Supplementary Materials

Full concentration gradient Li-rich layered oxides cathode materials via kinetic regulation of heterogeneous nucleation growth

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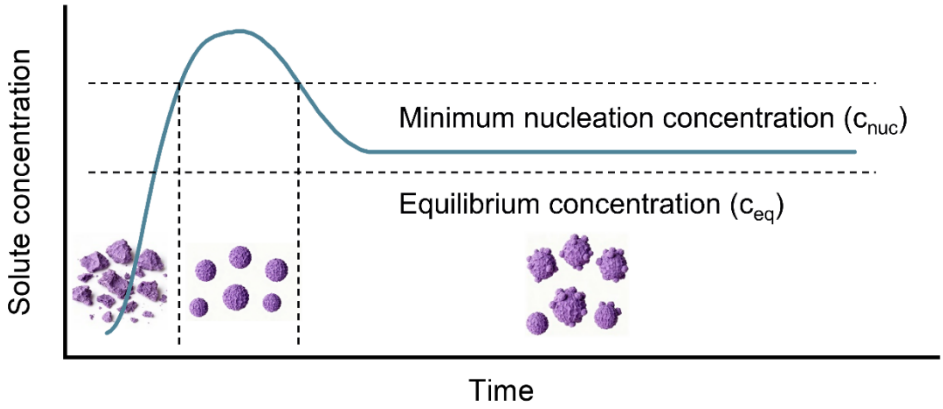
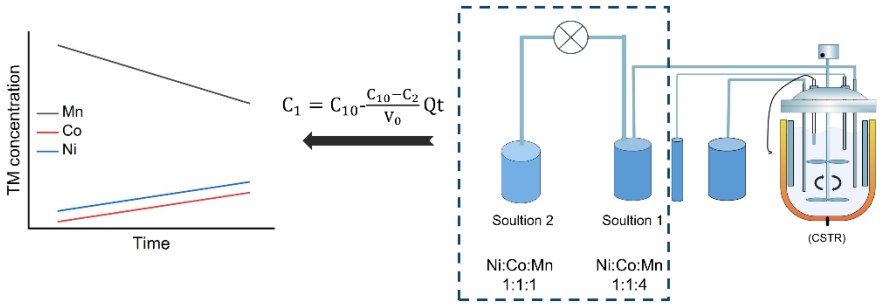
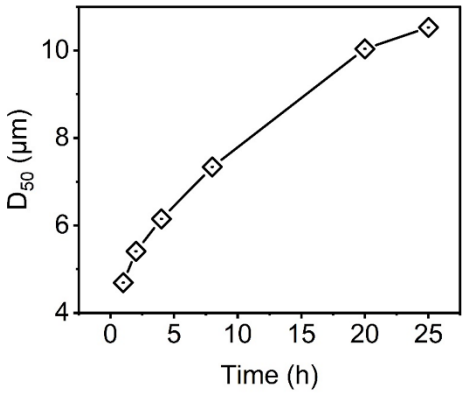


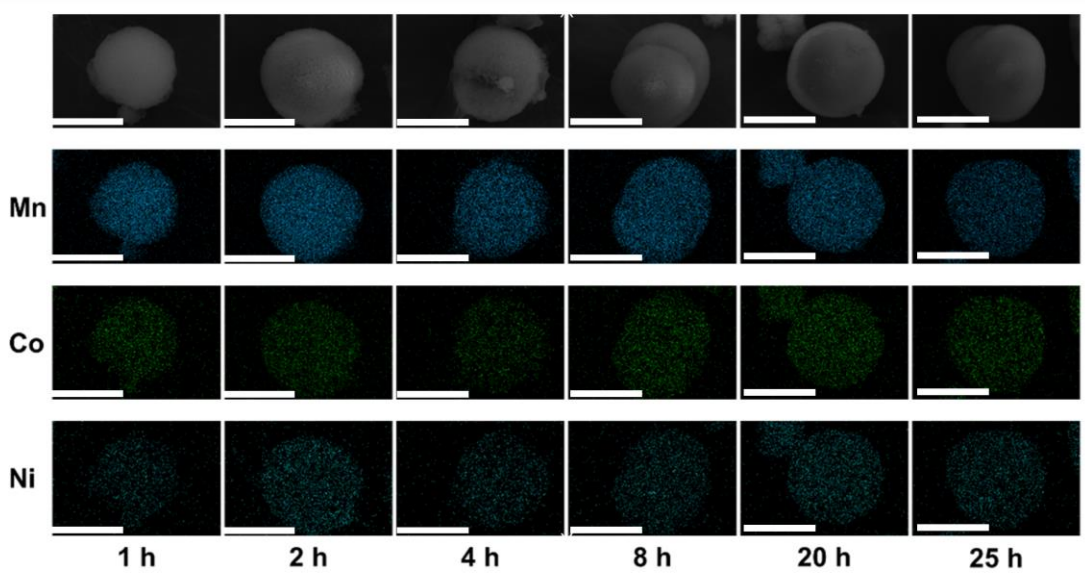
Supplementary Figure 1. The variation curve of solute concentration with time during nucleation.



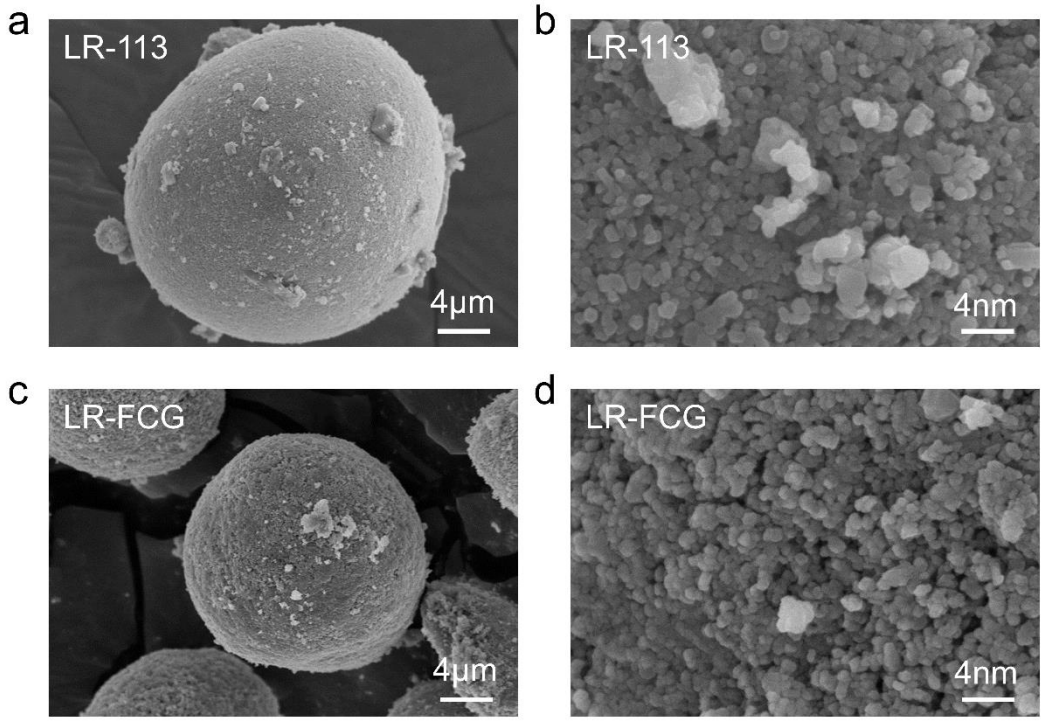
Supplementary Figure 2. Schematic illustration of the reaction setup. (C_1, real-time concentration of TM ions in Solution 1; C_{10}, initial concentration of TM ions in Solution 1; C_2, concentration of TM ions in Solution 2; V_0, initial volume of Solution 1; Q, flow rate of Solution 1 fed into the reactor.)



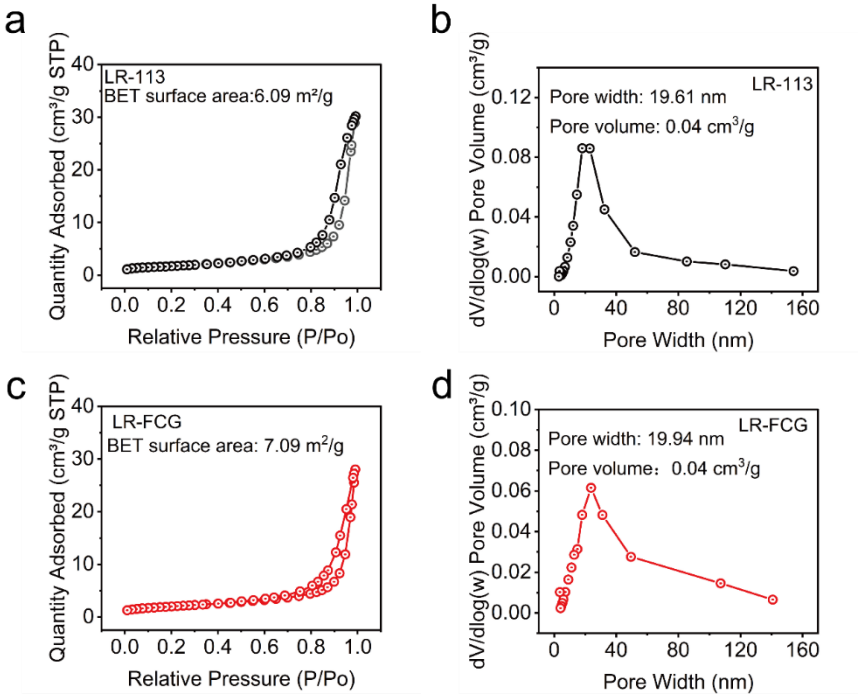
Supplementary Figure 3. Evolution of the median particle size (D_{50}) of the precursor at different reaction times during the coprecipitation process.



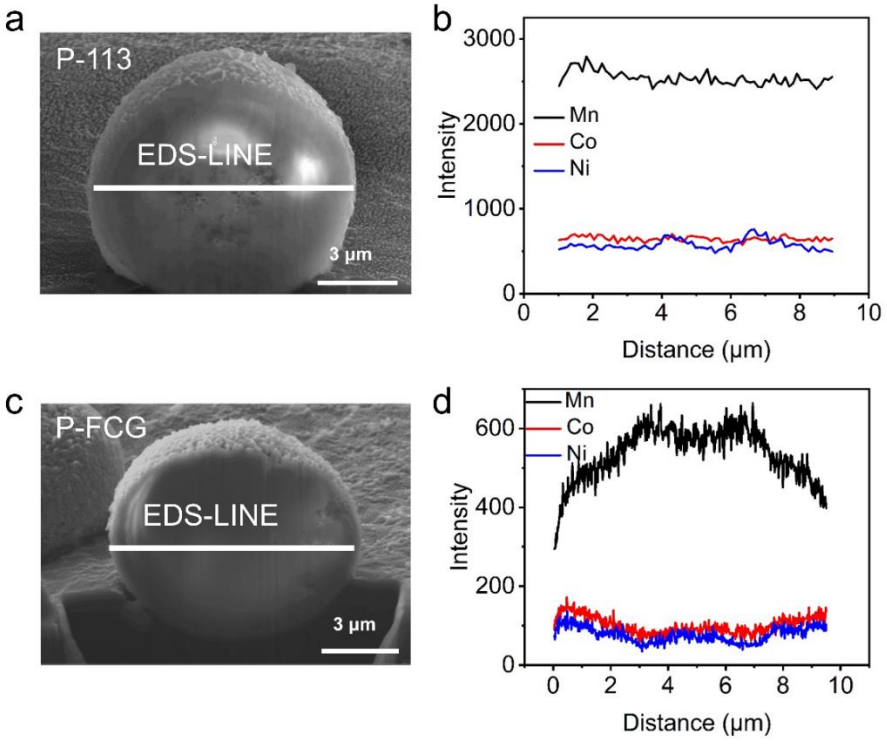
Supplementary Figure 4. Surface EDS spectra of the precursor at different reaction times during the coprecipitation process. The scale bar stands for 10μm.



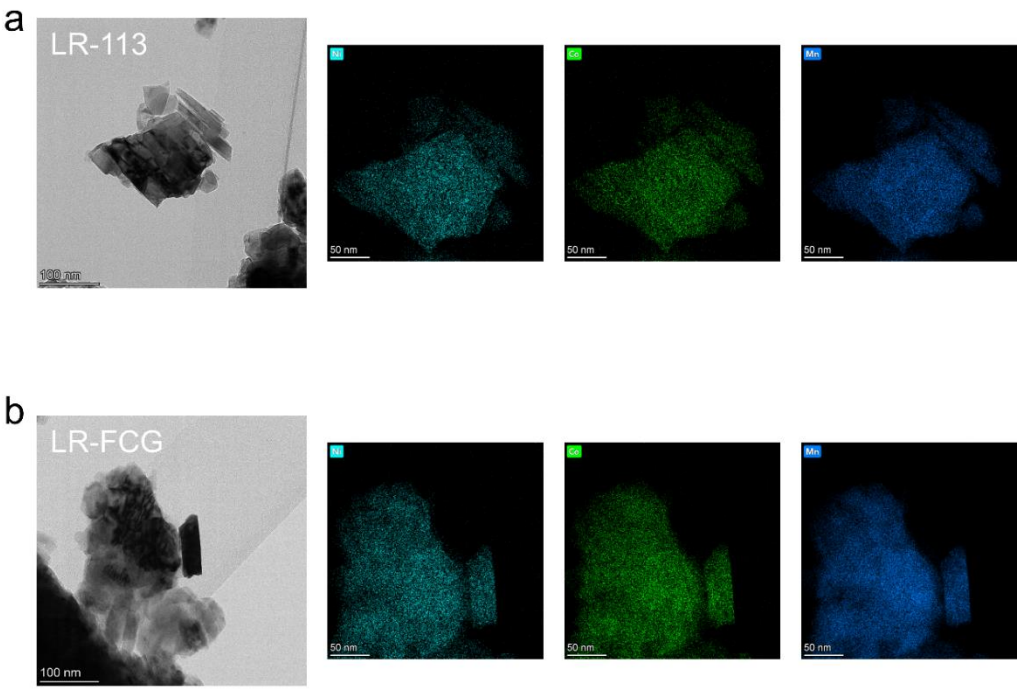
Supplementary Figure 5. (a and b) Low-magnification and corresponding high-magnification SEM images of the homogeneous sample LR-113. (c and d) Low-magnification and corresponding high-magnification SEM images of the full concentration gradient sample LR-FCG.



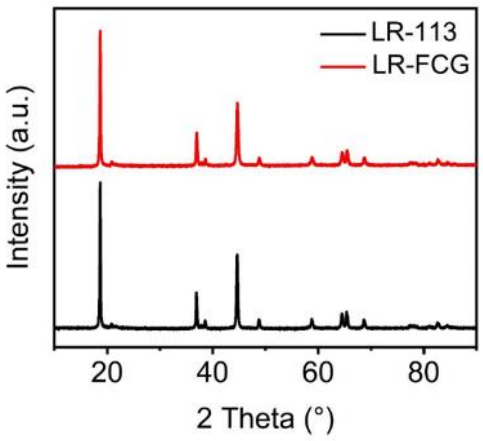
Supplementary Figure 6. (a) N_2 adsorption–desorption isotherms of LR-113. (b) Corresponding pore size distribution of LR-113. (c) N_2 adsorption–desorption isotherms of LR-FCG. (d) Corresponding pore size distribution of LR-FCG.



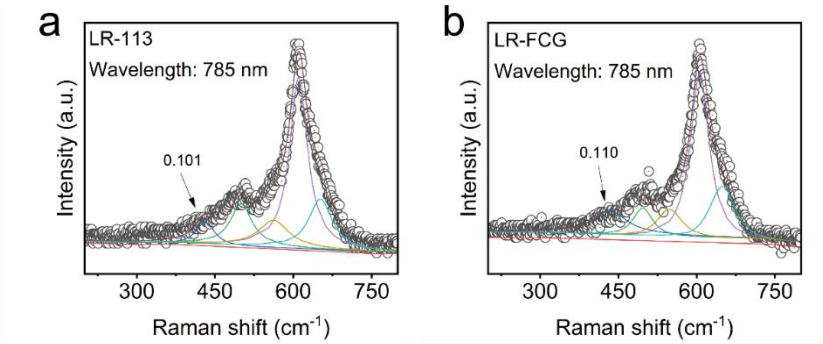
Supplementary Figure 7. (a) Cross-sectional SEM image of the homogeneous precursor P-113. (b) Corresponding EDS line-scan profiles of P-113 across the particle diameter. (c) Cross-sectional SEM image of the full concentration gradient precursor P-FCG. (d) Corresponding EDS line-scan profiles of P-FCG across the particle diameter.



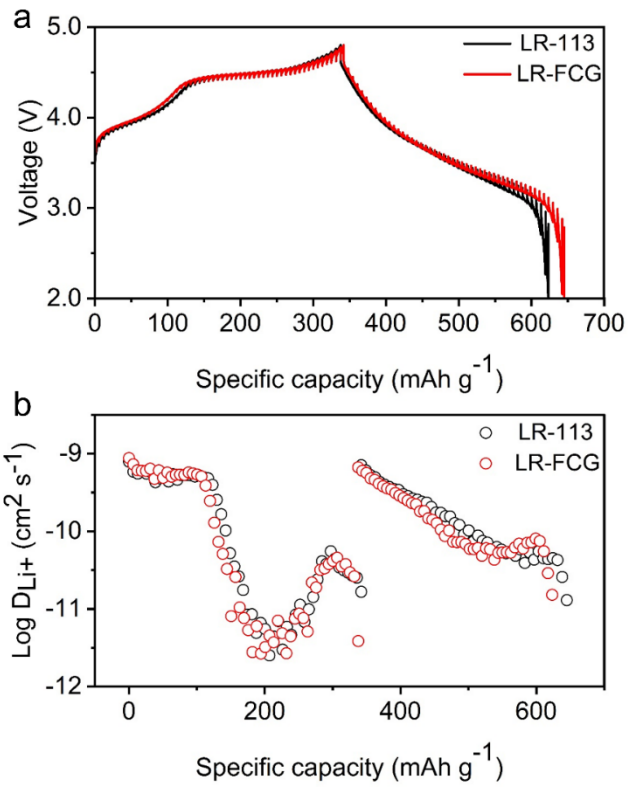
Supplementary Figure 8. (a) TEM image and EDS mapping of the homogeneous sample LR-113. (b) TEM image and EDS mapping of the full concentration gradient sample LR-FCG.



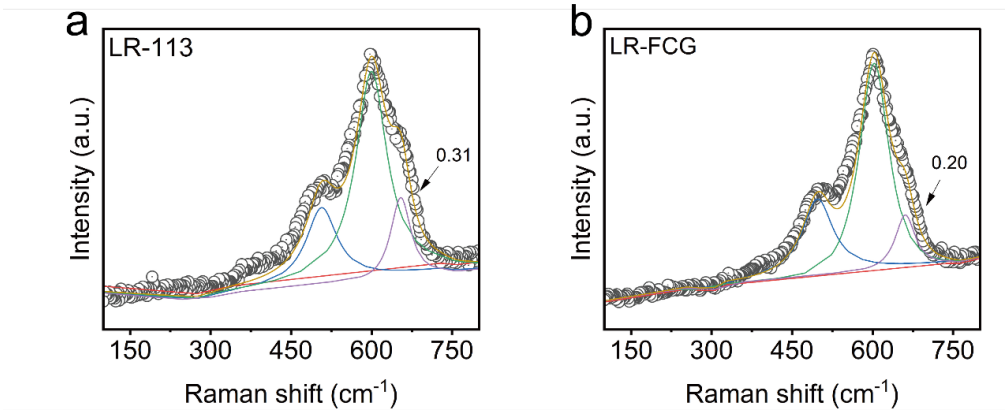
Supplementary Figure 9. XRD patterns of LR-113 and LR-FCG.



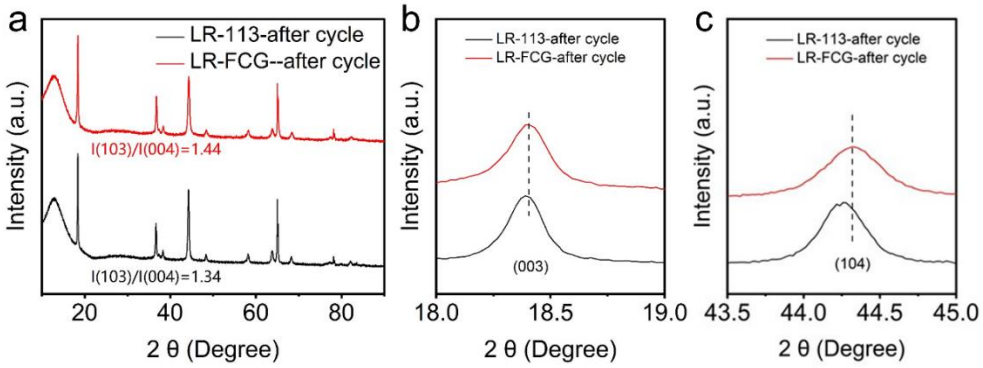
Supplementary Figure 10. Fitted Raman spectra collected using a 785 nm excitation laser: (a) LR-113; (b) LR-FCG.



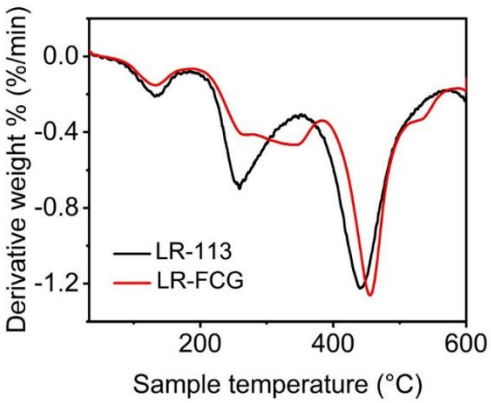
Supplementary Figure 11. (a) GITT curves obtained during the first cycle. (b) Comparison of the calculated Li^+ diffusion coefficients (D_{Li+}) during the charge and discharge processes.



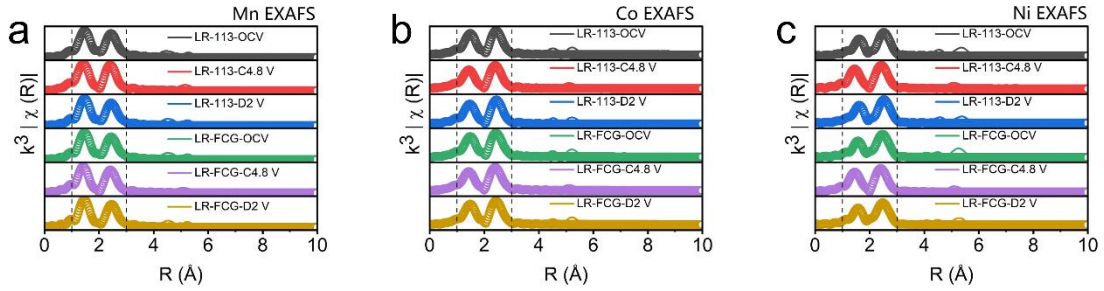
Supplementary Figure 12. Raman spectra of cycled cathode materials: (a) LR-113; (b) LR-FCG.



Supplementary Figure 13. *Ex situ* XRD patterns of cycled LR-113 and LR-FCG cathode materials: (a) full XRD patterns; (b) enlarged view of the (003) reflections; (c) enlarged view of the (104) reflections.



Supplementary Figure 14. DTG profiles of LR-113 and LR-FCG at fully charged state.



Supplementary Figure 15. (a) Mn EXAFS fitting patterns of LR-113 and LR-FCG at OCV, fully charged, and discharged states. (b) Co EXAFS fitting patterns of LR-113 and LR-FCG at OCV, fully charged, and discharged states. (c)Ni EXAFS fitting patterns of LR-113 and LR-FCG at OCV, fully charged, and discharged states.

Supplementary Table 1. ICP-OES of P-113 and P-FCG

Element / Sample	Ni (Atom.%)	Co (Atom.%)	Mn (Atom.%)
P-113	0.1951	0.2068	0.5981
P-FCG	0.1927	0.1992	0.6079

Supplementary Table 2. Refined structural parameters of rhombohedral R-3m phase for LR-113 and LR-FCG

	a[Å]	c[Å]	*V*[Å^3]	Z_{ox}	$S(MO_2)$ [Å]	$I(LiO_2)$ [Å]
LR-113	2.85222	14.24320	100.34697	0.24145	2.61742	2.13030
LR-FCG	2.8469	14.2341	99.9077	0.24170	2.60863	2.13606

Slab thickness: $S(MO_2) = 2[(1/3)-Z_{ox}]c$. Inter-slab space thickness:$I(LiO_z)=(c/3)-S(MO_2)$.