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A hydrophobic and abrasion-resistant MgF_2 coating with ultralow refractive index for double-layer broadband AR coating

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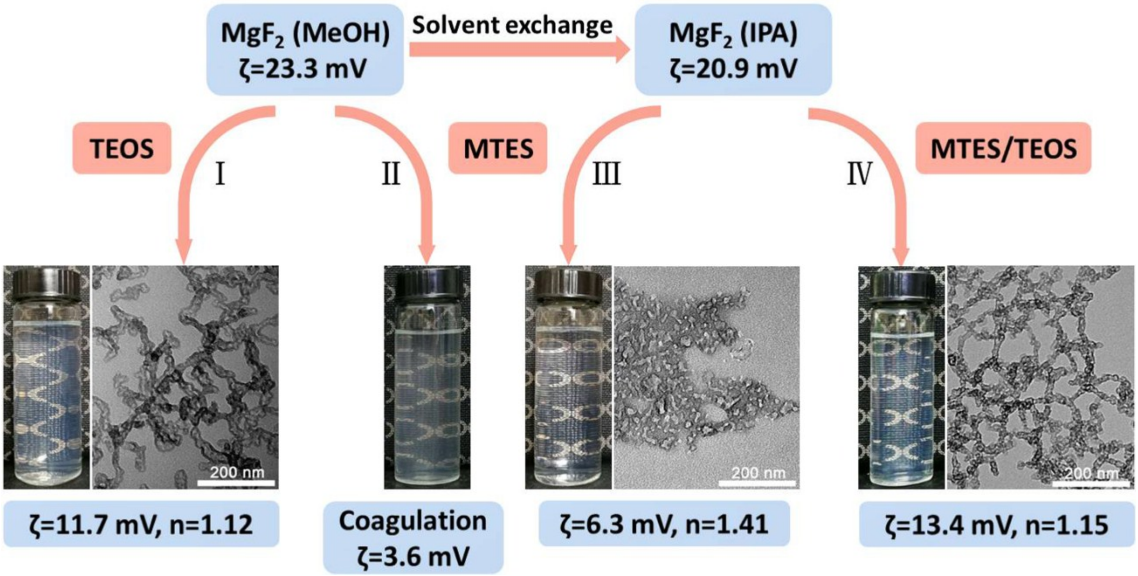

Fig. S1 Experimental design process to prepare the hydrophobic ultralow-index AR coating.

Route I shows that introducing TEOS to MgF_2-MeOH sol can assemble MgF_2 particles to form network structure with ultralow refractive index of 1.12. The zeta potential was measured as 11.7 mV.[1]

Route II shows that adding MTES to MgF_2-MeOH sol would quickly lead to the sol coagulation. What is causing the distinct result between MTES and TEOS? According to the theory of inductive effect analyzed in the book: sol gel science,[2] under acidic conditions, the hydrolysis rate increases with the degree of substitution of electron-providing alkyl groups because the electron-providing alkyl groups decreases the positive charge on the silicon atom and thus increases the negative charge on the oxygen, leading to the faster protonation of oxygen atoms.[3-5] So the rapid hydrolysis rate of MTES may be responsible for the coagulation result due to the strong interaction between locally enriched negative silanol groups and positively charged MgF_2 particles, which can be proved by the quickly decreased zeta potential from 23.3 to 3.6 mV.

To solve this coagulation problem, we proposed to exchange the dispersion medium of MgF_2 sol from MeOH to IPA, which larger steric hindrance may decrease the hydrolysis rate of MTES. But **route III** indicates that the MgF_2-MTES sol was transparent and stable, but cannot form the advantageous network structure, while form the dense structure with measured refractive index of nearly 1.41. The zeta potential also dramatically decreases from 20.9 to 6.3 mV. Thus this simple experimental route was infeasible to prepare the targeted hydrophobic ultralow-index AR coating due to the effect of the methyl group.

According to the relevant experimental result that the co-precursors of TEOS and MTES have lower hydrolysis-condensation rate than MTES and a copolymer with the methyl functionality randomly distributed can be obtained.[6] We attempted to introduce the MTES/TEOS co-precursors to MgF_2-IPA sol. **Route IV** indicates that this procedure can obtain the expected network structure. So in this paper, we conducted a series of experiments to study the mechanism of the structure-direction of MTES/TEOS co-precursors to MgF_2 particles and the hydrophobic and optical properties of fabricated MgF_2-$SiO_2(CH_3)$ coating.

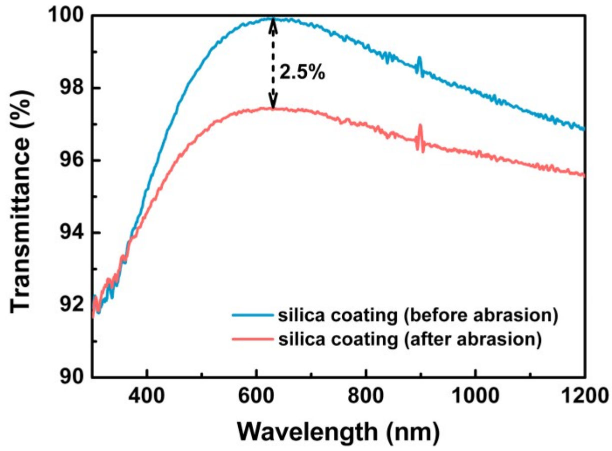

Fig. S2 Transmittance spectra of particle-like silica coating calcined at 250 °C before and after rubbing test.

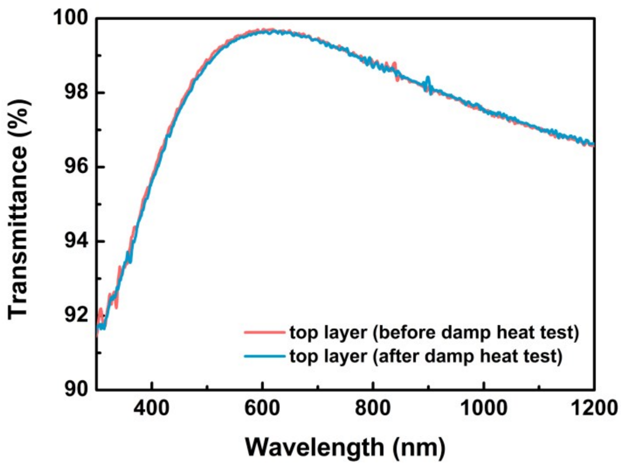

Fig. S3 Transmittance spectra of top layer before and after damp heat test.

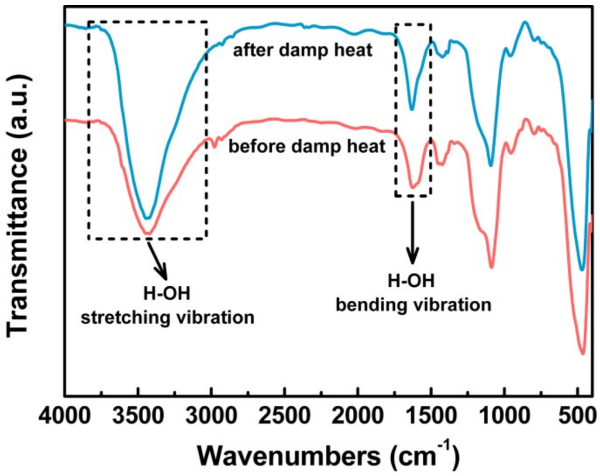

Fig. S4 IR spectra of MgF_2-SiO_2 xerogel before and after damp heat test.

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