Supporting Information

Hierarchical Cactus-Like Microsphere Network Membranes Engineered via Multiple Polyphenol-Mediated Complexation for Efficient Solar-Powered Water Purification

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Fig. S1 Digital photographs and SEM images of PPy@PVDF membrane.



Fig. S2 High-resolution C 1s spectra of (a) PVDF, (b) TA-Ti and (c) PPy@TA-Ti membrane.



Fig. S3 Evaporation rate of membrane under dark condition and 1 sun irradiation.

The thermogravimetric analysis (TG) was used to confirm the reduction of water evaporation enthalpy in membrane. 5 mg pure water and 5 mg membrane sample were synchronously located in a sealed crucible and measured under temperature of evaporation system. The TG program simulated a complete dehydration process of membrane. The water evaporation rate is calculated by the slope of each thermal gravimetric curve. The evaporation enthalpy of membrane confined water (E_{equ}) can be calculated according to the water mass changes and the known theoretical value of bulk water (Table S1) under identical power input (U_{in}).

$$U_{in} = E_{equ} m_g = E_0 m_0$$

where E_0 and m_0 refer to the evaporation enthalpy and mass change of bulk water in the dark without membrane, respectively; m_g is the water mass change within membrane sample.

Table S1. The theoretical evaporation enthalpy of bulk water at different temperature.

Temperature (°C) ^a	35	40	48	51
Evaporation enthalpy (J g ⁻¹)	2412.4	2401.1	2382.6	2378.1

^a Temperatures of bulk water are determined by the evaporation temperature on different membrane surface (Fig. S4).



Fig. S4 Surface temperature IR thermal images of membrane in evaporation system. The water evaporation enthalpy is related to the temperature of evaporation system in the steady state. The surface temperature of membrane was monitored by a thermal camera after illuminating 40 min. The surface temperature of PVDF, TA-Ti, PPy@PVDF, PPy@TA-Ti membrane was stabilized at 35 °C, 40 °C, 48 °C and 51 °C, respectively.

The cactus-like microsphere networks of PPy@TA-Ti PTMs served as localizations to confine water clusters in the nano-grooves and microcavities. The confined water

clusters possessed a higher saturated vapor pressure and a lower evaporation enthalpy compared to that of the flat liquid surface¹⁻⁴. Also, the cactus-like structure may also disrupt and disorganize the hydrogen bond distribution of water for faster evaporation⁵. Thus, the evaporation enthalpy of PPy@TA-Ti PTMs was 2028 J g⁻¹ which was smaller than the classic enthalpy of water (Fig. S5). The efficiency of membranes under 1 sun irradiation was calibrated (Fig. S5d).



Fig. S5 Thermograms of (a) pure water evaporation and (b) membrane confined water evaporation under the same evaporation temperature. (c) Equivalent enthalpy of confined water within different membrane. (d) Solar-to-vapor conversion efficiency of different membranes under 1 sun irradiation.

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