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Lithium Battery Recycling: Overview and a New Direction

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Effective recycling of lithium-ion battery waste can mitigate high-demand element supply chain problems, greenhouse gas emissions, and environmental hazards associated with the waste. For the cost-effectiveness of the recycling process, the metal separation step needs to be as simplified as possible. Lithium, an alkaline metal, differs significantly in terms of chemistry from transition metals such as nickel, manganese, and cobalt; therefore, its separation from cathode waste is straightforward. Separation of nickel, manganese, and cobalt from each other is possible; however, it requires energy and time to recover each element with high purity and yield. This review will illustrate the recent trend in lithium battery recycling and present a new direction explored in my lab at Auburn University.

Key words: Lithium-ion battery, Recycling, Materials chemistry

1. The Necessity of the Lithium-ion Battery Waste Recycling

Lithium-ion batteries (LIBs) are widely used in various electronic devices due to their numerous advantages, including high energy density, long lifespan, low self-discharge rate, and excellent charge-discharge efficiency. As a result, they account for 63% of the global market value in portable batteries [1]. Lithium-ion batteries are now used in electric cars (EVs) and hybrid electric vehicles (HEVs) to minimize petroleum use in transportation, increasing their usage [2,3]. As the demand for LIBs increases, the volume of spent LIBs will also grow. International Energy Agency (IEA) reported that the battery demand is at 1 terrawatt-hour (TWh) in 2023. Even in the most conservative scenario of IEA – Stated Policies Scenario (STEPS) – the battery demand will reach 6 TWh in 2035 and 9 TWh in 2050 [4]. The cathode materials in spent LIBs primarily consist of lithium, manganese, nickel, and cobalt oxides, and the transition metals contained in these materials are significant sources of environmental pollution. Raw material extraction and lithium-ion battery manufacturing processes pose substantial environmental challenges [5]. To mitigate these challenges, in a recent critical review, it was proposed to adopt lithium-ion battery cell design that is compatible with battery end-of-life recycling [6].

Traditionally, the two main benefits of spent LIB recycling have been recognized. The first is CO_2 emission mitigation by bypassing the outfield, long-range mining activities involving cobalt, nickel, manganese for cathodes and lithium for cathodes and electrolytes. By recycling spent LIB at the usage points for further use, we can eliminate transporting multiple elements. The second benefit of recycling is supplying chain resilience. According to International Energy Agency (IEA) [7], in 2030, 68% of mined nickel will be coming from Indonesia while 62% of mined cobalt will be coming from Democratic Republic of Congo. This high dominance of specific countries is not ideal considering possible political and economic conflicts. By eliminating dependence on certain nations and recycling domestically, we can self-control the supply chain.

2. Cost distibution of the Lithium-ion Battery

Cathodes comprise about 50% of the total battery cost because they contain valuable transition metals such as cobalt, nickel, and manganese (Fig. 1) [8]. Traditionally, the primary goal of spent cathode recycling is to recover individual transition metals. The European Parliament's 2027 recovery rate goal for nickel and cobalt is 90%. The goal for 2031 is 95% [9]. We can see that metal recovery technology is already quite mature. However, the problem lies in the

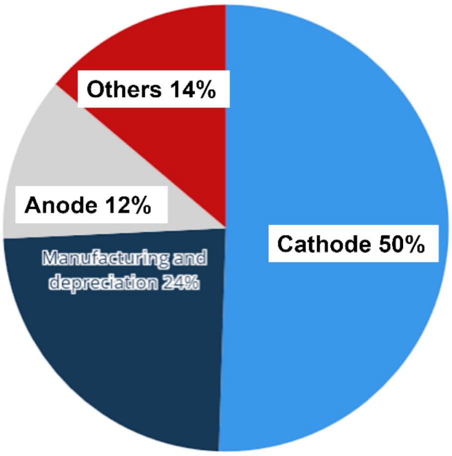

Fig. 1. Distribution of costs of lithium-ion battery cells used in electric vehicles worldwide in 2021, by battery component [8].

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environmental impact of the separation process and its associated costs.

3. Traditional Recycling Strategies

Traditionally, the focus of recycling was on how to extract the pure constituent metal out of the waste material. The market price of the extracted metal determines the feasibility of recycling. In this recycling strategy, the purity of the recovered metal is a critical factor.

3-1. Pyrometallurgy

The oldest recycling route doesn't differ too much from raw ore refining process. The traditional pyrometallurgy processes involve high-temperature treatment of materials, as the word "pyro" means heat. Fig. 2 shows an exemplary flowchart of a pyrometallurgy-based lithium battery recycling process.

3-2. Hydrometallurgy

To avoid the high-temperature processes of the pyrometallurgical approach, researchers have adopted wet-chemical methods, particularly leaching, to separate the metals of interest. For example, circulatory leaching between citric acid (leaching agent) and oxalic acid (precipitation agent) has been proposed to recover cobalt and lithium from $LiCoO_2$ [10]. Concentrated sulfuric acid was used to leach out cobalt, nickel, and manganese from the NMC ($LiNi_xMn_yCo_zO_2$, x+y+z=1) cathode while passivating aluminum foil [11]. As hydrometallurgical techniques improve, researchers sought milder chemicals such as tartaric acid, an effective leaching and precipitating agent [12]. In addition, an ultrasonic-assisted bioleaching process was recently reported for the simultaneous extraction of Co, Mn, Ni, and Li from lithium battery waste [13]. For cobalt recovery from spent $LiCoO_2$ cathode, a ternary deep eutectic solvent composed of choline chloride, ethylene glycol, and benzoic acid was shown to be effective, achieving almost 100% leaching efficiency at 170 °C in 3 h [14].

4. Direct Recycling

The key idea behind direct recycling is simple: retain the cathode crystal structure, which is logical considering the energy and effort required to synthesize the cathode from raw materials. Cathodes account for approximately 50% of the total cost of a lithium-ion battery. When this approach matures, it is expected to save energy and be more economical than previous pyrometallurgy or hydrometallurgy approaches (Fig. 3). This new process needs to be further developed, as its mass-scale applicability and industry-readiness have not been established yet.

4-1. ReCell Center's Efforts in Direct Recycling

US department of Energy's ReCell center conducted multiple projects regarding direct cathode recycling. This center consisted of three universities (UC San Diego, Michigan Technological University, and Worcester Polytechnic Institute) and three national laboratories (NREL, Argonne National Lab, Oak Ridge National Lab). Representative works from each institute are listed below:

4-1-1. Michigan Technological University = Cathode/cathode separation via froth flotation

Froth flotation is a separation technique that exploits hydrophobicity differences of particles. Hydrophobic particles will attach to the injected air bubbles in water and rise to the water surface. In contrast, hydrophilic particles attract water molecules rather than air bubbles to their surface, causing them to sink eventually. Dr. Lei Pan's lab tried froth flotation to separate NMC111 and lithium manganese oxide (LMO) [16]. This cathode/cathode separation is necessary to make the spent cathode waste ready for relithiation (Fig. 4). With a two-stage separation procedure, the authors achieved a 95% grade of NMC111 in the froth product (hydrophobic, rising to the top surface) and a 95% grade of LMO in the tailing product (hydrophilic, sinking to the bottom of the liquid cell). For effective separation, a commercial

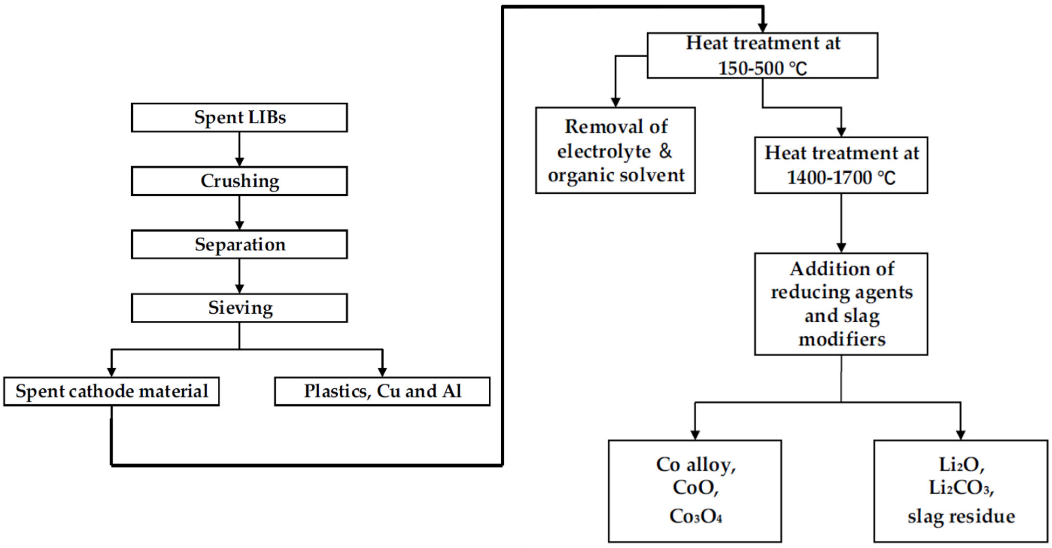

Fig. 2. Pyrometallurgy-based lithium-ion battery recycling flowchart. High process temperature is the main characteristic of the approach.

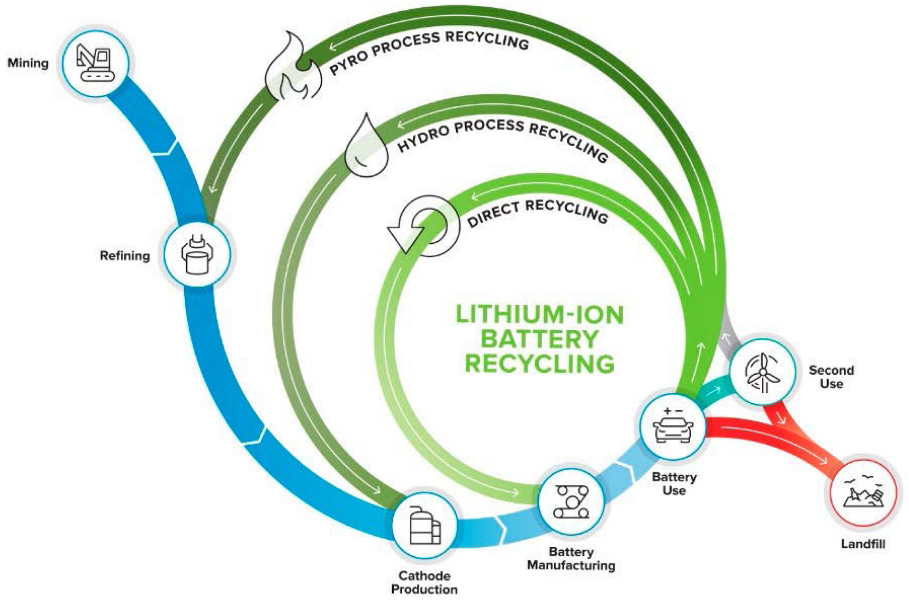

Fig. 3. The three different recycling approaches. Notice that the size of the circle decreases as newer technologies emerge. This figure is adopted from a paper that overviews ReCell center research activities [15].

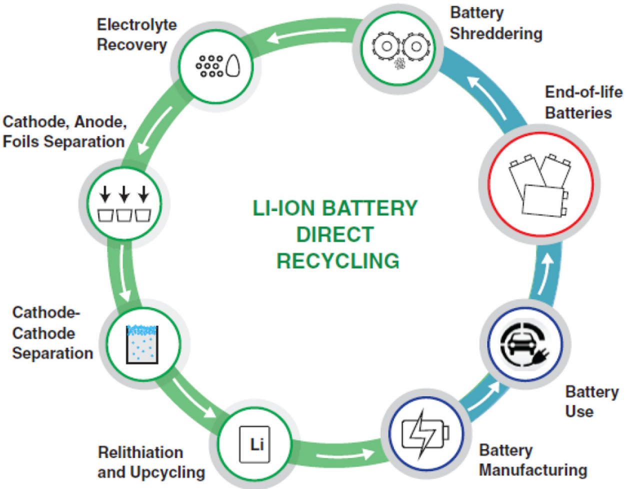

Fig. 4. A schematic for a direct recycling process [16].

collector, Atrac 922 (Nouryon), was used to make NMC111 hydrophobic, and methyl isobutyl carbinol was used to stabilize the froth. In addition to cathode/cathode separation, the authors demonstrated that the separation process caused no significant compositional change, resulting in only a negligible change in the electrochemical performance of the recovered cathode.

4-1-2. UC San Diego – Hydrothermal relithiation

Dr. Zheng Chen's lab took hydrothermal route to relithiate degraded cathode materials [17]. The degraded cathode lost lithium content after long-term use as a battery electrode, meaning that lithium vacancies are present within the crystal structure. Relithiation is a process that fills the lithium vacancies while maintaining the crystal structure intact. The hydrothermal process took place at as low as 100 °C with the help of a reducing agent such as ethylene glycol. Complete recovery

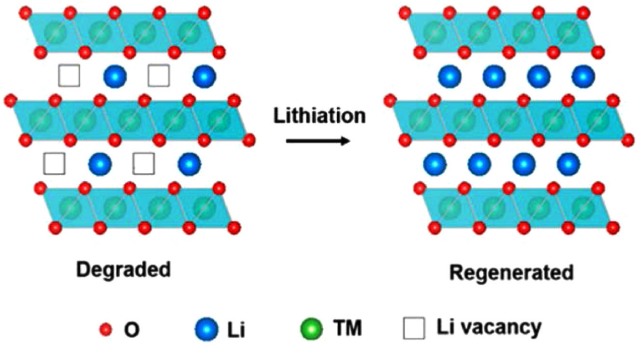

Fig. 5. Relithiation of degraded cathode [15]. The host lattice remains intact during lithium insertion. Layered structure is a common characteristic of many cathode materials.

of composition, crystal structure, and electrochemical performance was achieved for NMC622 and NMC111 cathodes. The authors reported 8% reduction in energy consumption when the hydrothermal reaction temperature was 100 °C rather than 220 °C.

4-1-3. Worcester Polytechnic Institute – Role of impurities in recycled cathodes

Dr. Yan Wang's lab conducted a systematic investigation into the impact of impurities on the cathode, as impurities inevitably emerge during the recycling of spent lithium-ion batteries. Especially, aluminum and copper will be highly relevant since they are used in the form of foil to collect currents from cathode and anode, respectively. The authors did not test the recycled cathode. To achieve precise control over aluminum impurity concentration, they had to fabricate NMC622 cathode particles freshly from raw precursors. $Al_2(SO_4)_3$ was the precursor for Al impurity. The authors found 0.2 at% Al impurity improved electrochemical performance of the cathode. At 5 at%,

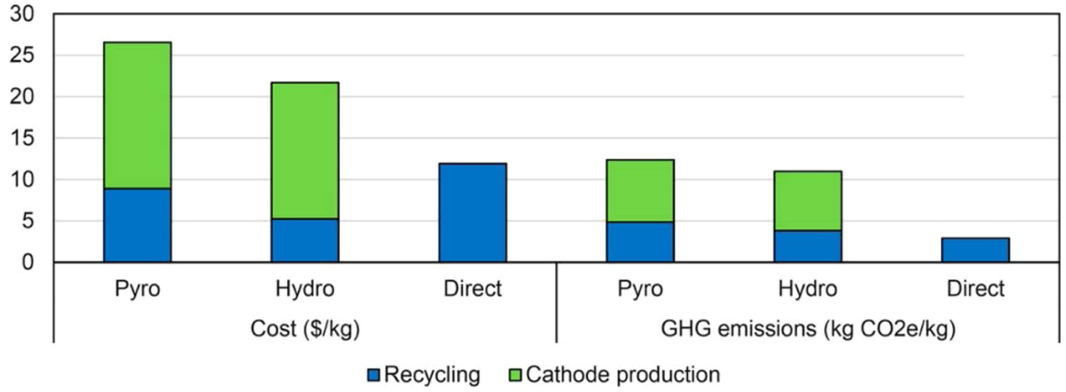

Fig. 6. Cost ($) and GHG emissions (kg CO2e) for 1 kg of recycled NMC111 [22]. Pyro- and hydrometallurgical approach have added cost (green bar) due to remaking of the cathode material from the recovered elements. The "Direct" sample was relithiated via the DTBQ redox mediator approach, eliminating the need for cathode remaking.

however, Al induced new phase formation at the cathode, resulting in a lower specific capacity and faster capacity fade [18]. For copper impurities, the same research team found that metallic copper impurities can easily short the battery, while copper ion impurities located at the nickel sites of the NMC622 unit cell enhanced the initial specific capacity [19].

4-1-4. Oak Ridge National Laboratory (ORNL) – Ionothermal relithiation

ORNL's Sheng Dai led the ionothermal relithiation project. Instead of typical LiOH aqueous solution, imidazolium ionic liquid was used as solvent with LiCl as lithium source [20]. Despite unique properties of ionic liquid, its cost is always an issue, so the ORNL team developed a solvent recycling scheme.

4-1-5. Argonne National Laboratory (ANL) – Solid state cathode upcycling

This ANL project's goal is to reuse spent low-Ni cathode after converting it to newer high-Ni composition cathode. For example, upcycling of recycled NMC622 to NMC811 was attempted by coating the surface of NMC622 with $Ni(OH)_2$-LiOH mixture layer and heating up the composite to 900 °C (2.5 h dwell time) [21]. The in situ X-ray diffraction and ex aitu transmission X-ray microscopy confirmed two phases, NMC and $LiNiO_2$, rather than single NMC phase with high nickel content.

4-1-6. National Renewable Energy Laboratory (NREL) – Redox mediator relithiation

NREL's Jaclyn Coyle led their efforts for direct recycling of lithium-ion batteries. They proposed a redox mediator, 3,5-di-tert-butyl-o-benzoquinone (DTBQ), to facilitate relithiation of spent NMC111 [22]. DTBQ molecules in the dimethoxyethane (DME) electrolyte act like lithium carriers that quickly release lithium to NMC cathode surface. The authors also demonstrated excellent electrochemical performance of the relithiated NMC111. Fig. 6 shows that the proposed relithiation process itself has higher cost than pyrometallurgical or hydrometallurgical process; however, the overall cost is lower for the direct recycling approach when the cost of remaking cathode is included for the traditional routes.

5. Repurposing

Dr. Tae-Sik Oh's lab at Auburn University is currently working on repurposing the spent NMC cathode as a dry methane reforming catalyst after selective lithium removal by oxalic acid leaching. Any chemical conversion that can be catalyzed by nickel and cobalt would be of potential interest for repurposing cathode waste. We will soon report the promising catalyst behavior.

6. Summary

In this review, the traditional pyrometallurgical and hydrometallurgical techniques for recycling lithium-ion batteries are briefly introduced. Recent developments in battery recycling have emerged from the concept of direct recycling, where researchers preserve the cathode crystal structure throughout the recycling process. Selected projects from US Department of Energy's ReCell center were introduced to familiarize the readers with direct recycling processes and challenges. The direct recycling strategy still needs further development to achieve its full potential. Repurposing of spent cathode material could be another way to handle lithium-ion battery waste.

References

1. Tarascon, J.-M. and Armand, M., "Issues and Challenges Facing Rechargeable Lithium Batteries," *Nature* **414**, 359-367(2001).
2. Goodenough, J. B. and Kim, Y., "Challenges for Rechargeable Li Batteries," *Chem Mater* **22**, 587-603(2010).
3. Armand, M. and Tarascon, J.-M., "Building Better Batteries," *Nature* **451**, 652-657(2008).
4. Recycling of Critical Minerals: Strategies to Scale Up Recycling and Urban Mining, International Energy Agency (2024).
5. Llamas-Orozco, J. A., Meng, F., Walker, G. S., Abdul-Manan, A.

F. N., MacLean, H. L., Posen, I. D. and McKechnie, J., “Estimating the Environmental Impacts of Global Lithium-ion Battery Supply Chain: A Temporal, Geographical, and Technological Perspective,” *PNAS Nexus*, **2**, 361(2023).

6. Thompson, D. L., Hartley, J. M., Lambert, S. M., Shiref, M., Harper, G. D. J., Kendrick, E., Anderson, P., Ryder, K. S., Gaines, L. and Abbott, A. P., “The Importance of Design in Lithium Ion Battery Recycling – a Critical Review,” *Green Chem*., **22**, 7585-7603(2020).
7. Global Critical Minerals Outlook 2025, International Energy Agency.
8. EV Outlook 2021, Bloomberg New Energy Finance.
9. www.europarl.europa.eu/pdfs/news/expert/2023/6/press_release/20230609IPR96210/20230609IPR96210_en.pdf Accessed on 07.25.2025.
10. Fan, B., Chen, X., Zhou, T., Zhang, J. and Xu, B. A., “Sustainable Process for the Recovery of Valuable Metals from Spent Lithium-Ion Batteries,” *Waste Management & Research* **34**, 474-481 (2016).
11. Fan, X., Song, C., Lu, X., Shi, Y., Yang, S., Zheng, F., Huang, Y., Liu, K., Wang, H. and Li, Q., “Separation and Recovery of Valuable Metals From Spent Lithium-ion Batteries Via Concentrated Sulfuric Acid Leaching and Regeneration of $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$,” *Journal of Alloys and Compounds* **863**, 158775(2021).
12. Chen, X., Kang, D., Cao, L., Li, J., Zhou, T. and Ma, H., “Separation and Recovery of Valuable Metals from Spent Lithium Ion Batteries: Simultaneous Recovery of Li and Co in a Single Step,” *Separation and Purification Technologies* **210**, 690-697 (2019).
13. Nazerian, M., Bahaloo-Horeh, N. and Mousavi, S. M., “Enhanced Bioleaching of Valuable Metals from Spent Lithium-Ion Batteries Using Ultrasonic Treatment,” *Korean J. Chem. Eng*., **40**(3), 584-593(2023).
14. Li, X., Li, Y., Qiao, Q., Wang, K. and Yu, H., “Recovery of Cobalt from Cathode of Lithium-Ion Battery Using Ternary Deep Eutectic Solvent,” *Korean J. Chem. Eng*., **41**(12), 3151-3161(2024).
15. Gaines, L.; Dai, Q.; Vaughey, J. T. and Gillard, S., “Direct Recycling R&D at the ReCell Center,” *Recycling* **6**, 31(2021).
16. Folayan, T.-O., Lipson, A. L., Durham, J. L., Pinegar, H., Liu, D. and Pan, L., “Direct Recycling of Blended Cathode Materials by Froth Flotation,” *Energy Technology* **9**, 2100468(2021).
17. Yu, X., Yu, S., Yang, Z., Gao, H., Xu, P., Cai, G., Rose, S., Brooks, C., Liu, P. and Chen, Z., “Achieving Low-Temperature Hydrothermal Relithiation by Redox Mediation for Direct Recycling of Spent Lithium-Ion Battery Cathodes,” *Energy Storage Materials* **51**, 54-62(2022).
18. Zhang, R., Zheng, Y., Yao, Z., Vanaphuti, P., Ma, X., Bong, S., Chen, M., Liu, Y., Cheng, F., Yang, Z. and Wang, Y., “Systematic Study of Al Impurity for NCM622 Cathode Materials,” *ACS Sustainable Chemistry and Engineering* **8**, 9875-9884(2020).
19. Zhang, R., Meng, Z., Ma, X., Chen, M., Chen, B., Zheng, Y., Yao, Z., Vanaphuti, P., Bong, S., Yang, Z. and Wang, Y., “Understanding Fundamental Effects of Cu Impurity in Different Forms for Recovered $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ Cathode Materials,” *Nano Energy* **78**, 105214(2020).
20. Wang, T., Luo, H., Bai, Y., Li, J., Belharouak, I. and Dai, S., “Direct Recycling of Spent NCM Cathodes through Ionothermal Lithiation,” *Advanced Energy Materials* **10**, 2001204(2020).
21. Nisbet, M. L., Luong, D., Allen, E., Park, S., Kinnibrugh, T. L., Stubbs, J. E., Eng, P. J., Vaughey, J. T. and Fister, T. T., “In Situ Diffraction and Ex Situ Transmission X-Ray Microscopy Studies of Solid-State Upcycling for NMC Cathodes,” *Advanced Energy Materials* 2500698(2025).
22. Park. K., Yu, J., Dai, Q., Frisco, S., Zhou, M. and Burrell, A., “Direct Cathode Recycling of End-Of-Life Li-Ion Batteries Enabled by Redox Mediation,” *ACS Sustainable Chemistry and Engineering* **9**, 8214-8221(2021).

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