Supplementary Materials

Polyurethane coatings reinforced with poly(MMA-co-HPMA)@$Mo_2Ti_2AlC_3$ as flexible EMI shielding solutions

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Characterization methods

1.1.1 ***FT-IR analyses*** were performed using a Spectrum Two FTIR spectrometer (PerkinElmer, Waltham, Massachusetts, United States) supplied with a MIRacleTM Single Reflection ATR (PIKE Technologies), at 4 cm^{-1} resolution, totaling 32 scans in the 4000–550 cm^{-1} wavenumber domain.

1.1.2 ***SEM morphology characterization*** of the polymer particles and of the polyurethane films were performed at 10 kV using a field emission gun scanning electron microscope (FEGSEM), Nova NanoSEM 630 (FEI) (Hillsboro, OR, USA).

1.1.3 **The Raman spectra** of the samples were collected by a Raman Spectroscopy Renishaw inVia Qontor Spectrometer, detector Centrus 4VHX97. Spectral imaging was recorded using a confocal Raman spectral imaging system with 633 nm NIR excitation and a 1200 nm laser edge. The incident laser power under the microscope objective (100X) was in the range of 5–10%, while the grating scan was at 1200 l/mm and extended mode (100-3200 cm^{-1}).

1.1.4 ***XRD analyses*** were carried out using a 9 kW Rigaku Smart Lab diffractometer, equipped with a Cu Kα1 source that provides a monochromatic beam with wavelength, $\lambda = 0.1546$ nm. The measurements were recorded in 2θ mode, while the incidence angle was fixed at 0.5° (grazing-incidence mode), using a speed of 5° min^{-1}.

1.1.5 ***Dynamic mechanical analysis (DMA)*** was conducted using a single cantilever mode to evaluate the viscoelastic properties of the polyurethane (PUR) films. The experiments were performed using a single cantilever clamp, with sample dimensions measuring 36 mm × 10 mm × 0.5–0.7 mm, at a constant frequency of 1 Hz. The temperature ramp ranged from −90 °C to +50 °C, with a controlled heating rate of 5 °C/min, facilitated by liquid nitrogen cooling. All

measurements were carried out using a Discovery DMA 850 apparatus (TA Instruments, New Castle, DE, USA), ensuring high precision and reliability in thermal and mechanical assessments.

1.1.6 ***Frequency-dependent shear measurements*** of the composite polyurethane (PUR) films were performed using the same Discovery DMA 850 instrument but equipped with a shear-sandwich clamp. The analysis was conducted in oscillatory mode, over a frequency range of 0.1 Hz–100 Hz at a constant strain of 10% and a temperature of 25 °C. The experimental method followed the ASTM D 945-92 Standard Test Methods for Rubber Properties in Compression or Shear, with appropriate adaptations for the composite materials under investigation.

1.1.7 ***Tensile tests*** were conducted using a Titan 10 Universal Strength Tester from James H. Heal & Co. Ltd, in accordance with the international standard ISO 37:2005. The testing procedure included the use of standard dumbbell tensile specimens, characterized by a narrow parallel section width of 5 mm, a total length of 100 mm, and a gauge mark distance of 20 mm, a load cell of 5000N. The rate of extension was set at 500 mm/min, with a jaw separation (using plain jaw faces) of 50 mm. For each type of polyurea tested, five tensile tests were carried out, and the average value was subsequently reported.

1.1.8 ***Evaluation of EMI shielding characteristics*** **-** To assess the electromagnetic interference (EMI) shielding characteristics at microwaves of the fabricated PUR films, we have employed a waveguide-based experimental setup (as previously described in our work[11]) to analyze the reflection and transmission properties between 8.2 and 12.4 GHz, i.e., the so-called X band. The latter is one of the most important frequency ranges, since most of the radars work in this band, which is also allocated for terrestrial and space communications, traffic light motion sensors, and the RF sources of particle accelerators. A common way to evaluate the shielding

properties of a material is to calculate (either from simulations or from measurements, where possible) the total EM shielding effectiveness (SE), which is given by the following formula[12]:

$$SE(dB) = SE_R(dB) + SE_A(dB) \quad (1)$$

in which SE_R is the shielding effectiveness related to EM reflections (R) and SE_A is the shielding effectiveness related to absorption (A) phenomena. SE_R and SE_A can be calculated using the scattering (S) parameters for a certain band of interest, namely the reflection (S_{11} or S_{22}) and transmission (S_{21} or S_{12}) parameters of a two-port setup, as follows [12]:

$$SE(dB) = SE_R(dB) + SE_A(dB) = 10log\left[\left(1 - |S_{11}|^2\right)^{-1}\right] + 10\log\left[\left(1 - |S_{11}|^2\right)/|S_{21}|^2\right] \quad (2)$$

From Eq. (1) and Eq. (2) we can identify two fundamental physical mechanisms to modulate *SE*: (i) EM reflections (increased in the case of metals, which exhibit a high conductivity and a very low absorption); (ii) absorption due to dielectric losses and low conductivity. *SE* cannot be reconfigured in the case of metals, while *SE* can be engineered in the case of composites (like polymer-based ones) by varying the concentration of their constituents and/or dopants.

Moreover, it is also possible to extract the high-frequency conductivity σ and skin depth δ of the materials under test (MUTs) by using the following relations[13]:

$$SE_R = 35.9 + 10log\left[\sigma/(2\pi f \mu_0 \mu_r)\right] \quad (3)$$

$$\delta = 1/\sqrt{\pi f \mu_0 \mu_r \sigma} \quad (4)$$

where f is the frequency, μ_0 is the vacuum permeability (equal to $4\pi\times10^{-7}$ N·A^{-2}), and μ_r is the relative permeability of the MUT. For non-magnetic materials (like in our case of study), $\mu_r = 1$.

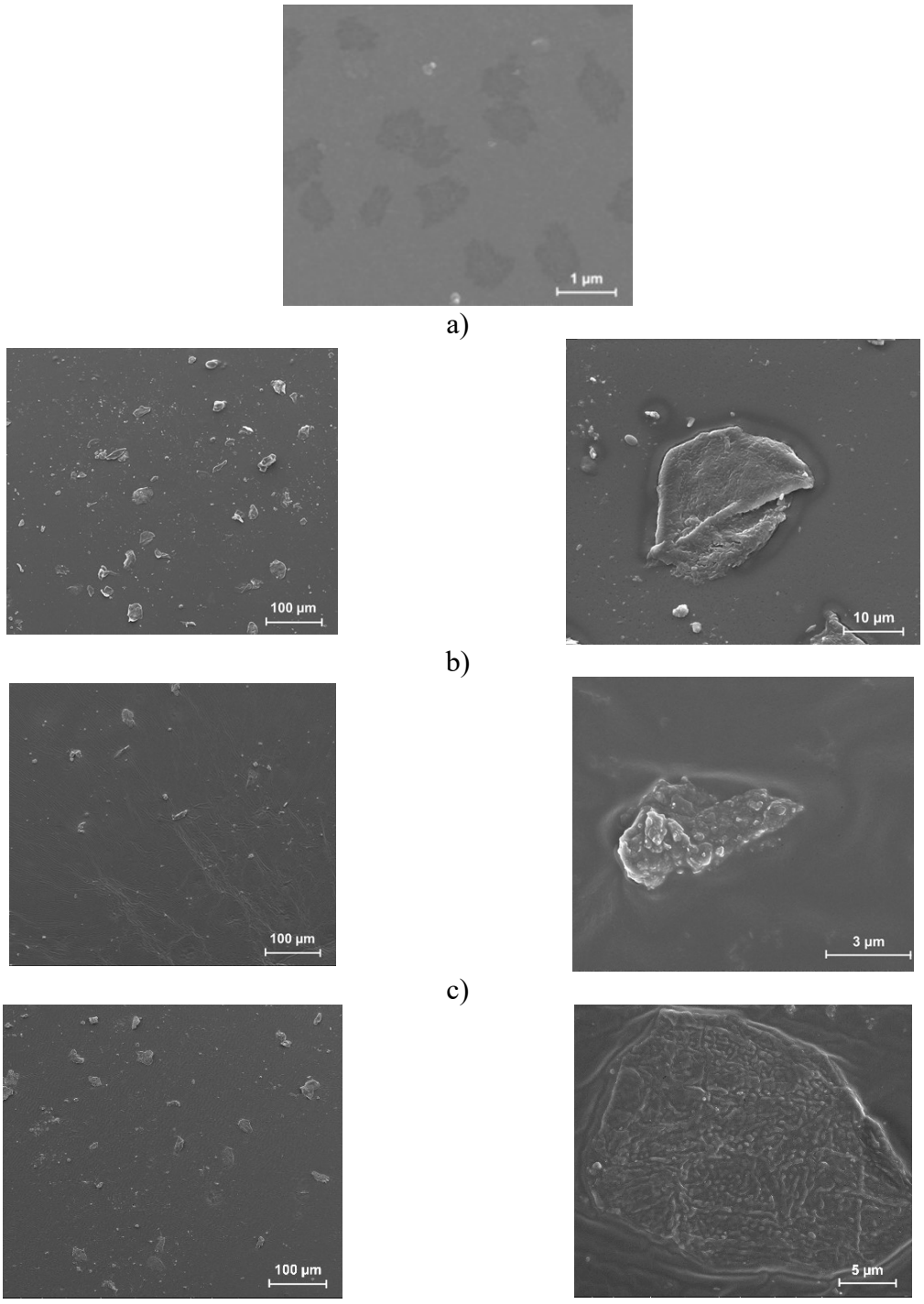

Figure S1. SEM images for the polyurethane coatings containing different concentrations (wt.%) of polymer particles containing MXenes:

a) PUR blank (0%); b) PUR 0.5 % MXene; c) PUR 1%-MXene; d) PUR 2 % MXene

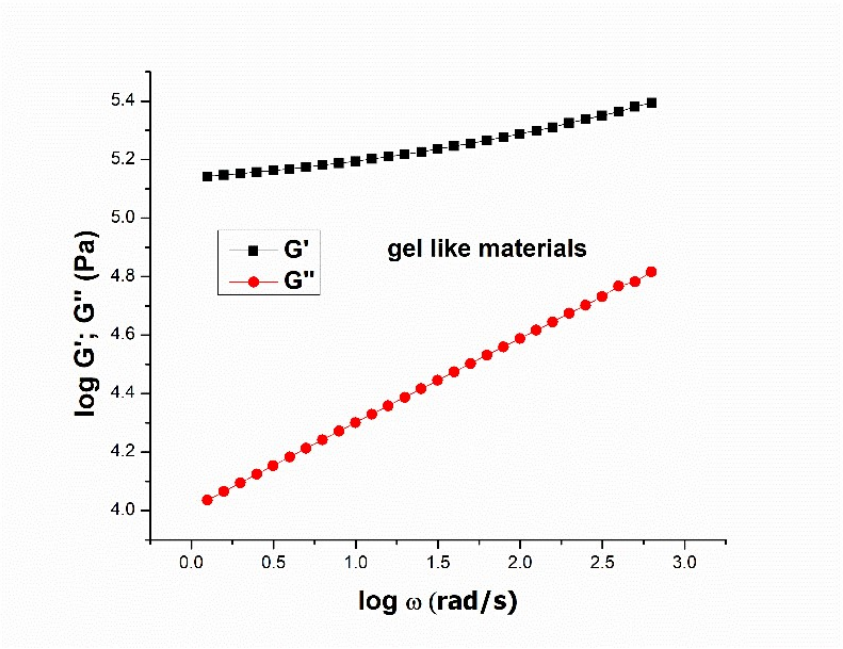

Figure S2: The dependence of log G', G'' on the log w for PUR

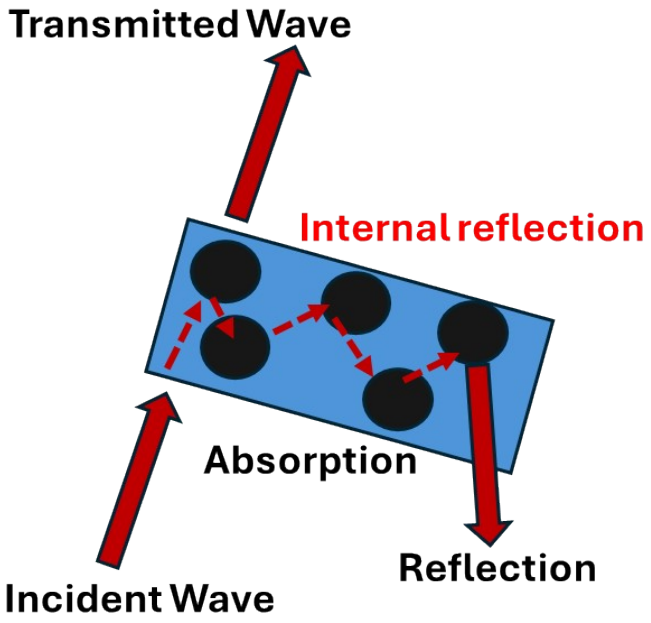

Figure S3: The mechanism of EMI shielding