

Supplementary Information

1

2

3 **Co-gel strategy for preparing hierarchically porous silica/polyimide** 4 **nanocomposite aerogel with thermal insulation and flame retardancy**

5 Xinhai Zhang,^a Xingxing Ni,^a Chenxi Li,^a Bo You,^{*a} and Gang Sun^b

6

7 ^a Department of Materials Science, Advanced Coatings Research Center of Ministry
8 of Education of China, Fudan University, Shanghai 200433, People's Republic of
9 China.

10 ^b Department of Aeronautics and Astronautics, Fudan University, Shanghai 200433,
11 People's Republic of China.

12 □ Corresponding author: Bo You

13 □ Corresponding Author E-mail: youbu@fudan.edu.cn

14

15

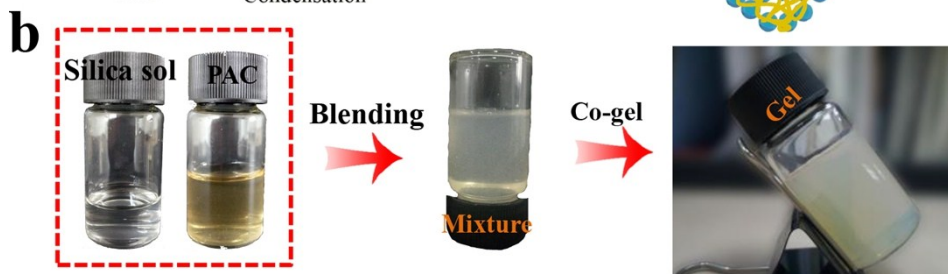
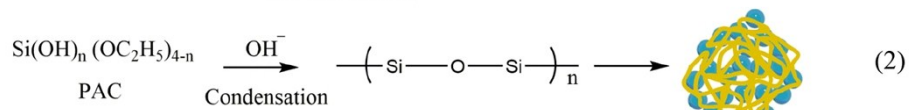
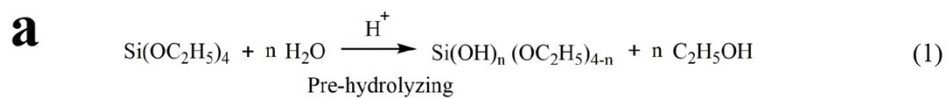
16

17

18

19

20



21

22 **Figure S1.** (a) Hydrolysis process for tetraethyl orthosilicate (TEOS, $n \geq 2$), (b) Digital images of

23 the *in situ* generated silica/PAC co-gel.

24

25

26

27

28

29

30

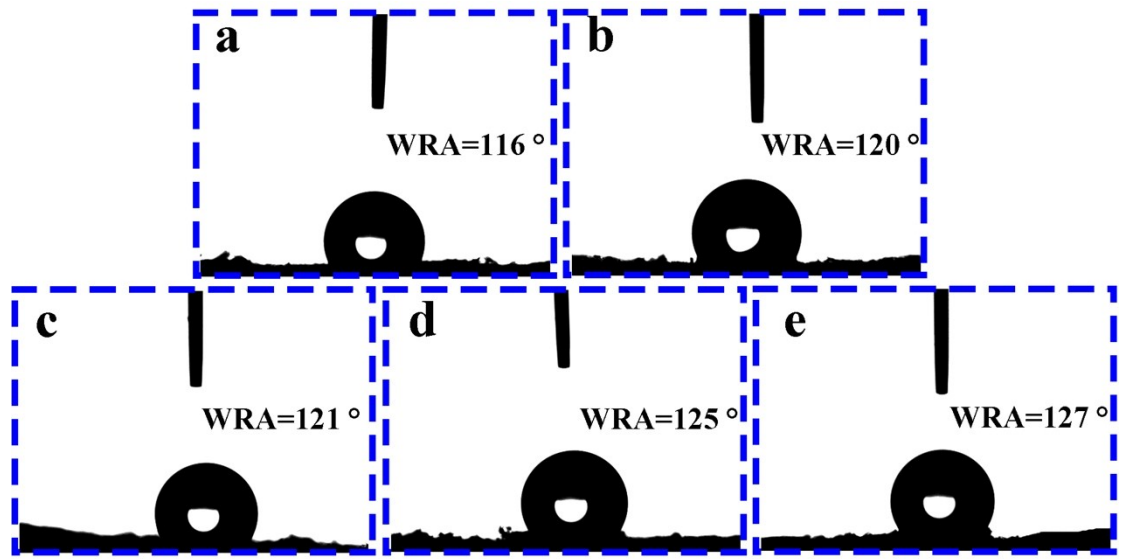
31

32

33

34

35



36

37 **Figure S2.** Water contact angle (WRA) of PI and SiO₂/PI-n aerogels. (a) PI aerogel. (b) SiO₂/PI-1

38 aerogel. (c) SiO₂/PI-2 aerogel. (d) SiO₂/PI-3 aerogel. (e) SiO₂/PI-4 aerogel.

39

40

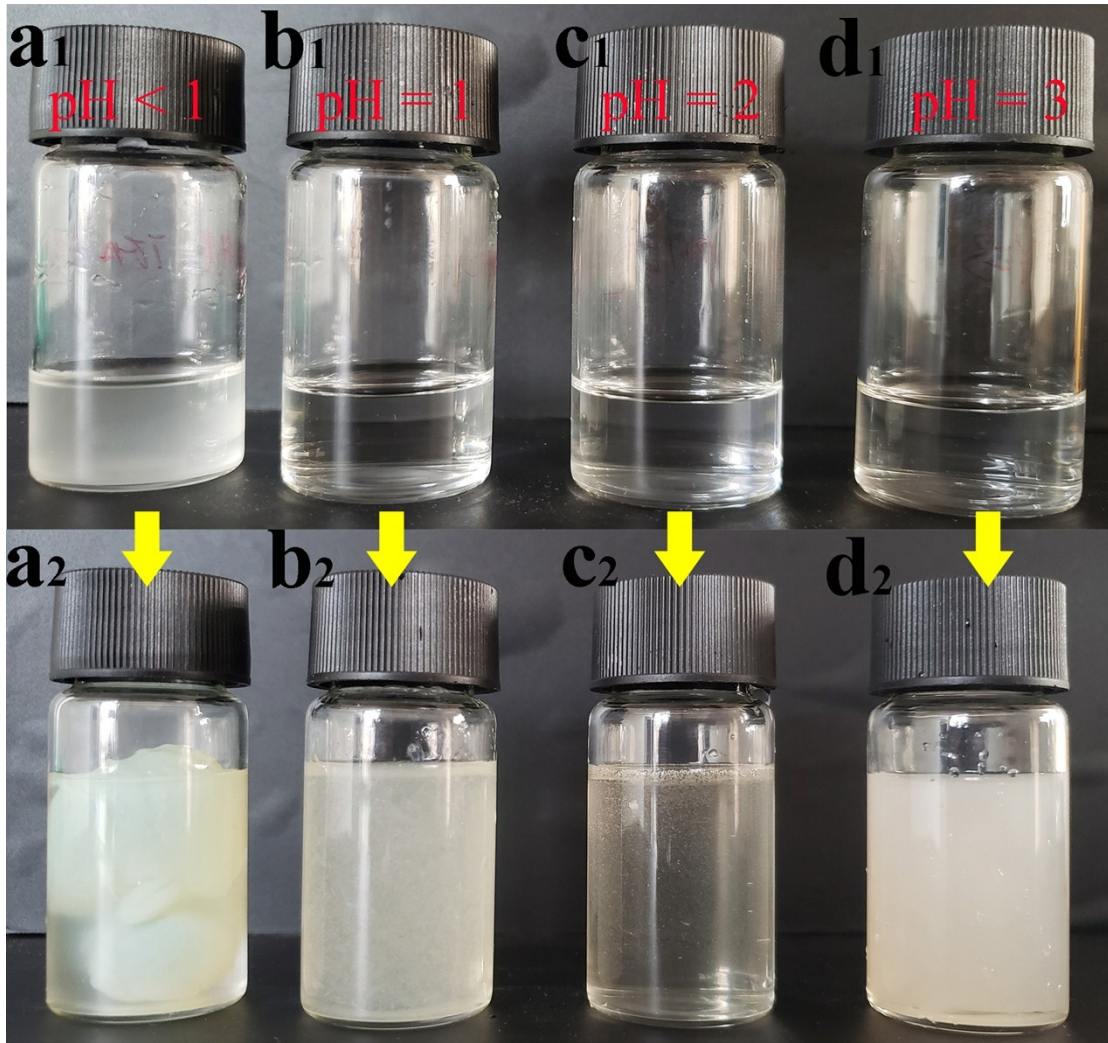
41

42

43

44

45



46

47 **Figure S3.** Digital images hydrolysate of TEOS under different pH value conditions and Digital

48 images of the mixture of PAC and hydrolysate of TEOS correspondently. (a₁ and a₂) Hydrolysis

49 pH value of TEOS is lower than 1. (b₁ and b₂) Hydrolysis pH value of TEOS is 1. (c₁ and c₂)

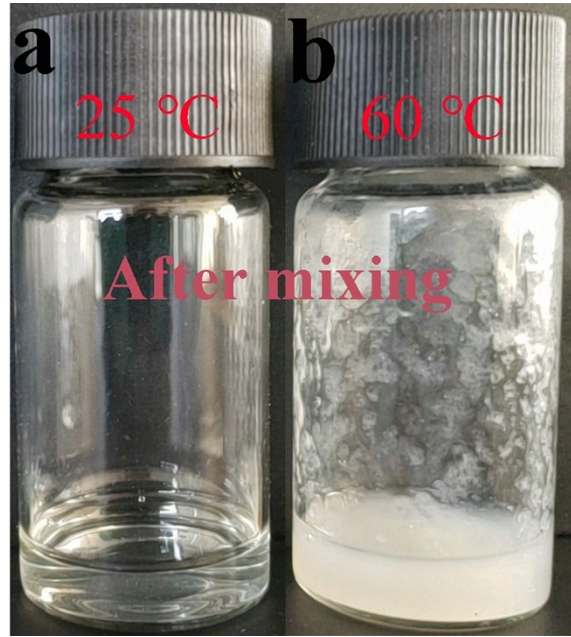
50 Hydrolysis pH value of TEOS is 2. (d₁ and d₂) Hydrolysis pH value of TEOS is 3.

51

52

53

54



55

56 **Figure S4.** Digital images of the mixture of water-soluble PAC and hydrolysate of TEOS under
57 different temperature conditions. (a) Hydrolysis temperature of TEOS is 25 °C. (b) Hydrolysis
58 temperature of TEOS is 60 °C.

59

60

61

62

63

64

65

66

67

68

69



70

71 **Figure S5.** Morphology images of PI aerogel. The PI aerogel showed a disordered honeycombs-

72 like porous structure and the wall of the pores exhibited smooth.

73

74

75

76

77

78

79

80

81

82

83

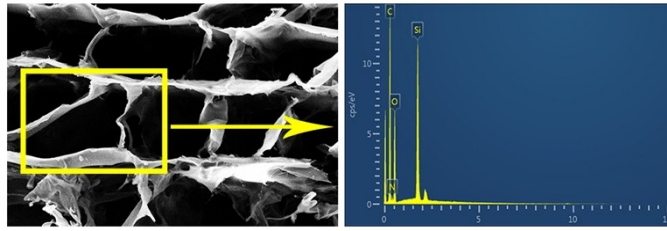
84

85

86

87

88



89

90 **Figure S6.** Morphology image of SiO₂/PI-3 aerogel for EDS mapping.

91

92

93

94

95

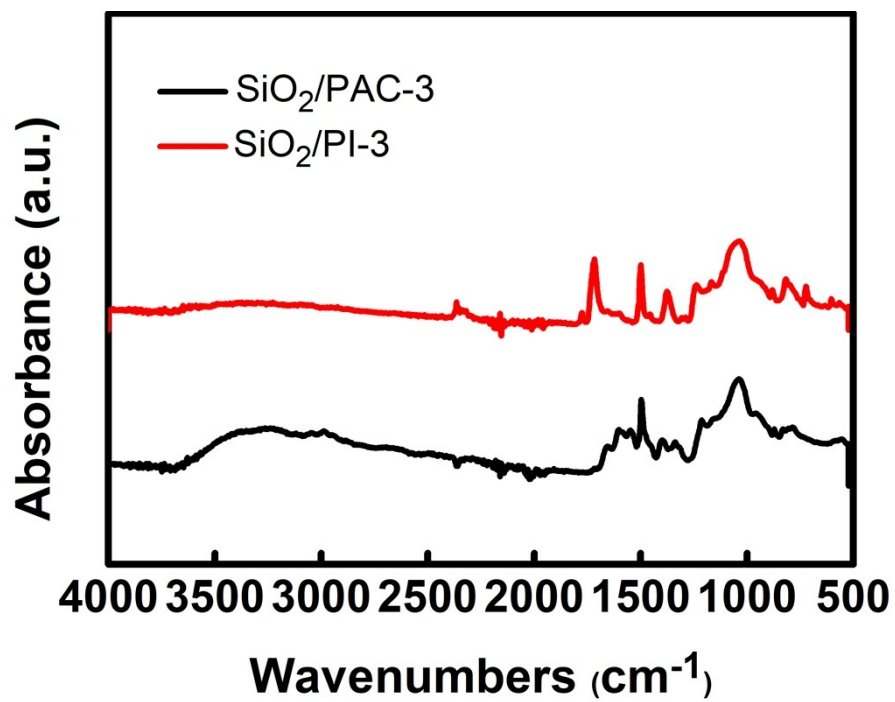
96

97

98

99

100



101

102 **Figure S7.** FTIR spectra of the SiO₂/PAC-3 and SiO₂/PI-3 aerogels.

103

104

105

106

107

108

109

110

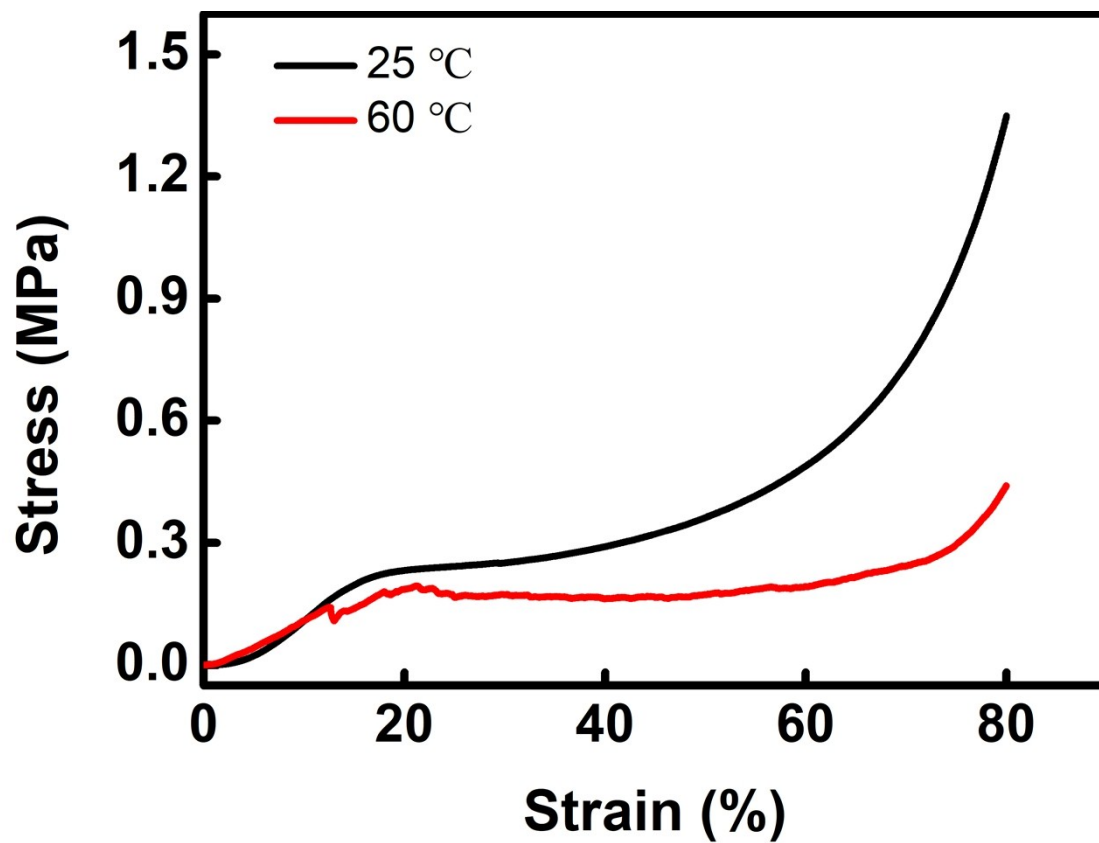
111

112

113

114

115



116

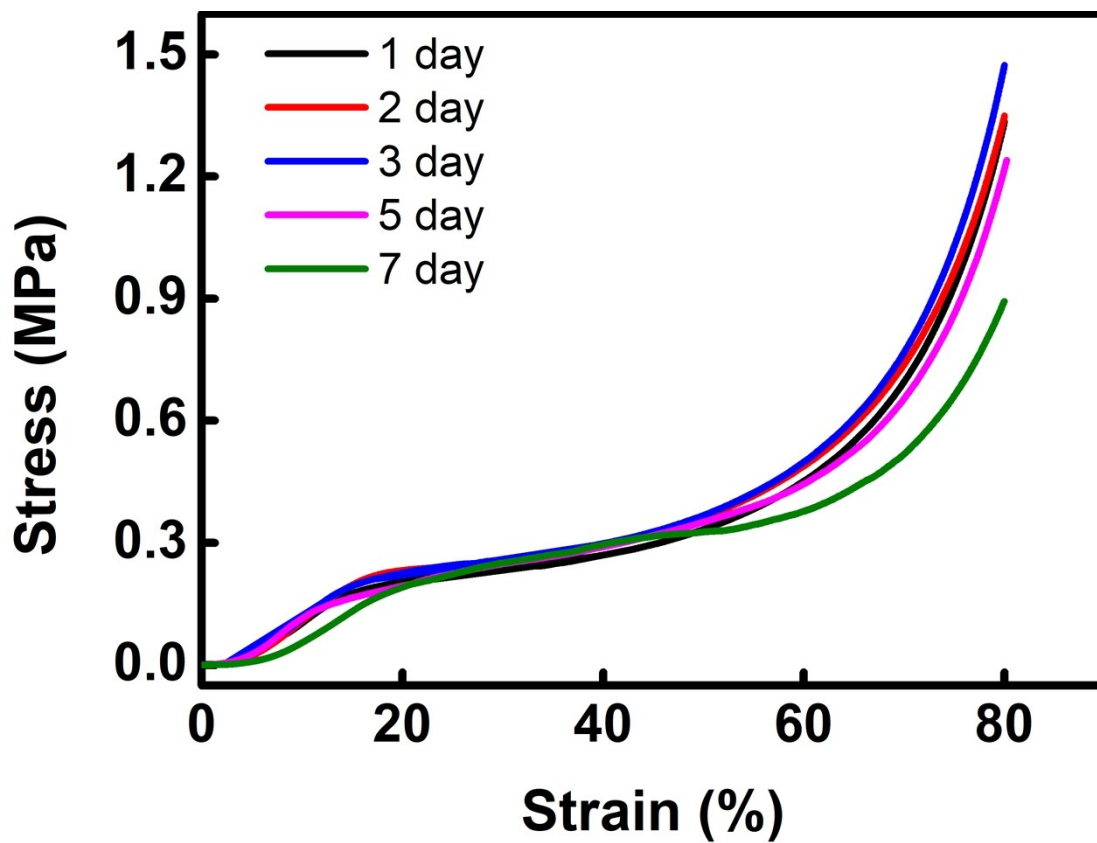
117 **Figure S8.** Compressive stress-strain (δ - ϵ) curves of SiO₂/PI-3 and SiO₂/PI-3-60 aerogels.

118

119

120

121



122

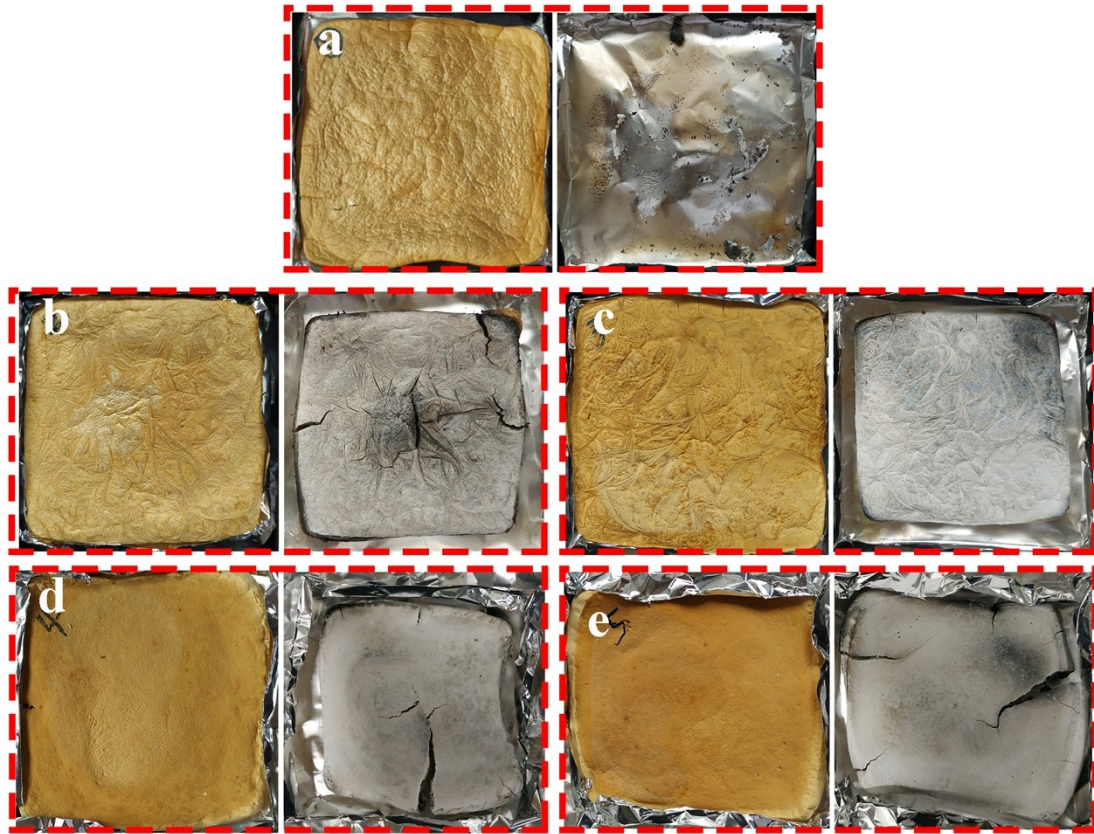
123 **Figure S9.** Compressive stress-strain (δ - ϵ) curves of SiO₂/PI-3, SiO₂/PI-3-1D, SiO₂/PI-3-3D,

124 SiO₂/PI-3-5D, and SiO₂/PI-3-7D aerogels.

125

126

127



128

129 **Figure S10.** Digital images of pre-test and post-test of samples for flame. (a) PI aerogel. (b)

130 SiO₂/PI-1 aerogel. (c) SiO₂/PI-2 aerogel. (d) SiO₂/PI-3 aerogel. (e) SiO₂/PI-4 aerogel.

131

132

133

134

135

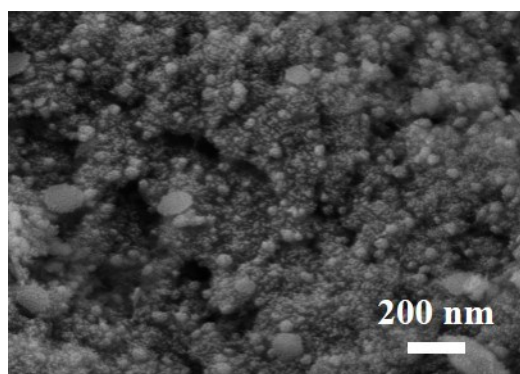
136

137

138

139

140



141

142 **Figure S11.** The morphology of the SiO₂/PI aerogel after burned.

143

144

145

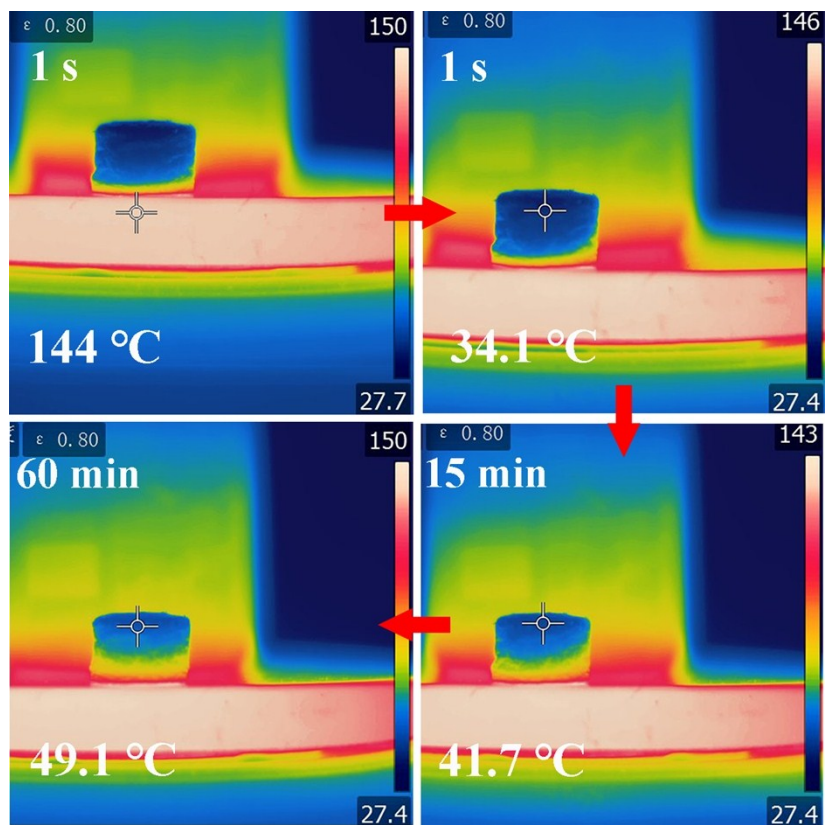
146

147

148

149

150



151

152 **Figure S12.** Infrared images of PI aerogel on a 144 °C heating stage.

153

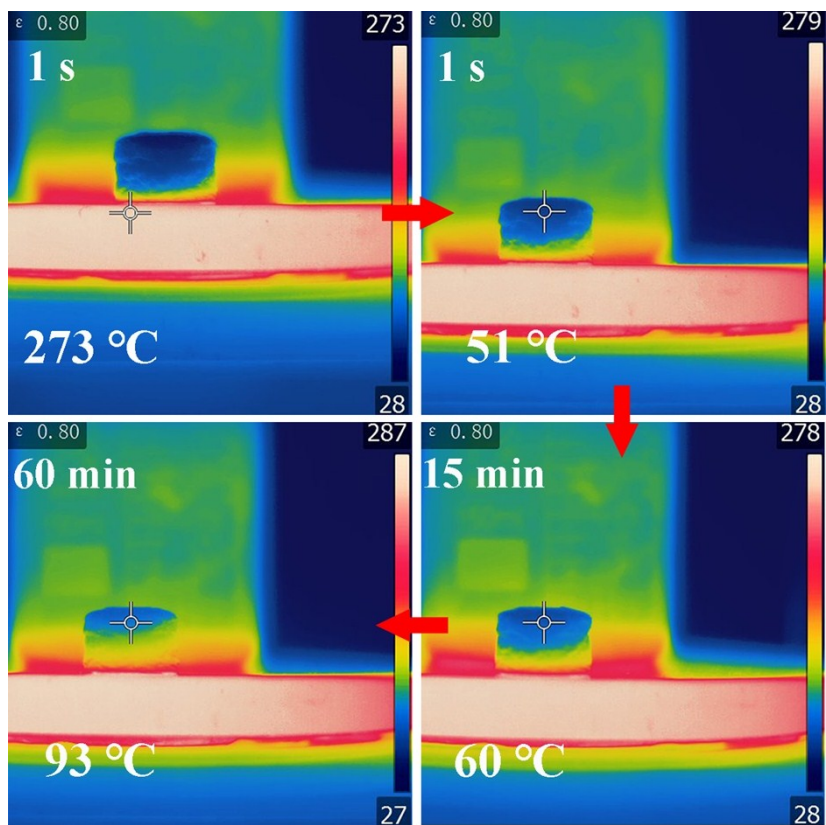
154

155

156

157

158



159

160 **Figure S13.** Infrared images of PI aerogel on a 273 °C heating stage.

161

162

163

164

165

166

167

168

169

170

171

172 **Table S1.** The detailed variable information for preparing silica/polyimide composite aerogels and
 173 the sample names.

Sample	TEOS (g)	pH value (water)	Hydrolysis temperature	Hydrolysis time
			(°C)	(day)
SiO ₂ /PI-1	0.348	2	25	2
SiO ₂ /PI-2	0.696	2	25	2
SiO ₂ /PI-3	1.044	2	25	2
SiO ₂ /PI-4	1.392	2	25	2
—	1.044	< 1	25	2
—	1.044	1	25	2
—	1.044	2	25	2
—	1.044	3	25	2
SiO ₂ /PI-3-60	1.044	2	60	2
SiO ₂ /PI-3-1D	1.044	2	25	1
SiO ₂ /PI-3-3D	1.044	2	25	3
SiO ₂ /PI-3-5D	1.044	2	25	5
SiO ₂ /PI-3-7D	1.044	2	25	7

174

175

176

177

178

179 **Table S2.** Comparison of flame retardant of various organic and inorganic composite flame-
 180 resistant materials.

Sample	LOI	Heat flux (kW m ⁻²)	PHRR (kW m ⁻²)	TPHRR (s)	FIGRA (W s)	THR (MJ m ²)	TSR (m ² m ⁻²)	Ref.
M5Pe5	35	50	80.1	65	1.2	19.7	~	1
PC/TiO ₂ @DPP5 0	29.7	50	412	65	6.3	20.5	~	2
RPUF-15	~	35	265.9	35	7.6	12.2		3
PU/Aerogel -0.7	60	50	220	36	6.3	19	964	4
PSi-70	~	35	19	~	~	0.55	~	5
FPU/Alag-20	~	50	71	14	5.1	3.8	38	6
PMMA/GAPPA	25	50	76	~	~	6.9	~	7
PI/G5/M10	55	50	52.5	52.5	1	10.7	18.4	8
A2.5C2.5-6	~	50	18.6	20	0.9	2.7	54.7	9
P5M3B	27.6	50	146	15	9.7	11.3	~	10
PI	34	50	84	40	2.1	8.6	84	This work
SiO ₂ /PI-1	43	50	52.9	90	0.6	6.2	59	
SiO ₂ /PI-2	44	50	42.2	105	0.4	5.2	44	
SiO ₂ /PI-3	47	50	36.6	115	0.3	4.1	29	
SiO ₂ /PI-4	48	50	30.7	130	0.2	3.2	20	

181

182

183

184 **References**

185 1 H. B. Chen, X. L. Li, M. J. Chen, Y. R. He, H. B. Zhao, *Carbohydr. Polym.*, 2019,
186 **206**, 609-615.

187 2 Y. X. Wei, C. Deng, H. Chen, L. Wan, W. C. Wei, Y. Z. Wang, *ACS Appl. Mater.*
188 *Interfaces*, 2018, **10**, 28036-28050.

189 3 S. X. Wang, H. B. Zhao, W. H. Rao, S. C. Huang, T. Wang, W. Liao, Y. Z. Wang,
190 *Polymer*, 2018, **153**, 616-625.

191 4 H. B. Chen, P. Shen, M. J. Chen, H. B. Zhao, D. A. Schiraldi, *ACS Appl. Mater.*
192 *Interfaces*, 2016, **8**, 32557-32564.

193 5 Z. L. Yu, N. Yang, V. Apostolopoulou-Kalkavoura, B. Qin, Z. Y. Ma, W. Y.
194 Xing, C. Qiao, L. Bergström, M. Antonietti, S. H. Yu, *Angew. Chem., Int. Ed.*, 2018,
195 **57**, 4538-4542.

196 6 H. Y. Xie, W. Yang, A. C. Y. Yuen, C. Xie, J. S. Xie, H. D. Lu, G. H. Yeoh, *Chem.*
197 *Eng. J.*, 2017, **311**, 310-317.

198 7 L. Wang, S. H. Wu, X. Y. Dong, R. Wang, L. Q. Zhang, J. L. Wang, J. Zhong, L. X.
199 Wu, X. Wang, *J. Mater. Chem. A*, 2018, **6**, 4449-4457.

200 8 L. Z. Zuo, W. Fan, Y. F. Zhang, L. Zhang, W. Gao, Y. P. Huang, T. X. Liu,
201 *Compos. Sci. Technol.*, 2016, 139, 57-63.

202 9 X. L. Li, M. J. Chen, H. B. Chen, *Compos. Part B: Eng.*, 2019, 164, 18-25.

203 10 K. Shang, D. D. Ye, A. H. Kang, Y. T. Wang, W. Liao, S. Xu, Y. Z. Wang,
204 *Polymer*, 2017, **131**, 111-119.

