



Energy materials for energy conversion and storage: focus on research conducted in Korea

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Received: 30 July 2021 / Revised: 13 September 2021 / Accepted: 28 September 2021 / Published online: 25 October 2021
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Abstract

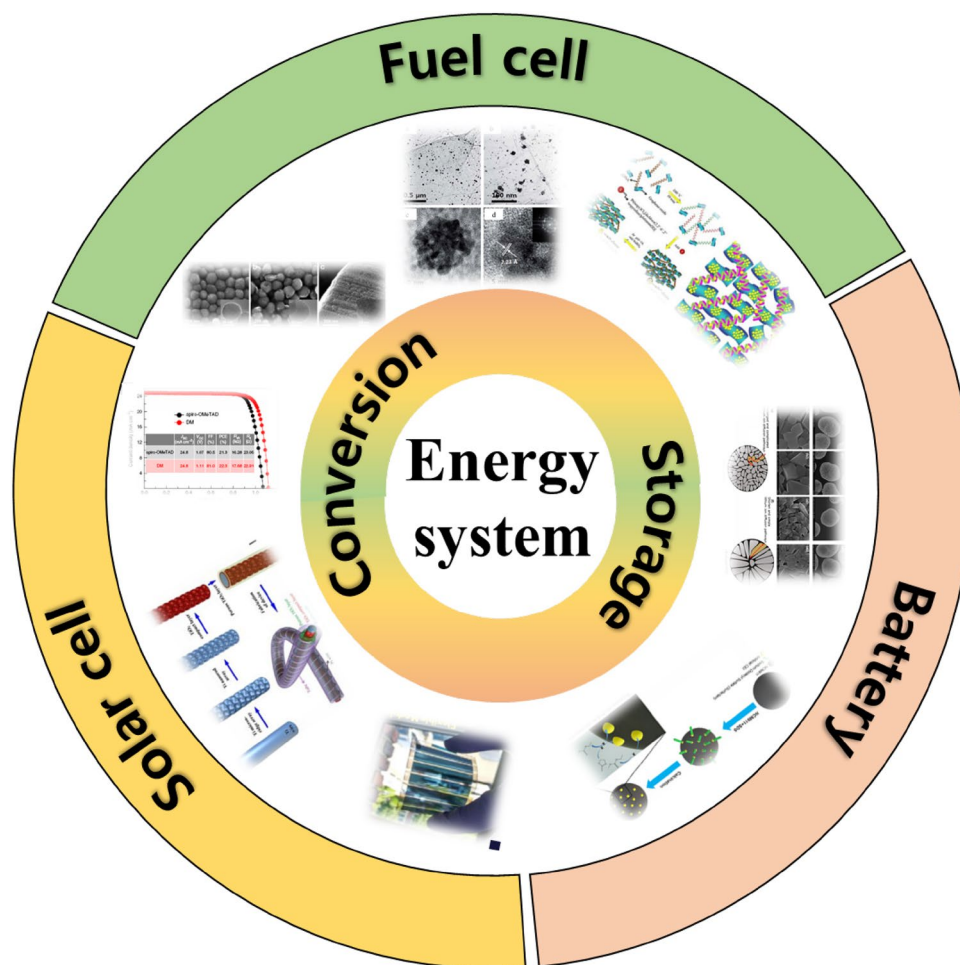
Fossil fuels are widely used around the world, resulting in adverse effects on global temperatures. Hence, there is a growing movement worldwide towards the introduction and use of green energy, i.e., energy produced without emitting pollutants. Korea has a high dependence on fossil fuels and is thus investigating various energy production and storage technologies for producing and using green energy. Renewable energy technologies are essential for producing green energy, and energy storage technologies are necessary for its effective use. In Korea, the renewable energy technologies of most interest are solar power generation and fuel cells, followed by energy storage, transportation. This review intends to provide information about the energy materials currently being researched to develop these energy technologies.

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Graphical abstract



Keywords Energy materials · Solar cell · Fuel cell · Lithium-ion battery

1 Introduction

As industrialization has proceeded over the past century, the dependence of all countries on fossil fuels, as their primary energy source, has increased. The continued abuse of fossil fuels in industrial systems has resulted in the emission of immense amounts of pollutants and carbon, which have triggered unprecedented global climate change [1–7]. Because of this, most countries are attempting to prevent environmental pollution as per international conventions, such as the Paris agreement (2016). The aim of the Paris agreement is to reduce fossil fuel usage by moving towards alternative energy technologies, such as solar, hydrogen, and wind energy, and by developing a sustainable energy production system [2, 5]. Korean industries consumed 280 million tons of fossil in 2018, ranking eighth in the world. Although Korean industries depend on imported energy

sources (primarily because fossil fuel deposits are rare in Korea), energy consumption is increasing each year as the industries continue to develop [3]. Such an increase in energy consumption does not correspond to the global trend of energy consumption, and the energy paradigm of Korean industries thus needs to change. Developed countries are operating large-scale programs to develop high-efficiency energy materials and change their national energy sources, and are providing full support for the development of energy materials and systematic research plans [8, 9]. Thus, Korea established the National Technology Road Map to emphasize the importance of energy materials and research on energy materials and systems [10]. The development of new energy materials has overcome the limitations of current energy technology, leading to advancements in the energy industry and the development of high-efficiency and high-performance, energy transport, storage, and savings

techniques. The new materials and components can be used in alternative energy devices to curb the use of fossil fuels, which are the predominant cause of environmental pollution [11]. Energy material technology is essential for the development of a comprehensive multidisciplinary energy system that involves new energy systems with high efficiency, high cleanliness, and future prospects [12].

As energy material technology is crucial for securing future energy sources, cooperation among countries is essential. Efforts are being made in Korea to continue research and development on various energy materials [13–16]. Energy materials have been actively researched and developed worldwide since the 2000s; therefore, the related technologies are extensive and diverse [17–19]. Among these, the areas of technology with the most promising active research and market growth trends are shown in Fig. 1. The introduction of solar cells and hydrogen fuel cells for eco-friendly energy production may be

accompanied by growth in materials technology through periodic investments, and the use of lithium-ion batteries (LIBs) for energy storage is also expected to increase [20–22].

Korea is paying considerable attention to energy-related topics for the following reasons [23–27]:

- (1) The solar power generation capacity is determined by the intensity and duration of sunlight. In 2017, the average annual sunlight duration in Korea was 2,557.3 h, indicating the possibility to generate solar power for an average of 7 h per day.
- (2) Because Korea has more daylight hours than many major countries (such as Germany), if the efficiency of energy generation is increased through technological development, solar power generation can become self-sufficient.

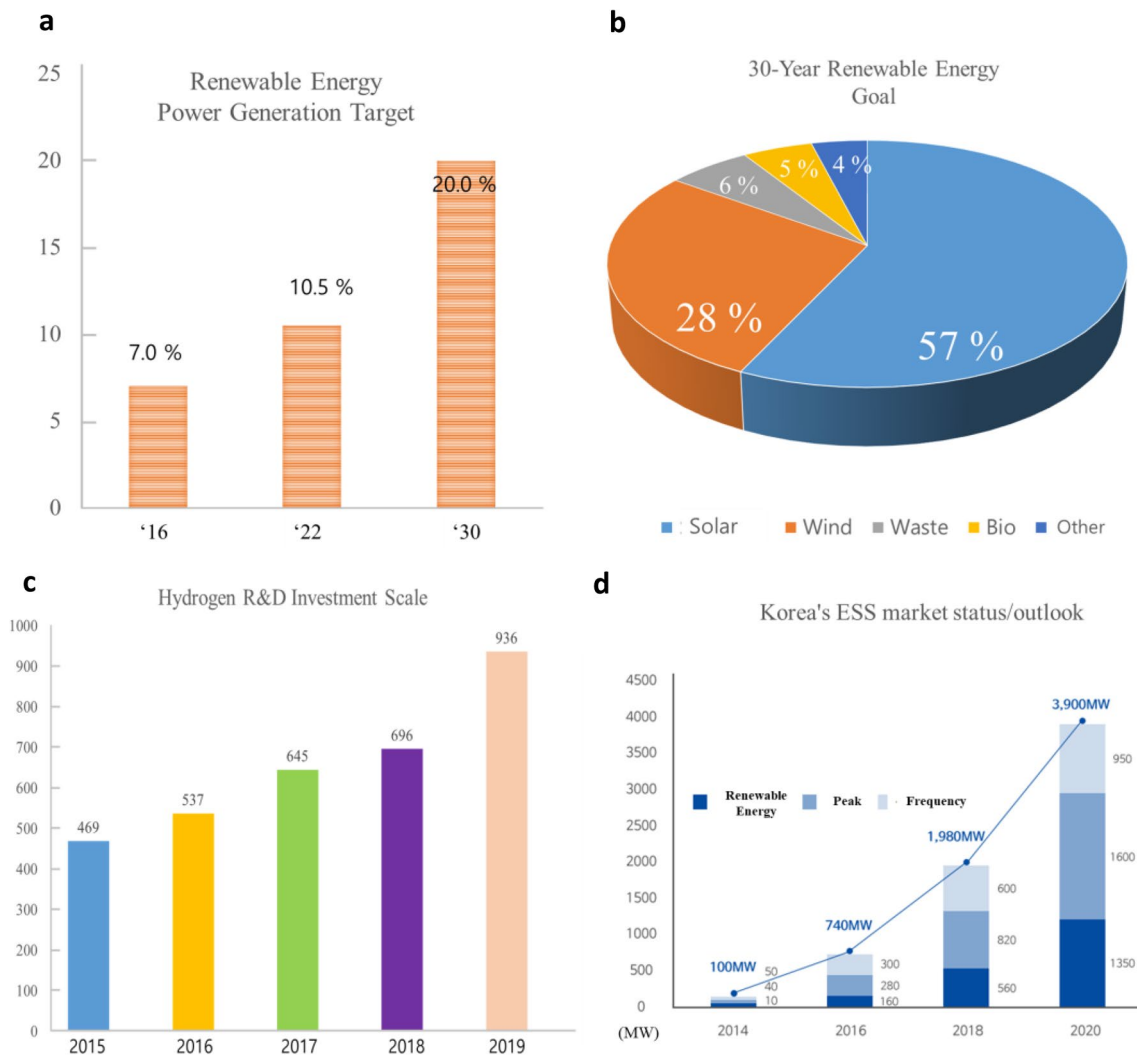


Fig. 1 Korea's energy market expansion plan and ESS market growth trend

- (3) Hydrogen can be used to produce a large amount of energy, and it is noise-free, pollution-free, and accessible through virtually infinite energy sources.
- (4) It is convenient to obtain electrical and chemical energy from fuel cells that use hydrogen as fuel; high energy efficiency can be achieved because chemical energy is directly converted into electrical energy.
- (5) Lithium batteries can be used to store energy produced through renewable sources for access at times of low production.
- (6) Stored energy can be made available to various locations because of its ease of transport.

Based on the above factors, the Korean government has increased the capacity of solar power facilities by 47% annually since 2012 (0.69 GW), with a particularly large increase in 2018. In addition, the government announced the specific goal of generating 15 GW, including 7 GW for export by 2022 and 8 GW for domestic consumption by 2040 (equivalent to approximately 18 large coal generators), by localizing 100% of the core parts of fuel cells [21–24]. Lithium secondary batteries have a higher energy density than conventional batteries; therefore, they are attracting progressively more attention as energy storage devices (ESSs). As renewable energy production increases, ESSs will become essential for stable energy storage and supply [25–27].

Various solutions are required to develop solar cells, hydrogen fuel cells, and secondary batteries suitable for application in Korea. Korea receives substantial daylight; however, due to its weather and geographical factors, it is necessary to study materials that can efficiently produce energy, even with low light. Hydrogen fuel cells commonly use platinum-based catalysts, which are expensive. Such systems are difficult to commercialize because the catalysts account for the majority of fuel cell production costs. Lithium secondary batteries exhibit high energy efficiency, but it is still necessary to develop better high-capacity materials for use as ESSs [7, 28].

The current research focus is on solving the aforementioned problems by employing high-efficiency solar cells, cheaper catalysts, high-capacity batteries, etc. In this paper, we describe research on solar cells, hydrogen fuel cells, and secondary batteries, which are currently among the most notable aspects of energy materials research in Korea.

2 Solar cells

Solar cells are a renewable energy technology through which electrical energy is generated by the photoelectric effect of a semiconductor made of a material such as a semiconductor dye or polymer [29, 30]. The principle of a solar cell is a structure in which an n-type semiconductor and

a p-type semiconductor with different electrical properties are bonded. When light is irradiated, electrons move toward the n-type semiconductor and holes move toward the p-type semiconductor, resulting in a potential difference. A current flows when a load, such as a light bulb, is connected [31, 32].

First-generation silicon solar cells have a narrow light absorption band such that the theoretical conversion efficiency does not exceed 30% in the case of a single-junction structure; therefore, there is currently a limit imposed on the materials and cost reduction, and the research and development of next-generation solar cells is necessary [33]. Halide perovskites have attracted considerable attention in the field of optoelectronics as next-generation solar cell materials because of their unique properties. Among these, lead-free halide perovskites are being studied because of their non-toxicity [34, 35]. Alternatively, research on oxide semiconductors, such as ZnO, has been actively conducted. However, ZnO has a unique oxygen vacancy defect that results in the production of p-type ZnO with high carrier concentrations. To solve this problem, an experiment was conducted in which the oxygen vacancy concentration (used as an oxygen source) was controlled during the production of ZnO from H₂O₂ via atomic layer deposition [28].

Among the solar cell materials currently being studied in Korea, materials exhibiting high effectiveness have been reported by Jeon et al. [36]. Their research team has achieved remarkable device performance improvements over a relatively short time through the development of various innovative technologies to realize high-quality perovskite thin films for producing next-generation solar cells. For commercialization, studies on long-term stability and large-area devices, in addition to studies on high efficiency, are essential. The existing spiro-OMeTAD is known to inhibit the stability of the device due to its low glass transition temperature (*T_g*) and the introduction of hygroscopic additives (Li-TFSI, tBP). To overcome this problem, a method has been developed to secure all three elements by chemically modifying the spiro-OMeTAD, which is the most thermally vulnerable component among the perovskite solar cell components. The end of the spiro-OMeTAD structure was designed and synthesized by introducing fluorene with high structural planarity and weak electron donation from methoxyphenyl. As shown in Fig. 2, fluorene-terminated HTM (*N*2,*N*2',*N*7,*N*7'-tetrakis(9,9-dimethyl-9H-fluorene-2-yl)-*N*2,*N*2',*N*7,*N*7'-tetrakis(4-methoxyphenyl)-9,9'-spirobi[fluorene]-2,2',7,7'-tetraamine) (DM), which has a relatively weak electron-donating ability, exhibits a higher occupied molecular orbital energy level than spiro-OMeTAD. This can be expected to reduce the energy loss due to the reduced energy offset of the valence band maximum of the perovskite layer. DM displayed a *T_g* that is 40 °C higher than that of spiro-OMeTAD, thus improving

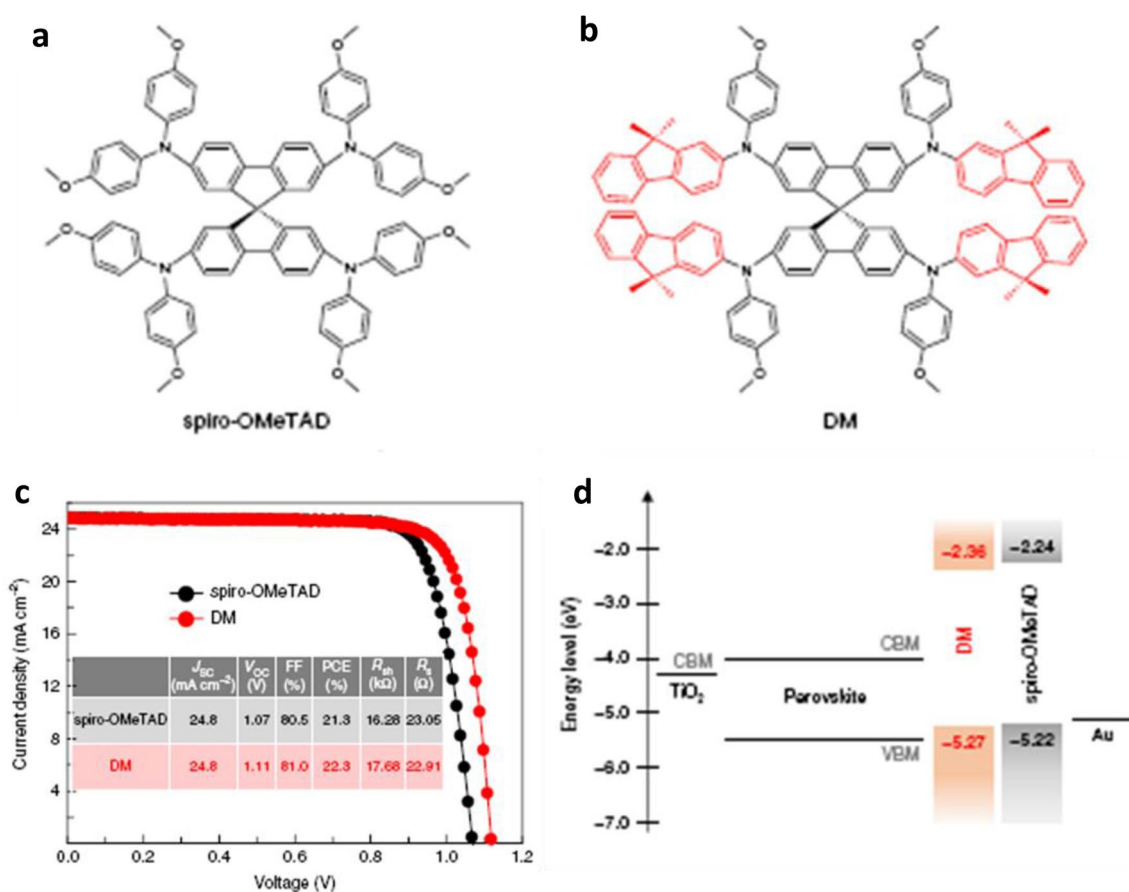


Fig. 2 Chemical structures of **a** spiro-OMeTAD and **b** DM. **c** J–V curves of a solar cell with spiro-OMeTAD and DM perovskite. **d** Energy level diagram of DM and spiro-OMeTAD. CBM and VBM represent the conduction band minimum and valence band maximum, respectively [36]

the thermal stability of the material. DM-based solar cells possess higher thermal stability at temperatures of 60–80 °C than solar cells with spiro-OMeTAD. As a result of confirming the long-term stability of the DM-based solar cell, the efficiency (21.2%) of the solar cell stored at 60 °C for 500 h demonstrated excellent thermal stability, with 95% efficiency retention compared to that of the initial solar cell (22.3%) [37].

Solar cells have problems in absorbing light from all areas and in converting it into electric energy in addition to their difficulty in converting weak incoming sunlight under cloudy conditions. To overcome these weaknesses, Lim et al. [38] developed a high-efficiency solar cell technology that enables normal operations on cloudy and rainy days or using indoor low-light sources using quantum cluster particles. The research team mixed gold quantum cluster particles with controlled atomic numbers inside an organic solar cell to fabricate a flexible and high-transmittance organic solar cell. The clusters used in the high-efficiency translucent solar cells could operate normally on cloudy or rainy days and improved the performance of the light-absorbing layer by absorbing excess light that cannot be absorbed by the basic

light-absorbing layer. In particular, quantum clusters are less affected by deficiencies in the light absorption environment, such as low illuminance and low altitude, and exhibit high quantum efficiency; thus, they effectively compensate for the light absorption characteristics of the basic light-absorbing layer. Therefore, energy conversion efficiency can be maintained even on cloudy or rainy days, as shown in Fig. 3 [38]. In addition, the structures of dye-sensitized solar cells have relatively less stringent requirements than those of silicon solar cells. Liu et al. [37] developed a dye-sensitized solar cell in the form of a yarn with excellent performance and free deformation by changing the surface characteristics of the Ti wire through a chemical reaction. A dye-sensitized solar cell that converts sunlight into electricity using organic dyes can be used as a window-type solar cell, which is usually manufactured on a flat surface and can be attached to buildings or windows. However, because of a lack of elasticity or flexibility, its use as a wearable solar cell is limited. To address this issue, researchers minimized the photoelectron transport distance between the titanium oxide layers and increased the contact area by electrochemically oxidizing the surface of a solar cell electrode made of Ti wire with a

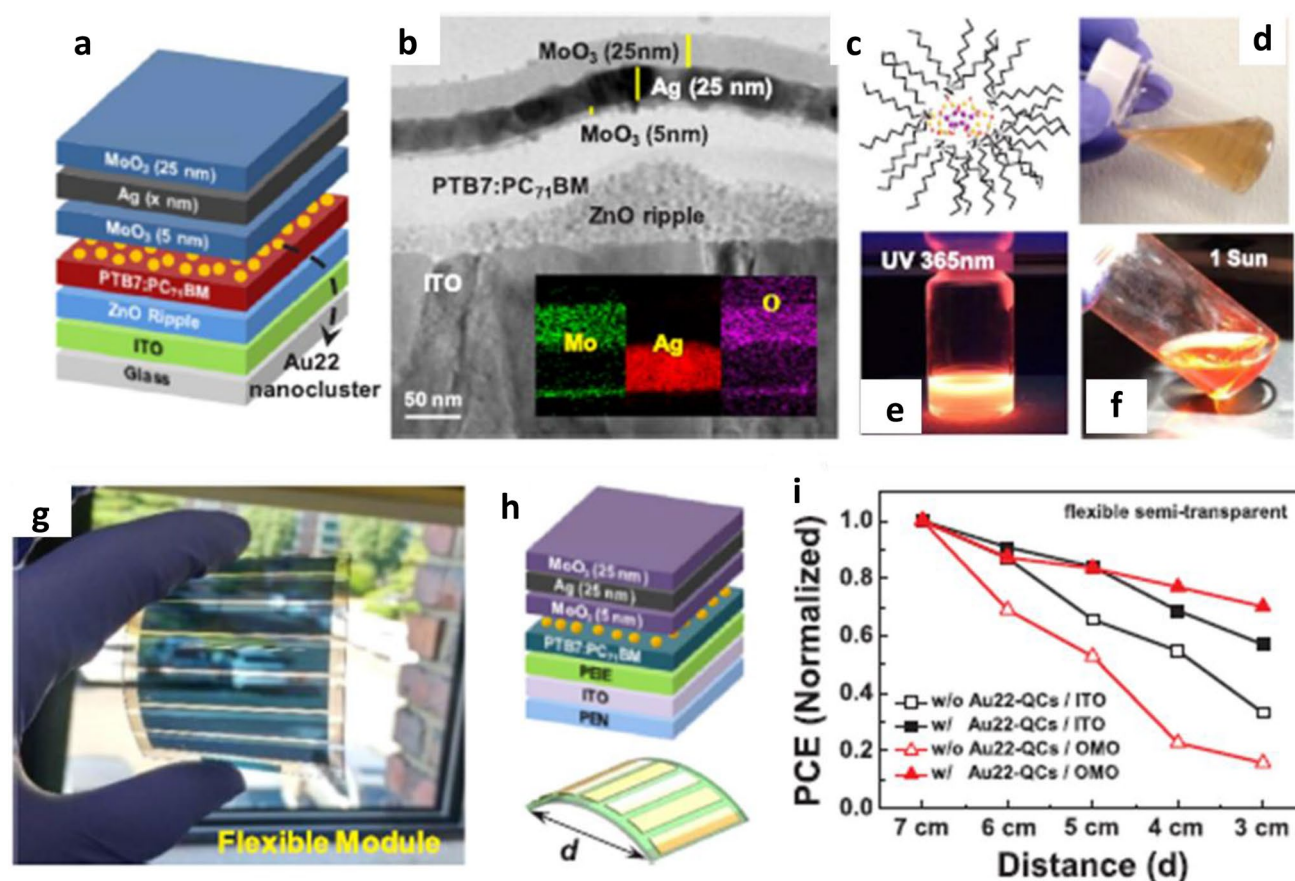


Fig. 3 Confirmation of the effect of Au22-QC on translucent solar cells. **a** Schematic diagram of the device structure using Au22-QC. **b** Cross-sectional TEM image of a translucent solar cell using OMO-25. Inserts show TEM-based elemental analysis for OMO stacking: Mo (green), Ag (red), and O (violet). **c** A schematic diagram of Au22-QC protected in a single layer of tetraoctylammonium. The core of the Au22-QC was obtained from Au22 (SG)18, where SG represents glutathione. **d** Au22-QC solution dispersed in CB. **e** Con-

firmation of photoluminescence emission of Au22-QC under 365 nm UV irradiation. **f** Confirmation of PL of Au22-QC under simulated solar irradiation. **g** Flexible mini-module fabricated on PEN substrate. **h** Schematic diagram of the structure of the flexible device. The PEIE ITO work function modifier was introduced instead of an electron transport layer based on oxide materials to improve flexibility. **i** PCE change according to the bending distance of the flexible module [38]

diameter of 250 μm using a sulfuric acid solution to increase the charge collection. Figure 4 shows the reaction principle and results of the fabricated wire-type dye-sensitized solar cells. Light incident on the titanium oxide layer was collected and scattered to improve efficiency. The fabricated dye-sensitized solar cell, in the form of a fiber, exhibited a photoelectric conversion efficiency of 8.128% and a photoelectric collection efficiency of 93.1%, which is far superior to the efficiencies of conventional nanostructure-based dye-sensitized solar cells. In addition, it was confirmed that this dye-sensitized solar cell with a yarn form has sufficient mechanically safe flexibility to maintain a solar cell performance of 95% or more, even when rolled to a diameter of 1 cm [37].

Solar cells have developed as a technology capable of producing eco-friendly energy, and the technology is being actively researched to increase its efficiency using perovskite

materials rather than silicon solar cells. Following the Renewable Energy 2020 Implementation Plan, Korea is currently focusing on research to advance the next generation of solar cell technology [39–43].

3 Fuel cells

A fuel cell is a device that supplies hydrogen as fuel and directly converts and generates electricity through an electrochemical reaction with oxygen in the atmosphere. Because there is no combustion process, the power generation efficiency is relatively high and there are no emissions of greenhouse gases, such as carbon dioxide. Hence, it is eco-friendly and has the advantage of simultaneously generating heat and electricity. In addition, it is a distributed power supply that can be installed on a small scale, which

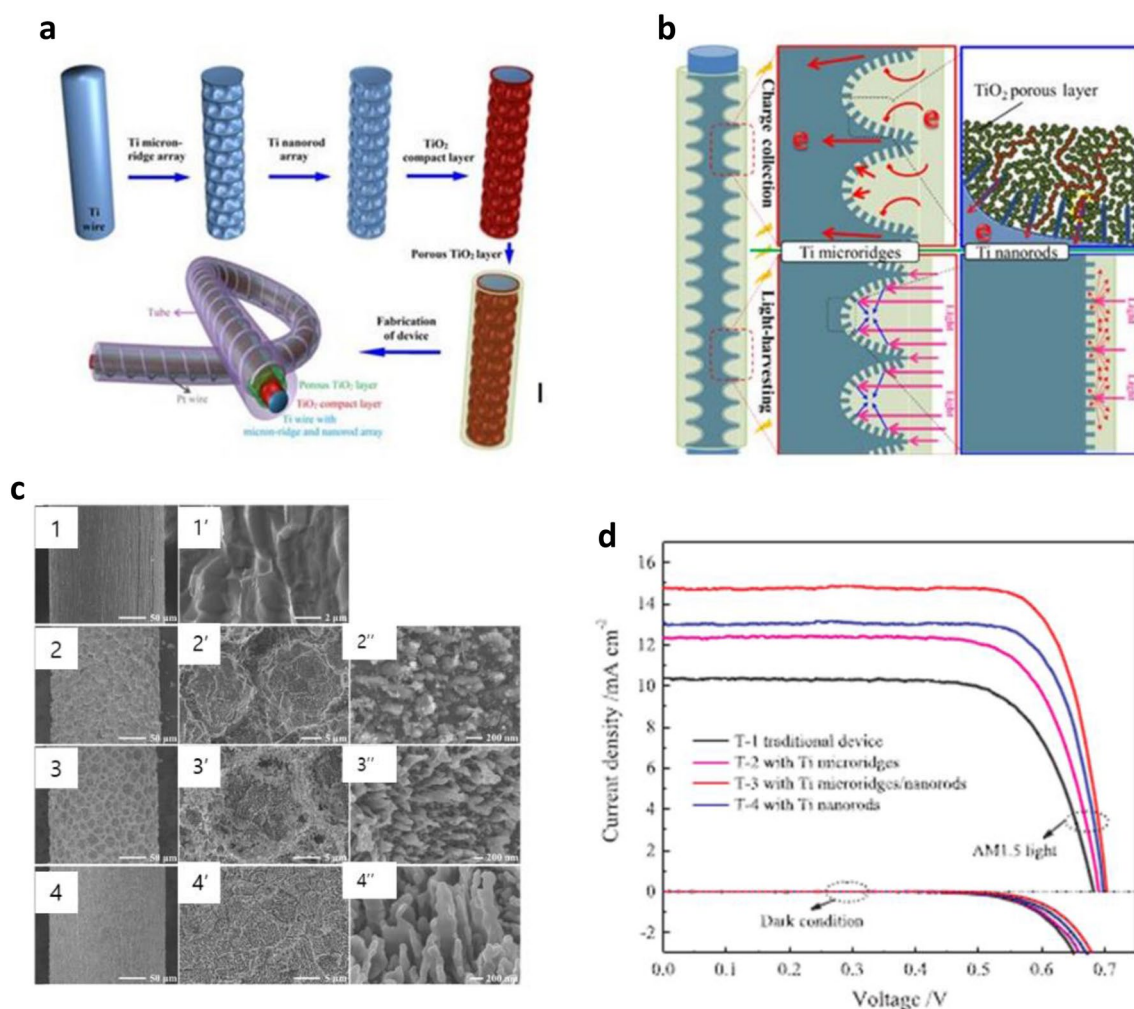


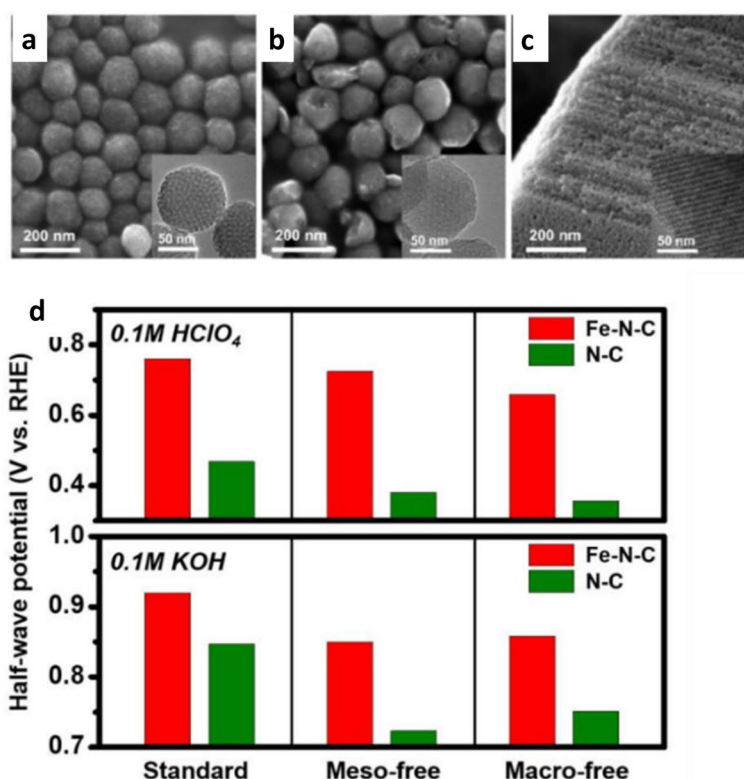
Fig. 4 **a** Schematic diagram of the fabrication of a fibrous dye-sensitized solar cell fabricated using hierarchical photons with good charge collection and harvesting capabilities. **b** Schematic diagram of the fabrication of dye-sensitized solar cells in a fiber shape. **c** SEM images of (1, 1') conventional titanium wire (T-1), (2, 2', 2'') titanium wire surface modified with a Ti microridge (T-2), (3, 3', 3'') titanium

wire surface modified with a Ti microridge/nanorod (T-3), and (4, 4', 4'') titanium wire surface modified with a Ti nanorod (T-4). **d** Current change characteristics for an AM 1.5 G solar irradiance of 100 mW cm^{-2} and for FDSSC based on Ti wire substrates synthesized under various conditions [37]

is a great advantage because it can be installed in the downtown areas of cities, which are major consumers. Small fuel cells are mainly used as power sources for automobiles or mobile devices, and large fuel cells can be used as distributed batteries or power generation cells in buildings [44–46]. However, Pt is used as a polymer electrolyte membrane fuel cell (PEMFC) catalyst and accounts for most of the fuel cell manufacturing cost because it has a price of approximately 10 million won per kg. Moreover, its performance significantly decreases over time. To overcome these problems, catalyst synthesis studies in which the use of carbon (M–N/C) and Pt catalysts is being minimized in favor of non-platinum catalysts are currently being conducted [47, 48].

Lee et al. [48] confirmed the difference in catalyst efficiency according to the pores of C to synthesize a highly efficient Fe–N–C catalyst. Based on the research results, the catalyst structure was designed with three types of pores, micropores, mesopores, and macropores, with diameters of less than 2 nm, 2–50 nm, and more than 50 nm, respectively. The structures of these pores were confirmed by TEM (Fig. 5). In the resulting structure, the mesopores serve to increase the electrochemical activity by increasing the surface area of the catalyst in which chemical reactions take place. The macropores enable the possibility of catalyst application by improving performance by rapidly transporting oxygen molecules participating in the reaction to the catalytically active region [48].

Fig. 5 Non-precious metal fuel cell catalysts with different porous structures: **a** 2 nm or less, **b** 2–50 nm, **c** 50 nm or more. **d** Catalyst activation improvement in various fuel cell driving environments [48]



Pt is the highest catalytic efficiency of all known catalysts but it is expensive. To date, improvements in the performance of non Pt catalysts has been limited. For this reason, researchers are working on developing catalysts with the minimal use of Pt. Existing electrocatalysts have a form in which Pt nanoparticles with sizes of 3–5 nm are dispersed on a carbon-based support. The fuel cell reaction occurs mainly on the surface of the Pt catalyst, and the inside surfaces of Pt are not sufficiently utilized. However, a study was conducted to maximize the utilization rate of Pt using Pd as the center of metal particles and coating Pt on the surface at an atomic thickness [49, 50]. The synthesized catalyst structures, for which different catalyst supports were used, are shown in Figs. 6 and 7. Kim et al. [51] (Fig. 7) employed two-dimensional graphene as a catalyst support to improve the dispersibility of the nano-sized catalysts, promote charge transport on the electrode surface, and maintain a stable catalyst structure. The catalyst properties improved in comparison with those of the Pt/C catalyst [50].

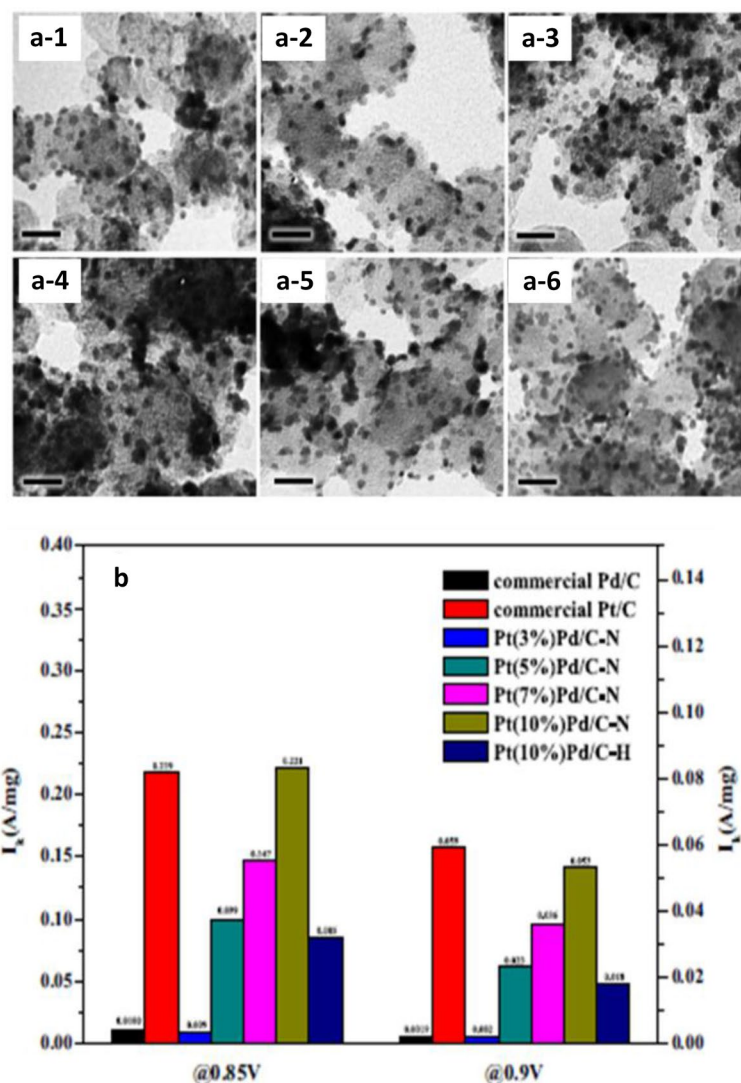
Lim et al. [52] developed a fuel cell catalyst that reduced Pt usage by 90% and improved its lifetime by two-fold. The research team aimed to improve the oxygen reduction activity and durability of existing Pt-based catalysts. First, a Pt–Ni alloy catalyst was synthesized and several metal elements were subsequently introduced to improve the performance. Ga was found to be the most effective at improving the performance. Therefore, the research team formed a Pt–Ni alloy catalyst into octahedral nanoparticles and added

Ga to the surface of the nanoparticles, improving the performance by a factor of 12 compared to the existing Pt catalyst. In addition to the aforementioned catalysts, various studies are underway to increase the fuel cell efficiency and reduce the use of Pt catalysts by different techniques. Tiwari et al. [53] revealed that their team succeeded in improving the catalytic performance by developing a hybrid material in which DNA and graphene were combined and much smaller Pt particles than those used in conventional catalysts, were attached to their surfaces, to increase the reaction surface area. The research team synthesized a composite material in which Pt particles with a size of 1 nm were uniformly dispersed in a hybrid material made of DNA and graphene. An image of the DNA-applied catalyst is shown schematically in Fig. 8. Smaller particles tend to become more clumped. Furthermore, the property of Pt ions to strongly bind to the bases of the DNA chain was used. Thus, 95% of the initial performance was maintained and the results were much better than those of commercial Pt nanoparticles (50%).

The aforementioned research can reduce the catalyst cost, which accounts for a significant part of the manufacturing cost for fuel cells. Accordingly, it is possible to reduce the overall production cost. Consequently, Korea can now advance as a leading country in the hydrogen economy by enabling transportation and power generation using hydrogen, an eco-friendly energy source.

A fuel cell that uses hydrogen energy can be interpreted as a simple energy storage system or not renewable energy.

Fig. 6 TEM images of the commercial Pd/C, Pt(x)Pd/C, and commercial Pt/C catalysts: **a-1** commercial Pd/C, **a-2** Pt(3%)Pd/C–N, **a-3** Pt(7%)Pd/C–N, **a-4** Pt(10%)Pd/C–N, **a-5** Pt(10%)Pd/C–H, and **a-6** commercial Pt/C. **b** Mass activity values at 0.85 and 0.9 V for the catalysts employed in this work. The activity values were calculated using the equation for obtaining kinetic current based on the total mass of metals used in this work [49]



In this review, fuel cells are investigated with renewable energy technologies because they can utilize greenhouse gas reduction by referring to the energy technology investment strategy announced by the Ministry of Science, ICT in Korea [2].

4 Lithium-ion batteries

LIBs are suitable for both short and long cycles because of their high energy density and easy modularization, which therefore enable their expansion to several tens of megawatts. They exhibit advantages of life and efficiency over other types of batteries [54]. In addition, there is significant potential for technological innovation in the future, and many companies are expanding their investment in LIB development, with an expectation of great improvements in performance and price. LIBs use the difference in

chemical energy between positive and negative electrode active materials. When an LIB is discharging, lithium ions in the negative electrode active material are spontaneously inserted into the positive electrode active material, with a relatively low chemical energy level in the electrolyte. As current flows, power is provided. In contrast to discharging, charging increases the energy level of the positive electrode active material through an external circuit, such that lithium ions are stored in the negative electrode active material. The capacity of the battery increases with the amount of lithium ions stored in the positive electrode active material. This has the greatest impact on battery performance, thereby prolonging battery life, but the crystal structure of the positive electrode active material must be maintained during long-term charging and discharging [55, 56].

As of 2018, nickel, cobalt, and manganese (NCM) accounted for the largest proportion (43%) of cathode materials, with a demand of approximately 156,000 tons.

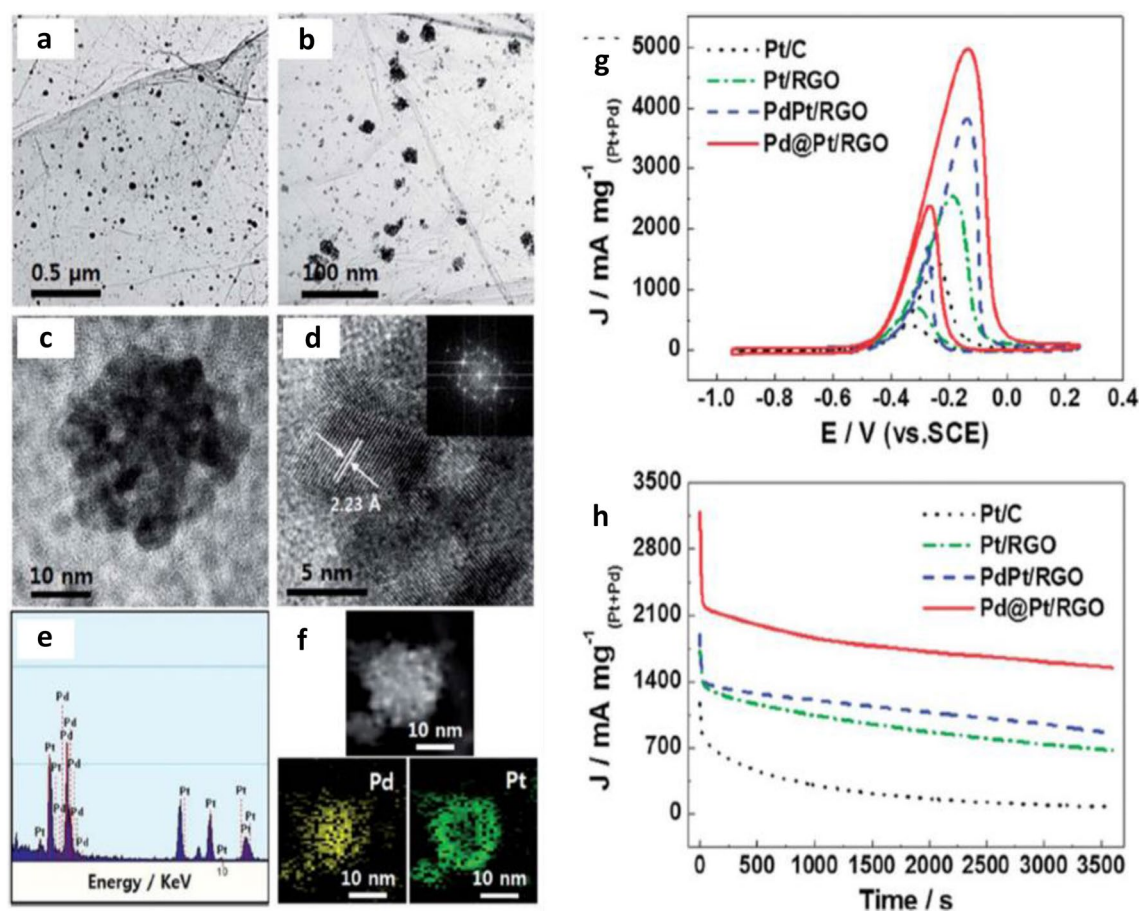


Fig. 7 **a** and **b** TEM images of PdPt/RGO at different magnifications, **c** and **d** HR-TEM images of an individual PdPt particle on RGO, **e** EDX spectrum of PdPt/RGO, and **f** HAADF-STEM images of PdPt/RGO and the corresponding elemental maps of Pd and Pt. The inset in (**d**) shows the FFT pattern of the HR-TEM image. **g** CV curves of

Pt/C, Pt/RGO, PdPt/RGO, and Pd@Pt/RGO in N_2 -saturated 0.5 M NaOH + 0.5 M methanol obtained at a scan rate of 50 mV s^{-1} . **h** CA curves of the catalysts in 0.5 M NaOH + 0.5 M methanol at a constant voltage of 0.25 V for 3600 s [51]

Following this, 85,000 tons (23%) of lithium, cobalt, and oxide (LCO), 58,000 tons (16%) of lithium, phosphate, and iron (LFP), and 55,000 tons of nickel, cobalt, and aluminum (NCA) (15%) were used. NCM use is expected to increase to 72% by 2025. It has a high capacity, and excellent life and safety characteristics. Due to these advantages, many secondary battery companies use NCM as a major cathode material for medium and large LIBs. The proportion of NCA use is expected to be 14% by 2025, similar to the current level. NCA is composed of more than 80% nickel and has a high energy density. Currently, Tesla's cylindrical secondary batteries, supplied by Panasonic, use NCA as a cathode material. Therefore, current trends in cathode materials are directing attention to Ni-rich systems, but their issues in stability during charging and recharging continues to be an issue. In the future, as the demand for LIBs increases due to the growth of the ESS market and the increasing demand for small batteries, it will be important to develop better positive

electrode materials by investigating high capacity, design conversion, and low price techniques [57, 58] (Fig. 9).

The capacity of NCM increases with an increasing Ni content, which is why development has moved in the direction of increasing the Ni content. The NCM types currently being studied for industrial use are 532 (Ni 50%, Co 30%, Mn 20%), 622 (Ni 60%, Co 20%, Mn 20%), and 811 (Ni 80%, Co 10%, Mn 10%). Currently, NCA possesses the most stable properties as $\text{LiNi}_{0.80}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$, which contains 0.05% Al [59]. For industrial use, various compositions of NCM and NCA are being researched and developed, and an increase in Ni content is essential for high-capacity cathode synthesis [60]. Table 1 summarizes the characteristics of various ratios of NMC and NCA. However, as the Ni content increases, the structure of the cathode material collapses during charging and discharging, resulting in poor battery characteristics. Accordingly, methods such as doping and surface treatment for NCM and NCA are being studied

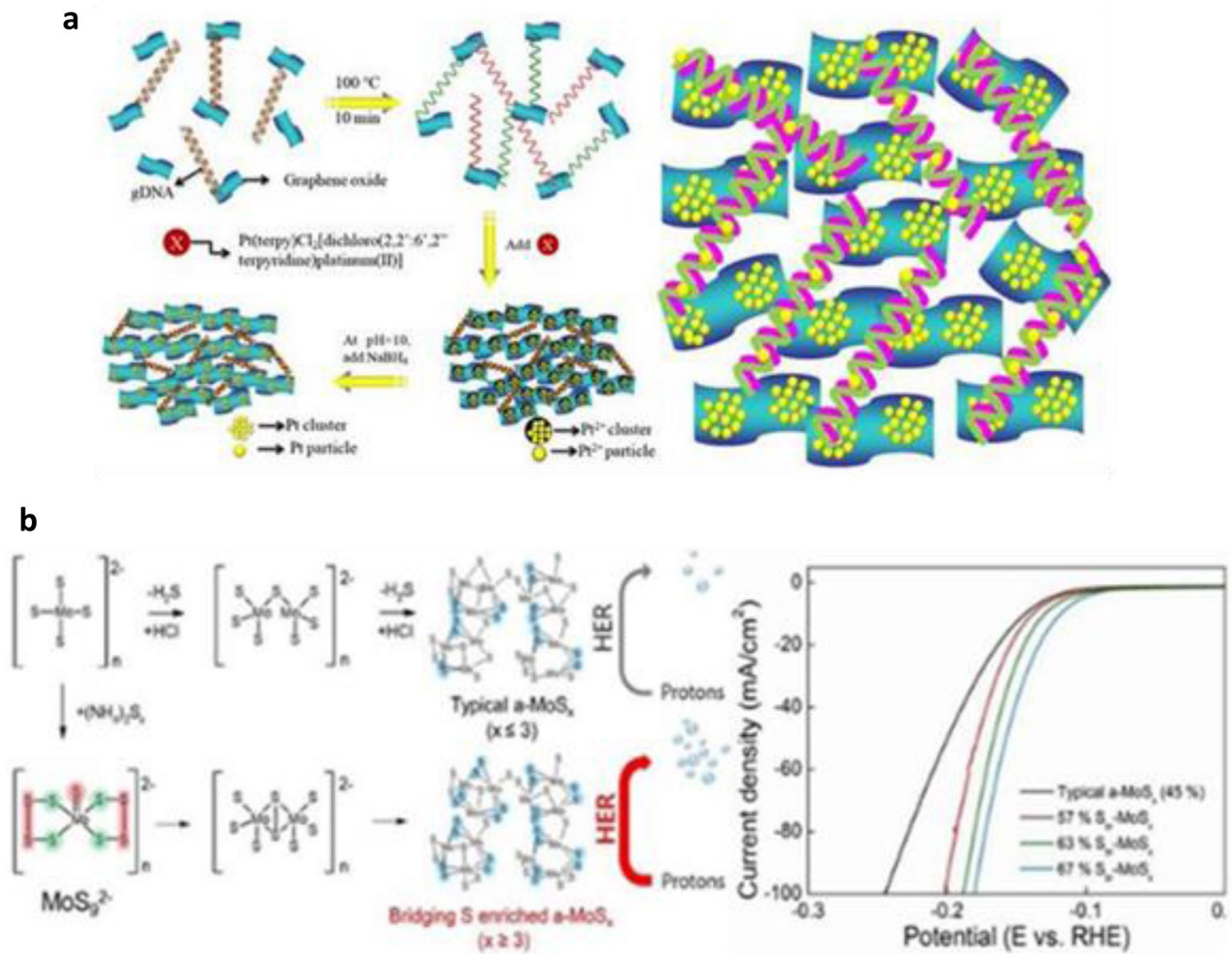


Fig. 8 **a** Schematic diagram of the synthesis of the platinum nanoparticle–DNA–graphene oxide composite material and **b** the growth principle of amorphous molybdenum sulfide and the hydrogen pro-

duction catalyst characteristics according to the content of specific bonds as current density [53]

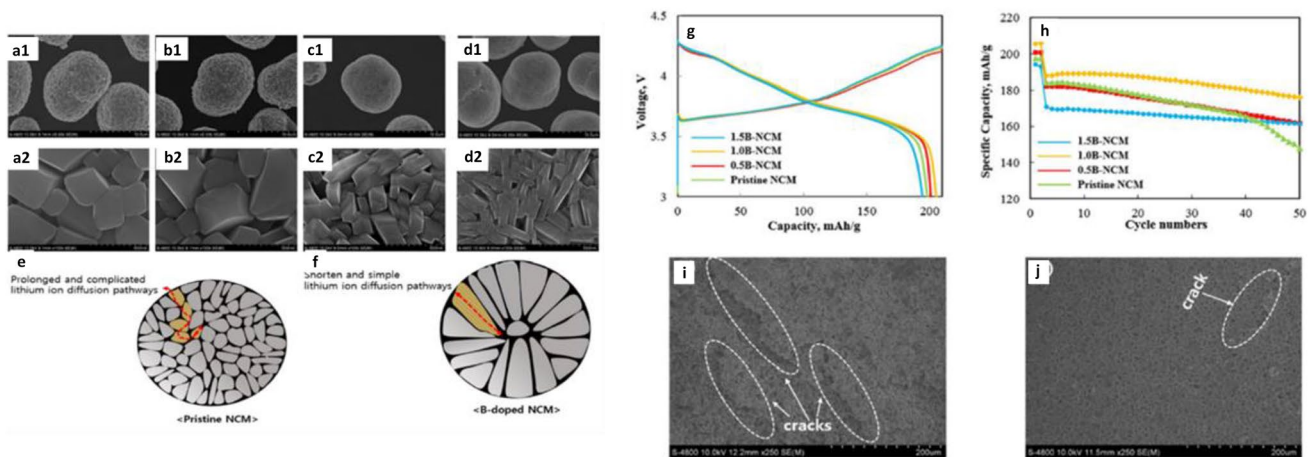


Fig. 9 FESEM images of **a** bare NCM and **b** 0.5B-, **c** 1.0B-, and **d** 1.5B-doped NCM. Schematic diagrams of **e** clean and **f** B-doped NCM. **g** Initial charge–discharge curves, and **h** cycle characteristics of clean and B-doped NCM. SEM images of **i** bare NCM and **j** 1.0B-doped NCM [62]

Table 1 List of experimentally confirmed properties of NCM and NCA cathode materials

Cathode materials	Initial discharge capacity (mAh g ⁻¹)	Voltage range (V)	Current density (mA cm ⁻²)
NCM333 [64]	169	3.0–4.3	0.1C
NCM523 [64]	175	3.0–4.3	0.1C
NCM622 [64]	187	3.0–4.3	0.1C
NCM811 [64]	203	3.0–4.3	0.1C
NCA8155 [65]	200	4.2	1C
NCA8164 [66]	202.5	2.7–4.3	0.1C
NCA88102 [66]	219.8	2.7–4.3	0.1C
NCA9541 [66]	236.8	2.7–4.3	0.1C

to achieve high stability [61]. Lee et al. [62] synthesized $\text{LiNi}_{0.84}\text{Co}_{0.10}\text{Mn}_{0.06}\text{O}_2$, a high-capacity positive electrode material, by increasing the Ni content. The disadvantage of the Ni-rich cathode is that cations are mixed as the Ni content increases, causing structural instability and capacity reduction. To address this problem, they used B^{3+} to inhibit ion migration and stabilize the crystal structure of Ni-rich NCM. As a result, 1.0 wt% boron-doped NCM (1.0B-NCM) exhibited the best electrochemical performance. The results are presented in Fig. 10. 1.0B-NCM was confirmed to have a discharge capacity of 205.3 mAh g⁻¹ at 0.5 C and maintained 93.1% of its capacity after 50 cycles, while maintaining 87.5% capacity at 2 C [62]. This suggests that the 1.0B-NCM cathode can be applied to next-generation LIBs. In addition, Sattar et al. [63] investigated the possible enhancements by doping Mg into $\text{LiNi}_{0.84}\text{Co}_{0.11}\text{Mn}_{0.05}\text{O}_2$. All Mg-doped samples exhibited significant improvements in structural stability and electrochemical properties, such as capacity stability and rate ability. With 1 wt% Mg-doped $\text{LiNi}_{0.84}\text{Co}_{0.11}\text{Mn}_{0.05}\text{O}_2$, a high discharge capacity of 196.7 mA g⁻¹ was obtained at 0.1 C, and a capacity retention of 85.95% was observed after 80 cycles. In addition, it was shown that the Mg-doped samples were more electrochemically stable at high voltages than the undoped samples [63].

Seo et al. [67] produced multi-doped (Ga,B) $\text{Li}[\text{Ni}_{0.885}\text{Co}_{0.100}\text{Al}_{0.015}]\text{O}_2$ to synthesize a high-stability NCA. The researchers synthesized bare NCA, Ga-NCA, and Ga&B-NCA to confirm the effect of multi-doping and measured the properties. To confirm the effect of Ga, doping of 0.5–1.5 mol% was conducted, and Ga:B = 0.75:0.25 Ga&B-NCA was synthesized. As a result, the NCA89 doped with 0.25 mol% of B and 0.75 mol% of Ga was found to have an initial capacity of 222.2 mAh g⁻¹ at 0.1 C, maintaining 91.7% of its initial capacity after 100 cycles, demonstrating excellent cycle characteristics. Furthermore, 83.4% of the initial capacity was maintained after 500 cycles. This indicates the role of mitigating the anisotropic internal deformation caused by the phase transition in the state of high charge

by modifying the surface energy of B and, because Ga provided structural and chemical stability to the O–TM–O slab, thereby improving the recharging characteristics [67]. In the case of such a Ni-rich cathode, in which structural change occurs easily as a result of charging and discharging, studies to improve the structural stability using additives are necessary and are underway.

As mentioned earlier, an increase in Ni content is inevitable to increase the capacity of NCM and NCA; however, the efficiency of the battery decreases during charging and discharging. To counteract this effect, studies have been conducted to mitigate the structural changes during charging and discharging by mixing different cathode materials. Kim et al. [68] synthesized a material with a Ni content of 89% in NCMA. The research team confirmed the possibility of a hybrid cathode material of Ni-rich NCA and NCM, a four-element system composed of $\text{LiNi}_{0.89}\text{Co}_{0.05}\text{Mn}_{0.05}\text{Al}_{0.01}\text{O}_2$. The aim was to increase the structural stability of the cathode material by forming Al–O from the Al in the synthesized cathode material. The synthesized material displayed better cycling and thermal stability properties than NCA89 and NCM90 with similar Ni contents. This was the case because NCMA has a high structural stability and incurs little change in the c-axis during charging and discharging, thereby suppressing the occurrence of internal microcracks charge cycling. The proposed Ni-rich NCMA cathode had a capacity of 228 mAh g⁻¹ and a long-term cycling stability (85% after 1000 cycles) [68].

In addition to doping ions to improve the performance of the cathode material, research on improving the structural stabilization by modifying the cathode surface is also underway. Various materials, such as Al_2O_3 [69], $\text{Li}_4\text{P}_2\text{O}_7$ [70], and $\text{Al}_2\text{O}_3/\text{LiAlO}_2$ [71], have been used for surface modification. In a recent study by Chae and Yim [72], a one-step efficient surface modification process using an organic surfactant (SDS) with a sulfuric acid functional group was developed to improve the surface stability of a Ni-rich NCM material (NCM811). The surface modification formed a cathode–electrolyte interphase layer on the Ni-rich NCM surface containing SO_x . Cells treated with 0.5% SDS–Ni-rich NCM exhibited 91.9% capacity retention, which was better than the result for untreated cells of NCM811; this is due to the stabilization of the surface of the NCM811 cathode by SDS [72]. In a study by Moon et al. [73], the surface of NCA was treated with pluronic acid (F127) and polyvinyl alcohol (PVA) as polymer templates. The researchers confirmed that the positive electrode material produced HF during cycling at > 4.3 V in an electrolyte containing LiPF_6 and that the HF, formed because of damage to the positive electrode material, prevented the deterioration of electrochemical performance. Samples were prepared by combining the two materials in various ratios, and it was confirmed that the synthesized positive electrode had the best electrochemical

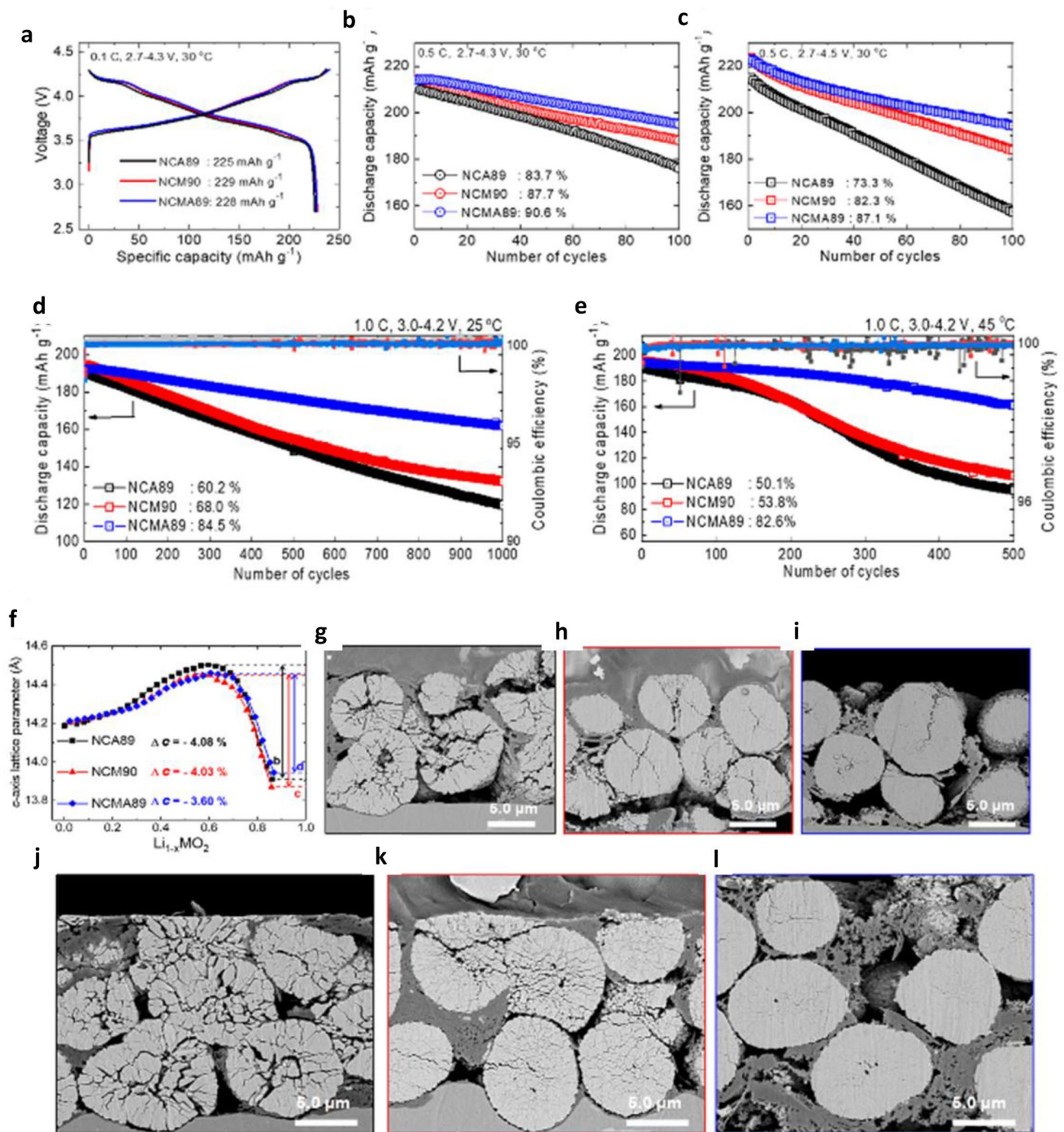


Fig. 10 Comparison of the electrochemical performances of NCA89, NCM90, and NCMA89 cathodes: **a** First cycle voltage curves for the three cathodes. All cells used Li metal as the anode and measured CV in the voltage range of 2.7–4.3 V at 0.1 C and 30 °C in a half cell. Charge–discharge performance of the three positive electrodes tested at 0.5 and 30 °C, at voltage ranges of **b** 2.7–4.3 V and **c** 2.7–4.5 V. Long-term charge and discharge performances of NCA89, NCM90, and NCMA89 cathodes using anodes of cellomethocarbon

microbead graphite in the voltage range of 3.0–4.2 V at 1.0 C and **d** 25 °C and **e** 45 °C. **f** *c*-axis lattice parameters of the NCA89, NCM90, and NCMA89 cathodes as a function of extracted lithium. Cross-sectional SEM images of fully charged cathodes in the first cycle ($\text{Li}_{1-x}\text{MO}_{2x}=0.88$): **g** NCA89, **h** NCM90, and **i** NCMA89. Cross-sectional SEM images of fully discharged cathodes after 1000 cycles: **j** NCA89, **k** NCM90, and **l** NCMA89 [68]

result for the cathode in LIB, which was prepared with the optimum ratio of 3:2 for F127 to PVA [73] (Fig. 11).

Lithium-ion battery technology is an essential element in current plug-in electric vehicles and mobile devices. Furthermore, research and development will continue because interest in energy storage devices, along with eco-friendly energy generation, is increasing. For practical applications, energy storage devices must be manufacturable with a high capacity at low cost. Because the high-nickel cathode mentioned above is attracting attention as a material that can satisfy these criteria, research on this cathode is expected to continue in the future.

The aforementioned battery-related content referred only to cathode studies that are important for capacity determination in the secondary battery composition. However, the direction of secondary battery research these days is drawing attention to studies on solid state batteries that do not use liquid electrolytes to improve safety [74, 75]. However, studies on solid state batteries are extensive and therefore not mentioned in this review.

5 Future policy and directions of research

Globally, energy conversion is emerging as a vital endeavor to mitigate the problem of climate change caused by environmental pollution. Because a new climate agreement will be implemented from 2021, in place of the Kyoto Protocol (which expires in 2020), countries participating in the Climate Change Convention will be obliged to establish and implement their goals. Accordingly, Korea has a legal plan in place for the energy sector, which is established every 5 years in accordance with the 2006 Framework Act on Energy to promote energy policy and provide energy demand and supply forecasts for the next 20 years from the time of establishment, including energy security and supply measures. It provides guidance for managing technological development and manpower training plans. Such a plan could constitute a new energy paradigm for the future. The first and second plans suggested the movement towards a sustainable energy

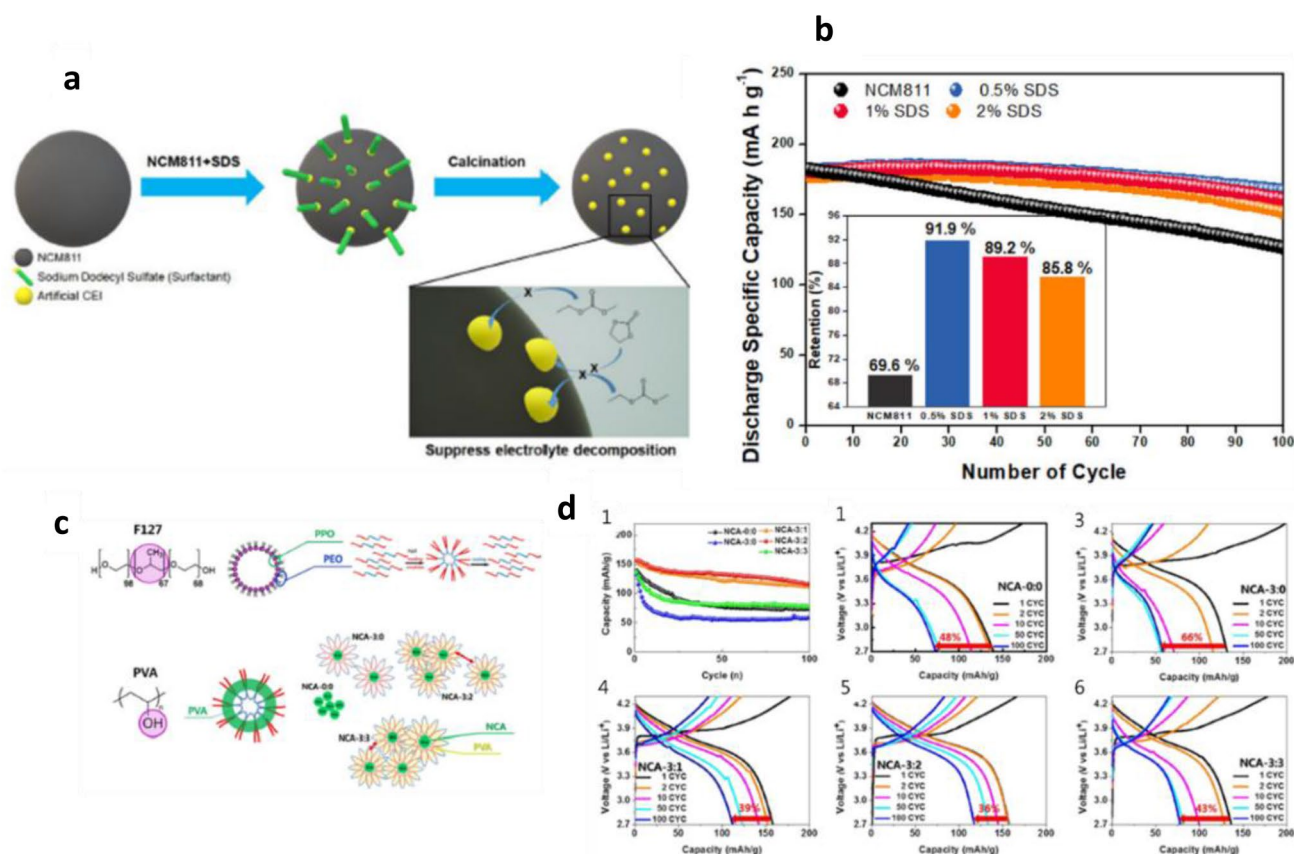


Fig. 11 **a** Schematic diagram of the surface-modified NCM811 cathode by SDS and **b** specific capacities of NCM811 (black), 0.5% SDS-treated NCM811 (blue), 1.0% SDS-treated NCM811 (red), and 2.0% SDS-treated NCM811 (orange) [71]. **c** Schematic diagram of the mechanism of formation of an NCA cathode prepared using a

polymer template of F127 and PVA. **d-1** Charging and discharging of NCA at a current density of 20 mA g⁻¹ for 100 cycles. Charge/discharge characteristic curves of Li/Li⁺ at 2.7–4.3 V measured at a current density of 20 mA for **d-2** NCA-0: 0, **d-3** NCA-3: 0, **d-4** NCA-3: 1, **d-5** NCA-3: 2, and **d-6** NCA-3: 3 [72]

system, and the third plan, implemented in 2019, focused on the innovative transformation of the energy system at all stages of energy production, distribution, and consumption while maintaining this system [76].

For this innovative energy change, Korea's focus is on expanding decentralized and participatory energy systems and realizing a hydrogen economy to secure global competitiveness in the energy industry. The first plan was to expand distributed and participatory energy systems. Currently, Korea uses a system that transmits power produced by large-scale power plants, and an energy transfer imbalance occurs during this process. To solve this problem, distributed power systems using solar power and fuel cells have been implemented so that each region can produce energy. The number of distributed power plants in Korea is currently planned to increase from 220,000 (as of 2018) to 1.9–2.7 million by 2040. Currently, the most important part of Korea is the renewable energy base, which will secure new momentum for industrial innovation and greenhouse gas reduction by utilizing hydrogen as a primary energy source. Hydrogen-based technology can be used for transportation and power generation because it is easy to use and does not pollute the environment. Therefore, Korea is planning extensive growth in technology that can generate energy using hydrogen, such as fuel cells [77].

6 Summary

Various attempts have been made to address the current problem of global climate change. Representative policies include the application of carbon credits and renewable energy support between countries. The purpose of the latter policy is to increase the use of renewable energy and introduce a method of producing clean energy instead of energy based on carbon. Therefore, research on energy materials useful for producing and storing energy is expected to continue. The energy materials being developed should have the following characteristics:

- Innovative functions to maximize high-efficiency and high-performance energy-saving techniques.
- Combinations of advanced functions that can be used in energy storage and supply systems for stable storage and energy supply.
- High performance, highly functional, and environmentally friendly characteristics as a core factor necessary for the development of clean energy technology.

The most important aspect in the field of energy materials is securing a high-performance system that can facilitate

highly efficient energy conversion and storage to ensure stable supply [64].

To increase energy conversion efficiency, solar cells can be utilized over a wide area or energy can be produced from a small amount of light by changing the bandgap of the energy materials. In addition, fuel cells have been noted to be an important technology for energy production from non-fossil sources. In general, catalysts are difficult to employ in energy conversion devices as they require expensive materials. To mitigate this problem, research is being conducted to develop catalysts that do not contain precious metals or to synthesize effective catalysts using small amounts of expensive materials [62, 63, 67, 68].

Energy storage materials are eco-friendly, and Ni-rich cathode materials have been confirmed to exhibit high capacity and high performance. Research has been extensively conducted to improve the characteristics of NCM and NCA, which are increasingly used industrially. As the Ni content is increased, the structural stability of the cathode decreases. To prevent this loss of stability, the cathode can be doped with B, Ga, and Mg, which can increase the structural stability. In addition, because structural changes are caused by reactions with the liquid electrolyte, research has been conducted on improving stability through surface treatment. Accordingly, researchers in Korea are investigating the increase in binding stability caused by graphene to produce high-performance electrode materials. Therefore, research on energy materials is being widely conducted in the current situation, with the focus on energy production and storage techniques.

Acknowledgements This work was supported by Defense Acquisition Program Administration and Agency for Defense Development under the contract (UD190006GD) and Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea government(MOTIE)(2021400000690, GCU-AU Next Generation Fuel Cell Development Program).

Funding This work is supported by Gachon University research funding in 2021 (GCU-2021-03050001).

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