# An organosilicon-loaded plastic scintillator synthesized for neutron/gamma discrimination

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## Abstract

In this work, we report a scintillator used for neutron/gamma discrimination, which consists of a polyvinyltoluene matrix loaded with monomers of dimethoxydiphenylsilane (DDS), and fluor dyes, namely 2,5-diphenyloxazole and 7-diethylamino-4-methylcoumarin as the primary fluorophore and wavelength-shifting, respectively. The DDS is an organosilicon compound containing double benzene rings that facilitates fluorescence emission and radiation resistance. Several measurements were performed to explore the scintillators optical and detection properties, including $^{238}$Pu-Be neutron radiation and relative light yield conducted by 137Cs gamma-ray source. It is found that the synthesized plastic scintillators are capable of excellently discriminating between neutron and gamma-rays. They exhibit a light output that are comparable to the commercial scintillator (EJ-200). The scintillators are low-cost productive, highly transparent, and somewhat soft but hard enough for post-production processing (machine cutting and polishing). This design may contribute a new strategy for highly efficient neutron/gamma discrimination organic scintillators.

## Full Text

## Preamble

### An Organosilicon-Loaded Plastic Scintillator Synthesized for Neutron/Gamma Discrimination

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In this work, we report a scintillator for neutron/gamma discrimination consisting of a polyvinyltoluene matrix loaded with dimethoxydiphenylsilane (DDS) monomers and fluor dyes—specifically 2,5-diphenyloxazole and 7-diethylamino-4-methylcoumarin as the primary fluorophore and wavelength-shifter, respectively. DDS is an organosilicon compound containing double benzene rings that facilitates fluorescence emission and radiation resistance. We performed several measurements to explore the scintillators' optical and detection properties, including $^{238}$Pu-Be neutron radiation exposure and relative light yield measurements using a $^{137}$Cs gamma-ray source. The synthesized plastic scintillators demonstrate excellent discrimination between neutrons and gamma-rays and exhibit light output comparable to commercial scintillator EJ-200. The scintillators are cost-effective to produce, highly transparent, and somewhat flexible yet hard enough for post-production processing (machining and polishing). This design may contribute a new strategy for highly efficient neutron/gamma discrimination in organic scintillators.

**Keywords:** Neutron detection, Plastic scintillation, Pulse shape discrimination, Polymethyl-methacrylate, Polystyrene

## Introduction

Organic scintillator-based neutron detection has been employed effectively for decades in nuclear and particle physics experiments, particularly within high-radiation environments [1–3]. However, neutron detection is always accompanied by gamma-rays caused by neutron activation of surrounding materials. Consequently, the capability to distinguish between neutrons and gamma-rays—known as pulse shape discrimination (PSD) [4,5]—is essential for reliable neutron spectroscopy using organic scintillators, especially when detecting high-energy neutrons in the presence of gamma-ray background [6]. For many years, neutron detection has predominantly relied on $^3$He proportional counters due to their excellent gamma rejection and high detection efficiency for thermal neutrons [7–9]. However, the extremely high cost of $^3$He has prevented widespread usage and stimulated intensive searches for relatively inexpensive alternative materials.

Among PSD-capable organic scintillators, plastic scintillators have recently emerged as promising candidates for large-scale scientific infrastructure, border security systems, and reactor monitoring applications [10]. Unlike single crystals constrained by manufacturing costs or liquid scintillators limited by toxicity, flammability, and handling challenges for large-area detectors, plastic scintillators offer distinct advantages: low production cost, inherent safety, ease of fabrication, and the ability to be manufactured in customizable geometries

[11,12].

The most widely used plastic scintillators are ternary systems composed of polystyrene (PS) or polyvinyltoluene (PVT) matrices doped with primary fluorophores and secondary wavelength shifters [13–15]. These aromatic hydrocarbon polymers contain benzene rings bonded to methyl ($CH_3$-) and vinyl ($CH_2=CH$-) groups. When irradiated by high-energy particles, molecular chains in such polymers undergo damage through scission and cross-linking [1], altering their chemical structure and generating free radicals that form color centers [16,17]. These radiation-induced transformations correlate with bond dissociation energies in the polymer backbone: 3.59 eV for C–C bonds, 3.71 eV for C–O bonds, and 4.61 eV for Si–O bonds [17].

Polysiloxane, also known as organosilicon with a backbone of alternating Si–O–Si bonds and organic substituents, exhibits high bond energy and excellent radiation resistance. Studies demonstrate that polysiloxane-based scintillators show no color yellowing at radiation doses of 100 kGy [18] and are capable of discriminating neutrons in gamma-ray background using time-of-flight (TOF) method [19] or PSD technique [20]. However, they are soft due to their elastic nature. Although cross-linking strategies can improve fluorescence scintillation [21], they exhibit inherently inferior PSD performance compared to conventional PS/PVT-based systems.

In this work, we have developed a novel PSD organic scintillator that combines the advantages of both pure PVT and polysiloxane matrices via copolymerization of 2,4-diphenyl-1,3,2-dioxasilolane (DDS) monomers with PVT resins, doped with 2,5-diphenyloxazole (PPO) and 7-diethylamino-4-methylcoumarin (MDAC) as the primary fluorophore and wavelength-shifting component, respectively. Through low-density self-polymerization, the DDS monomers form linear polysiloxane segments that interpenetrate the PVT cross-linked network, resulting in bulk scintillators with some elasticity while retaining adequate rigidity for mechanical machining. The synthesized scintillators are highly transparent, cost-effective, and demonstrate outstanding PSD performance, making them promising candidates for large-scale radiation detection applications.

## II. Samples Fabrication and Characterization

Vinyltoluene monomers ($C_9H_{10}$, >98%, m- and p-mixture, containing stabilizer TBC) were purchased from TCI Chemicals (Shanghai) Industrial Development Co., Ltd. DDS, MDAC, and the initiator 2,2'-Azobis(2-methylpropionitrile) (AIBN, >98%) were obtained from Shanghai Macklin Biochemical Co., Ltd. PPO was purchased from Shanghai YuanYe Biotechnology Co., Ltd., and divinylbenzene (DVB, containing stabilizer) was obtained from Meryer (Shanghai) Chemical Technology Co., Ltd.

The synthesis procedure for the scintillator samples is similar to that described in our previous work [13]. Prior to sample preparation, the vinyltoluene monomers and divinylbenzene were passed through an aluminum oxide column

to remove stabilizers, then mixed and stirred at room temperature for three minutes. The mixture was sonicated until a homogeneous, clear solution was obtained. Subsequently, fluorophores (30 wt% PPO, 0.2 wt% MDAC), radical polymerization initiator (0.05 wt% AIBN), and cross-linking agent (2 wt% DVB) were added to the mixture, which was stirred for an additional 2–5 minutes and transferred to flat-bottom glass vials for ultrasonication until the dopants were completely dissolved.

The mixed solution was poured into glass vials and treated repeatedly at least three times through a process of fast freezing with liquid nitrogen, vacuum degassing (3–5 min), and thawing at room temperature to completely remove dissolved oxygen. The vials were sealed in an oxygen-free atmosphere and heated in a thermostat to promote radical thermal polymerization. To prevent explosive polymerization, the thermostat temperature was maintained at 50.0 °C for two days, then slowly raised from 80.0 °C to 110 °C at a rate of 3 °C/h and maintained for 24 h. To prevent crack and bubble formation during annealing, the polymers were slowly cooled to room temperature at 3 °C/h, then placed in a constant temperature environment at 50.0–60.0 °C for internal stress release. Plastic scintillators with different DDS loadings (RK1 to RK5 corresponding to 20, 25, 30, 35, and 40 wt% DDS, respectively), measuring approximately 20.0 mm in diameter and 10.0 mm in thickness, were obtained by carefully breaking the vials after cooling to room temperature.

The scintillators were cut and polished to the same size as control samples (EJ-200, EJ-276). Thickness differences between samples were measured to be no more than 100 μm using a Mitutoyo micrometer (Mitutoyo Corp.). Shore hardness was evaluated using a portable digital durometer (Shore A hardness meter, Taizhou AI Measurement Instrument Co., Ltd.), with five equidistant points measured on a flat side of each sample and averaged to give the hardness value. Elastic recovery testing and excitation-emission spectra were performed using a metallographic microscope (BX53M, Olympus Corp.) and an FLS1000 fluorescence spectrometer (Edinburgh Instruments), respectively. Ultraviolet-visible transmission spectra were recorded using a UV-Vis spectrophotometer (PerkinElmer Lambda 750) over an emission wavelength range of 250–800 nm. Thermal decomposition behavior was studied from 30 °C to 800 °C using a thermogravimetric analyzer (STA 449 F3 Jupiter, Netzsch GmbH, Germany), and Fourier-transform infrared spectra (FTIR) were obtained using a Fourier transform infrared spectrometer (Nicolet 6700, Thermo Fisher Scientific, USA) to analyze structural and functional group information.

The scintillators were irradiated with a 20 Ci $^{238}$Pu-Be neutron source installed at the University of South China to study their PSD performance using an 8-channel 14-bit 500 MS/s flash ADC waveform digitizer (CAEN DT5730). All samples were wrapped in three layers of polytetrafluoroethylene (PTFE) tape on every surface except the one coupled to a Hamamatsu H1949-51 photomultiplier tube (PMT) with optical dimethyl silicone (Dow Corning, PMX-200). Signals collected from the PMT anode were converted to digital waveforms and analyzed

by CAEN DPP-PSD firmware, which combines digital QDC (charge integration) and shape discrimination functions for particle identification. A $^{60}$Co source (1.17 MeV, 1.33 MeV) in tandem with $^{137}$Cs (662 keV) was used for electron-equivalent energy calibration by Gaussian fitting to the Compton edge position of each source.

The light output (LO) of the scintillators, determined using the maximum position of the Compton edge in the $^{137}$Cs pulse height spectra, was compared with that of commercially available three-year-stored EJ-200 to obtain relative LO. To explore sample stability in terms of relative LO and PSD performance, measurements were carried out three times at two days, 96 days, and 172 days after preparation. Conditions including digitizer settings, PMT operation at -1600 V by high voltage power supply (ISEG Corp.), 10 cm distance from neutron source to samples, and lead brick shielding were kept constant to ensure consistency between measurements for each sample.

## III. Results and Discussion

### A. Physical Properties

DDS is a cost-effective organosilicon compound with double benzene rings in which organic dyes can be efficiently dissolved. Organosilicon compounds have demonstrated superior radiation resistance over conventional PVT- or PS-based scintillators [1,22–25]. Therefore, we expect that PVT-DDS based PSD organic scintillators will exhibit enhanced radiation stability relative to pure PVT-based scintillators. The aromatic matrices (PVT and DDS) inherently exhibit specific optical properties: they emit fluorescence primarily in the ultraviolet region (250–380 nm) [26], and their absorption spectra show noticeable absorption starting around 380 nm, increasing sharply below 350 nm [12]. Specifically, PVT exhibits excitation and emission peaks at 266 nm and 310 nm, respectively, while DDS shows corresponding peaks at 302 nm and 353 nm (Fig. S1, Supplementary Materials). To minimize self-absorption by the matrix, primary fluorophore PPO is added to absorb photons from matrix emission and emit photons with a peak at 393 nm. To maintain scintillation in the blue-green region of the optical spectrum that matches most PMT spectral sensitivity, a secondary fluorophore dye—wavelength-shifter MDAC—is chosen to shift the scintillator emission to 410–500 nm. As the primary fluorophore, PPO has high solubility in aromatic solvents, while MDAC, a secondary dye with high light output, is routinely used in plastic preparation. This PPO-MDAC dye pair likely leads to excellent PSD capability in PVT-DDS based scintillators from the perspective of basic energy transfer principles [27].

With 20–40 wt% DDS, the synthesized scintillators are colorless and optically transparent without yellowing or surface dye precipitation visible to the naked eye even six months after preparation (Fig. 1 Figure 1: see original paper). However, when DDS content exceeds 40 wt%, formation of microcracks, cracking, and color yellowing are observed (Fig. 1(b)-Top). In contrast to pure

PVT-based samples (0 wt% DDS), scintillators containing 10–30 wt% DDS and 50 wt% PPO remain optically transparent after 15 days of storage (Fig. 1(b)-Bottom), revealing that fluorophore dissolution stability is significantly improved by DDS monomers. This may be understood through molecular isolation within network subdomains constructed by such functional monomers, where spatial separation of fluorophores minimizes intermolecular interactions. Combined with our previous research [21], we conclude that any small amount of functional monomers loaded into the host matrix of a scintillator that participate in cross-linked network formation will always aid dye (solute) dispersion and prevent dye aggregation.

The mechanical hardness of the scintillators (RK1–RK5) was measured to be $45.5\pm0.8 HD, 41.4\pm0.5 HD, 39.7\pm0.3 HD, 39.3\pm0.9 HD, and 37.2\pm0.5 HD, respectively, showing a general decre seeoriginalpaper], the 30 wt^{-1}$ corresponding to Si–O–Si vibration. The absence of both new characteristic peaks and peak shifts suggests that DDS incorporation does not chemically modify the polymer chain structure but rather establishes a physically compatible blended system.

We speculate that during thermal polymerization, low-molecular-weight siloxane chains derived from DDS monomers were incorporated into the PVT-based cross-linked network. This structural integration, combined with the inherent elasticity of DDS siloxane components, likely contributes to the reduced mechanical hardness. Such reduction is not necessarily detrimental from an elastic recovery perspective. To confirm this, a blunt needle with 0.79 mm diameter was used to press the scintillator sample (30 wt% DDS) for one minute. The time evolution of surface morphology for sample RK3 is shown in Fig. 3 [Figure 3: see original paper] and Fig. S2 (Supplementary Materials). The initial indentation exhibited cylindrical geometry with 1.5 mm depth. Remarkably, within 30 seconds post-compression, the indentation underwent near-complete recovery to a planar surface. After 60 seconds, residual deformation or surface extrusion traces were barely observable, confirming that DDS introduction imparts elastic recovery properties to the scintillator. This phenomenon may be attributed to high mobility of siloxane main chains facilitated by exceptionally low energy barriers for Si–O–Si bond bending and torsional motions [28].

Thermal stability was evaluated by thermogravimetric analysis (TGA) under protective nitrogen atmosphere, with temperature increased at 10 °C/min from 30 °C to 800 °C. Results are shown in Fig. 4 [Figure 4: see original paper]. No mass loss occurred below 120 °C. The synthesized scintillators exhibited similar thermogravimetric profiles during the first degradation stage (120–260 °C), where samples underwent inter- or intrachain reactions generating low-molecular-weight products including volatile compounds, linear oligomers, and fluorophores (PPO, MDAC). Compared to the 0 wt% DDS scintillator (decomposition temperature ~160 °C), DDS-containing scintillators began decomposition at lower temperatures, perhaps due to phase-separated domains created between DDS and PVT constituents. Above 210 °C, the thermal degradation rate showed slight dependence on DDS content in a complex manner requiring

future study. In the second stage (260–380 °C), DDS-containing samples exhibited faster weight loss than pure PVT-based scintillators, indicating reduced thermal stability of DDS short-chain structures. During the third stage (380–800 °C), degradation continued, with 2–5% solid residue primarily consisting of SiOC carbonaceous substance from mineralization [29].

## B. Optical Performance

The synthesized scintillators utilize PVT and DDS as matrices. Critically, the emission spectra of both matrices overlap significantly with the absorption spectrum of fluorophore PPO, enabling efficient Förster resonance energy transfer (FRET) from matrices to PPO. To simultaneously excite both matrices, a 270 nm UV source was selected. This wavelength lies well below the 310–330 nm absorption peaks of both PPO and MDAC fluorophores, thereby minimizing direct fluorophore excitation and allowing specific investigation of resonance energy transfer from excited matrices. Figure 5(a) presents the excitation-emission matrix of RK3 scintillator (30 wt% DDS). Under 270 nm excitation, its emission spectrum (scanned from 400–600 nm) shows a distinct 434 nm peak. The excitation spectrum over 250–400 nm was monitored at 434 nm wavelength. The prominent emission peak at 434 nm signifies characteristic fluorescence of MDAC dye, and its presence under 270 nm excitation provides strong evidence for efficient energy transfer from excited matrices to MDAC dye.

Photoluminescence (PL) spectra excited at 270 nm are shown in Fig. 5 Figure 5: see original paper and Fig. S3 (Supplementary Materials). Increasing DDS content promotes π–π stacking of benzene moieties, enhancing exciplex formation and resulting in broadened emission peaks with 10–30 nm red shifts. PL intensity initially rises with DDS concentration but declines above 30 wt% DDS. At high concentrations, fluorescence emission is likely dominated by aggregation-caused quenching (ACQ), where dense π–π stacking creates compact molecular proximity. This facilitates electron energy level rearrangement in the matrix, primarily through splitting of ground-state and first excited-state levels. Consequently, concentration-dependent PL intensity correlates with scintillator optical transparency, as both phenomena originate from photon loss mechanisms in the aggregated system.

Optical transparency of the scintillators is very high (Fig. 5(c)). All samples exhibit transmittance values between 70% and 87%, comparable to commercial scintillator EJ-276 (Fig. S4, Supplementary Materials). Within the visible spectrum, transmittance initially increases with DDS content up to 30 wt%. However, further DDS increases lead to decreased transmittance. This non-monotonic behavior is attributed to two factors: self-absorption within the matrix material and dye molecule aggregation at higher DDS concentrations. Below 30 wt% DDS, monomers were sparsely dispersed; most did not participate in thermal polymerization and acted as photon scattering centers, reducing transparency. As DDS approaches 30 wt%, dispersed monomers polymerized into linear-chain segments, enhancing transparency. However, above ~30 wt%

DDS, high benzene ring density promoted strong $\pi$–$\pi$ stacking interactions that induced fluorophore aggregation [30]. At these concentrations, numerous low-molecular-weight DDS chains embedded within the cross-linked PVT network created phase-separated domains with mismatched plastic deformation behaviors. This structural incompatibility generated interfacial shear stresses and microcracks at phase boundaries (Fig. 1(b)-Top), causing observed transparency reduction.

## C. PSD and Relative Light Output

The synthesized scintillators were irradiated with a $^{238}$Pu-Be source to study PSD performance. The PSD method identifies particles based on different proportions of prompt and delayed components in scintillation pulses. When energetic particles pass through a scintillator, $\pi$ electrons of scintillator molecules are ionized and recombine to produce both singlet and triplet excited states, which promptly de-excite to the lowest singlet ($S_1$) and triplet ($T_1$) states [31]. Transition from $S_1$ to ground state ($S_0$) with fluorescence emission on a timescale of $10^{-9}$–$10^{-8}$ s produces the prompt component. Direct transition from $T_1$ to $S_0$ is spin-forbidden; instead, bimolecular triplet–triplet interaction and annihilation ($T_1 + T_1 \rightarrow S_1 + S_0$), namely the TTA process, occurs kinetically with subsequent $S_1$ to $S_0$ de-excitation ($S_1 \rightarrow S_0 + h$) over a longer timescale ($10^{-4}$ s) producing the delayed component. $T_1$ state density depends on the ionizing power of incident particles. For a given incident energy, heavy charged particles produce more $T_1$ states per unit path length than light particles, resulting in more pronounced slow scintillation components. This difference in the ratio of integrated charges between prompt and delayed components forms the basis of the PSD method.

PSD capability was evaluated using the charge integration method [32]. For each digital waveform, integrations over a short-time gate ($Q$ , fast component) and a long-time gate ($Q$ ) yield the PSD parameter: $PSD = 1 - Q /Q$ , where $Q$ is the charge integral from waveform start to the prompt peak, and $Q$ represents the full-waveform integral. Two-dimensional plots of PSD versus $Q$ are shown in Fig. 6 [Figure 6: see original paper] and Fig. S5 (Supplementary Materials) for prepared scintillators (RK1–RK5 and 0 wt% DDS sample) two days after synthesis, alongside EJ-276. Excellent separation between neutron and gamma-ray signals is observed. The locus with higher PSD values corresponds to neutrons, while lower PSD values correspond to gamma-rays. Notably, neutron and gamma bands exhibit distinct curvature, potentially arising from PMT non-linearity under high-voltage operation (Fig. S6, Supplementary Materials) [33,34]. Separation capability is quantified by the figure of merit (FOM), defined as $FOM = S/(\sigma + \sigma)$, where S represents separation between gamma and neutron distribution peaks, and $\sigma$ ($\sigma$ ) denotes full width at half-maximum (FWHM) of gamma (neutron) peaks in one-dimensional PSD distribution. These parameters are obtained through Gaussian fitting. Consequently, PSD FOM depends on two factors: neutron/gamma peak separation

(S) and their respective widths. Increasing light output reduces FWHM of both distributions, thereby enhancing PSD FOM [27].

To quantitatively assess PSD dependence on DDS concentration, we extracted PSD values within the Q range of 0.2–1.2 MeVee (MeV electron equivalent energy) from 2D distributions. Fig. 7 Figure 7: see original paper plots calculated FOM values against DDS concentration, while Fig. 7(b) shows FOM values calculated within each interval of Q $\pm$0.1 bins (in MeVee) for prepared scintillators (black dots). As clearly demonstrated in Fig. 7(b), each scintillator achieves superior neutron/gamma separation with FOM of 1.27 at Q values down to 0.15 MeVee. This value of 1.27 is generally recognized as the FOM threshold for efficient pulse shape discrimination [11].

Light output of prepared scintillators was studied using a $^{137}$Cs source by comparing Compton edge positions in pulse height spectra with those of EJ-200 standard (Fig. S7, Supplementary Materials). As shown in Fig. 7(c), LO relative to EJ-200 exhibits an overall decreasing trend within calculated uncertainties as DDS monomer concentration increases. Conversely, neutron/gamma PSD capability, quantified by FOM, shows slight concentration-dependent enhancement before reaching a plateau (Fig. 7(a)). This apparent inverse correlation between PSD performance and LO likely results from formation of a more efficient triplet ($T_1$) state energy transfer network arising from gradual PVT substitution with DDS, which increases benzene ring density required for triplet energy migration. Furthermore, enhanced fluorophore dissolution facilitated by DDS incorporation likely affects local $T_1$ excitation density and TTA process efficiency governing the neutron/gamma separation parameter S.

Long-term stability was evaluated through repeat measurements at 96 and 172 days after synthesis (Fig. 7(a) and (c)). After approximately six months of aging, synthesized scintillators exhibited 9–16% reductions in FOM and 27–41% reductions in LO relative to initial values. This degradation highlights a significant challenge. While DDS incorporation offers potential advantages in radiation resistance and PSD performance, observed aging effects—likely stemming from fluorophore aggregation, potential matrix instability at higher DDS loadings (as suggested by Fig. 1(b)), or chemical degradation pathways—currently limit practical long-term utility. Addressing this stability issue, potentially through optimized fluorophore selection, stabilization additives, or refined matrix composition, is crucial for realizing the promising PSD performance of PVT-DDS based scintillators in demanding applications.

## IV. Summary

Organic scintillators based on PS and PVT are widely used in many fields, from nuclear physics experiments to safeguard equipment. However, these materials suffer from poor radiation resistance. Breaking of C–C and C–H bonds generates free radicals and hydrogen release from the polymer, causing color yellowing and reductions in light yield and transparency. One approach to circumvent

this problem is to replace or partially replace these materials with polysiloxane polymers having higher radiation hardness.

In this work, we successfully developed a novel elastic scintillator by incorporating 20–40 wt% siloxane compound DDS into the PVT matrix for neutron/gamma discrimination. The obtained organosilicon-loaded plastic scintillators comprise a mixture of PVT and siloxane-based matrix (DDS). They are transparent, mechanically hard enough for machining and polishing, and capable of excellently discriminating neutrons from background gamma-rays. Performance characterization shows that DDS addition imparts elasticity and improves dye molecule solubility by preventing aggregation. While DDS-PVT based scintillators exhibit excellent PSD performance, they also show decreased light yield (98–86% LO relative to EJ-200) as DDS concentration increases (RK1–RK5), implying that radioluminescence in such scintillators is less favorable than in PVT-based scintillators. Measurements at different times after preparation indicate that stability in terms of PSD and LO still requires improvement. Our future work will focus on investigating the cross-linked network in scintillators synthesized through copolymerization and homopolymerization, aiming to understand singlet and triplet exciton migration required for efficient PSD capability in organosilicon-loaded scintillators. Nevertheless, this newly designed organosilicon-loaded plastic scintillator is promising for fabrication of next-generation neutron detectors used under high-dose irradiation.

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## Author Contributions

All authors contributed to this study. Weirong Liu, Yingdu Liu, and Jie Bao designed and prepared the scintillators. Weirong Liu, Zhiyun Zhu, and Siyu Zhang performed experimental measurements. Weirong Liu and Pengcheng Shi completed data analysis. Weirong Liu and Pengcheng Shi wrote the first draft, and all authors commented on previous manuscript versions. All authors have read and approved the final manuscript.

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