Supplementary Information

Experimental

Synthesis of HfO_2 and ZrO_2 based Nanoparticles

The HfO_2 and ZrO_2 based nanoparticles were synthesized by the modified sol-gel method as reported previously.[1] A fixed amount of hafnium isopropoxide (Aldrich, 99.9%) (or zirconium isopropoxide for the synthesis of ZrO_2nanoparticles) was firstly dissolved into30 mL methacrylic acid (MAA, Sigma Aldrich, 99%) and was stirred at 65 ºC for 5-10 minutes to obtain a clear solution. A solution composed of 9 mL methacrylic acid and 1 mL water was added dropwise into the precursor solution. The mixture was then aged at 65 ºC for 18 hours. The white precipitate was centrifuged and collected after washing with water and acetone multiple times. The final product HfO_2/MAA (denoted as HfMAA) was obtained after overnight drying in the vacuum oven at 60ºC and stored for further use. HfO_2/trans-2,3-dimethylacrylate (denoted as HftDMA), HfO_2/2-methylbutric acid (denoted as Hf2MBA), HfO_2/isobutyric acid (denoted as HfIBA) were synthesized with the same procedure with the corresponding acids replacing methacrylic acid. ZrO_2 based nanoparticles were synthesized using a similar procedure.

Solubility Tests

25 mg of the appropriate nanoparticle system and a fixed amount of photoacid generator (PAG) N-hydroxynaphthalimidetriflate(1 and 5 wt%, respectively) were dissolved into propylene glycol monomethyl ether acetate (PGMEA, Sigma-Aldrich, 99.5%) with a total mass up to 250 mg. The solution was filtered with a 0.2 μm syringe filter to remove large particles and was then spin-coated on pre-cleaned silicon wafers (WRS materials) at 2000 rpm for 60 seconds. The thickness of the deposited photoresist film was about 100 nm. The film was heated at 110 ºC for 60s to remove excess solvent and then exposed to 254 nm wavelength deep UV light with a dosage of 150 mJ/cm^2 through a quartz mask. An ABM mask aligner was used for photo-patterning. The silicon wafers before and after exposure were cut into 5 mm×5 mm pieces and were immersed into 5 mL of different solvents for solubility testing.

Materials Characterization

Infrared spectra were collected by attenuated total reflectance with a Nicolet iZ10 ATR-FTIR spectrometer. 64 scans with 4 cm^{-1} resolution were used for analysis. Thermogravimetric analysis and differential thermal analysis (TG/DTA) were performed with a Seiko Instruments TD/DTA 6200. The thermal analysis was conducted under N_2 atmosphere with a heating rate of 10 ºC/min from room temperature to 580ºC. Particle size and particle size distribution were obtained with the Malvern Zetasizer. Scanning electron microscopy images were collected on Zeiss LEO 1550 FE-SEM with an accelerating voltage of 1 kV.

Table S1. Weight percentage of different hybrid nanoparticles after heating from room temperature to 580 ºC with a heating rate of 10ºC/min.

Materials	**HfO_2**				**ZrO_2**			
	MAA	tDMA	2MBA	IBA	MAA	tDMA	2MBA	IBA
Weight (%)	59	64	51	64	62	54	47	49

Table S2. The Hansen solubility parameters and dielectric constant (ε) of solvents used for the solubility test and the corresponding parameters for the hybrid nanoparticles.

Solvent	δ_d (MPa$^{1/2}$)	δ_p (MPa$^{1/2}$)	δ_h (MPa$^{1/2}$)	ε
Water	15.6	16	42.3	80.1
Methanol	15.1	12.3	22.3	32.4
Acetonitrile	15.3	18	6.1	37.5
Isopropyl alcohol	15.8	6.1	16.4	19.92
Dimethylformamide	17.4	13.7	11.3	37.1
Acetone	15.5	10.4	7	20.7
Methyl ethyl ketone	16	9	5.1	18.5
Ethyl acetate	15.8	5.3	7.2	6.02
4-methyl-2-pentanol	15.4	3.3	12.3	10.4
PGMEA	16.1	6.1	6.6	8.3
Methyl isobutyl ketone	15.3	6.1	4.1	13.11
n-butyl acetate	15.8	3.7	6.3	5.01
Methylenechloride	18.2	6.3	6.1	9.1
Chloroform	17.8	3.1	5.7	4.8
Chlorobenzene	19	4.3	2	5.62
Toluene	18	1.4	2	2.24
Decalin	18.4	0	0	2.2
Hybrid Nanoparticles				
ZrMAA	16.6	8.6	12.2	-
HftDMA	16.7	8.5	11.1	-
ZrtDMA	16.7	8.5	11.1	-
HfIBA	18.4	8.5	4.6	-
ZrIBA	20.0	7.7	10.3	-
Hf2MBA	21.1	7.6	9.2	-
Zr2MBA	21.1	7.6	9.2	-

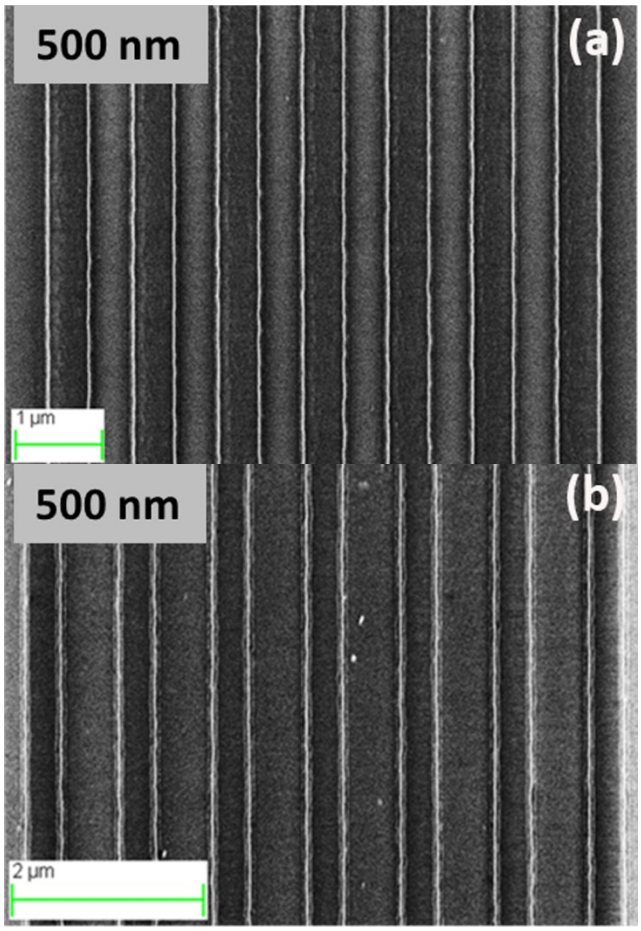

Figure S1.SEM images of photo-patterns by 254 nm deep UV lithography from (a) ZrMMA and (b) HfMMA with addition of 1 wt% PAG in 4-methyl-2-pentanol developer.

Reference

[1] L. Li, S. Chakrabarty, K. Spyrou, C. K. Ober, E. P. Giannelis, Chem. Mater. 2015, 27, 5027.