1 **Supporting Information**

2 **Catalytic behavior of Mn during molten salt oxidation of cationic**

3 **exchange resins in Li_2CO_3-Na_2CO_3-K_2CO_3 melt**

4

5 **Supporting method**

6   Switching capacity test method: Firstly, 0.25, 0.5, 1, 1.5, 2, and 4 g of $MnCl_2$ were

7 weighed to make 50 ml of exchange solution at concentrations of 0.039, 0.079, 0.158,

8 0.317, 0.238, and 0.635 mol/L, respectively. Subsequent exchange analysis tests were

9 performed using the pH titration method and ICP exchange efficiency method.

10   (1) The pH titration method: The pH values of the solutions were measured before

11 exchange as "a", and then the exchange solution was mixed with the resin and placed

12 in a shaker for 48h to complete the exchange. The pH value of the exchanged solution

13 was measured as "b", and the pH value was adjusted back to "a" by using 1 mol/L

14 sodium hydroxide solution, and the volume of sodium hydroxide consumed was "V_1".

15 Since the sulfonic acid group is a Lewis acid, the effect of ionization of the sulfonic

16 acid group on the pH value should be excluded from the process, and therefore, a blank

17 experiment was set up (the resin was exchanged on a shaking table by mixing it with

18 distilled water, and then the filtrate was adjusted back to its starting value by consuming

19 volume of sodium hydroxide solution as "V_2"). Then the Mn content exchanged onto

20 the resin is:

21   Molarity of Mn exchange = $(V_1-V_2)/2$

22   (2) ICP measurement: ICP is applied to measure the concentration of metal ions in

23 solution. After the resin has been exchanged, the filtrate is diluted to the corresponding

24 concentration and tested by ICP. The concentration of the test result is considered as

25 "c_1". The concentration of the exchange solution before exchange is considered as "c_2".

26   Exchange efficiency = $(c_2-c_1)/c_2 \times 100\%$

27   According to the test results of pH titration method, the maximum exchange

28 concentration should be between 0.1587 and 0.6349 mol/L, as shown in Fig. S1(a).

29 After continuing to increase the exchange concentration, although the number of moles

30 exchanged still increased, it could be due to two reasons: (1) hydrolysis of unexchanged

31 Mn^{2+} lowered the pH, which was counted in the volume of sodium hydroxide consumed

32 and thus increased the test results. (2) Mn^{2+} is not exchanged into the sulfonic acid

33 groups of the resin, but is adsorbed on the surface of the resin particles, thus increasing

34 the result. According to the ICP test results in Fig. S1(b), the exchange efficiency must

35 meet the requirements of the nuclear level exchange efficiency. Therefore, the

36 concentration of 0.158 mol/L exchange solution was used in this experiment in order

37 to achieve a better match with the actual treated waste resin.
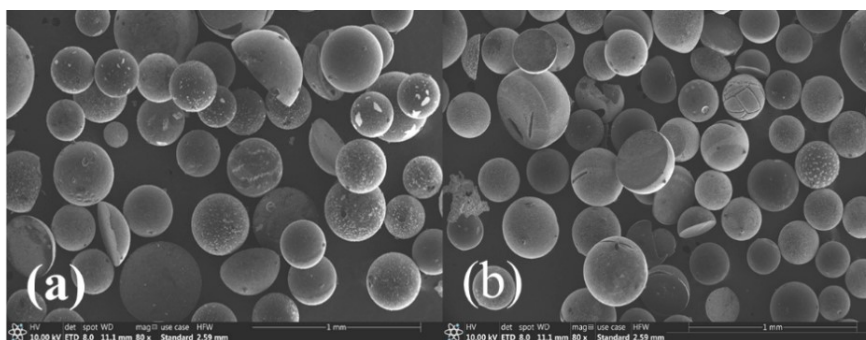
38 **Supporting Fig**
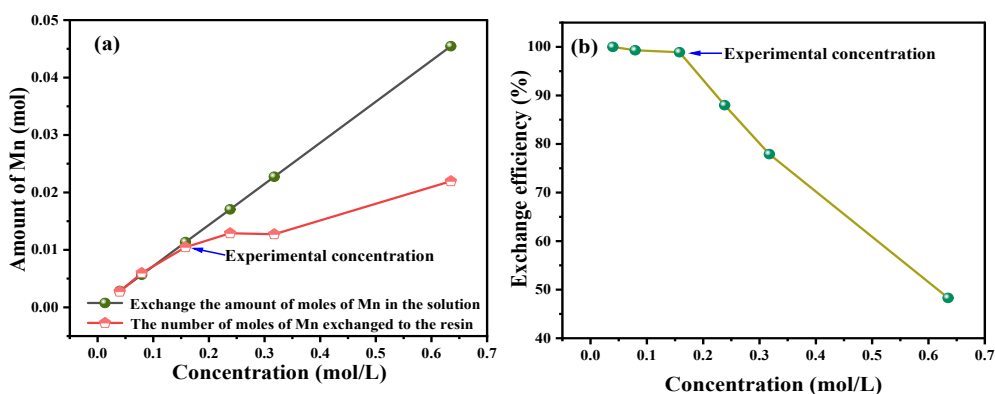
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40 Fig. S1 Exchange adsorption tests for solutions with different Mn ion concentrations: (a) pH titration

41 method, (b) ICP exchange efficiency method.

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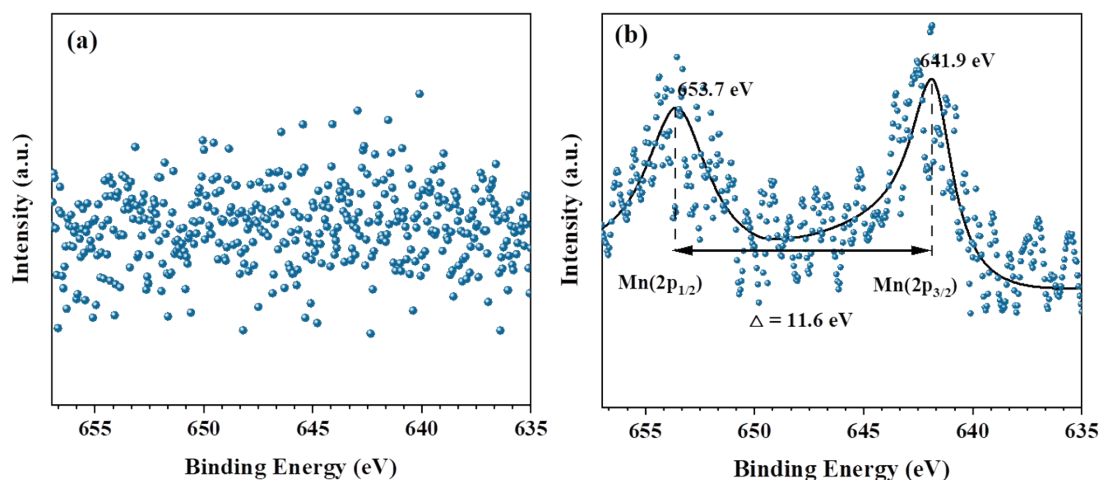
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44 Fig. S2. XPS spectra of samples from molten salt oxidation at 650°C: (a) resin residue, (b) waste
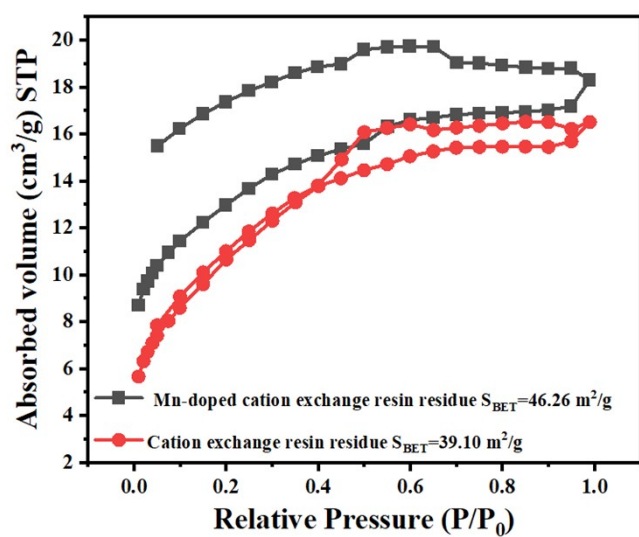
45 salt.

46

48 Fig. S3. The SEM image of cation exchange resin (a) and Mn-doped cation exchange resin (b) at
49 650 °C.

50

52 Fig. S4. The N_2 adsorption isotherm of the undoped and Mn doped resin residues after molten salt
53 oxidation.