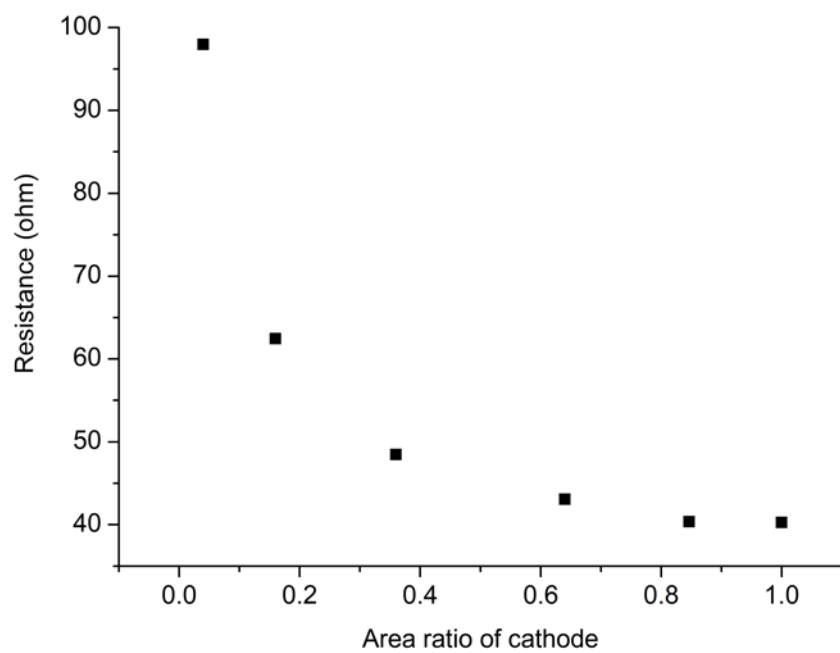


19

20 **Fig. S1.** Stack configuration of the large-scale RED stack used for experiments. Black and red
 21 meshes are Pt-coated Ti mesh, and polymer-coated Ti mesh, respectively.

22

23

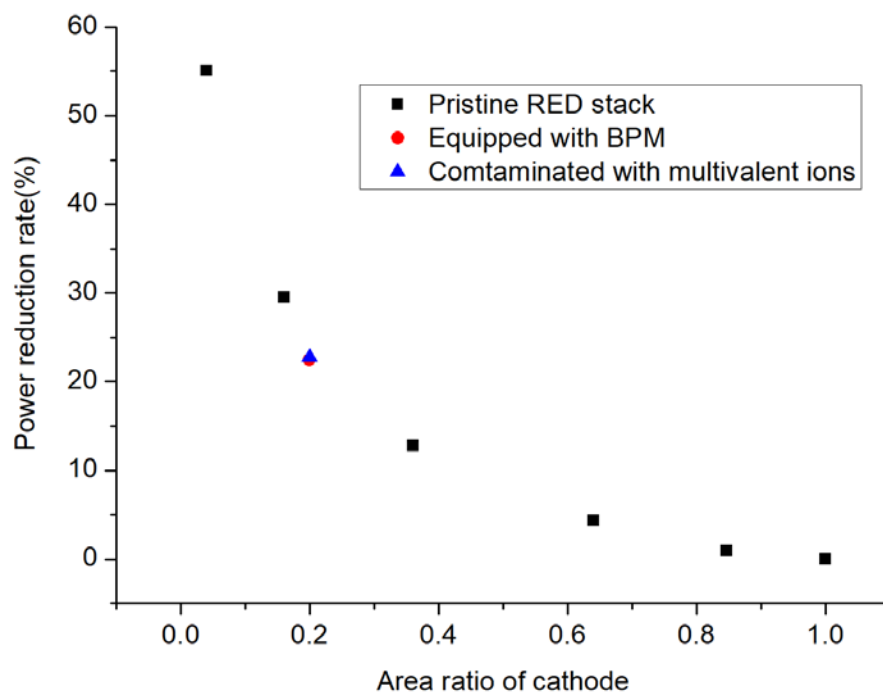


24

25 **Fig. S2.** Internal resistances with varied area ratios of cathode in the lab-scale RED stack with
26 100 cell pairs.

27

28

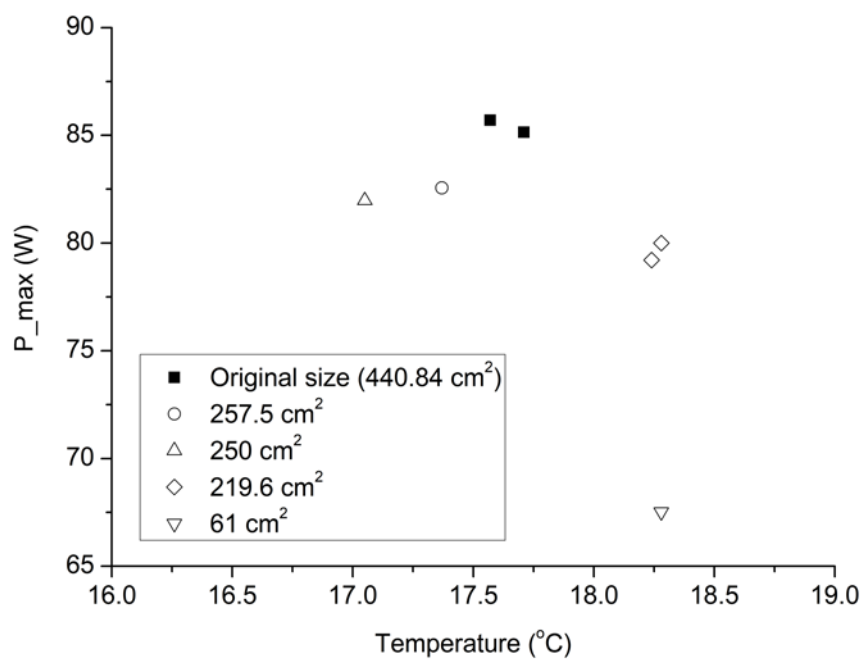


29

30 **Fig. S3.** Power-reduction rates of the lab-scale RED stacks contaminated with multivalent ions
 31 (blue triangle) or those equipped with a bipolar membrane (BPM, Fumatech BWT GmbH,
 32 Germany) as the shielding membrane (red circle). The other data (black squares) are the same
 33 as that in Figure 1d.

34

35



36

37 **Fig. S4.** P_{max} of the large-scale RED stack with 1,000 cell pairs as a function of temperature
 38 of the feed solutions.

39

40

41

42

43

44

45

46

47

48 **Calculation of permselectivity (α)**

49 The anion transport number (t_-^m) in the membrane phase (AEM), was calculated from the
50 measured membrane potential (E_m).

51
$$t_-^m = \frac{\left[E_m / \left(\frac{RT}{F} \ln \frac{a_1}{a_2} \right) \right]}{2} + 1$$

52 where R is the gas constant (8.314 J/(mol K)), F is the Faraday constant (96490 C/mol), T is
53 the absolute temperature (K), and a_1 and a_2 are the activities of the concentrated and the
54 diluted NaCl solutions (mol/L), respectively.

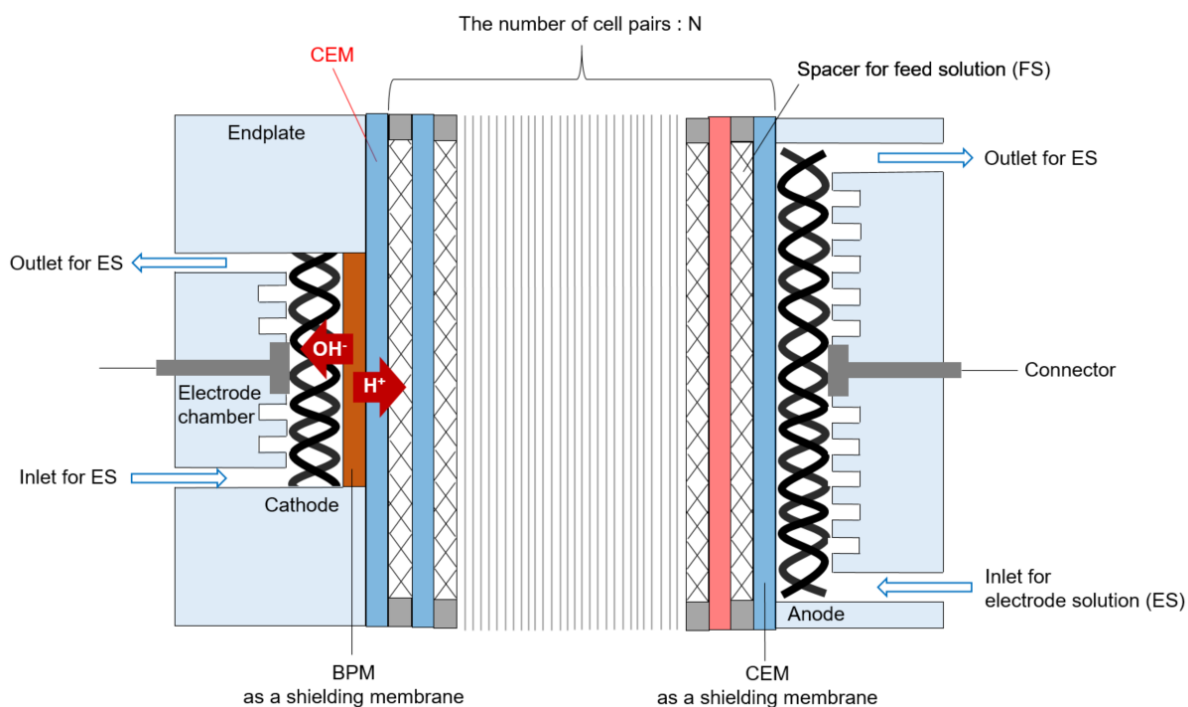
55 The α can be defined in terms of transport numbers (Strathmann 2011).

56
$$\alpha = \frac{t_-^m - t_-^s}{t_+^s}$$

57 where m refers to the membrane phase and s refers to the solution phase, t_+^s refers to cation
58 transport number in the solution phase, and t_-^s refers to the anion transport number in the
59 solution phase.

60

61



62

63 **Fig. S5.** Schematic of asymmetrical electrode system with bipolar membrane (BPM) for
 64 suppressing inorganic scaling around the cathode in a large-scale RED system. To prevent
 65 leakage between the BPM and the corresponding spacer, the CEM can be additionally inserted
 66 next to the BPM, and it served the purpose for both acting as a physical barrier for bulk fluid,
 67 and also as the ion-selective layer for protons and cations.

68

69

70 Strathmann, H. (2011) Introduction to membrane science and technology, Wiley-VCH, Weinheim.

71