## A Novel Isocyanate-free Strategy towards Preparation of Polyurea by

## **Ring-opening Reaction**

Xuejie Wang<sup>a,b,c</sup>, Chongwen Yu<sup>b,c</sup>, Jiaqi Sun<sup>b,c</sup>, Chenzhong Mu<sup>d,\*</sup>, Yujie Song<sup>b,c,\*</sup>.

<sup>a</sup> School of Material Science and Chemical Engineering, Ningbo University, Ningbo 315211, China;

<sup>b</sup> Engineering Laboratory of Advanced Energy Materials, Ningbo Institute of Materials Technology

& Engineering, Chinese Academy of Sciences, Ningbo 315201, China.

E-mail: songyujie@nimte.ac.cn

° Qianwan Institute of CNiTECH, Zhongchuangyi Road, Hangzhou Bay district, Ningbo, Zhejiang,

315336, China;

<sup>d</sup> State Key Laboratory of Special Functional Waterproof Materials, Beijing Oriental Yuhong Waterproof Technology Co., Ltd., Beijing 100123, China.

E-Mail: <u>mucz@yuhong.com.cn</u>



SI-Fig. 1 The picture of the sample for tensile test.



**SI-Fig. 2** FT-IR spectra of the various curing agents (X-1) and their corresponding polyurea products: (A-2) 1,3-D based PUA, (B-2) 1,5-D based PUA, (C-2)1,8-D based PUA, (D-2) 1,3-C based PUA, (E-2) 4,4-D based PUA, (F-2) *m*-X based PUA and (G-2) 4,4-Dpt based PUA.



SI-Fig. 3 TGA curves of PUA films, (A) 1,3-D based PUA; (B) 4,4-D based PUA and (C) 4,4-Dpt based PUA.



SI-Fig. 4 DMA diagram of (A) 1,5-D based PUA; (B) 1,8-D based PUA; (C) 4,4-D based PUA.



**SI-Fig. 5** DSC diagram of four representative PUAs, (E) Glass transition temperatures of soft segments (Tgs) determined by DSC and DMA, (F) Glass transition temperatures of hard segments (Tgh) determined by DSC and DMA.

Two Tgs for all the samples also appear in DSC images, which correspond to DMA. However, the values of DSC test are generally lower than those of DMA, which is because the test principles are different. DMA is a mechanical test and the accuracy is not as high as DSC.



**SI-Fig. 6** Stress-strain curves of PUA films, (A) PUAs containing *n*-alkanes; (B) PUAs containing cycloalkanes; (C) PUAs containing benzene rings; (D) PUAs containing siloxanes.