



Achieving ultra-low magnetic loss in Fe-based nanocrystalline soft magnetic composites via interface-domain coupling

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ABSTRACT

Soft magnetic composites (SMCs) are promising materials for enabling high-frequency, high-power-density magnetic components in power conversion systems for AI computing infrastructure. However, conventional SMCs suffer from substantial core loss (P_{cv}) under high-frequency operation, which significantly degrades the efficiency of magnetic components and limits their application in high-performance power converters. In this work, a $\text{Fe}_{74.8}\text{Si}_{9.5}\text{B}_{10}\text{P}_4\text{Cu}_{0.7}\text{Nb}_{1.5}$ composition with high B_s and amorphous forming ability (AFA) was designed, followed by the fabrication of amorphous spherical powders via a multi-stage atomization process, aiming at developing of ultra-low loss nanocrystalline SMCs through a dual-engineering strategy that couples interfacial insulation with magnetic-domain regulation. Controlled hydrolysis of tetraethyl orthosilicate (TEOS) was employed to synthesize a uniform SiO_2 insulating layer, yielding an optimized TEOS-0.12 SMC that exhibits an ultra-low P_{cv} of 12.3 W/cm^3 at 0.1 T and 1 MHz and a high saturation magnetic flux density (B_s) of 1.08 T, outperforming not only conventional FeNiMo and FeSiAl SMCs but also reported Fe-based amorphous and nanocrystalline SMCs to date. The synergy between high-density ultrafine nanocrystals and the SiO_2 interfacial insulating layer produces regular and wide magnetic domains with reduced domain-wall density, which facilitates coherent rotation and smooth domain-wall motion, leading to the simultaneous suppression of hysteresis and excess losses. This work establishes a generalizable method for designing high-efficiency magnetic cores tailored for next-generation AI servers, data-center power modules, and wide-bandgap-based MHz power electronics.

1. Introduction

Driven by the explosive growth of artificial intelligence (AI), cloud computing, and high-performance graphics processing units (GPUs), modern data centers are confronting unprecedented power challenges [1]. Therefore, it is urgent to develop advanced soft magnetic materials with high saturation magnetic flux density (B_s), low core loss (P_{cv}) and MHz-level frequency stability to facilitate the development of data center power modules towards higher power density and conversion efficiency [2,3]. Traditional ferrites offer excellent electrical resistivity and low loss but suffer from intrinsically low B_s , which limits the achievable current and power density. Conversely, metallic Fe-based alloys provide high B_s but are subject to severe eddy currents at high frequencies, preventing their direct use above several tens of kilohertz

[4–6]. Soft magnetic composites (SMCs), which integrate metallic powders with electrical insulation, represent the most promising pathway to simultaneously achieving high B_s , high electrical resistivity and improved frequency characteristics [7,8]. Among the metallic candidates, Fe-based nanocrystalline alloys (e.g., Fe-Si-B-Nb-Cu, Fe-Si-B-P-Cu) are particularly attractive because the exchange-averaging effect suppresses local magnetic anisotropy and overcomes the trade-off between P_{cv} and B_s typically faced in traditional crystalline alloys [9,10]. Despite their superior properties, the preparation of high B_s nanocrystalline alloy powders by atomization remains a significant challenge due to their limited amorphous forming ability (AFA) [11,12].

In addition to the inherent material challenges, MHz range loss in Fe-based SMCs remains exceptionally high, dominated by hysteresis loss (P_h) of the magnetization and reversal, eddy current loss (P_e) of Joule

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heating and excess loss (P_{exc}) of micro-current from domain wall motion [5]. To suppress P_{cv} , extensive efforts have focused on the introduction of insulating interfacial layers [13], particularly inorganic oxides such as Al_2O_3 , MgO , TiO_2 , ZnO , Fe_2O_3 and SiO_2 , which are typically prepared through methods including mechanical ball milling [14], sol-gel [15, 16], oxidation [17,18], and hydrolysis-precipitation [19]. Among them, SiO_2 is especially attractive due to its chemical inertness, high thermal stability and high resistivity up to $10^{13} \sim 10^{14} \Omega \cdot \text{m}$, which effectively suppress eddy currents between contacting powders [20]. Zhou et al. [21]. Reported that tuning the tetraethyl orthosilicate (TEOS)-derived SiO_2 coating can effectively reduce the loss of FeSiBCCr SMCs. However, the microscopic mechanisms, particularly the relationship between the SiO_2 interfacial structure, magnetic domain structure, and high-frequency excess loss (P_{exc}), remain insufficiently understood. Given that the pinning and motion of domain walls directly govern energy dissipation at high frequencies, elucidating and tailoring the domain structure is therefore important to suppressing P_{cv} and extending the operational frequency range [22].

Herein, we propose a synergistic interface-domain engineering strategy to achieve ultra-low MHz losses in high B_s Fe-based nanocrystalline SMCs. The $\text{Fe}_{74.8}\text{Si}_9\text{B}_{10}\text{P}_4\text{Cu}_{0.7}\text{Nb}_{1.5}$ (at.%) alloy, previously reported by the authors [23], was systematically designed to achieve high B_s and low coercivity (H_c) while ensuring sufficient AFA for powder fabrication and good thermal processability, making it a promising candidate for high-performance SMCs. In this alloy, the high Fe content enhances B_s , while Si, B, and P stabilize the amorphous structure [22]. Cu promotes the heterogeneous nucleation of α -Fe nanocrystals, whereas Nb enhances the AFA of the precursor and suppresses α -Fe grain growth during annealing owing to its large atomic radius and sluggish diffusion rate [17,24,25]. A high-resistivity SiO_2 insulating layer was constructed on the surface of the powder particles to modulate inter-particle coupling and further optimize the magnetic properties of SMCs, while optimized annealing induced a high density of ultrafine nanocrystals within the particles, thereby promoting the formation of well-defined planar domains with low domain wall density. The synergy between interfacial insulation and domain structure enables the simultaneous suppression of hysteresis, eddy-current, and excess losses under high-frequency conditions. This strategy provides an effective route for developing next-generation high-efficiency Fe-based nanocrystalline SMCs for MHz power conversion in point-of-load (POL) and voltage regulator module (VRM) applications for AI processors, GPUs, and data center power systems.

2. Experimental

Alloy ingots with nominal compositions of $\text{Fe}_{74.8}\text{Si}_9\text{B}_{10}\text{P}_4\text{Cu}_{0.7}\text{Nb}_{1.5}$ (at.%) were prepared by induction melting in a highly purified argon atmosphere. The corresponding amorphous powder was produced via a multistage crushing atomization process with a melt superheat of 150°C and a gas pressure of 2 MPa, during which high-speed rotary disk shearing (8000 rpm) further promoted secondary droplet breakup, resulting in a median particle size of approximately $15 \mu\text{m}$ and facilitating rapid solidification. A precursor solution was prepared by dissolving 0.5 g of polyvinylpyrrolidone (PVP, Aldrich) in 10 mL of anhydrous ethanol under mechanical stirring. The amorphous powder was then dispersed in the PVP solution and stirred at room temperature. TEOS (>99%, Aldrich) was subsequently added at predetermined content of $n \text{ mL g}^{-1}$ (where $n = 0.06, 0.12, 0.24$), representing the volume ratio of TEOS to powder weight, and the mixture was stirred continuously using a dropping funnel. The reaction yielded FeSiBPCuNb powders uniformly coated with SiO_2 , denoted as TEOS- n . Afterwards, TEOS- n powders were washed, dried and then mixed uniformly with 2 wt.% epoxy resin. Then, the insulated powders were compacted into toroidal TEOS- n SMCs ($\Phi 12.7 \text{ mm} \times \Phi 7.6 \text{ mm} \times 2 \text{ mm}$) by cold pressing at 1800 MPa. Based on DSC analysis (Fig. S1), the pressed TEOS- n SMCs were vacuum-annealed at 520°C for 30 min to induce

nanocrystallization. Meanwhile, for comparison, a control sample (TEOS-0 SMC) was prepared under identical condition without TEOS treatment.

X-ray diffraction (XRD, Bruker D8-Discover) was used to characterize the phase structures of the atomized powders. Scanning electron microscopy (SEM, Crossbeam 350) was used to observe the surface morphology of the TEOS- n powders. X-ray photoelectron spectrometer (XPS, Thermo Kalpha) was used to investigate the composition of the insulating layer. Transmission electron microscope (HRTEM, Talos F200X) was used to characterize the microstructure of the insulation layer. The contact potential difference (V_{CPD}) of the TEOS- n SMCs was measured by atomic force microscopy (AFM) in Kelvin probe force microscopy mode (AFM-KPFM, Bruker Dimension Icon). In KPFM, the V_{CPD} between the conductive probe and the sample surface is obtained by nullifying the electrostatic force through an applied DC bias, which reflects the local work function difference. Therefore, the V_{CPD} mapping was used to evaluate spatial variations in surface potential across different regions of the sample. The scanned area was set to $30 \times 30 \mu\text{m}^2$ to cover multiple powder particle boundaries. Before testing, all samples were subjected to vibratory polishing to eliminate the effect of surface roughness. Magnetic domains were observed using the magneto-optical Kerr microscope (Evico Magnetics GmbH, em-Kerr-highres). To minimize the effect of stress generated during polishing on the magnetic domains, all samples were vibratory polishing prior to measurement. The density of the TEOS- n SMCs was measured by the Archimedeian method, and the relative density was calculated as the ratio of the measured bulk density to the true density of the corresponding powder. Vibrating sample magnetometer (VSM, Lake Shore 7407) was used to measure the hysteresis loops of the magnetic powders. The permeability spectra of the TEOS- n SMCs were measured from 10 kHz to 120 MHz using an impedance analyzer (Keysight E4990A). The resistance of TEOS- n SMCs was measured using an LCR meter (TongHui, TH2838H), and the resistivity was calculated using the equivalent volume method. An AC B-H analyzer (Iwatsu, SY-8218) was used to measure the core loss of SMCs. Simulation results of magnetic loss distribution are obtained using COMSOL Multiphysics software with Maxwell equation.

3. Results and discussion

3.1. Morphology and microstructure

Multistage-atomized $\text{Fe}_{74.8}\text{Si}_9\text{B}_{10}\text{P}_4\text{Cu}_{0.7}\text{Nb}_{1.5}$ powders exhibit a predominantly spherical morphology with relatively smooth surfaces and a median particle size of $\sim 15 \mu\text{m}$ (Fig. 1a and b), which possess a complete amorphous structure (Fig. S2). Afterwards, a uniform SiO_2 coatings were successfully formed on the powder surfaces via a sol-gel process using TEOS as the precursor. It was found that as the TEOS content increased, the coating layer evolved from discontinuous to a continuous dense film (Fig. 1c-e). At an optimal TEOS content of 0.12 mL/g, a homogeneous encapsulating layer was obtained, whereas further increases led to partial SiO_2 agglomeration (Fig. 1f). As displayed in the XRD patterns (Fig. S3), all coated powders exhibit a broad diffuse peak around 45° , indicating that the alloy powder retains a fully amorphous structure. Notably, a secondary low-angle diffuse hump centered at approximately 23° is observed, which is assigned to the amorphous SiO_2 coating, consistent with the known amorphous nature of TEOS-derived SiO_2 .

The surface chemical composition and valence states of the TEOS-0.12 insulating layer was further investigated by XPS. The full XPS spectrum in Fig. S4a confirms the existence of Fe, Si, and O in the TEOS-0.12 powder sample. The corresponding $\text{Fe}2p_{3/2}$ spectrum (Fig. S4b) shows two main peaks at 706.6 eV ($2p_{3/2}$) and 711.2 eV ($2p_{3/2}$), which are attributed to Fe^0 and Fe^{3+} , respectively. The Si 2p spectrum in Fig. S4c displays two peaks at 99.2 and 102.7 eV, corresponding to the metallic Si^0 and oxidized Si^{4+} , respectively. According to the O 1s spectra, two main peaks attributed to the oxygen in metal oxides,

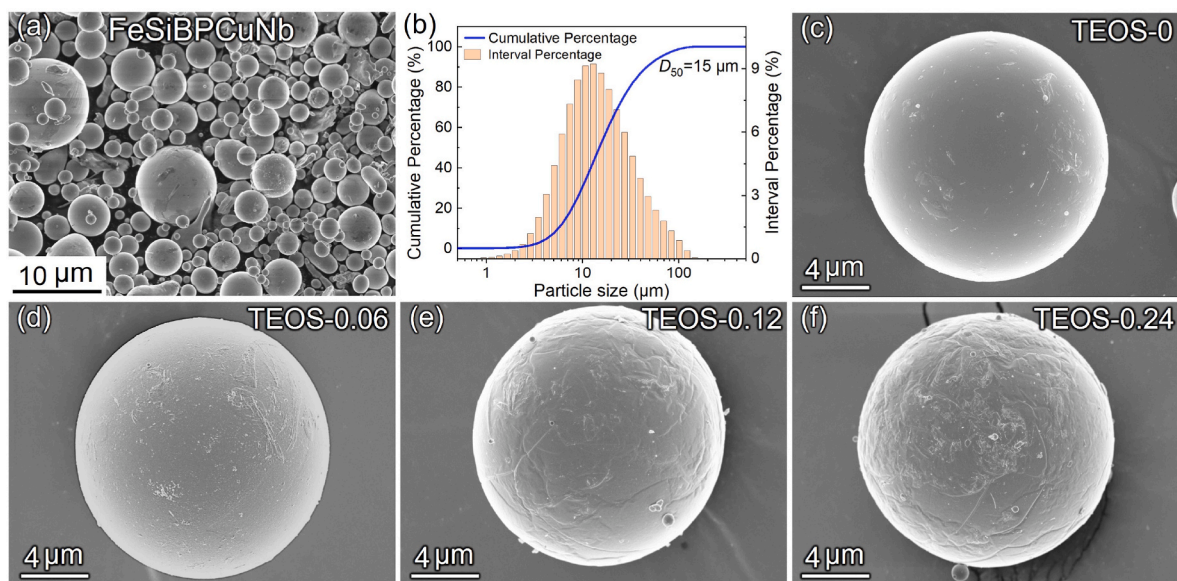


Fig. 1. (a) The morphology and (b) particle size distribution of ultrafine FeSiBPCuNb amorphous powder, (c-f) SEM images of TEOS-n particles.

hydroxyl groups and surface-absorbed water molecules, can be detected in all four samples. Combined with the peak at 530.6 eV and 532.2 eV of

the O 1s spectrum (Fig. S4d), these results confirm the existence of SiO₂ and a small amount of Fe₂O₃.

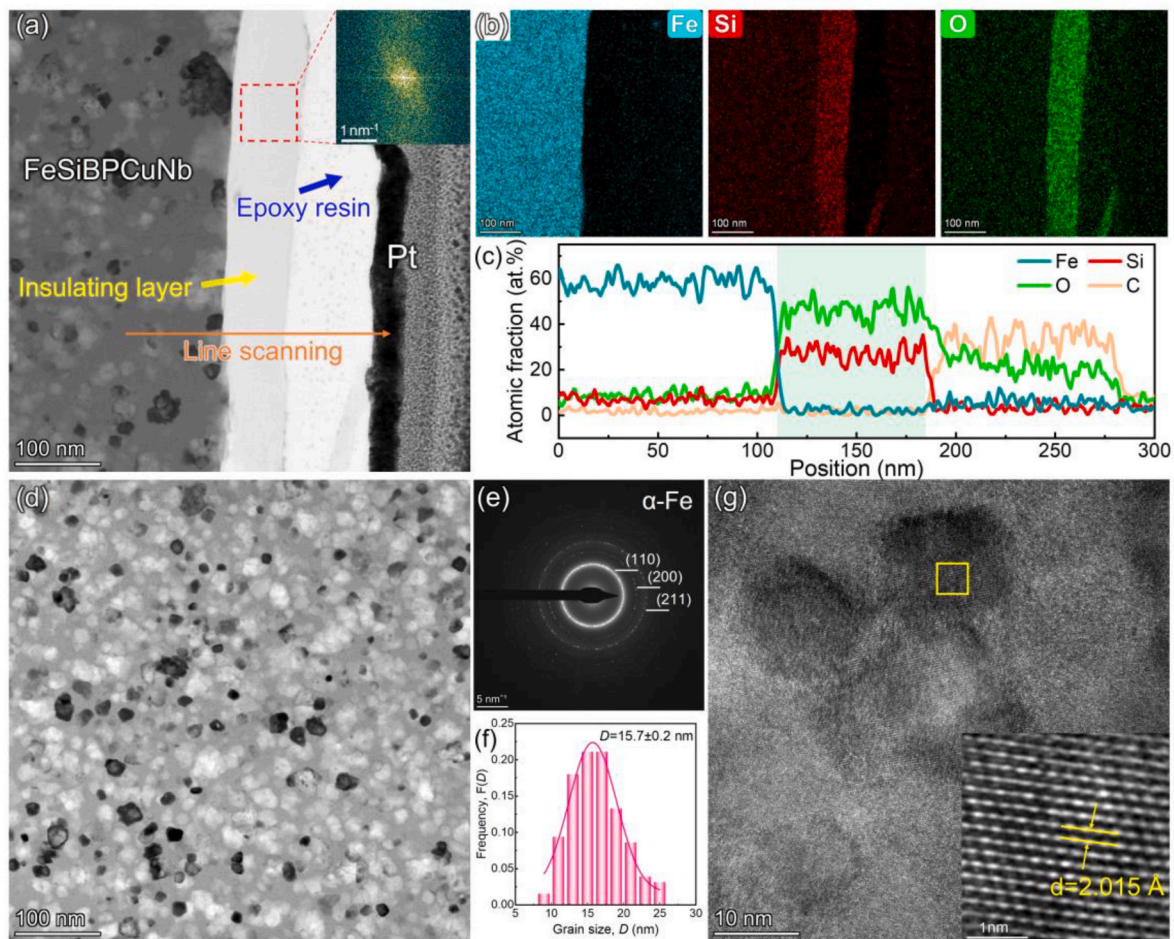


Fig. 2. (a) STEM image of the TEOS-0.12 SMC interface, the inset shows the Fast Fourier transform (FFT) pattern from the region marked by the red dotted box, indicating an amorphous structure. (b) EDS elemental mapping showing Fe, Si, and O element distribution. (c) Atomic concentration profiles along the orange line in (a). (d) Bright-field TEM image of FeSiBPCuNb nanocrystalline matrix, along with the (e) corresponding SAED patterns and (f) grain size histogram. (g) HRTEM image of the nanocrystalline matrix, inset shows the inverse Fourier transform of the highlighted yellow region.

The interfacial and nanostructure characteristics of TEOS-0.12 SMC were further investigated. As shown in Fig. 2a, a uniform and compact interfacial insulating layer with thickness of ~ 74 nm is clearly observed at the boundary of FeSiBCuNb matrix. The fast Fourier transform (FFT) pattern obtained from the highlighted red square shows no diffraction spots, clearly indicating an amorphous structure. EDS elemental mapping (Fig. 2b) reveals that the inner insulating layer is continuous and rich in Si and O elements. The corresponding line scan profiles (Fig. 2c) across the interface display a sharp compositional transition from Fe-rich to Si/O-rich regions, verifying the existence of an amorphous SiO_2 layer. The microstructure of the FeSiBCuNb matrix is depicted in Fig. 2d, revealing an amorphous-nanocrystalline dual-phase structure composed of uniformly distributed ultrafine nanocrystalline grains with average size of ~ 15.7 nm (Fig. 2f). Selected-area electron diffraction (SAED) pattern (Fig. 2e) exhibits diffraction rings corresponding to the (110), (200), and (211) planes of α -Fe, confirming the formation of bcc nanocrystals. High-resolution TEM imaging (Fig. 2g) further identifies lattice fringes with an interplanar spacing of 2.015 Å for the (110) planes of α -Fe, corroborating the SAED observations.

3.2. Soft magnetic properties

The influence of interfacial insulation layer on the magnetic hysteresis loops of TEOS-n SMCs is observed in Fig. 3a. The FeSiBCuNb SMCs without insulation (TEOS-0) exhibits a B_s of 1.09 T. As SiO_2 introduced, B_s gradually decreases to 1.06 T for TEOS-0.24, reflecting magnetic dilution by non-ferromagnetic coating [26]. However, H_c shows a non-monotonic variation, which is closely related to the microstructure. The corresponding magnetic properties obtained from the hysteresis loops are summarized in Table S1. Due to inevitable presence of pores in SMCs, the relative density of TEOS-n SMCs is only $\sim 80\%$, which leads to a lower B_s compared to their bulk counterparts with the same alloy composition (Fig. S5). In addition, these pores serve as distributed air gaps and reduce the frequency dependence of permeability. As shown in Fig. 3b, all samples maintain excellent frequency stability across the range of 10 kHz to 1 MHz, ensuring reliable

operation under high-frequency conditions. Although the effective permeability (μ_e) of TEOS-n SMCs gradually decreases with increasing TEOS content, the TEOS-0.12 SMC still exhibits high value of 63. The P_{cv} behavior further highlights the competing effects of insulation and exchange coupling. It decreases initially and then increases with increasing TEOS content, and TEOS-0.12 SMC exhibits minimum values of 0.41 and 12.3 W/cm 3 at 0.1 T/100 kHz and 0.1 T/1 MHz, respectively (Fig. 3c). To further evaluate its performance at elevated frequencies, the P_{cv} of the TEOS-0.12 SMC was measured up to 3 MHz under various magnetic flux density (B_m) conditions. As illustrated in Fig. S6, P_{cv} generally rises with increasing B_m and frequency. At relatively low B_m , P_{cv} exhibits a slow growth trend across the entire frequency range up to 3 MHz. Although the loss increment in the 1-3 MHz range becomes more pronounced when B_m is increased to 30 mT, the overall core loss at 3 MHz remains effectively suppressed and maintains a relatively low value, with P_{cv} as low as 7.66 W/cm 3 at 30 mT and 3 MHz, which indicates the effectiveness of our interface-domain coupling strategy in sustaining ultra-low loss across the MHz regime. Since SMCs are widely used in alternating conditions with DC-bias fields, their DC-bias performance is a critical merit for practical applications in power conversion systems. Fig. S7 presents the DC-bias performance of the TEOS-n SMCs, measured as the permeability retention ratio ($\mu\%$) under superimposed DC magnetic fields up to 100 Oe. It can be seen that all samples exhibit a stable and gradual decline in effective permeability with increasing DC bias field. Meanwhile, under the 100 Oe bias condition, the DC-bias performance improves progressively with increasing TEOS content, elevating the percent permeability ($\mu\%$) from 50% for the TEOS-0 SMC to 54% for the TEOS-0.24 SMC. This excellent DC-bias characteristic is primarily attributed to the high inherent B_s of the nanocrystalline matrix, which provides a fundamental threshold against early magnetic saturation. In addition, the SiO_2 insulation layer introduces additional distributed air gaps between powder particles, which increase the demagnetizing field and effectively shift the operating point away from saturation, further enhancing the DC-bias performance.

In addition to the TEOS content, the nanocrystalline structure (such as the grain size and volume fraction of α -Fe) plays an important role in

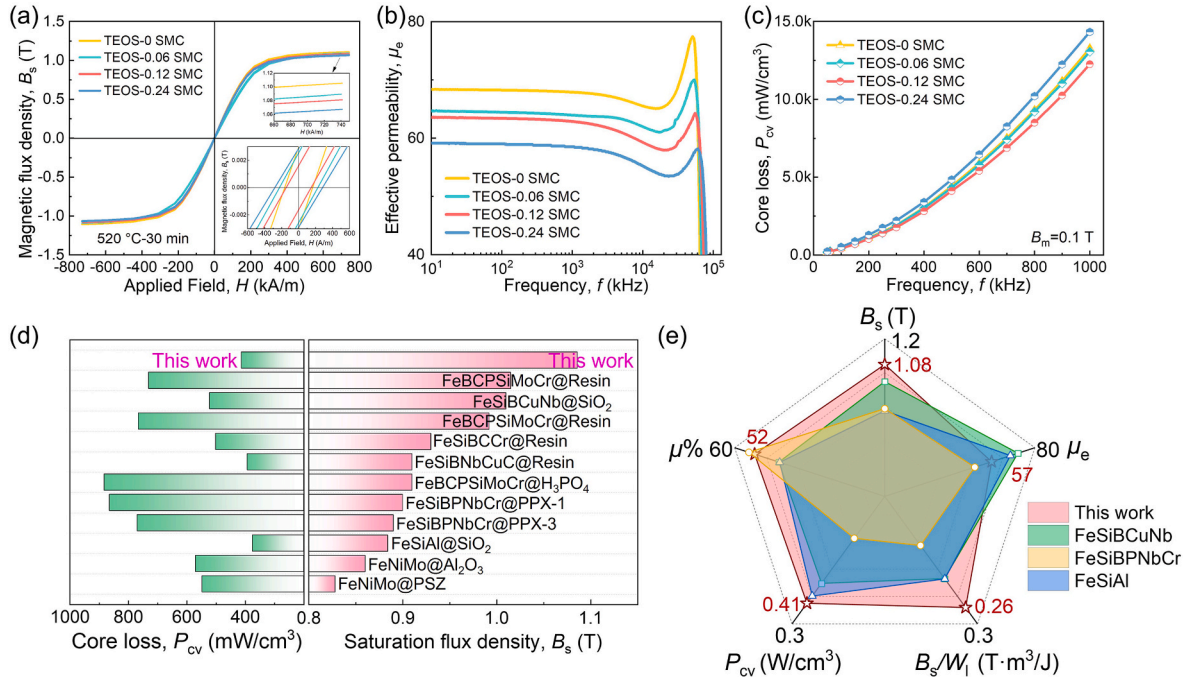


Fig. 3. Magnetic properties of TEOS-n SMCs: (a) hysteresis loops (inset shows enlarged view near saturation), (b) frequency dependence of μ_e , and (c) core loss ($B_m = 0.1$ T) versus frequency. (d) summary of B_s and P_{cv} between TEOS-0.12 SMC in this work and reported Fe-based amorphous, nanocrystalline, and conventional SMCs, (e) comparison of B_s , μ_e , P_{cv} (0.1 T/100 kHz), $\mu\%$ (100 Oe), and B_s/W_l of the TEOS-0.12 SMC and representative crystalline FeSiAl, amorphous FeSiBCuNb, and nanocrystalline FeSiBCuNb SMCs.

governing the magnetic softness and B_s of SMCs (Fig. S8). Optimal annealing temperatures (520 °C) promote the formation of high-density α -Fe nanocrystals, which effectively average the magnetocrystalline anisotropy and consequently contribute to a reduction in P_{cv} . In contrast, low annealing temperatures (480 °C) lead to insufficient crystallization (Fig. S9). Although a comparable grain size (~ 14.5 nm) is obtained, the reduced volume fraction of nanocrystals results in weak exchange coupling among α -Fe grains, giving rise to increased effective magnetic anisotropy and ultimately leading to higher P_{cv} . To highlight the prominent comprehensive soft magnetic properties of the TEOS-0.12 SMC, previously reported data including B_s and P_{cv} (0.1 T/100 kHz) of the SMCs fabricated under similar molding processes are summarized and compared with those of the present study in Fig. 3d. It is worth noting that the B_s values gradually increase from conventional FeNiMo and FeSiAl SMCs to Fe-based amorphous/nanocrystalline SMCs [4,8,27–33]. Remarkably, the TEOS-0.12 SMC developed in this work exhibits a high B_s of ~ 1.08 T, which can be attributed to the intrinsically high B_s of the $\text{Fe}_{74.8}\text{Si}_{9.8}\text{B}_{10}\text{P}_{4.0}\text{Cu}_{0.7}\text{Nb}_{1.5}$ nanocrystalline alloy, while also achieving one of the lowest P_{cv} values among the reported Fe-based SMCs. The combination of high B_s and low P_{cv} results in a markedly higher B_s/W_l ratio (where $W_l = P_{cv}/f$), indicating superior energy-conversion efficiency. Fig. 3e presents a radar plot comparing various soft magnetic properties of the TEOS-0.12 SMC in this study with those of representative crystalline FeSiAl, amorphous FeSiBPnCr, and nanocrystalline FeSiBCuNb SMCs. It is notable that the TEOS-0.12 SMC possesses an excellent comprehensive balance of soft magnetic properties, including ultralow P_{cv} , high B_s and B_s/W_l , large μ_e and μ_0 (100 Oe), which obviously outperforms other materials and offers significant potential for high-frequency, high-efficiency power devices.

3.3. Loss separation analysis

In high-power-density and high-current application scenarios, such as integrated inductors, reducing P_{cv} of nanocrystalline SMCs can effectively reduce heat generation and enhance the overall efficiency and current-carrying capability of power modules, thereby serving as a critical foundation for achieving device miniaturization and high reliability [34]. It is commonly known that P_{cv} rises sharply as frequency increases into the MHz range. However, the TEOS-0.12 SMC exhibits the slowest rate of increase, suggesting the effectiveness of the optimized interfacial insulation. To clarify the underlying mechanisms, a loss separation analysis was performed based on the classical loss separation theory [35], in which P_{cv} consists of P_h , P_e , and P_{exc} components [4,36]:

$$P_{cv} = P_h + P_e + P_{exc} = K_h B_m^x f + (K_e^{inter} + K_e^{intra}) B_m^2 f^2 + K_{exc} B_m^y f^z \quad (1)$$

where P_h can be obtained by the intercept (Eq. (1), hysteresis loss per cycle) of the fitted P_{cv}/f curves under different B_m conditions as depicted in Fig. 4a and b. P_e can be calculated according to the inter-particle (P_e^{inter}) and intra-particle (P_e^{intra}) eddy current losses equation in reference [37]. The P_{exc} is then determined by subtracting P_h and P_e from P_{cv} . Finally, the parameters K_{exc} , y , and z can be fitted using the nonlinear least squares method [38]. The simulated results of P_{cv} as a function of frequency and the B_m for the TEOS-0.12 SMC is shown in Fig. 4c. The measured loss values show well agreement with the simulated results, which confirms that the simulation method is reliable. The individual loss components (Fig. 4d) reveal that P_h dominates below 270 kHz, while P_{exc} becomes predominant at higher frequencies when $B_m = 0.1$ T. Moreover, P_e remains negligible across the entire frequency

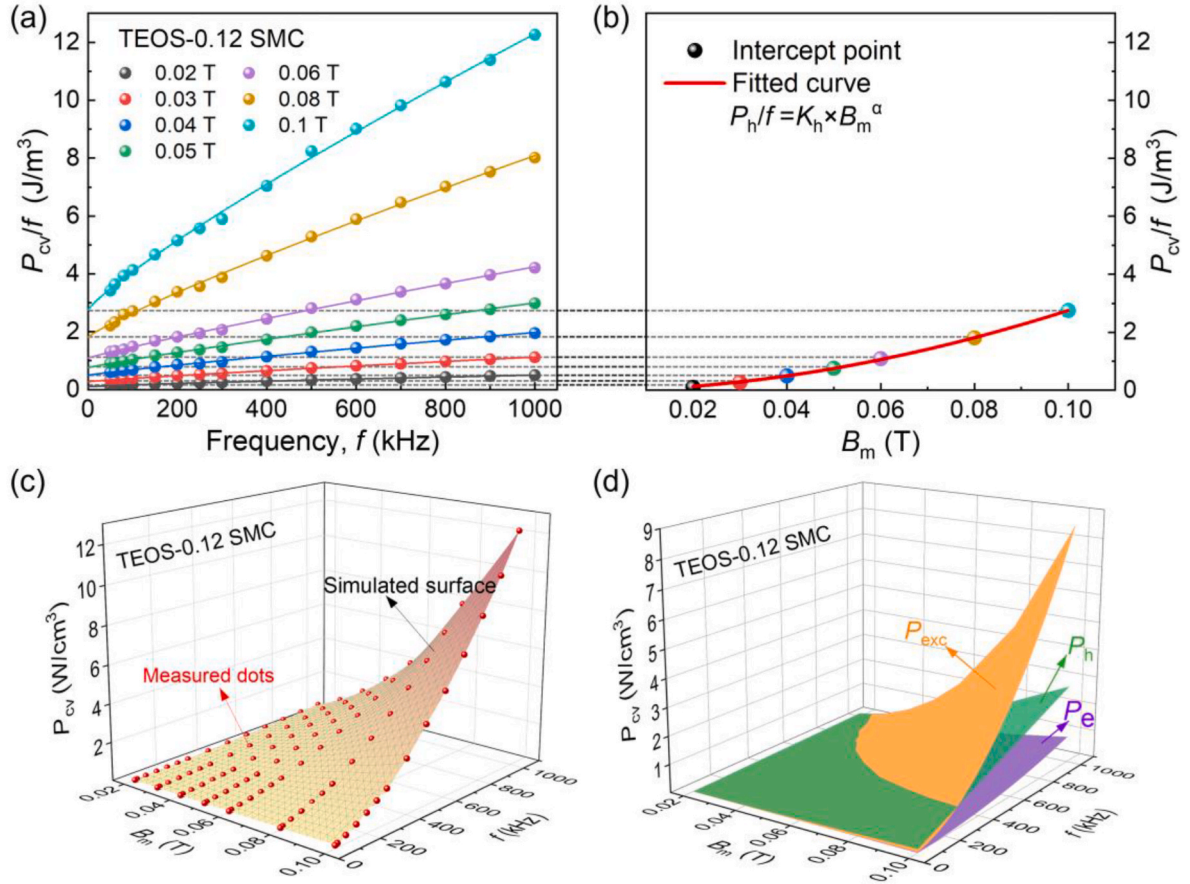


Fig. 4. (a) Experimental and simulation results for P_{cv}/f curve of TEOS-0.12 SMC under various B_m . (b) The intercepts of (a) on the vertical coordinate with dots and the fitted curve (red line) for quasi-static hysteresis loss versus B_m . (c) Mesh surface for P_{cv} as a function of f and B_m for TEOS-0.12 SMC. (d) Decomposed loss components: hysteresis loss (P_h), eddy current loss (P_e) and excess loss (P_{exc}), respectively.

range, which can be attributed to two factors: Firstly, interparticle eddy currents are suppressed by the SiO_2 interfacial insulating layer; Secondly, intraparticle eddy currents are confined by the ultra-fine powder and the high resistivity arising from the amorphous-nanocrystalline dual-phase structure. The loss separation is used to deeply explore the effects of the SiO_2 insulating layer content on the specific losses of TEOS-n SMCs (Fig. S10). From the loss separation results (Fig. S10a) and the measured H_c values, it can be seen that H_c and P_h exhibit a positive correlation, which can be attributed to the fact that both are governed by magnetic domain wall motion and the associated pinning effects. A higher H_c indicates stronger domain wall pinning and increased irreversibility during magnetization reversal, which results in an increased hysteresis loop area and thus higher P_h . Notably, the TEOS-0.12 SMC exhibits the lowest P_h , primarily ascribed to its uniform and dense SiO_2 insulating layer, which reduces the surface magnetic anisotropy [39]. In contrast, excessive TEOS content (e.g., 0.24 mL/g) induces SiO_2 agglomerates, which act as a surface defect and increase P_h . With increasing TEOS content, P_e^{inter} initially decreases and then rises (Fig. S10b). TEOS-0.12 SMC exhibits the lowest P_e^{inter} , which is attributed to the uniform SiO_2 interface insulating layer with high resistivity, while P_e^{intra} remains unchanged due to the same particle diameter [40]. The correlation between interfacial insulation and electrical resistivity was further confirmed by Kelvin probe force microscopy (KPFM, Fig. 5a and b). The TEOS-0 SMC exhibits a potential difference of ~ 21.6 mV between particles and boundaries, indicative of easier charge transport (Fig. 5c). Conversely, for the TEOS-0.12 SMC, the potential difference increases to ~ 42 mV, confirming enhanced electrical insulation. The KPFM potential trends are consistent with bulk resistivity measurements (Fig. 5d), corroborating the effectiveness of the interface engineering approach. Similarly, with an increase in TEOS content, P_{exc} decreases initially and then increases, reaching a minimum at TEOS-0.12 (Fig. S10c). It is clearly seen that P_{exc} makes a major contribution to P_{cv} in the MHz frequency range for all TEOS-n SMCs, while P_h and P_e together account for less than 35% (Fig. S10d). Therefore, minimizing high-frequency P_{exc} through controlled interfacial structures is essential

for achieving ultra-low loss in MHz applications.

3.4. Magnetization mechanism

The core advantage of the prepared nanocrystalline SMCs lies in their low power loss, which is intrinsically dictated by their magnetization dynamics. Magnetic spectroscopy serves as a crucial bridge connecting macroscopic losses with microscopic magnetization dynamics. Therefore, the magnetic spectrum fitting was performed to further elucidate the mechanisms responsible for the loss variations in TEOS-n SMCs. According to previous studies, the complex permeability of SMCs is commonly considered to consist of contributions from domain wall motion and spin rotation, which can be fitted using the equation given in reference [36]. The corresponding fitting parameters for all TEOS-n SMCs are summarized in Table 1. Based on these parameters, the real part of the permeability spectra was calculated. As a representative example, the simulated result for the TEOS-0.12 SMC was shown in Fig. 6a. It can be observed that the permeability contributed by magnetization rotation is higher than that by domain wall motion, indicating the dominant role of rotation in the high-frequency magnetization process. This dominance is from the distinct relaxation times inherent to each mechanism, which control their dynamic response. As the frequency increases into the MHz range, domain wall motion needs a longer relaxation time, which lags behind the rapid oscillations of the external magnetic field. In addition, the magnetic domains are confined within individual particles in terms of metal SMCs, and domain walls are strongly pinned by defects on the particles surface. This inherently suppresses large-scale domain wall motion. Consequently, the magnetization process primarily relies on magnetization rotation (with short relaxation time), which can respond rapidly to a high-frequency external field [41]. The relative contribution of domain wall motion (K_{dw}) in the magnetization process can be derived from domain wall motion static susceptibility χ_{d0} and spin rotation static susceptibility χ_{s0} through the formula $K_{\text{dw}} = \chi_{\text{d0}}/(\chi_{\text{d0}} + \chi_{\text{s0}})$ (Table 1). K_{dw} and P_h exhibit an inverse correlation for TEOS-n SMCs, indicating that domain wall motion is

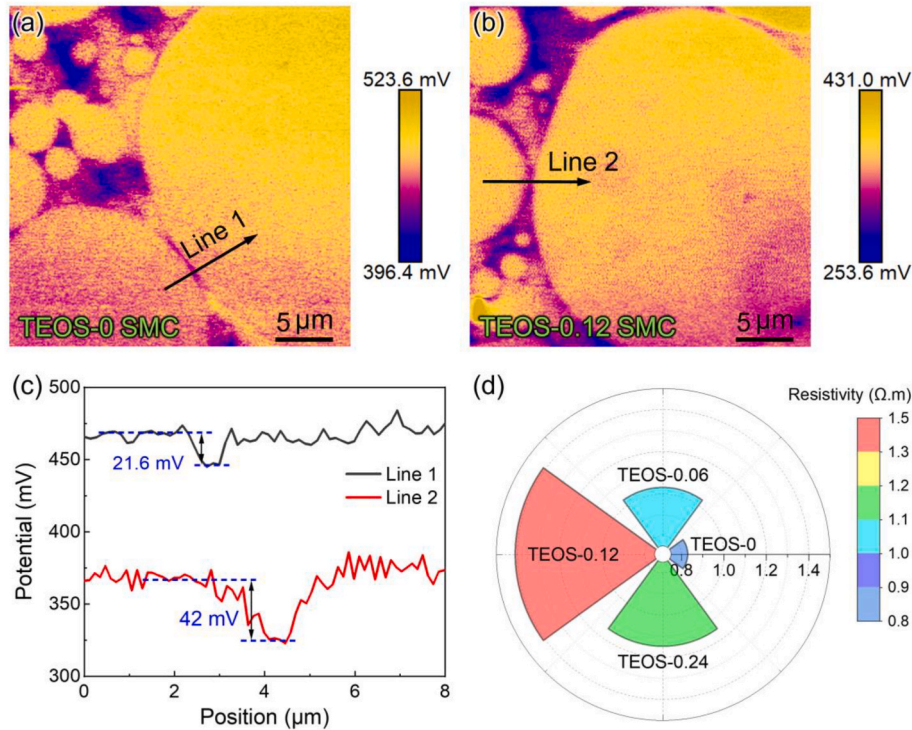
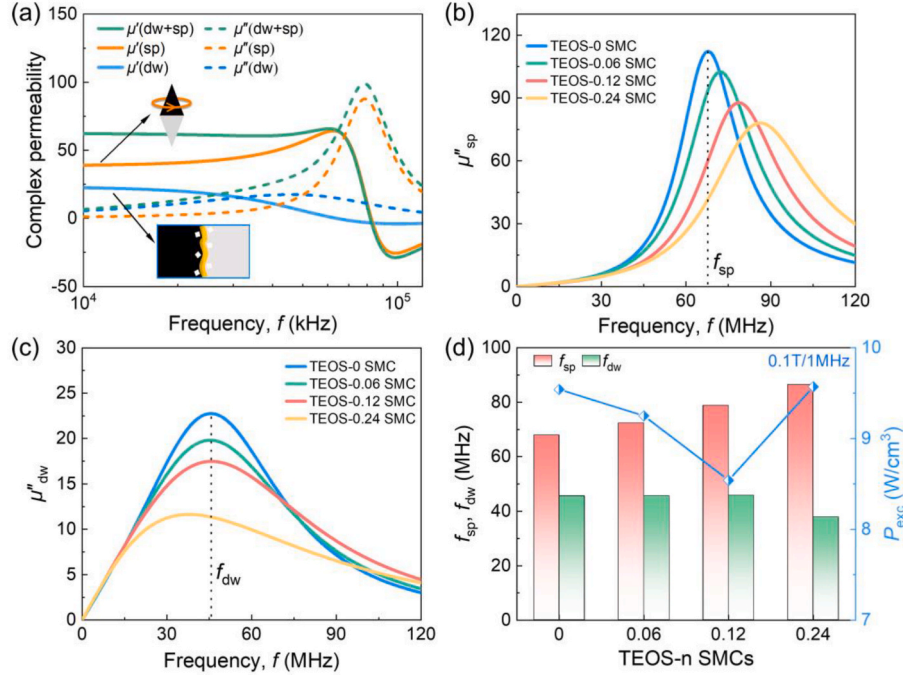


Fig. 5. KPFM maps of (a) TEOS-0 and (b) TEOS-0.12 SMCs, (c) corresponding potential profiles along the black arrows, (d) resistivity comparison of the SMCs with different TEOS contents.

Table 1

The simulation parameters in permeability spectra of TEOS-n SMCs.

TEOS-n SMCs	Domain wall component			Spin relaxation component			$K_{dw} \chi_{d0}/(\chi_{d0} + \chi_{s0})$
	χ_{d0}	ω_d	β	χ_{s0}	ω_s	α	
n = 0	24.42	3.45×10^8	4.05×10^8	41.82	4.34×10^8	0.189	0.369
n = 0.06	23.41	3.62×10^8	4.79×10^8	41.71	4.63×10^8	0.208	0.360
n = 0.12	23.03	3.99×10^8	6.15×10^8	38.23	5.07×10^8	0.223	0.376
n = 0.24	18.65	4.50×10^8	9.82×10^8	39.55	5.61×10^8	0.262	0.320

**Fig. 6.** (a) Simulated complex permeability spectra of TEOS-0.12 SMC. (b-c) The μ''_{sp} and μ''_{dw} in the imaginary part of complex permeability for TEOS-n SMCs. (d) Evolution of f_{sp} , f_{dw} , and P_{exc} (at 0.1 T/1 MHz) versus TEOS content.

more favorable than magnetization rotation for achieving low P_h . [42]. For TEOS-n SMCs, the measured spin rotation resonance frequencies f_{sp} for TEOS-n SMCs are 67.9 MHz, 72.3 MHz, 78.8 MHz, and 86.5 MHz, while the domain wall resonance frequencies f_{dw} are 45.5 MHz, 45.6 MHz, 45.8 MHz, and 37.8 MHz (Fig. 6b and c). This difference in resonance frequency correlates closely with P_{exc} . It is widely recognized that P_{exc} arises from two mechanisms: local eddy currents induced by non-uniform domain wall motion and resonance loss associated with domain walls and spin [22,43]. The evolution of these resonances is key to understanding high-frequency excess losses. As listed in Table 1, χ_{s0} is much higher than χ_{d0} , which indicates that spin rotation dominates dynamic magnetization for TEOS-n SMCs [44]. With the increase in TEOS content, f_{sp} increases gradually, resulting in a significant decrease in P_{exc} . However, when the TEOS content reaches 0.24 mL/g, P_{exc} increases instead (Fig. S10d). This is due to the strong damping of the domain wall, which leads to a lower f_{dw} , resulting in an increase in the resonance loss of the domain wall [36], as shown in Fig. 6d. Overall, the optimized TEOS-0.12 SMC achieves a balance between interparticle insulation and magnetic coupling, effectively minimizing high-frequency P_{exc} through controlled domain-wall and spin dynamics.

3.5. Magnetic domains and dynamic evolution

To explore the reason why TEOS-0.12 SMC exhibits the lowest core loss, the shape and response of magnetic domains in TEOS-0.12 SMC under varying magnetic fields were observed (Fig. 7). The TEOS-0.12 SMC exhibits a regular magnetic domain morphology, which is

induced by the low effective magnetic anisotropy resulting from strong exchange coupling between ultrafine nanocrystals (Fig. 7a1). Such regular planar magnetic domains facilitate coherent rotation and smooth domain wall motion, enabling excellent soft magnetic properties through reduced magnetization resistance [37,45]. As the applied field increases from 0 Oe to 1000 Oe, the magnetic domains gradually expand and eventually integrate into a single-domain state at 1000 Oe, indicating magnetic saturation (Fig. 7a2-a7). In contrast, the TEOS-0 SMC and TEOS-0.24 SMC exhibit incomplete magnetization under the same field (Fig. 8a and b), with residual unmagnetized regions resulting from a more significant pinning effect. These differences confirm that the TEOS-0.12 SMC reaches saturation with the smallest driving field, indicating that minimal domain wall resistance and the lowest P_h are achieved. In addition, TEOS-0.12 SMC exhibits a multi-domain structure separated by 180° domain walls, yet the number of domain walls was significantly reduced due to the synergistic effect of the insulating layer on the powder surface and the high-density ultrafine nanocrystals inside the powder [7]. A reduced domain wall density promotes magnetization rotation at high frequencies, aligning with the magnetic spectrum analysis results presented above. Consequently, the dominance of magnetization rotation leads to the effective suppression of P_{exc} , which is associated with localized micro-currents generated by non-uniform domain wall motion [5,46]. On the contrary, the TEOS-0 SMC displays a higher density of irregular domain walls, and these irregular domain configurations imply pronounced spatial non-uniformity during the magnetization process. Under high-frequency condition, these domain walls are subjected to pinning and depinning events, resulting in a

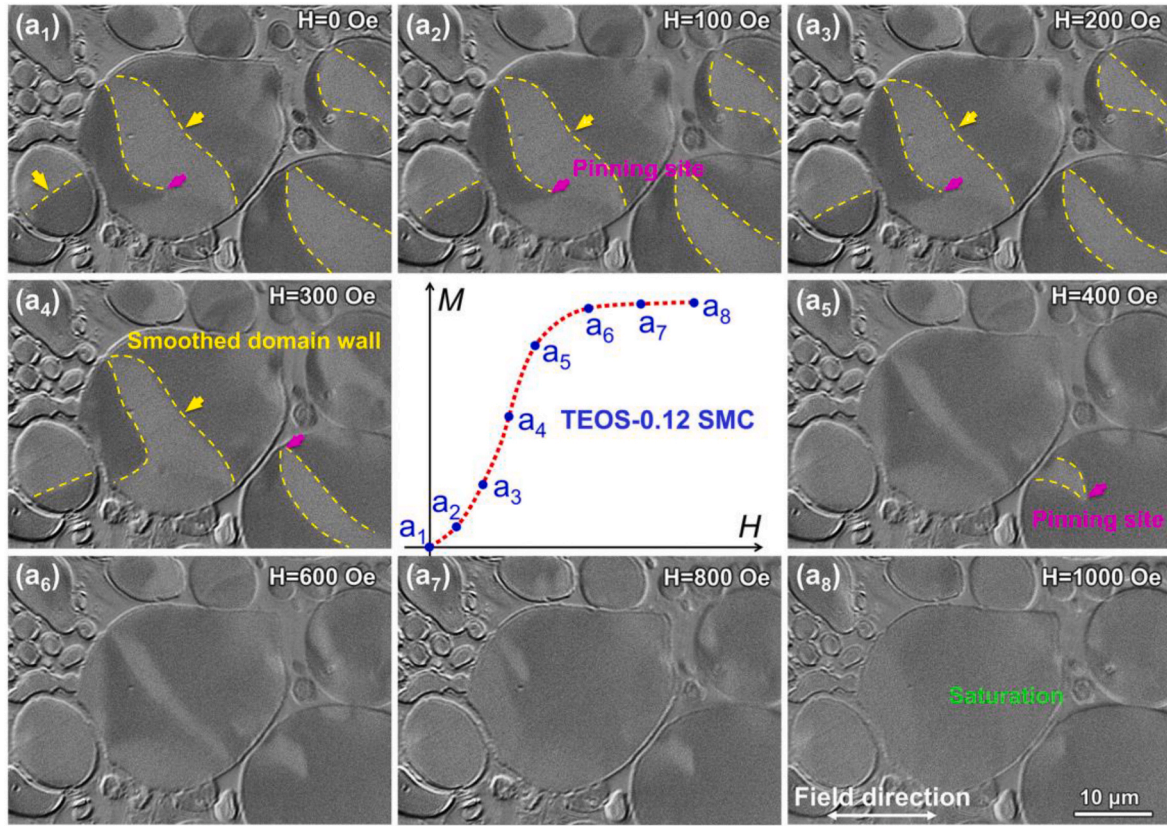


Fig. 7. Magneto-optical Kerr effect (MOKE) images of domain evolution in TEOS-0.12 SMC under varying magnetic fields.

highly discontinuous magnetization response. Such non-uniform motion also triggers intense localized fluctuations in magnetic flux density, thereby inducing localized micro-eddy currents. These localized dissipation events lead to an excessive conversion of magnetic energy into thermal energy, ultimately magnifying P_{exc} within the MHz regime. More critically, distinct trans-granular domains are observed in the TEOS-0 SMC, confirming a robust inter-granular exchange coupling that bypasses particle boundaries. Although such coupling enhances permeability at quasi-static frequencies, it would provide disadvantages on high-frequency P_{exc} , which provides a pathway for macro-scale eddy current loops and deteriorates dynamic energy dissipation. This extended magnetic response allows power loss to propagate across multiple particles, ultimately resulting in the sharp escalation of both classical P_e and P_{exc} . These contrasting domain behaviors, ranging from the exchange-coupled trans-granular domains in TEOS-0 SMC to the isolated micro-domains in TEOS-0.12 SMC, agree well with the macroscopic electrical properties, thereby mutually validating the effectiveness of interface structure regulation. The electrical resistivity of the TEOS-0.12 is higher than that of the TEOS-0 SMC (Fig. 5d). This increased resistivity signifies the formation of a more integrated and robust SiO_2 insulating layer on the powder surfaces, which acts as an electrical and magnetic barrier. This barrier not only shortens long-range electron transport to minimize classical eddy current loss, but also effectively decouples the inter-granular exchange interactions. Consequently, the constructed insulation layer not only severs macro-scale eddy current paths but also disrupts the magnetic continuity, thereby decoupling the magnetization responses between adjacent particles and minimizing total P_{cv} of the TEOS-0.12 SMC in the MHz regime. Despite the contribution of the insulating layer to reducing P_{cv} , the volume fraction of nanocrystals also has a great impact on P_{cv} . A low annealing temperature would lead to insufficient nanocrystal formation, which in turn leads to high effective magnetic anisotropy. This leads to an irregular magnetic domain structure that pins the domain wall and

consequently results in high core loss (Fig. S11).

To visualize the spatial distribution of magnetic loss in the toroidal cores, finite-element electromagnetic simulations were conducted (Fig. 9a–d). The loss of TEOS-0 SMC localized in the inner circumference, especially under high frequency (100 kHz, 1 MHz), which originates from poor electrical insulation between magnetic particles. These local “hot zones” of magnetic dissipation act as micro-sources of excess loss, consistent with their high P_{exc} components. In contrast, TEOS-0.12 SMC exhibits a much radially uniform loss distribution, the homogeneous color gradients across the cross-section indicate that the desired SiO_2 interfacial layer promotes uniform magnetic flux distribution by enhancing the resistivity of the SMC, which in turn enables uniform magnetization and suppresses high-frequency P_{exc} [47]. The simulations further corroborate that the low-loss performance of TEOS-0.12 SMC results from microscale flux homogenization and eddy-current confinement, highlighting the critical role of interface engineering in achieving high-efficiency SMCs for MHz-range applications.

The present study elucidates the fundamental mechanisms by which interface and magnetic domain engineering enhance the performance of Fe-based nanocrystalline SMCs. By adjusting the microstructure, the energy balance of the system is changed, thereby inducing the formation of a low-loss magnetic domain morphology. As illustrated in Fig. 10, three representative microstructures result in distinct soft magnetic properties due to different energy competition mechanisms. Without insulating layer (Fig. 10a), Fe-based nanocrystalline powders contact direct facilitates macro-scale eddy currents across the entire cross-section. Moreover, the irregular magnetic domain structures induce non-uniform magnetization response and accelerate P_{cv} at high frequencies. Even with insulation layer, Fe-based nanocrystalline powders with low volume fraction of α -Fe nanocrystals, have limited ability to average the magnetocrystalline anisotropy, resulting in a relatively large effective anisotropy (Fig. 10b). This in turn induces irregular and refined domain structures with strong domain wall pinning, thereby

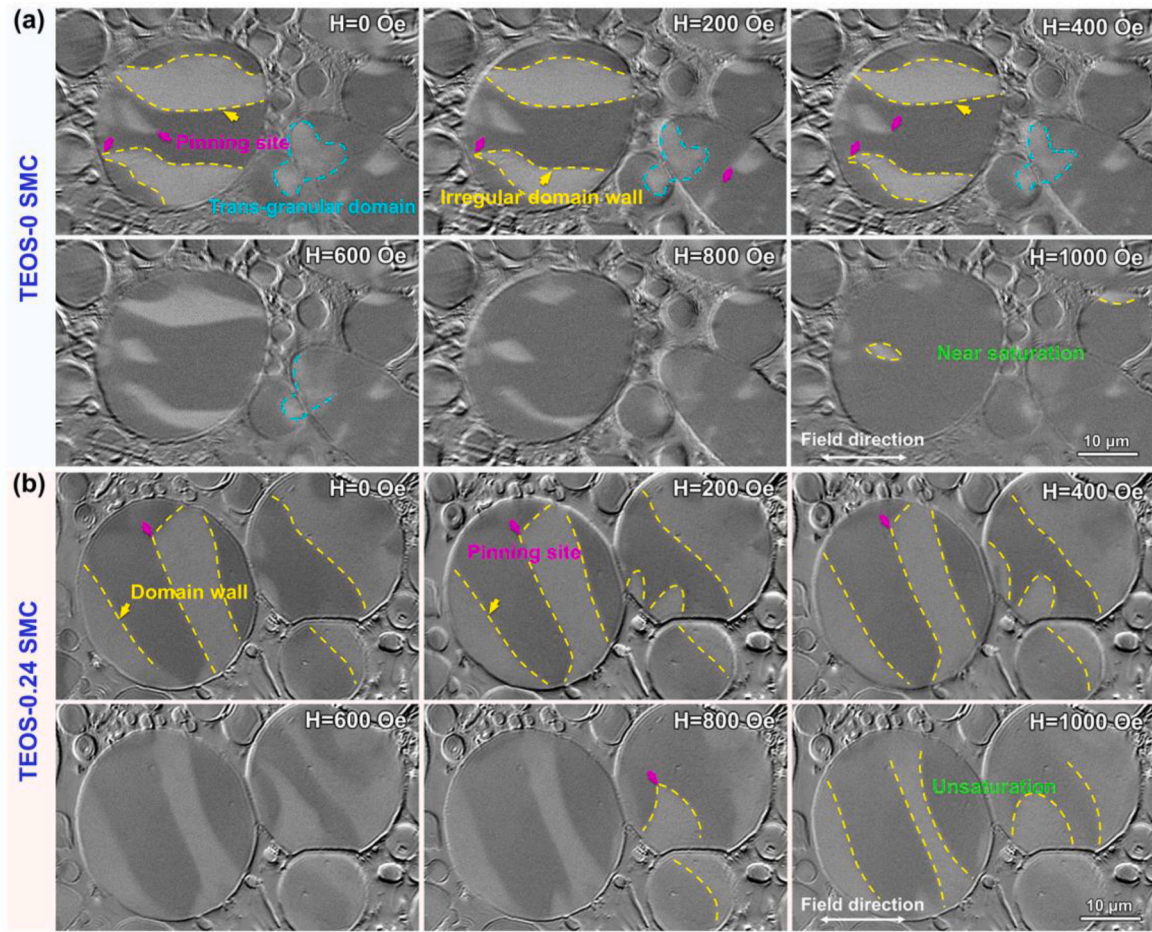


Fig. 8. Magnetic domain evolution under the magneto-optical Kerr effect (MOKE) of the TEOS-n SMCs. (a) TEOS-0 SMC. (b) TEOS-0.24 SMC.

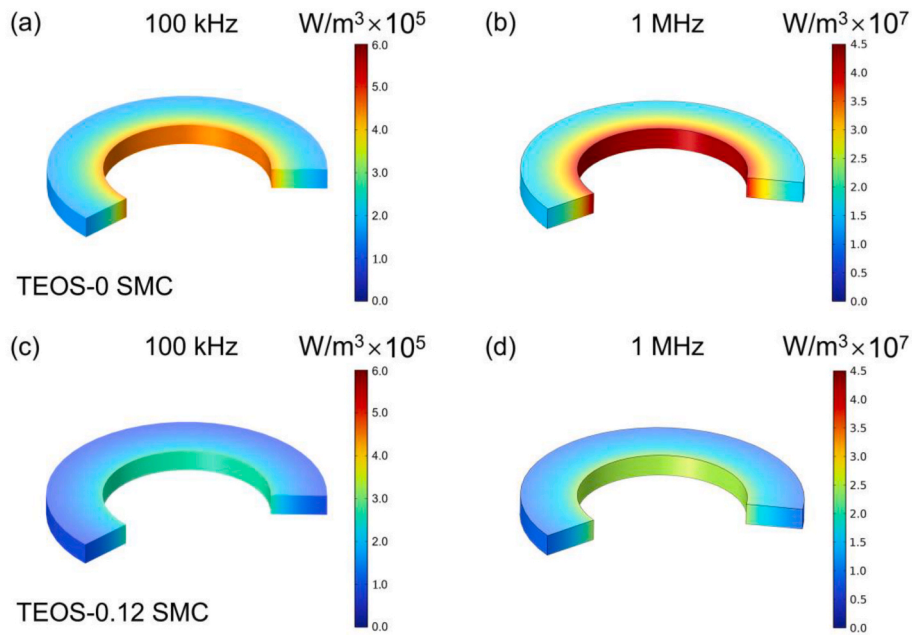


Fig. 9. Simulated magnetic core losses distribution in (a) TEOS-0 SMC and (b) TEOS-0.12 SMC at 100 kHz, and 1 MHz.

deteriorating the soft magnetic properties. When the SMCs possess optimal insulation and heat treatment (Fig. 10c), they exhibit lower P_{cv}

owing to the cooperative effect of interface insulation, strong exchange coupling among nanocrystals, and the formation of regular planar

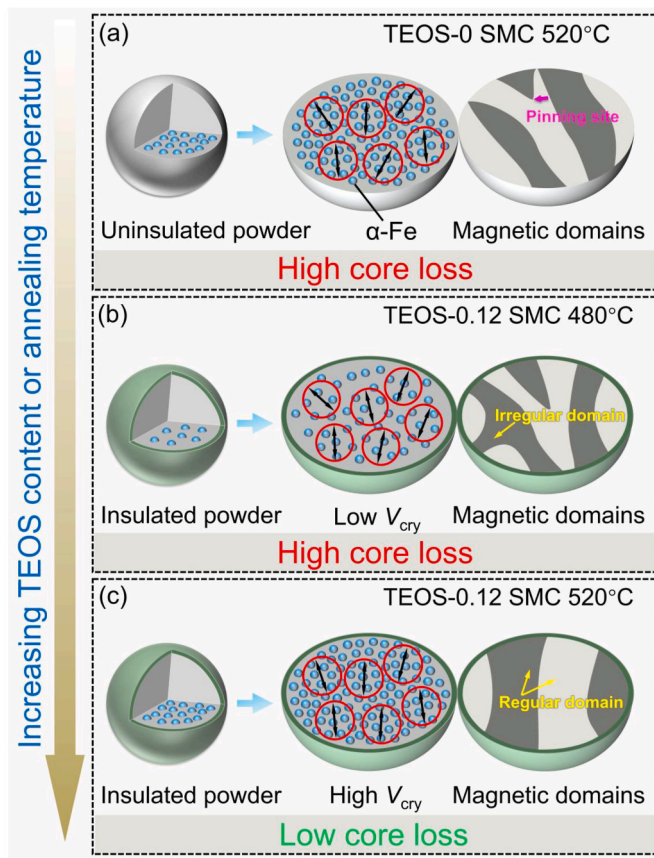


Fig. 10. Three representative scenarios illustrating the correlation among insulation and magnetic domain configuration with core-loss behavior in Fe-based nanocrystalline SMCs. (a) Fe-based nanocrystalline SMCs without insulation exhibit particle contact and irregular multi-domain configurations, resulting in high P_{cv} ; (b) The low crystallization volume fraction within the powder particles leads to a high effective magnetic anisotropy, forming high-density magnetic domains with irregular walls, which results in high P_{cv} ; (c) In Fe-based nanocrystalline SMCs with both optimal insulation and crystallization volume fraction, a transformation to regular planar domains with low wall density occurs, significantly reducing P_{cv} .

domains with low wall density.

4. Conclusions

This work establishes a comprehensive interface-domain co-engineering method that enables Fe-based nanocrystalline soft magnetic composites (SMCs) to achieve ultra-low MHz-range core loss, high flux density and unprecedented energy-conversion efficiency. The optimized TEOS-0.12 SMC exhibits an ultra-low P_{cv} of 12.3 W/cm³ at 0.1 T/1 MHz and a high B_s of 1.08 T, outperforming not only conventional FeNiMo, and FeSiAl composites but also previously reported Fe-based amorphous and nanocrystalline SMCs. These properties collectively deliver markedly enhanced power-conversion efficiency, enabling lower temperature rise and higher effective inductance in MHz-class power modules. Loss-separation analysis reveals that P_h governs the low-frequency region, whereas P_{exc} dominates above several hundred kHz, accounting for more than 65% of the total loss in the MHz range. The SiO₂ interfacial layer formed by controlled TEOS hydrolysis effectively interrupts interparticle eddy-current paths, thus reducing P_e . Concurrently, the amorphous-nanocrystalline dual-phase structure promotes exchange coupling, generating regular planar domains with low resistance to coherent rotation and domain-wall motion that minimize P_h . Magnetic spectrum fitting confirms that spin rotation is the primary magnetization

mechanism at high frequencies, and its high spin rotation resonance frequency contributes to the reduction of P_{exc} . In addition, the significantly reduced 180° domain walls density minimizes P_{exc} arising from micro-currents induced by non-uniform domain wall motion. Through the cooperative suppression of P_h , P_e , and P_{exc} , the TEOS-0.12 SMCs deliver not only the lowest MHz-range loss but also the highest reported MHz-level magnetic-energy conversion efficiency among Fe-based SMCs. Beyond setting a new performance benchmark, this work provides a universal design paradigm for next-generation high-efficiency magnetic cores suitable for AI server VRMs, high-density data-center power modules, and wide-bandgap semiconductor power electronics.

CRediT authorship contribution statement

Yanzhou Fan: Data curation, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. **Zhijun Guo:** Methodology, Supervision, Writing – review & editing. **Jinhong Fu:** Formal analysis, Investigation. **Changlong Jin:** Formal analysis, Investigation. **Fan Hu:** Formal analysis, Investigation. **Qianqian Wang:** Writing – review & editing. **Yaqiang Dong:** Data curation. **Qikui Man:** Data curation. **Zhe Jia:** Writing – review & editing. **Qiang Luo:** Writing – review & editing. **Baolong Shen:** Funding acquisition, Project administration, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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