Supplementary information

Table S1 Specification of batteries, conditions, and results of submerged deactivation of (a) lithium-ion batteries and (b) cells dismantled from LIBs for a hybrid electric vehicle with lime water in N_2 flow.

(a)	**Specification of battery sample**				**Approximate volume of lime water (L)**	**N_2 flow[*5] (L/min)**	**Gas evolution**		
Exp. #.	**W×L×D (mm)**	**Positive electrode**	**SOC (%)**	**OCV (V)**			**H_2**	**O_2**	**White smoke**
S01[*1]	30×40×7	NCA[*3]	100	4.14	0.30	4.5	Detected	n.d.c.[*8]	n.o.[*9]
S02[*1]	30×40×7	NCA[*3]	100	4.13	0.30	4.5	Detected	n.d.c.[*8]	n.o.[*9]
S03[*1]	30×40×7	NCA[*3]	0	2.35	0.30	4.5	-[*7]	-[*7]	n.o.[*9]
S04[*1]	40×21×5	NMC[*4]	100	4.13	0.30	4.5	Detected	n.d.c.[*8]	n.o.[*9]
S05[*1]	88×46×9	LFP[*5]	100	3.58	0.67	4.5	Detected	n.d.c.[*8]	n.o.[*9]
S06[*2]	30×40×7	NCA[*3]	100	4.13	114	15–20	-[*7]	-[*7]	n.o.[*9]
S07[*2]	30×40×7	NCA[*3]	100	4.13	190	15–20	Detected	n.d.c.[*7]	Observed

[*1] The LIB was cut into two pieces in lime water in N_2 atmosphere.

[*2] The biaxial crusher in Fig. 2 was used

[*3] $LiNi_xCo_yAl_zO_2$

[*4] $LiNi_xCo_yMn_zO_2$

[*5] $LiFePO_4$

[*6] The distance between the nitrogen gas inlet and the crusher differs in the prismatic cell experiments and the small cell experiments.

[*7] Measurement / observation was not carried out.

[*8] not detected clearly.

[*9] not observed.

(b)	**Specification**		**Approximate volume of lime water (L)**	**N_2 flow (L/min)**	**Gas evolution**		
Exp. #.	**SOC (%)**	**OCV (V)**			**H_2**	**O_2**	**White smoke**
S08	100	4.10	222	15–20	Detected	n.d.c.[*2]	Observed

S09	90	4.05	222	15–20	Detected	n.d.c.*[2]	Observed
S10	50	3.68	222	15–20	Detected	n.d.c.*[2]	n.o.*[4]
S11	25	3.30	222	15–20	n.d.c.*[2]	n.d.c.*[2]	n.o.*[4]
S12	100	4.09	222	15–20	-*[3]	-*[3]	Observed
S13	100	4.09	222	15–20	-*[3]	-*[3]	Observed
S14	-	0.56	222	15–20	n.d.c.*[2]	n.d.c.*[2]	n.o.*[4]
S15	-	0.97	222	15–20	n.d.c.*[2]	n.d.c.*[2]	n.o.*[4]
S16	100	4.09*[1]	222	15–20	Detected	n.d.c.*[2]	-*[3]
S17	100	4.09*[1]	222	15–20	Detected	n.d.c.*[2]	Observed
S18	100	4.07	222	15–20	-*[3]	n.d.c.*[2]	Observed
S19	100	4.07	222	15–20	Detected	n.d.c.*[2]	Observed
S20	100	4.07	222	15–20	Detected	n.d.c.*[2]	Observed
	100	4.08					

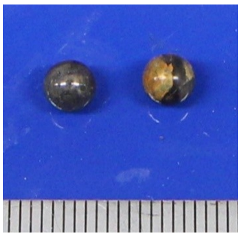
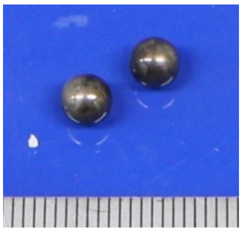
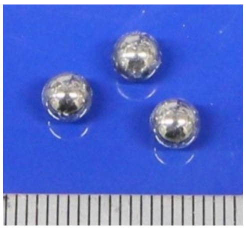
*[1] OCV was measured one month before comminution.

*[2] not detected clearly. O_2 concentration in the hopper was constant lower than 0.2 %.

*[3] Measurement / observation was not carried out.

*[4] not observed.

Table S2 The results of corrosion tests of steel in hydrofluoric acid, sodium chloride (NaCl) water, and lime water. Hydrofluoric acid is selected because Li salts such as $LiPF_6$ and $LiBF_4$ in batteries are hydrolyzed to produce HF in the solution. Steel balls (Diameter: 5 mm, SUJ-2) were immersed into 100 mL of 0.01 mM hydrofluoric acid, 0.25 M NaCl water, or lime water where 1 g of $Ca(OH)_2$ was added. The photographs of steel balls and solutions were shown. The color of steel balls in hydrofluoric acid and NaCl water was turned black one day later. In addition, orange-brown precipitates were formed on the steel balls in the solutions after 7 days. On the other hand, the obvious corrosion was not observed in lime water because color change of steel balls and formation of precipitate were not observed. This is explained with passivation film on the surface.[1,2] This point is one of reasons why we recommend to use lime water. But, the passivation film could be broken by Cl^- ion.[1,2] We still need to be careful about the corrosion of equipment.

Elapsed Time (day)	**Sample**	**Aqueous solution for corrosion test**		
		1.0×10^{-5} M hydrofluoric acid	**0.25 M NaCl water**	**Lime water ($Ca(OH)_2$ suspended)**
1	Steel ball			
7	Steel ball			
	Steel ball with deposit in solution			

1) Y. Tsuru, Y. Tamae, Y. Nagai, T. Nakahata, & K. Matsui, Effects of solution pH, sodium chloride and amino acids on the corrosion of a steel plate in lime water, *J. Soc. Mat. Sci., Japan(Zairyo),* 2002, **51**, 1197–1202.
2) V. K. Gouda, *British Corrosion Journal*, 1970, **5**, 198–203.

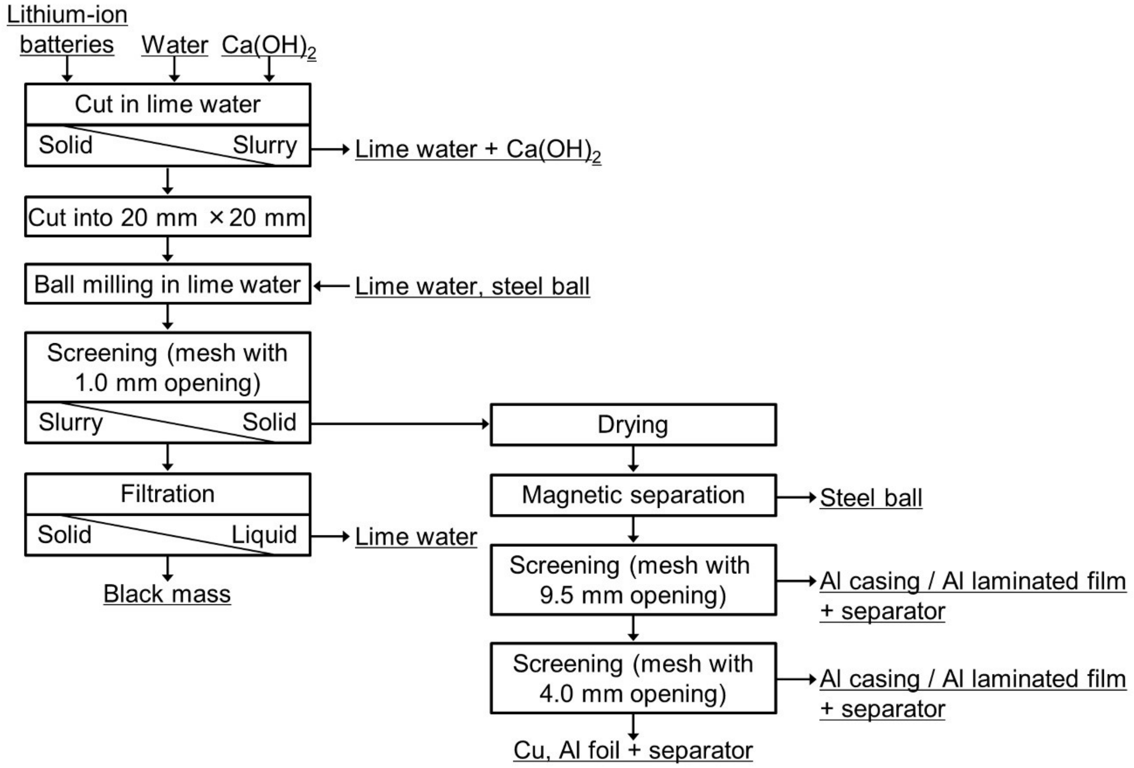


Fig. S1 Flowchart of submerged comminution and physical sorting of lithium ion batteries examined in Fig. S3(A) and (B). Two types of lithium ion batteries using organic electrolyte (called as small LIB in this paper) and polymer electrolyte (called as Li-Po) were examined. Small LIBs are prismatic batteries with NCA positive electrode (Capacity: 895 mAh) or LFP positive electrode (Capacity: 2000 mAh), and Li-Po is Li-ion polymer battery with NMC positive electrode (Capacity: 400 mAh). These batteries were fully charged with the enclosed charger. These were submerged and cut with nippers into two pieces in 300 mL or 670 mL of lime water, which was prepared from deionized water and $Ca(OH)_2$ of more than saturation limit, in a polycarbonate glove box under 4.5 L/min N_2 flow (O_2 conc. < 1 %) in Exp. #S01 to #S05. The sampled gas was analyzed by gas chromatography (Shimadzu Corp., GC-8A). The conditions and results are summarized in Table S1. The batteries after comminution were salvaged from lime water. The batteries were cut into approximately 20 mm × 20 mm and pulverized by ball milling with 100 mL of lime water and steel balls (Diameter: 9 mm) in a polypropylene bottle for 10–72 h at 650 rpm. The milled samples were separated from slurry by mesh with 1.0 mm opening and dried at 60 °C. The solid fraction was then subjected to screening by mesh with 4.0 mm and 9.5 mm opening and magnetic separation. These Al casing, separator, and Cu pieces were sorted by vibrating meshes for approximately 10 min. The slurry fraction was filtrated to separate black mass from lime water. Each component was dissolved in mixtures of 1 M hydrochloric acid and 0.82 M hydrogen peroxide at 40 °C for 72 h and analyzed by inductively coupled plasma-atomic emission spectroscopy (ICP-AES, SII Nano Technology Inc., SPS3520 UV). The distribution fraction of each component was analyzed by sample weight and ICP-AES analysis.

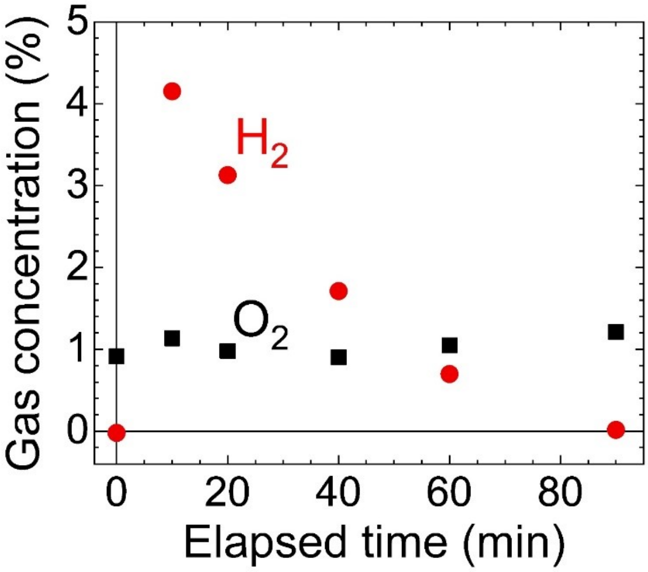


Fig. S2 Profiles of hydrogen and oxygen concentration during cutting in lime water (Exp. #S02). The LIB was cut into two pieces in lime water in N_2 atmosphere, and gas evolution was observed. About 60 cm^3 of gas was sampled at the position of 10–20 mm above water level after 0, 10, 20, 40, 60, and 90 min of the cutting. The sampled gas was analyzed by gas chromatography (Shimadzu Corp., GC-8A). The oxygen concentration stayed almost constant around 1 %. On the other hand, the concentration of hydrogen was increased to approximately 4 % just after the cutting. These results indicate that the evolved gas was not oxygen but mainly hydrogen. The hydrogen evolution was also observed in the cases of LIB with LFP positive electrode (Exp. #S05) and Li-Po (Exp. #S04) by gas chromatography.

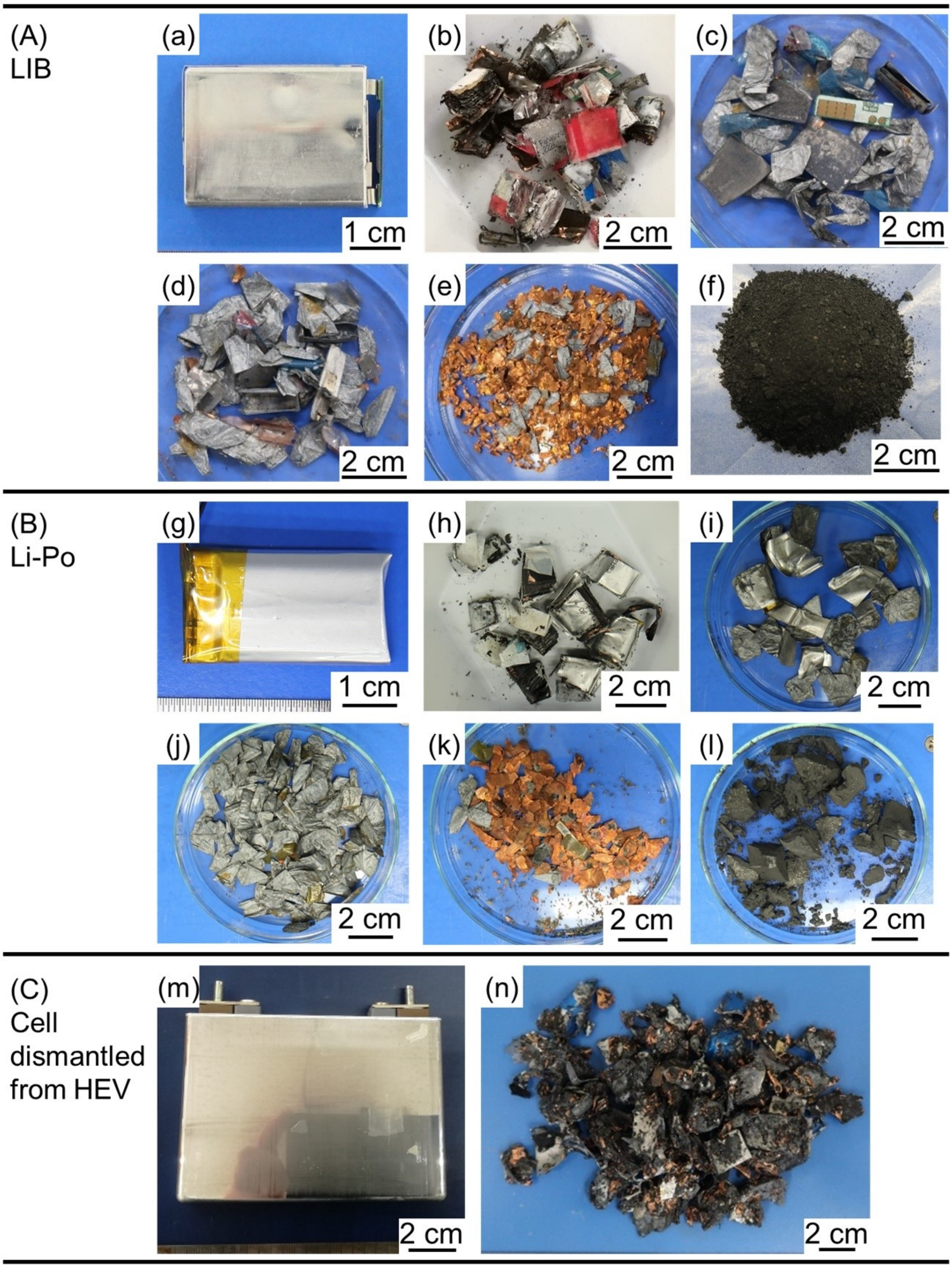


Fig. S3 (A) (a) LIB before experiment (b) after cut into 20 mm × 20 mm (Exp. #S02, hereafter the same), (c) separator and Al casing pieces on mesh with 9.5 mm opening and (d) 4.0 mm opening, (e) Cu and separator pieces under mesh with 4.0 mm opening, and (f) black mass mainly composed of NCA positive electrode material and graphite. These

Al casing, separator, and Cu pieces were sorted by vibrating meshes for approximately 10 min. By ICP-AES analysis, 97 % of Ni and 99 % of Co in LIB were concentrated in the black mass. 90% of Cu was obtained in metallic form under mesh with 4.0 mm opening. The distribution fraction of Li to black mass was 97% at Exp. #S02 and 67 % at Exp. #S01. (B) (g) Li-Po before experiment, (h) after cut into 20 mm × 20 mm (Exp. #S04, hereafter the same), (i) aluminum laminated film and separator on mesh with 9.5 mm opening, (j) separator on mesh with 4.0 mm opening, (k) Cu and separator pieces under mesh with 4.0 mm opening, and (l) black mass mainly composed of NMC positive electrode material and graphite. 88.2 % of Li, 100 % of Ni, 100 % of Co, and 99.7 % of Mn were concentrated in the black mass. Additionally, 90% of Cu was obtained under mesh with 4.0 mm opening. (C) (m) The dismantled cell before and (n) after submerged comminution (Exp. #S08).

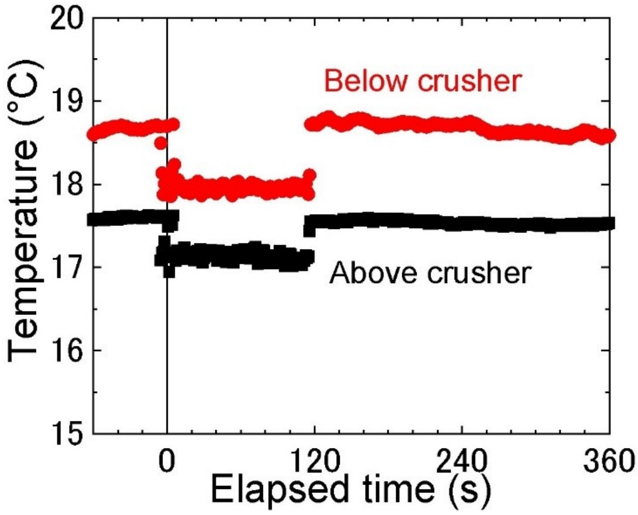


Fig. S4 Water temperature measured with thermocouples during the submerged comminution of LIBs in lime water under N_2 flow (Exp. #S08). The water temperature did not rise by comminution. The temperature showed a slightly lower value when the crusher started running and returned to the original value when the crusher finished running at 120 seconds. The reason for this behavior is unknown.

Movie S1.

Submerged comminution of cell dismantled from LIB for a hybrid electric vehicle (SOC = 25 %, Cell voltage: 3.30 V) with lime water in N_2 flow, as seen in Fig. 3 (a) (Exp. #S11).

Movie S2.

Submerged comminution of cell dismantled from LIB for a hybrid electric vehicle (SOC = 50 %, Cell voltage: 3.68 V) with lime water in N_2 flow, as seen in Fig. 3 (a) (Exp. #S10).

Movie S3.

Submerged comminution of cell dismantled from LIB for a hybrid electric vehicle (SOC = 100 %, Cell voltage: 4.10 V) with lime water in N_2 flow, as seen in Fig. 3 (a) (Exp. #S08).