/cm³, the tensile strength drops from 124.68 MPa to 71.19 MPa, the interfacial Vickers hardness slightly increases from 83.51 HV to 84.9 HV, and the electrical resistivity rises from 7.31 μΩ·cm to 24.28 μΩ·cm. These results indicate that the 550 °C pre-oxidation process generates high-resistance and low-wettability oxide phases at the interface, thus degrading the initial electrical conductivity and tensile strength of the Ag-based composites. The existence of high-hardness oxide phases, however, probably improves the composite's interface hardness.

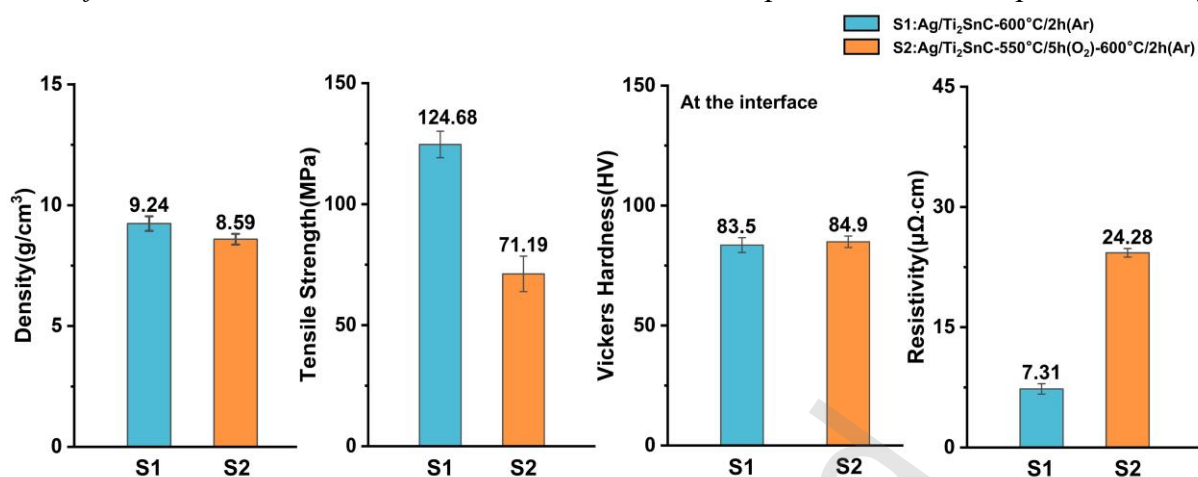


Fig. 4 Physical properties of sample S1 and S2, bulk density, tensile strength, interfacial Vickers hardness, and electrical resistivity from left to right.

Fig 5 presents the mass changes of the cathode contact and the anode contact in samples S1 and S2, respectively, as well as their total mass loss percentage before and after 10,000 discharge cycles. After arc erosion, in general, the decreasing mass for the anode contact and the increasing mass for the cathode contact were observed in both samples S1 and S2, demonstrating a material transfer behavior from the anode to the cathode on the contact surface during the arc discharge process. Specifically, the total mass loss of 2.10% in sample S1 is significantly higher than that of 1.20% in sample S2, indicating the superior arc resistance of Ag/TSC@TiO₂ compared with Ag/TSC.

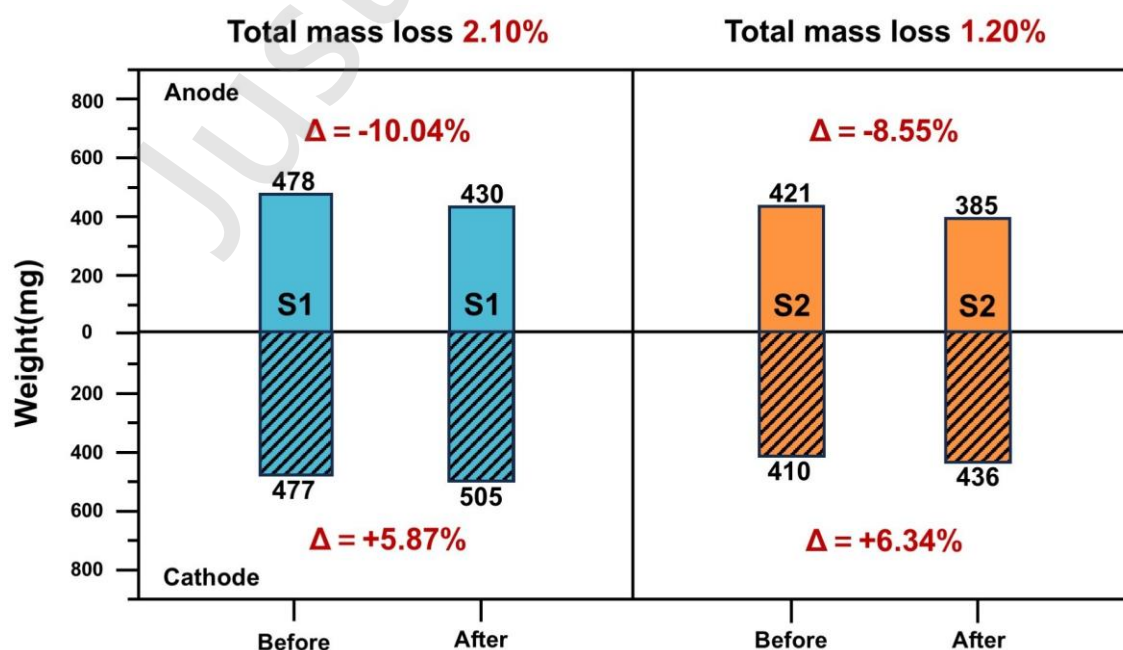


Fig. 5 Mass changes of the anodes and cathodes and total mass loss percentage for samples S1 and S2 before and after 10,000 discharge cycles.

The arc erosion process reflects the interaction between the contact material and the electrical arc [34]. Fig 6 compares the arc time, arc energy, contact resistance, and welding force of samples S1 and S2 throughout the discharge cycles. The sample S2 exhibited significantly lower arc time of 35.9 ms and arc energy of 5876.7 mJ, respectively, compared to those of the S1 contact sample (39.1 ms and 7783.6 mJ). Especially, the relatively low arc time (22.9~32.5 ms) and arc energy (3500.0~5777.0 mJ) during the initial and intermediate stages of arc erosion (1~5,000 cycles) facilitate the formation of excellent arc erosion resistance of the Ag/TSC@TiO₂ composites. Although, the average value of 26.79 mΩ and 0.50 N in contact resistance and welding force of sample S2, respectively, are slightly higher than those of sample S1 (22.43 mΩ and 0.44 N). When arc discharge comes to the late stages of 5,000~10,000 cycles, the rapidly decreasing contact resistance of 23.81~26.32 mΩ and welding force of 0.19~0.34 N make Ag/TSC@TiO₂ remarkable advantages in reducing temperature rise and adhesion failure compared to Ag/TSC. Therefore, the electrical contacting experimental results indicate that introducing oxides on the TSC surface can effectively reduce the interaction time of the electrical arc with the Ag-based electrical contact and simultaneously lower the overall energy of the electrical arc. Moreover, in the later stage of arc erosion, the TSC@TO reinforcing phase can significantly reduce the contact resistance and welding force.

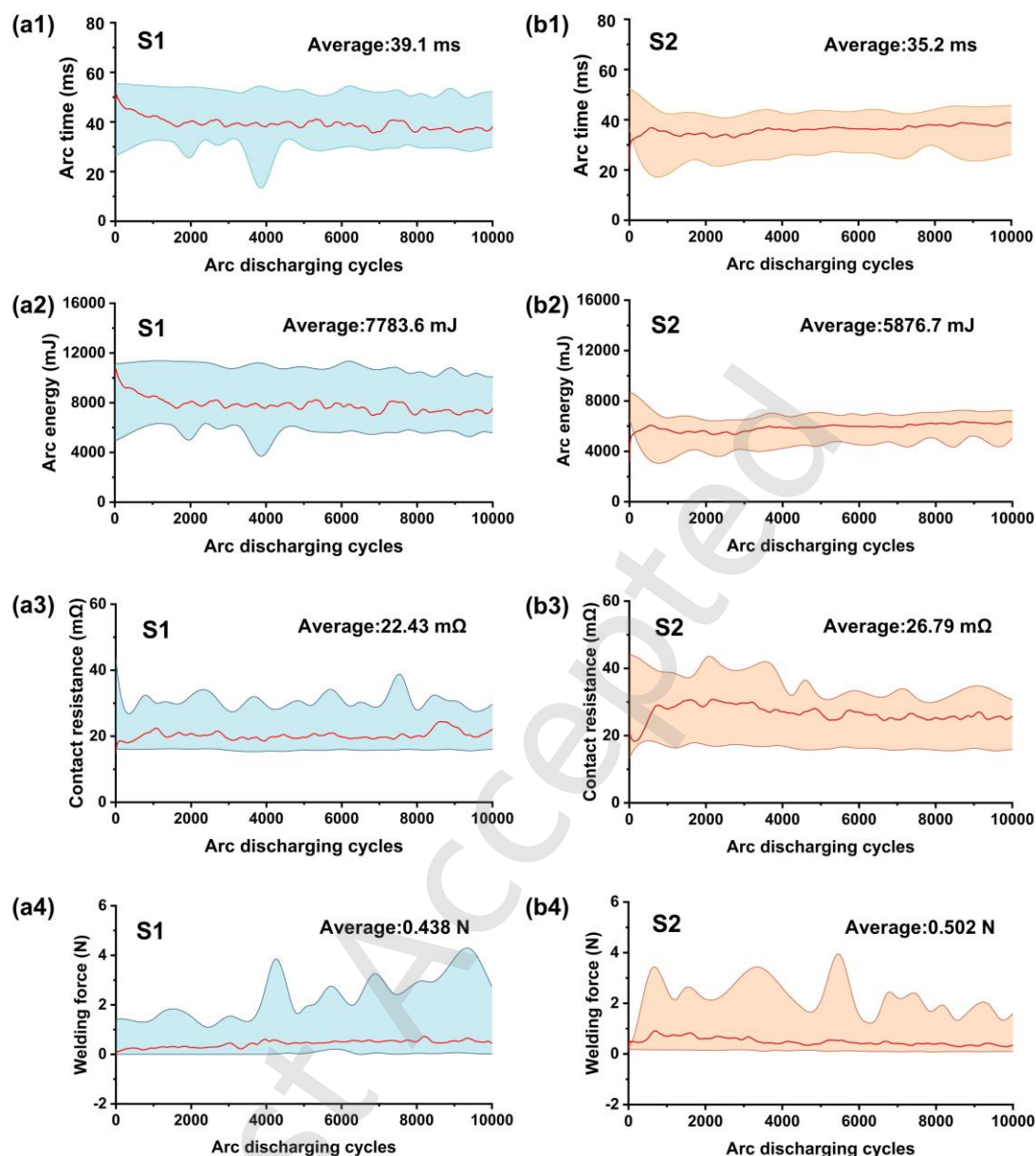


Fig. 6 Arc time, arc energy, contact resistance, and welding force of samples S1 and S2 during 10,000 discharge cycles. Solid lines represent the mean values for each test stage, and the shaded regions indicate the corresponding minimum-to-maximum ranges.

4 Discussion

4.1 Interfacial diffusion behavior

EDS point analyses were performed under high-magnification SEM to compare the elemental compositions of the reinforcing phases, interfaces, and Ag matrix in S1 and S2 (Fig. 7). As shown in Fig. 7a1, the Ti/Sn atomic ratio at point 3 within Ti_2SnC in S1 is approximately 2.04, close to the

theoretical stoichiometric ratio, indicating that the internal composition of the reinforcing phase remains intact. At the interface (point 4), the Ti/Sn ratio increases to 2.49, and a small amount of Ag is detected, indicating limited compositional variation and elemental interdiffusion near the interface. In the Ag matrix, a small amount of Sn is detected only at point 5, approximately 1 μm from the interface, whereas no Sn is detected at farther positions, suggesting that Sn diffusion in S1 is mainly confined to the region adjacent to the interface. In contrast, for S2, the Ti/Sn ratio in the Ti_2SnC region near the interface (point 3) increases to 2.12, indicating a certain degree of compositional change in the surface region of the reinforcing phase after pre-oxidation. At the interface (point 4), the Ti/Sn ratio further increases to 4.99, accompanied by detectable O, Ag, and Sn, indicating pronounced oxidation and compositional reconstruction in this region. More importantly, Sn remains detectable in the Ag matrix at distances of approximately 1, 2, and 3 μm from the interface (points 5–7), indicating that Sn remains detectable at a distance of approximately 3 μm from the interface in S2. This result demonstrates more pronounced Sn diffusion from Ti_2SnC into Ag matrix after pre-oxidation.

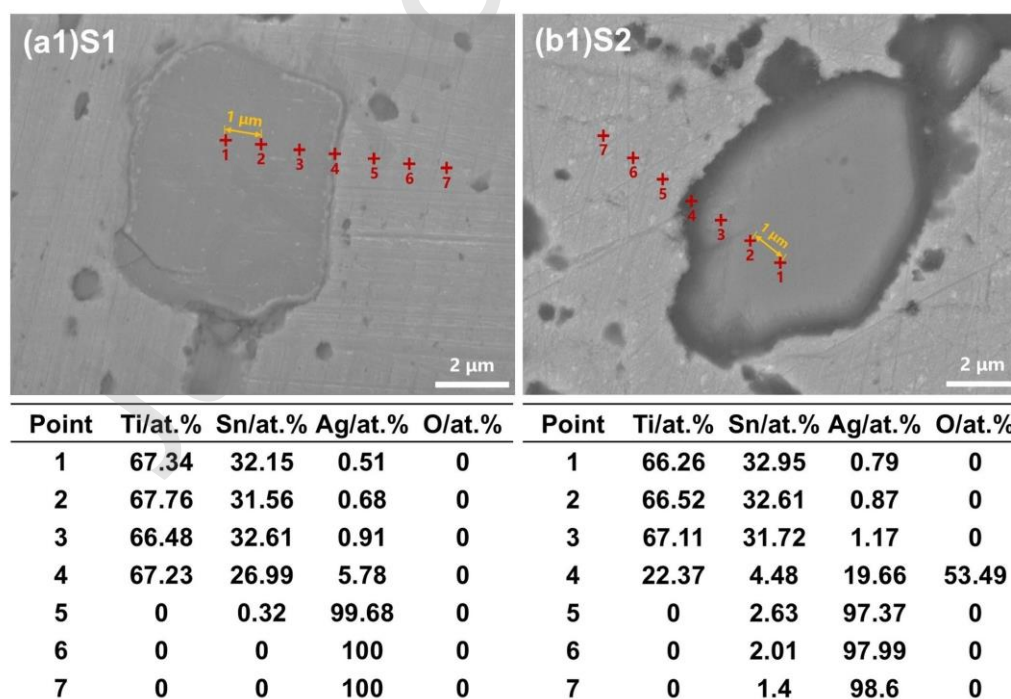


Fig. 7 SEM images of the Ag/TSC interfaces and corresponding EDS point analysis results of the Ag-based composites: (a1) sample S1 and (b1) sample S2.

To further verify the TSC surface oxide layer and its spatial continuity, EDS line-scan analyses were performed on S1 and S2. As shown in Fig. 8a3, the Ag, Ti, and Sn signals in S1 exhibit distinct gradients across the Ag matrix/TSC interface, but the elemental transition region is relatively narrow. Meanwhile, the O signal remains weak throughout the scanned region, indicating limited interfacial interdiffusion in the non-pre-oxidized sample. In contrast, the line-scan result of S2 (Fig. 8b3) shows that the Ag signal extends from the matrix toward the TSC side, whereas the Ti and Sn signals extend toward the Ag matrix, forming more pronounced elemental concentration gradients across the interface. Meanwhile, an O signal appears within the interfacial transition region. Compared with S1, S2 exhibits a broader elemental transition region, particularly with a greater extension of Sn into the Ag matrix, supporting enhanced interfacial elemental diffusion after pre-oxidation.

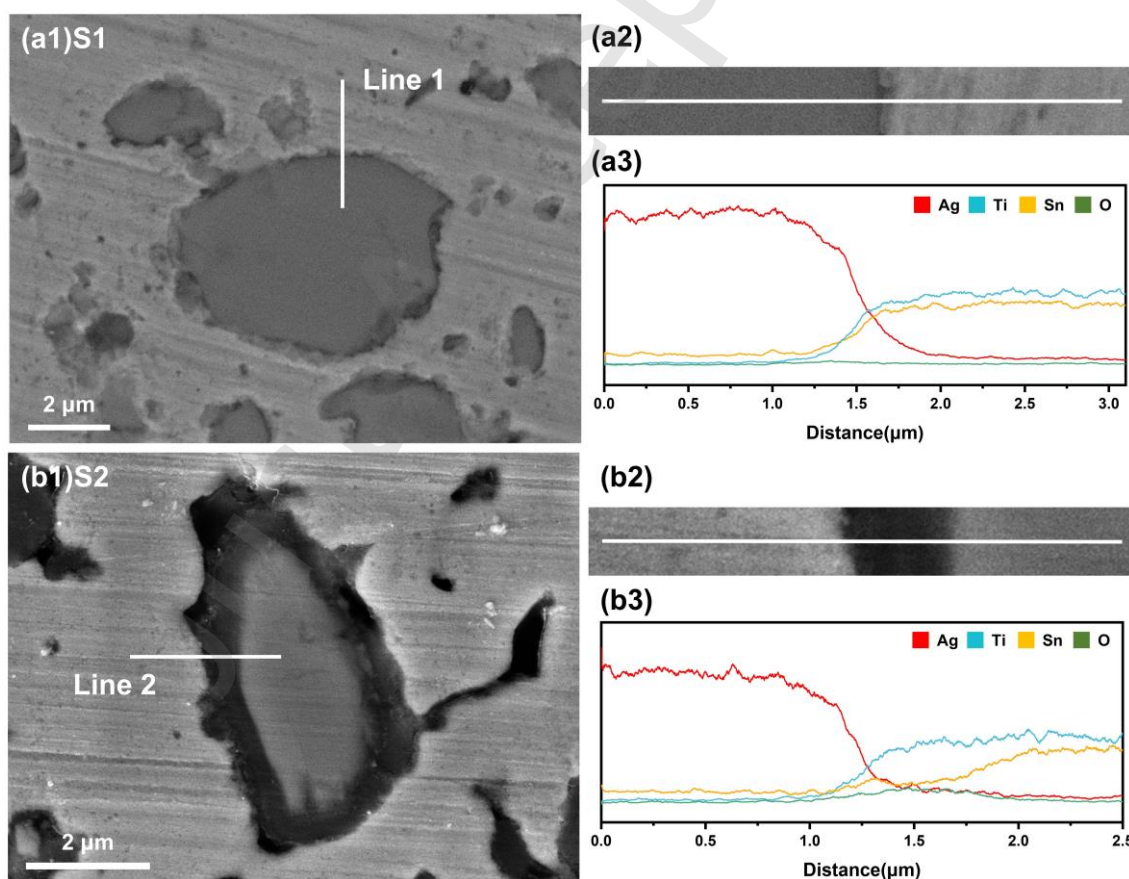


Fig. 8 Interfacial SEM images and corresponding EDS line-scan results of the Ag-based composites: (a1) SEM image, (a2) EDS line-scan path, and (a3) corresponding elemental profiles of sample S1; (b1) SEM image, (b2) EDS line-scan path, and (b3) corresponding elemental profiles of sample S2

Elemental mapping further reveals the spatial distribution of interfacial elements in the two samples.

As shown in Fig. 9a, Ti and Sn in S1 are mainly confined within the TSC particles, while Ag is predominantly distributed in the matrix. Only limited elemental overlap is observed at the particle boundaries, and no obvious O-enriched region is present, consistent with the relatively weak interfacial interdiffusion in S1. In contrast, S2 (Fig. 9b) exhibits pronounced O enrichment around the Ti_2SnC particles, accompanied by a broader distribution of Ti and Sn toward the surrounding matrix and a more evident extension of Ag toward the particle side, indicating pre-oxidation-induced surface reactions and more extensive elemental redistribution.

Combined with the EDS point analyses, line scans, and phase and chemical-state characterization, these results support the formation of a spatially continuous TiO_2 -rich interfacial oxide layer around the TSC particles in S2. Based on these observations, the interfacial structure of S1 can be described as $\text{Ag}/(\text{Ag}+\text{Sn})/(\text{Ti}_2\text{Sn}_{1-x}\text{C}+\text{Ag})/\text{Ti}_2\text{SnC}$ (Fig. 9c1), whereas S2 develops a more complex $\text{Ag}/(\text{Ag}+\text{Sn})/(\text{TiO}_2+\text{Ag}+\text{Sn})/(\text{Ti}_2\text{Sn}_{1-x}\text{C}+\text{Ag})/\text{Ti}_2\text{SnC}$ interfacial structure (Fig. 9c2).

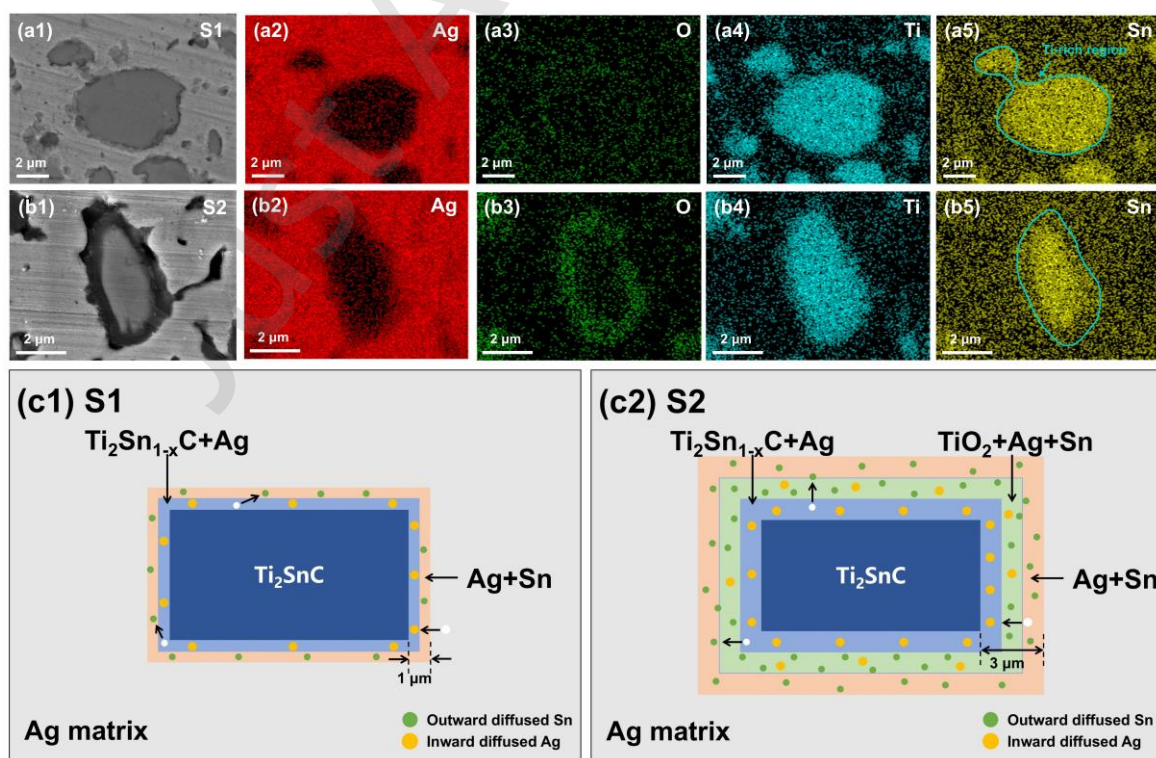


Fig. 9 Interfacial elemental distributions and schematic illustrations of the Ag-based composites: (a1) SEM image and corresponding EDS elemental maps of (a2) Ag, (a3) O, (a4) Ti, and (a5) Sn for sample S1; (b1) SEM image and corresponding EDS elemental maps of (b2) Ag, (b3) O, (b4) Ti, and (b5) Sn for sample S2; schematic illustrations of the interfacial diffusion behavior in (c1) S1 and (c2) S2.

4.2 Micro/nanomechanical behavior

To analyze the influence of interfacial composition and diffusion behavior on the physical properties of the Ag-based composite material, the Vickers hardness testing was performed on the Ag matrix of both samples S1 and S2, as shown in Fig 10. Obviously, compared with sample S1 in the OM images of Figs 10a1 and a2, the Vickers hardness indentations on the Ag matrix in sample S2 are significantly smaller, reflecting its relatively higher hardness (Figs 10b1 and b2). Moreover, as shown in Fig 10c, statistical results of numerous hardness tests show that the average value of the Ag matrix in sample S2 is nearly 25.3% higher than that in sample S1. Furthermore, the 2D nanoindentation hardness maps in Figs. 10d1 and d2 further reveal the spatial differences in local hardness within the Ag matrix. Under the same hardness scale, the deep-blue low-hardness regions are more extensive in S1, with local hardness mainly distributed at approximately 0.4–1.1 GPa. In contrast, these low-hardness regions are less extensive in S2, whereas the light-blue to green regions corresponding to approximately 1.4–2.0 GPa are more widely distributed, indicating an overall shift of the local hardness toward higher values. Notably, the yellow-to-red high-hardness regions adjacent to the reinforcing phase are more pronounced in S2 and extend farther into the surrounding Ag matrix than in S1, indicating a more evident local strengthening effect in the interfacial Ag matrix. Combining the elemental-distribution and hardness results, the increased hardness of the Ag matrix in S2 is consistent with the more pronounced Sn diffusion and the resulting solid-solution strengthening in Ag [35]. In the immediate interfacial region, the high-hardness TiO₂-rich phase and local interfacial microstructural evolution may also contribute to the measured hardness.

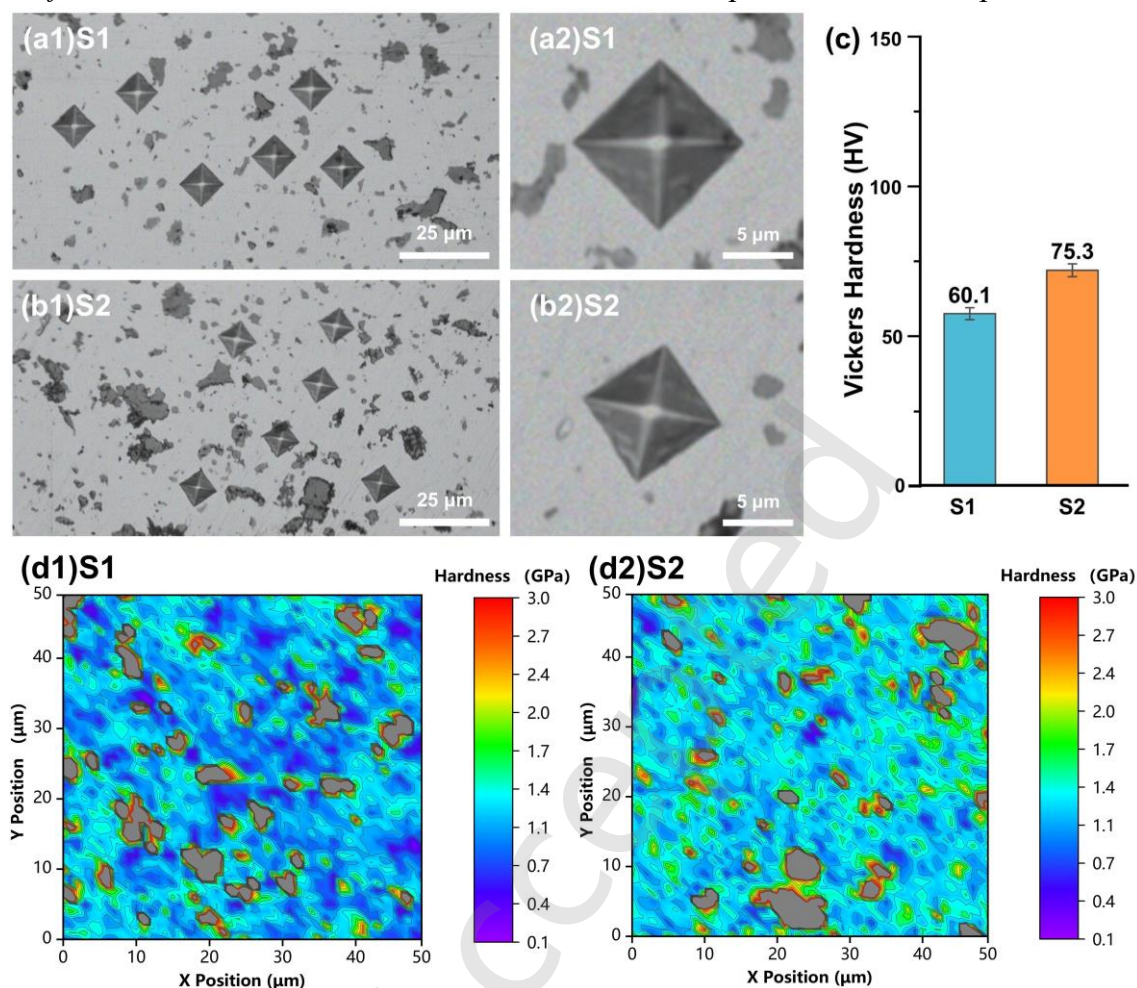


Fig. 10 Indentation morphologies and size on the Ag matrix of the (a) sample S1 and the (b) sample S2, as well as the statistical results of Vickers hardness; and spatial distributions of nanoindentation hardness for (d1) S1 and (d2) S2.

Subsequently, the nanoindentation combined with EDS characterization was further analyzed to determine the intrinsic correlation between elemental distribution and surface nanohardness from the TSC reinforcing phase across the interface to the Ag matrix in samples S1 (Fig. 11) and S2 (Fig. 12) [36]. In the sample S1, the Ti/Sn ratio within the Ti_2SnC reinforcing phase slightly increases from 2.10 to 2.21 (from Point 3 to Point 1, Fig. 11a), with the corresponding nanohardness gradually decreasing from 2.299 GPa to 1.229 GPa (Figs. 11b, 10c). At the interface of point 0, the Ti/Sn ratio rapidly rises to 2.49, and the corresponding nanohardness further drops to 1.015 GPa (Fig. 11c), indicating that the partial decomposition of the Ti_2SnC surface leads to a degradation in nanomechanical properties at the interface. Furthermore, at the 1 μm distance from the interface to the Ag matrix, the Sn/Ag ratio is

0.31, corresponding to a surface nanoindentation hardness of 0.901 GPa. In the Ag matrix beyond 1 μm from the interface, the Sn/Ag ratio is 0, and the surface nanoindentation hardness remains within a relatively low range of 0.882~0.763 GPa, which further confirms the relatively weak diffusion of Sn into the Ag matrix in the Ag/TSC composite.

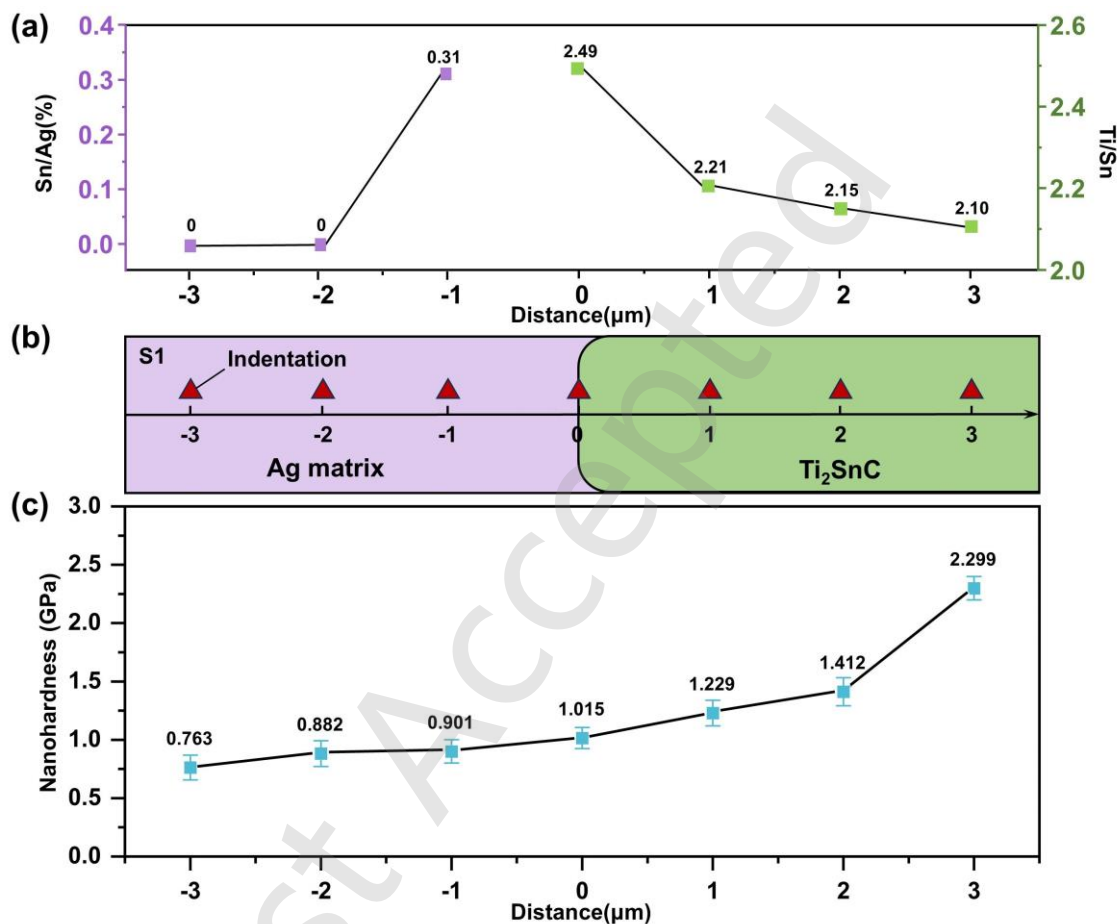


Fig. 11 (a) the elemental ratios of Sn/Ag and Ti/Sn at different positions, (b) schematic of indentation positions crossing the interface of sample S1, and (c) the nanohardness value at the corresponding position.

In sample S2, the Ti/Sn ratio from Point 3 to Point 1 within the Ti_2SnC reinforcing phase also exhibits a slight upward trend (2.11 to 2.15), while the Ti/Sn ratio at the interface (Point 0) reaches an exceptionally high value of 4.99 (Fig. 12a), which indicates that the Ti_2SnC surface structure undergoes severe decomposition. Nonetheless, the generation of a high-hardness TiO_2 layer (~ 10 GPa) [37] and distinct Ag-Sn diffusion at the interface of Ag/TSC@TO still increases the interface nanohardness to 1.358 GPa (Figs. 12b, 10c). Compared to sample S1, the Sn/Ag ratios from point -3

to point -1 in the Ag matrix of sample S2 are verified with higher values from 2.73 to 1.41, corresponding to a higher surface nanoindentation hardness from 1.219 GPa to 0.953 GPa (Fig. 12c), once again confirming the significant diffusion behavior of Sn into the Ag matrix and solid-solution strengthening effect.

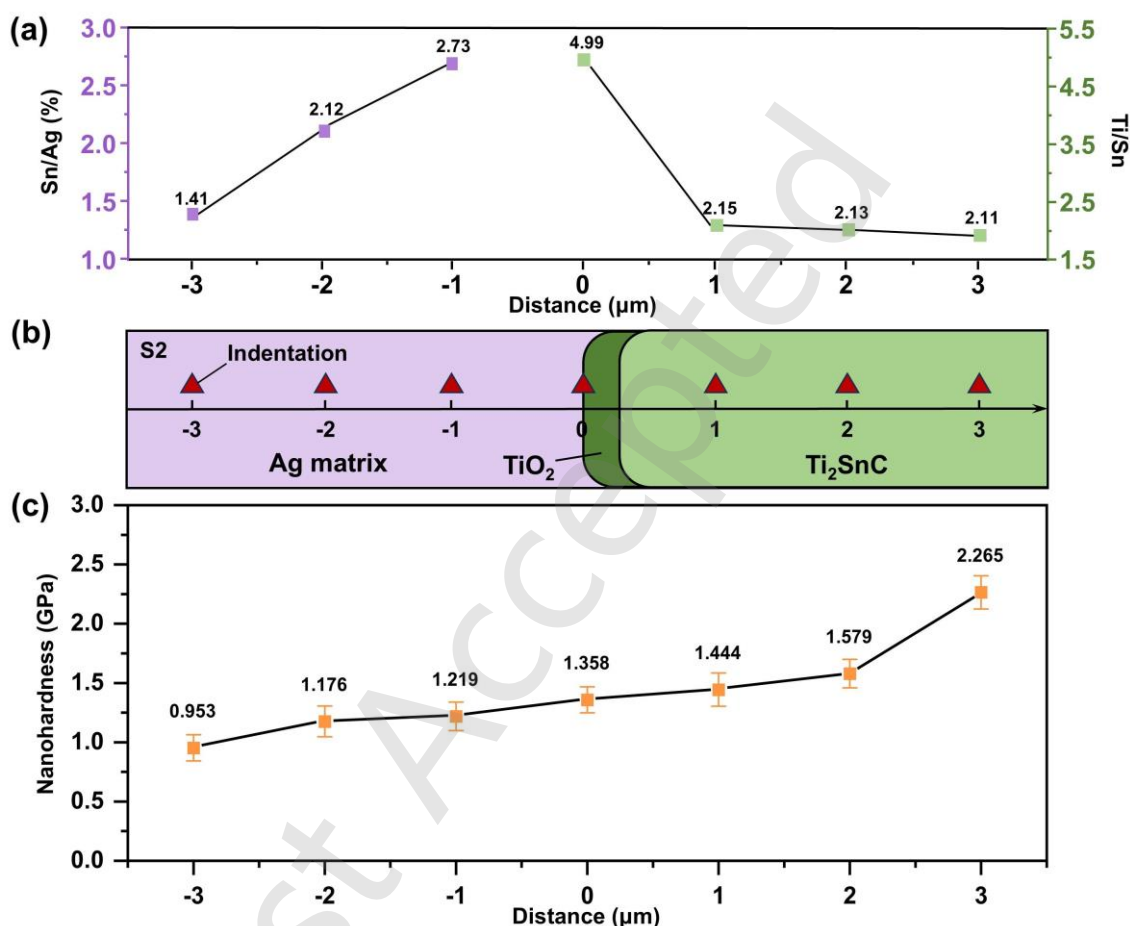


Fig. 12 (a) the elemental ratios of Sn/Ag and Ti/Sn at different positions, (b) schematic of indentation positions crossing the interface of sample S2, and (c) the nanohardness value at the corresponding position.

4.3 Arc erosion behavior of the contact surface

Long-term arc exposure can lead to progressive cumulative erosion of the contact surface and ultimately affect its service performance [38]. To more clearly examine the influence of pre-oxidation on the early-stage erosion behavior, the surface morphologies of S1 and S2 after 50 arc-discharge cycles were characterized by SEM, as shown in Fig. 13. The low-magnification images show that S1 mainly exhibits a relatively concentrated major erosion region (Fig. 13a1), whereas the erosion regions

of S2 are distributed over a broader surface area and appear as multiple spatially separated major erosion regions (Fig. 13b1). At higher magnification, the major erosion region of S1 shows more evident localized melting and pore formation (Figs. 13a2 and a3), whereas the multiple erosion regions in S2 are more spatially dispersed (Figs. 13b2 and b3). These early-stage erosion features are consistent with a more spatially dispersed arc–material interaction after pre-oxidation rather than arc action concentrated within a single localized region.

With increasing arc-discharge cycles, these early-stage differences are further reflected in the long-term surface degradation. As shown in Fig. 14, after 10,000 arc-discharge cycles, the S1 surface exhibits obvious cracks, pores, and regions of severe localized erosion (Fig. 14a2). Higher-magnification SEM observations further reveal numerous deep pores together with irregular Ag deposits and agglomerates in the eroded region of S1 (Fig. 14a3), indicating more pronounced local material transfer and surface degradation. Such severe surface defects and localized material transfer may also adversely affect the contact resistance and welding force during the intermediate and late stages of arc erosion [39]. In comparison, S2 retains a relatively intact surface morphology after 10,000 cycles, with fewer obvious cracks and large erosion pits (Figs. 14b1–b3). Therefore, the more spatially dispersed early-stage erosion features in Fig. 13 are consistent with the less severe localized erosion observed after long-term cycling in Fig. 14, indicating a more dispersed erosion pattern and a lower degree of localized surface damage in S2.

The EDS results further show that the Sn content in the eroded regions of S1 is relatively low, ranging from 0 to 0.56 at.%, whereas higher Sn contents of 1.46–13.30 at.% are detected in the corresponding regions of S2. This result is consistent with the more pronounced Sn diffusion and redistribution into the Ag matrix in S2 discussed above and supports a possible contribution of Sn diffusion and the associated solid-solution strengthening to the improved local erosion resistance of the Ag matrix [40,41]. It should also be noted that the porosity introduced during pre-oxidation may adversely affect electrical conduction, which is consistent with the higher initial electrical resistivity

of S2. However, the less severe localized surface degradation during repeated arc cycling may help maintain relatively stable electrical contact behavior at the later stage. Therefore, the long-term electrical contact performance of S2 results from the combined effects of the initial changes in electrical properties and the subsequent evolution of surface erosion. Overall, the more spatially dispersed erosion regions after 50 cycles together with the less severe localized erosion after 10,000 cycles suggest that pre-oxidation may alter the spatial distribution of arc–material interactions and thereby help mitigate localized concentrated erosion and material loss.

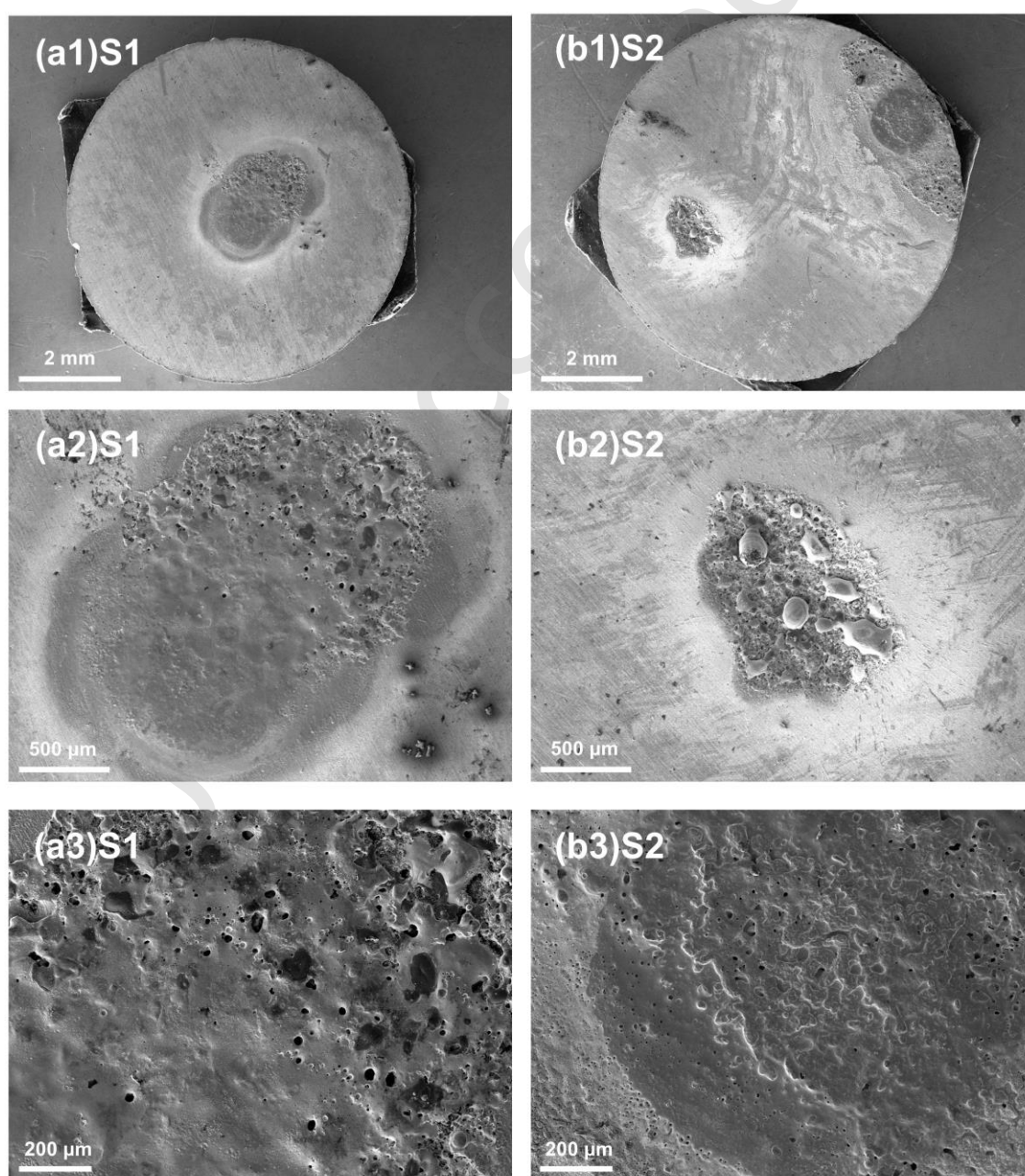


Fig. 13 SEM surface morphologies of samples S1 and S2 after 50 arc-discharge cycles: (a1–a3) S1 and (b1–b3) S2 at different magnifications.

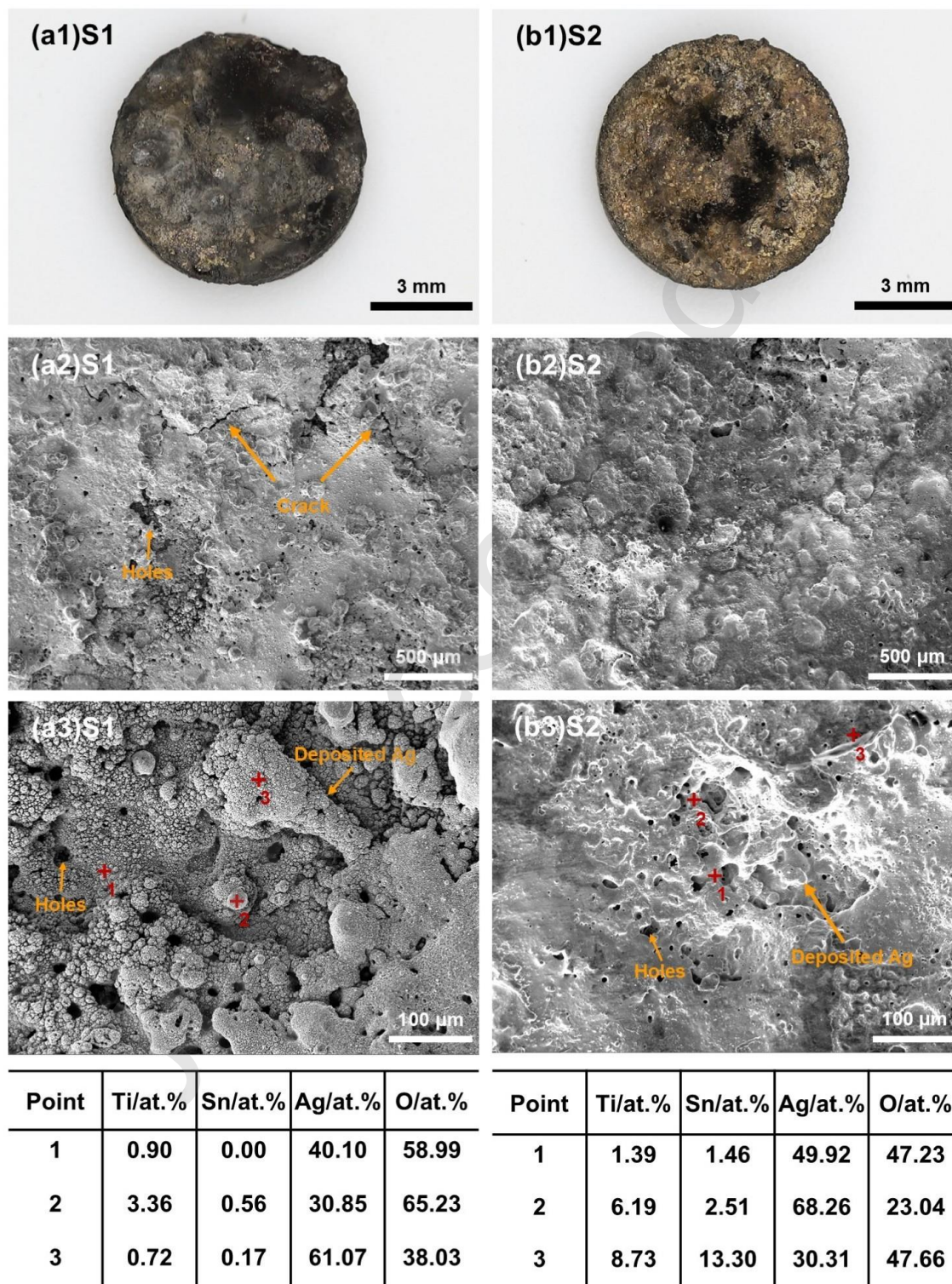


Fig. 14 OM images, surface morphologies and elemental compositions of the different regions in the (a) samples S1 and (b) sample S2 after 10,000 arc discharge cycles.

4.4 Mechanism of resisting arc erosion

Based on the post-erosion microstructures, EDS results, mass-loss data, and electrical contact properties, Fig. 15 presents a proposed interpretation of the possible arc erosion evolution of Ag/TSC and Ag/TSC@TO composites. For the sample S1 of Ag/TSC in Fig 15a1, in the early stage of electrical arc discharge, the electrical arc primarily acts on the Ag matrix with a relatively lower work function (4.26 eV) [42], thereby causing material transfer behavior such as vaporization and spattering of Ag due to a lack of significant Sn diffusion in the Ag matrix. However, the thin Ag+Sn layer formed at the interface between TSC and the Ag matrix strengthens the interface bonding between the two, further reducing the damage caused by the electric arc to the reinforcing phase and the interface. As shown in Fig 15a2, in the mid-stage of electrical arc discharge, in addition to the continuously gradual destruction of the Ag matrix and the accelerated transfer of liquid Ag inside the molten pool, the high temperature of the electrical arc also triggers the "gradual decomposition-slow oxidation" behavior of the TSC surface structure, as well as the slight interdiffusion between the TSC and the Ag matrix. The oxidized product on the TSC surface begins to interact with the electrical arc, dispersing the electrical arc and absorbing its partial energy [43], thereby reducing the damage to the Ag matrix and molten pool. Since the formation of the oxide layer lags behind the initial melting and splashing of the Ag caused by the electrical arc, its ability to suppress electrical damage and material loss in the early and mid-stage is limited. Therefore, when entering the later stage of arc erosion (Fig. 15a3), although TSC continues to decompose and oxidize, its surface oxide layer is simultaneously consumed under electrical arc damage. Moreover, due to the reduction of TSC and the formation of the oxide layer, there is almost no Sn-Ag diffusion between the reinforcing phase and Ag matrix, leading to rapid deterioration of interfacial bonding and consequently intensified material transfer and mass loss. Finally, Ag/TSC contact exhibits higher mass loss and leaves behind poor surface morphologies with a lot of cracks, holes, erosion pits and Ag blocks (Fig. 13).

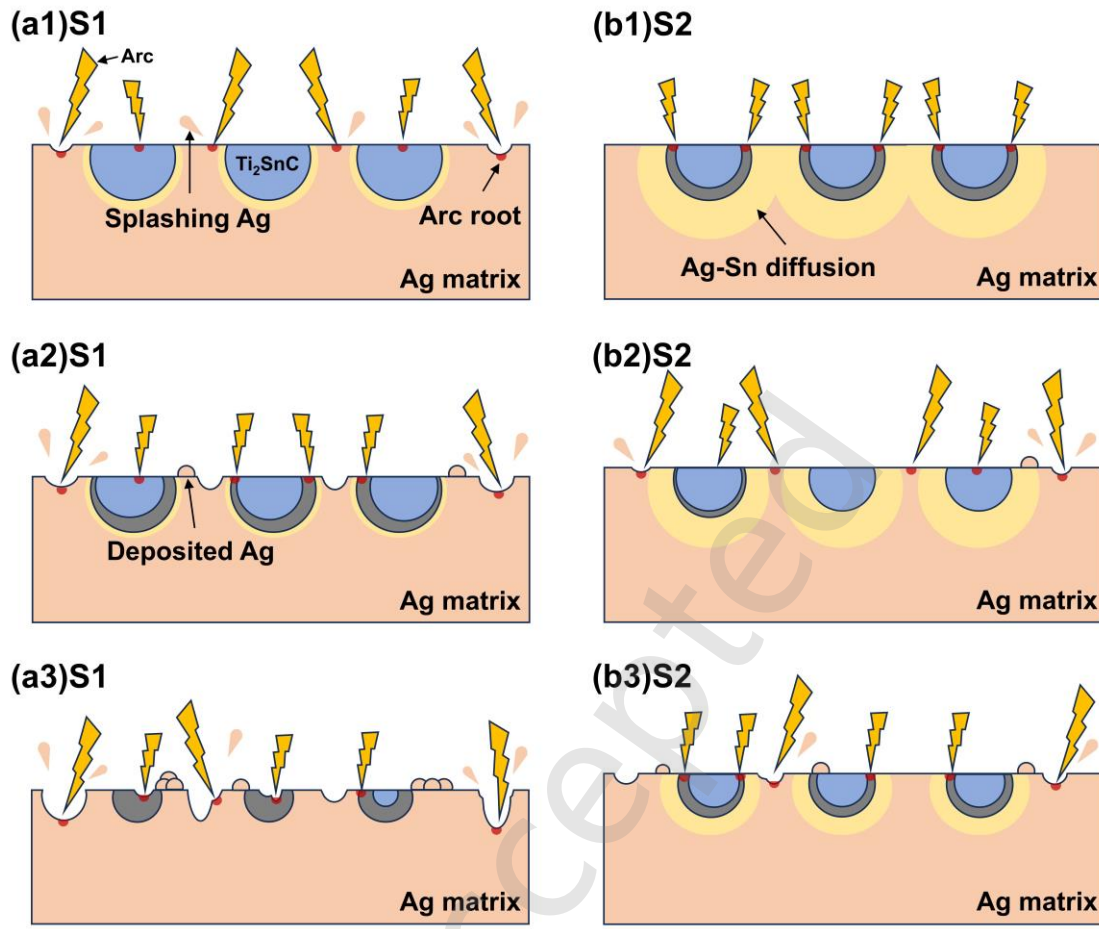


Fig. 15 Proposed schematic illustration of the possible arc erosion evolution of (a) sample S1 and (b) sample S2; 1, 2, and 3 represent the early, intermediate, and late stages of arc erosion, respectively.

For the sample S2 of Ag/TSC@TO, the pre-oxidation process formed a primarily TiO_2 -based oxide layer on the surface of the TSC reinforcing phase, and enhanced the interface bonding between the reinforcing phase and the Ag matrix by creating multiple diffusion layers of $(\text{Ag}+\text{Sn})/(\text{TiO}_2+\text{Ag}+\text{Sn})/(\text{Ti}_2\text{Sn}_{1-x}\text{C}+\text{Ag})$ that cross the interface. Therefore, during the early stage of arc erosion (Fig. 15b1), the preformed oxide layer and the Sn–Ag diffusion zone may influence the local arc–material interaction, which could contribute to the lower arc energy and reduced damage to the Ag matrix observed for S2 [44]. This explains that in the intermediate stage of arc erosion (Fig. 15b2), as the pre-formed oxide interface layer gradually depletes in resisting the damage from the electrical arc, the exposed TSC and Ag matrix simultaneously interact with the electrical arc. This continues to disperse the electrical arc and reduce its energy, thereby minimizing arc-induced damage

to the Ag matrix and reducing material transfer. Although the preformed TiO_2 -rich interface layer may be progressively consumed during continuous arc erosion, the oxide-containing interface of Ag/TSC@TO may undergo continued evolution during arc erosion (Fig. 15b3). The subsequently exposed Ti_2SnC may still undergo a “gradual decomposition–slow oxidation” process under the high-temperature arc and air environment, resulting in the formation of new oxidation products that may further contribute to arc erosion resistance [45]. This possible evolution of the oxide-containing interface may influence the spatial distribution of arc attachment and local arc–material interaction, thereby reducing the tendency for severe localized melting and large-sized Ag melt pool formation. Therefore, in contrast to the 2.10% mass loss of Ag/TSC composite in sample S1, sample S2 with Ag/TSC@TO demonstrates stronger resistance to arc erosion and extremely low material loss of 1.20% (Fig. 5). Furthermore, due to the still existence of the strengthening effect on Ag matrix from Sn–Ag interdiffusion and the persistent dispersion of electric arc by oxidation products, the fluidity of liquid Ag is effectively restricted and the splashing and migration of Ag molten pool is largely suppressed. Thus, the Ag/TSC@TO electrical contact has fewer defects and is smoother than the Ag/TSC electrical contact. As a result, it exhibits lower contact resistance and welding force in the later stage of arc erosion (Fig. 6).

5 Conclusions

To reveal the role of preformed oxide layers in the arc erosion of Ag-based electrical contact materials, Ag/TSC and Ag/TSC@TO composites were prepared and comparatively investigated. The main conclusions are as follows:

(1) Pre-oxidation induced surface decomposition and oxidation of Ti_2SnC particles, promoted Sn diffusion into the Ag matrix, and formed a TiO_2 -dominated transition layer at the Ag/TSC interface. Accordingly, the original $\text{Ag}/(\text{Ag}+\text{Sn})/(\text{Ti}_2\text{Sn}_{1-x}\text{C}+\text{Ag})/\text{Ti}_2\text{SnC}$ interface evolved into an $\text{Ag}/(\text{Ag}+\text{Sn})/(\text{TiO}_2+\text{Ag}+\text{Sn})/(\text{Ti}_2\text{Sn}_{1-x}\text{C}+\text{Ag})/\text{Ti}_2\text{SnC}$ composite interface. The oxide layer disrupted interfacial electrical continuity and introduced porosity, reducing the density and tensile strength from

9.24 to 8.59 g·cm⁻³ and from 124.68 to 71.19 MPa, respectively, while increasing the electrical resistivity from 7.31 to 24.28 μΩ·cm. Nevertheless, enhanced Sn–Ag interdiffusion and local compositional reconstruction produced slight solid-solution strengthening, increasing the interfacial hardness from 83.51 to 84.90 HV.

(2) Both composites exhibited anodic mass loss and cathodic mass gain during arc erosion, indicating an identical direction of material transfer. Compared with Ag/TSC, Ag/TSC@TO exhibited lower total mass loss, primarily because of reduced anodic erosion. Pre-oxidation decreased the average arcing time and arc energy from 39.1 to 35.9 ms and from 7783.6 to 5876.7 mJ, respectively, but increased the average contact resistance and welding force from 22.43 to 26.79 mΩ and from 0.44 to 0.50 N. Thus, although pre-oxidation compromises the initial contact properties, it markedly enhances arc erosion resistance.

(3) The experimental results are consistent with the possibility that the preformed and subsequently evolving oxide-containing interface contributes to the improved arc erosion resistance of Ag/TSC@TO by influencing the spatial distribution of arc attachment and local arc–material interaction. Such interfacial regulation may help disperse localized arc action, mitigate local heat accumulation, and suppress melt-pool expansion and liquid Ag splashing.

In summary, the pre-oxidation strategy utilizes the intrinsic oxidation behavior of Ti₂SnC to pre-establish an oxide-containing interface without introducing additional coating materials or foreign reinforcing phases, providing a distinct interfacial pre-design approach for Ag/MAX electrical contacts. The improved arc erosion resistance demonstrates its potential engineering value, although further optimization of the oxidation conditions, oxide-layer characteristics, and Sn diffusion is still required to balance the initial mechanical and electrical properties with long-term arc erosion performance.

Declarations

During the preparation of this manuscript, generative AI and AI-assisted technologies were used to assist with English language polishing and translation, as well as the preparation of the graphical

Acknowledgements

Not applicable.

Funding

This work was supported by the National Natural Science Foundation of China (52431003, 52171033), the Enterprise-University Cooperation Project (RH2500000024, RH2400002823 and RH2200003061), the College Students' Innovation and Entrepreneurship Training Program (202610360082 and S202610360429) and the Anhui Provincial Natural Science Foundation (2408085JX008)

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The individual contributions of authors to the manuscript should be specified in this section. The Journal recommends authors to use CRediT statements (the Contribution Roles Taxonomy).

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Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Declarations

Declaration of generative AI and AI-assisted technologies in the writing process and figure preparation

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Just Accepted