## Experimental details

### *Synthesis of $HfO_2$ nanoparticles*

Hafnium isopropoxide (6.0 g, 12.6 mmoles) was dissolved in 30.0 ml acetic acid and stirred at 65 °C for 15 min to form a clear solution. Then 10.0 ml of a solution comprised of 9.0 ml acetic acid and 1.0 ml $H_2O$ (1.0 g, 55.5 mmoles) was added causing the formation of a white precipitate. The reaction mixture was stirred at 65 °C for 18 h before 10 ml of a similar acetic acid – water solution was added followed by stirring for 3 h at the same temperature. The white precipitate was centrifuged at 8,000 *g* for 10 min and then washed once with acetic acid and twice with acetone. The final product was dried overnight to give a fine white powder.

### *Synthesis of (s)-(+)-tetrahydrofurfuryl -O-O'-diacetyl-(2R,3R)-hydrogen tartrate (TDHT)*

To a stirred mixture of racemic tetrahydrofurfuryl alcohol (4.7g) and pyridine (0.4 g) solid diacetyltartaric anhydride (10 g) was added. Stirring was continued for 30 minutes at room temperature, then 5 ml of ethyl acetate was added, and the temperature was raised to 80 °C. After 2 hr heating, 10 ml of ethyl acetate was added, and the mixture was allowed to cool to room temperature, then kept for 10 hrs at 5 °C. The crystals were filtered off, washed with ethyl acetate (10 ml) and dried. 7 g of crude ester was obtained. The crude product was recrystallized three times from ethyl acetate to yield 1.8 g of pure TDHT. IR (KBr, $cm^{-1}$): ν(C-O):1069, 1212; ν(C=O): 1750, 1759; ν($CH_3$): 2605, 2944; ν(OH): 3495. $^1H$ NMR (Acetone-$d_6$): δ 5.74-5.68 (2H, m), 3.62-4.26 (5H, m), 2.09-2.17 (6H, s), 1.18-2.08 (4H, m) ppm.

### *Ligand exchange reaction and purification*

$HfO_2$-acetate x (x=0.04~0.12g) was mixed with ligand 0.2 g + 1 ml of PGMEA in 25 ml round flask. After ultra-sonication of the solution for 5 minutes, the cloudy solution was heated at 130 °C for 10 minutes. After ligand exchange, the solution became clear. To eliminate any residue of acetate or acetic acid, an excess of acetone/water/ether co-solvent was added to the solution. The solution was centrifuged at 10,000 rpm for 10 min, and then washed three times with a mixture of acetone/water/ether. The residual powder was redissolved in water/acetone (50/50) mixture. The solution was purified by dialysis using semipermeable membrane (molecular cutt off, 500) for 3 days and vacuum dried at 50 °C.

### *Film preparation*

193 nm photoresist powder poly(methyladamantane methacrylate-*co*-alpha-methacryloxy-gamma-butyrolactone) (PMAdMA-*co*-GBLMA) (Mitsubishi Rayon America, Inc.) 50 mg and photoacid generator (PAG) triphenylsulfonium triflate 2.5 mg were dissolved in 1 ml of PGMEA. A desired amount of $HfO_2$-TDHT is added to the photoresist solution. After sonication and heat treatment, the solution became transparent. After being filtered through 0.2 μm syringe filter, the solution was spin-coated onto 2 inch BARC-coated or HMDS primed Si wafers at 2000 rpm for 60 s. The film was baked at 130 °C for 60 s after spin coating to remove residual solvent in the film. The BARC-coated Si wafer (~90 nm thickness) was supplied by SEMATECH (ARC29A-8) and was baked at 120 $^{o}$C for 60 s for the dehydration and was cooled to room temperature prior to resist coating. The film thickness and the roughness of each film are listed in table S1.

| $HfO_2$-TDHT content in PMAdMA-*co*-GBLMA | Film thickness (nm) | Roughness (nm) |
|---|---|---|
| 0% | 130±16 | 0.287±0.005 |
| 4% | 180±10 | 0.276±0.004 |
| 8% | 210±7 | 0.539±0.013 |
| 12% | 220±8 | 0.485±0.022 |

**Table S1.** Film thickness and roughness of PMAdMa-*co*-GBLAMA + $HfO_2$-TDHT film after spin-coated at 2000 rpm for 60 s on BARC coated film.

### *Lithographic Evaluation*

Deep-UV patterning was done by exposing films through a quartz mask using an ABM mask aligner at a wavelength of 254 nm (measured power 8.7 mW/cm$^2$). High-resolution patterning was done using a Leica VB6-HR electron-beam lithography system operating at 100 kV, followed by a post-exposure bake of 115 °C for 60 sec.

**Supporting figures**

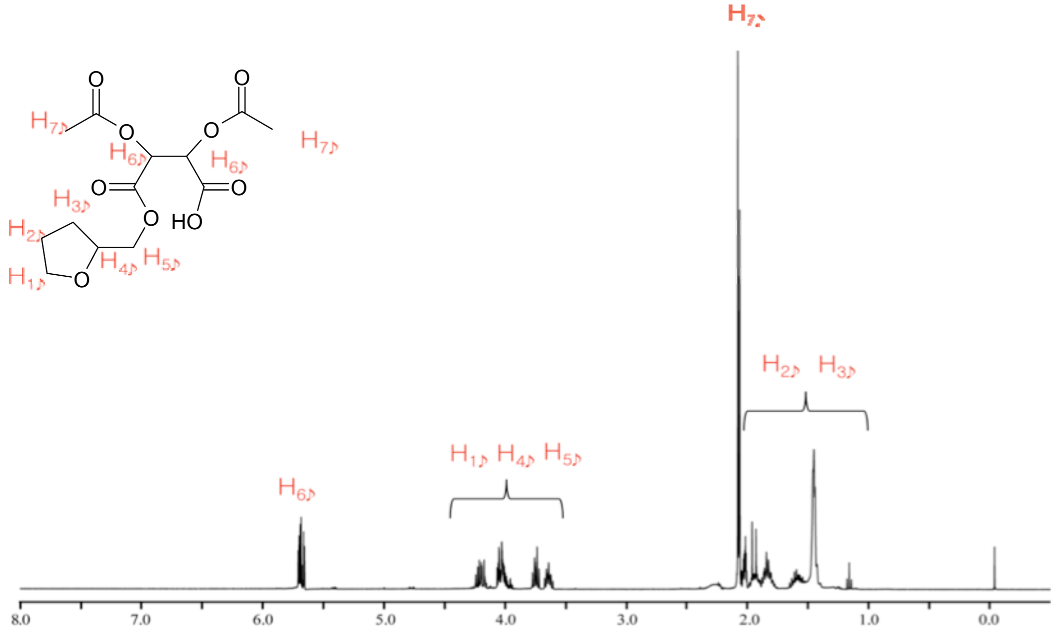

**Figure S1.** $^1$H NMR spectrum of TDHT

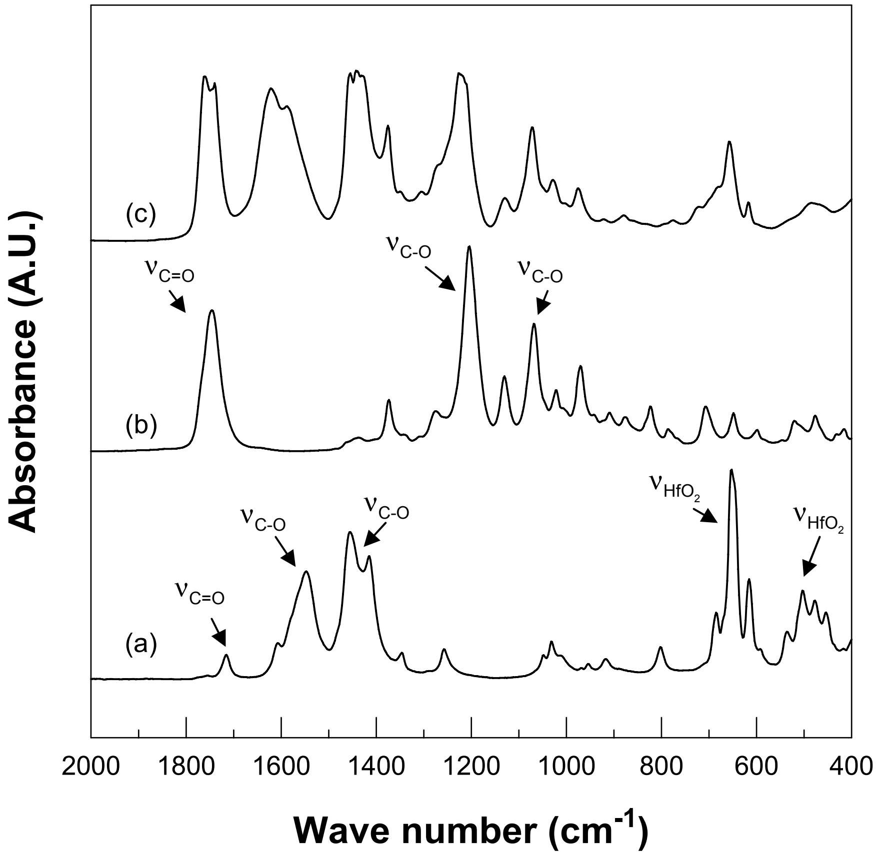

**Figure S2.** FTIR spectra of (a) $HfO_2$-aceatate, (b) TDHT, (c) $HfO_2$-TDHT

**Figure S3.** Optical micrograph of P(MAdMA-*co*-GBLMA)/ HfO$_2$–TDHT nanoparticles (8 wt%).

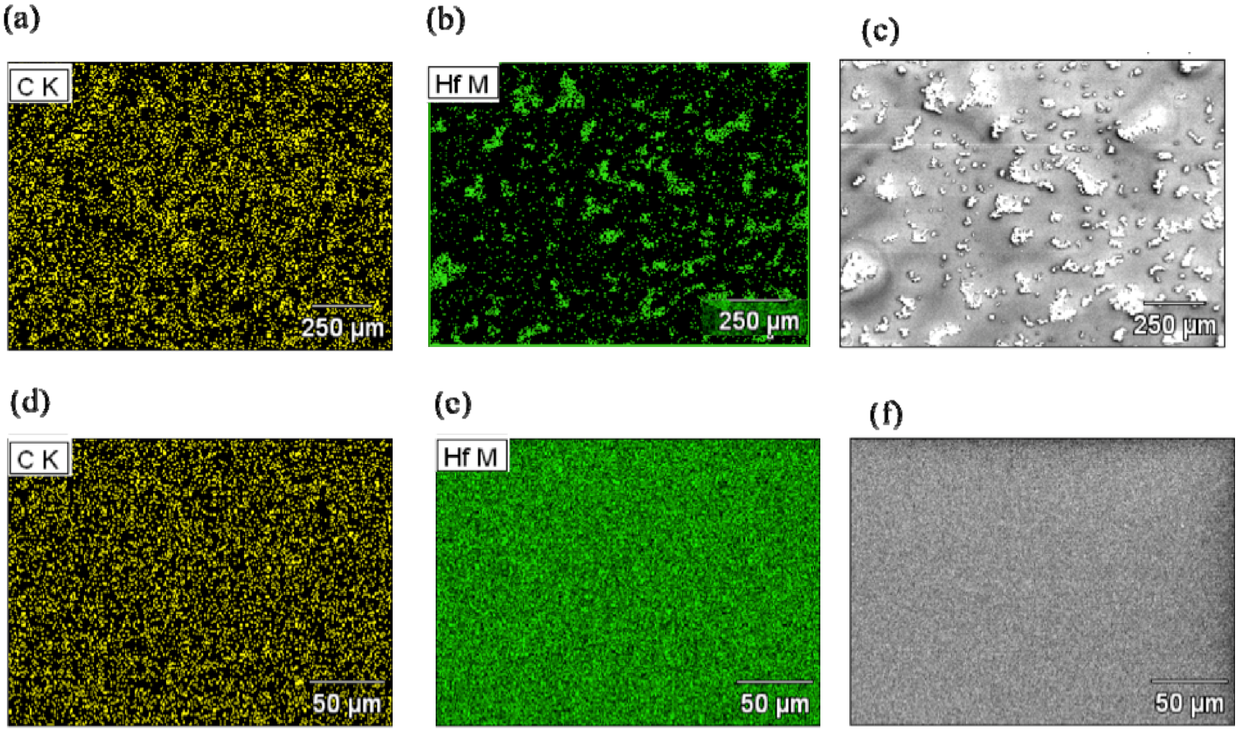

**Figure S4.** SEM images of (c) P(MAdMA-*co*-GBLMA)/ HfO$_2$–acetate (8wt%) and (f) P(MAdMA-*co*-GBLMA)/ HfO$_2$–TDHT (8wt%) and corresponding elemental maps (a), (d) C at K peak and (b), (e) Hf at M peak.

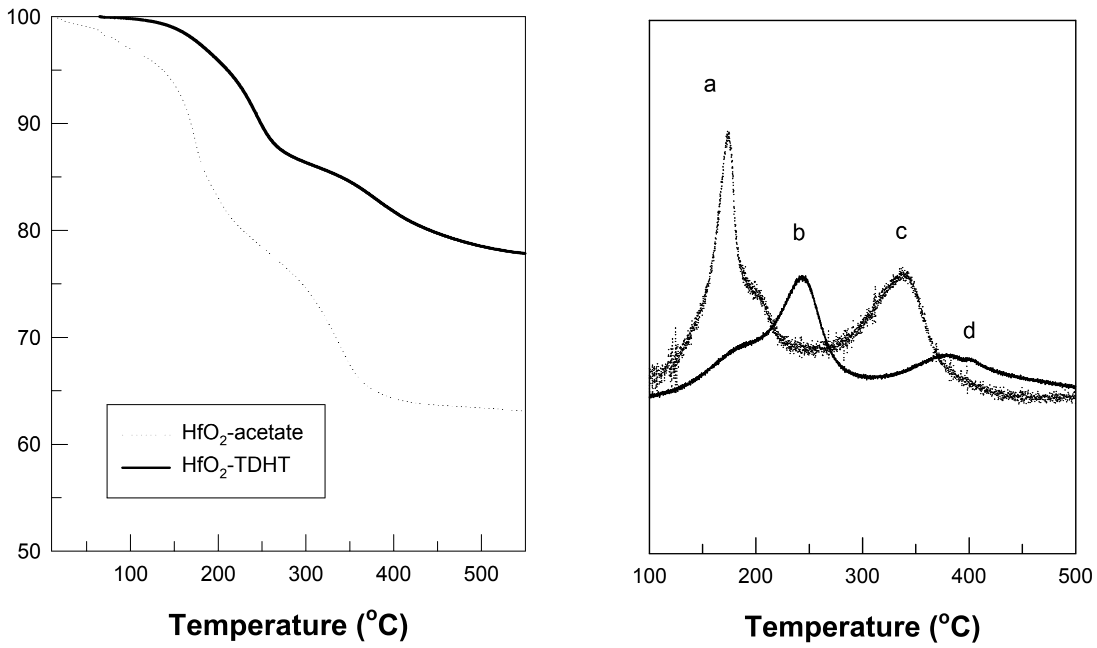

**Figure S5.** TGA thermogram for HfO$_2$-acetate and HfO$_2$-TDHT (left) and 1st derivative curve of the left graph (right). The peak a is attributed to the disconnection of crystal water and the shoulder next to the peak a is related with the reaction: $CH_3COO \rightarrow CO_2$ + solid residue.[S1] The peak b is related with the dissociation of TDHT group. The Peak c and d are attributed to the decomposition of organic residue.

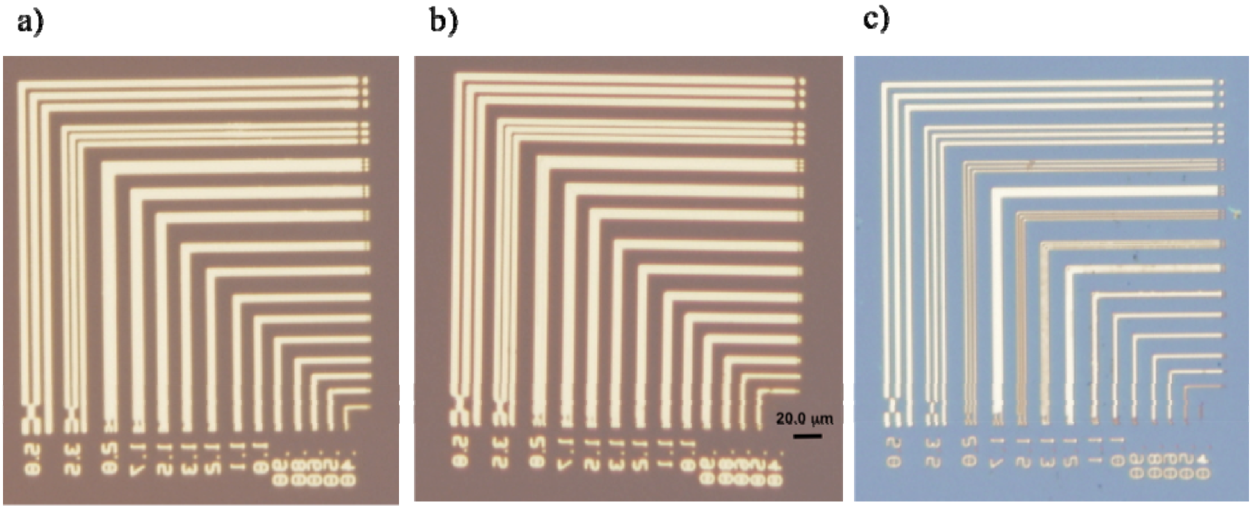

**Figure S6.** Optical microscopy images of (a) P(MAdMA-*co*-GBLMA), (b) P(MAdMA-*co*-GBLMA)/ $HfO_2$–TDHT nanoparticles (8 wt%) and (c) P(MAdMA-*co*-GBLMA)/$HfO_2$-TDHT nanoparticles (12 wt%), patterned by DUV contact aligner.

## References

S1 A. Bakalova, *J. Therm. Anal. Cal.* 2009, **2**, 593.