Electronic Supplementary Material (ESI) for RSC Advances.
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This version of the ESI published 17 June 2021 replaces the original version published on 08 June 2021. Three of the values in the 5% KPS column of Table S1 have been updated.

Supporting Information

Experimental Details

Table S1. Raw material ratio and preparation conditions of hydrogels and surface area of aerogels (30% and 50% are the molar ratio of AAm : AMPSL, 2‰ is the molar ratio of MBA : monomers)

Sample	AMPS (g)	LiOH (g)	AAm (g)	MBA (g)	5% KPS (μL)	Total Dose (kGy)	SA_{BET} (m^2 g^{-1})
I1	2.0	0.231	0.206 (30%)	0.0039 (2‰)	68		88.39
I2	2.0	0.231	0.343 (50%)	0.0045 (2‰)	78		80.04
I3 (I-gel)	4.0	0.462	0.412 (30%)	0.0077 (2‰)	136		102.43
I4	4.0	0.462	0.686 (50%)	0.0090 (2‰)	156		95.31
I5	6.0	0.692	0.618 (30%)	0.0116 (2‰)	204		-
I6	6.0	0.692	1.029 (50%)	0.0134 (2‰)	234		-
R1	4.0	0.462	0.412 (30%)	0.0077 (2‰)		20	90.42
R2	4.0	0.462	0.412 (30%)	0.0077 (2‰)		30	105.07
R3	4.0	0.462	0.686 (50%)	0.0090 (2‰)		30	95.32
R4 (R-gel)	4.0	0.462	0.412 (30%)	0.0077 (2‰)		40	114.18
R5	4.0	0.462	0.686 (50%)	0.0090 (2‰)		40	99.83
R6	4.0	0.462	0.412 (30%)	0.0077 (2‰)		50	87.44
R7	4.0	0.462	0.686 (50%)	0.0090 (2‰)		50	76.62
R8	4.0	0.462	0.412 (30%)	0.0077 (2‰)		70	40.66

Firstly, a series of CTS/P(AMPSL-*co*-AAm) double-network hydrogels initiated by KPS initiators were prepared according to different raw material ratios (Tab. S1). Detailed synthesis methods are shown in 2.3. The surface aera of I1 to I4 were characterized. The surface area of I1 and I2 does not increase significantly (or even decrease) compared with that of O-gel, probably due to the low monomer concentration in prepolymer solutions, which is difficult to form a uniform and extensive second network in O-gel. As the monomer concentration increases (I3, I4), the surface area of the aerogels increases. It may indicate that the effective construction of the uniform second network makes the I-gel with double-network structure have good porosity.

However, as the monomer concentration continues to increase (I5, I6), the proportion of the more hydrophilic second network increases after polymerization. After the hydrogels were washed and swelled by deionized water, the swelling was particularly obvious, and even the macroscopical phase separation of P(AMPSL-*co*-AAm) and CTS occurred.

Then a group of CTS/P(AMPSL-*co*-AAm) double-network hydrogels initiated by radiation were prepared. The impacts of different radiation dose and the ratio of AMPSL : AAM on the surface area of aerogels were analyzed (Fig. S1). As the radiation dose increases from 20 kGy to 70 kGy, the surface area of the aerogels increases and then decreases. When the radiation dose is 40 kGy, the surface area of aerogel (R-gel) is the largest, which may be due to the radiation-initiated *in situ* free radical polymerization, a stable polymer double network is well-constructed in the hydrogel. However, as the radiation dose continues to increase, the surface area of aerogels decreases significantly. It is probably because that high radiation dose starts to cause polymer degradation.

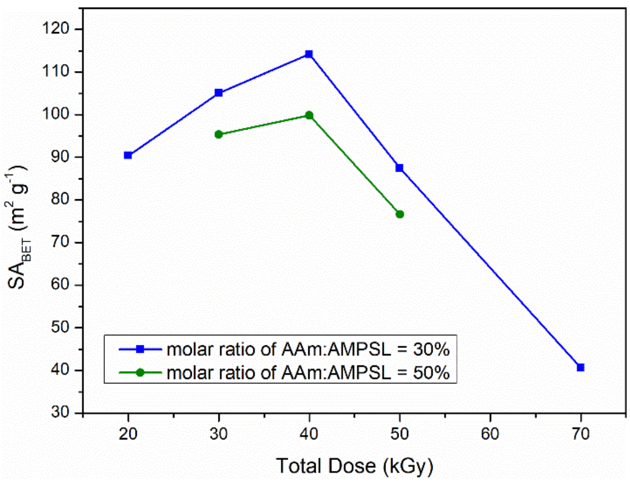

Figure S1 Surface aera of freeze-dried radiation-initiated hydrogels with different total dose.

Swelling Behavior Characterization

The SR of O-gel, P-gel, I-gel and R-gel was calculated by gravimetric method (Fig. S2).

The SR of O-gel and P-gel are 17.9 and 37.8 (g/g), respectively. After the introduction of the P(AMPSL-*co*-AAm) second network, the SR of the double-network hydrogels increases obviously compared to O-gel. The SR of I-gel and R-gel is 25.1 (g/g) and 28.9 (g/g), respectively. The higher SR of the double-network hydrogels is probably result from the more hydrophilic P(AMPSL-*co*-AAm) second network. Moreover, the SR of R-gel is larger than I-gel, which may due to the radiation-initiated double network structure of R-gel is more uniform. The uniform polymer network can contact with more water molecules during swelling.

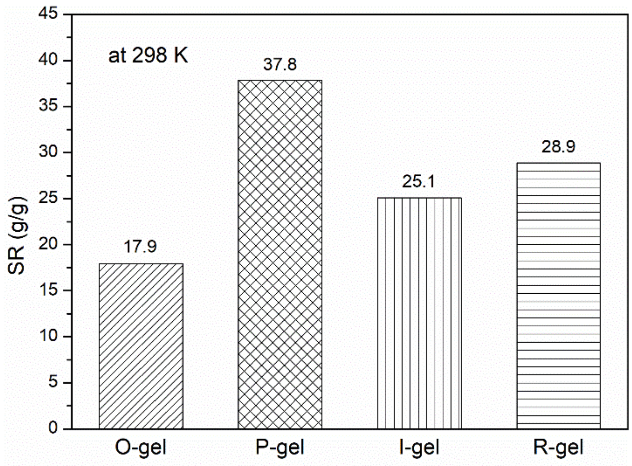

Figure S2 Swelling ratio of O-gel, P-gel, I-gel and R-gel.