Bioethanol Conversion to Gasoline-range Fuels: Guerbet Reaction and Ethylene Oligomerization Pathways

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Abstract: The upgrading of ethanol into gasoline-range biofuels relies critically on efficient carbon–carbon coupling strategies, among which the Guerbet reaction and ethylene oligomerization pathways play central and complementary roles. This review focuses on these two mechanistic platforms as core routes for controlled carbon chain growth from ethanol-derived intermediates. The Guerbet reaction proceeds via a redox-neutral hydrogen-borrowing mechanism involving ethanol dehydrogenation, aldol condensation, dehydration, and subsequent hydrogenation to form higher alcohols such as n-butanol. Its bifunctional catalytic requirements—metal sites for reversible hydrogen transfer and basic sites for C–C coupling—enable atom-economical chain extension with minimized external hydrogen demand. Catalyst design principles emphasizing metal–base interfacial proximity, balanced basicity, and hydrothermal stability are discussed, alongside process intensification strategies including tandem reactor systems and water management. In contrast, ethylene oligomerization—following ethanol dehydration—proceeds predominantly through carbocation-mediated chain propagation within acidic porous frameworks, offering a petrochemically mature pathway to C5–C12 olefins and paraffins. The roles of Brønsted acidity, pore confinement, branching control, and coke suppression are analyzed in relation to gasoline selectivity. Together, these two upgrading paradigms illustrate how rational catalyst engineering and reaction network control can transform renewable ethanol into infrastructure-compatible hydrocarbon fuels.

Keywords: Bioethanol; Biofuel; Fermentation; Catalysis; Conversion

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1 Introduction

The transition from fossil-derived transportation fuels to renewable carbon resources represents one of the central technological challenges of the twenty-first century [1]. Global reliance on petroleum-based gasoline has enabled unprecedented mobility and economic development, yet it has simultaneously intensified greenhouse gas emissions [2], geopolitical vulnerabilities, and long-term resource insecurity [3]. While electrification is advancing rapidly in passenger transport, liquid hydrocarbon fuels will remain indispensable for sectors requiring high energy density [4], rapid refueling, and compatibility with existing infrastructure [5]. In this context, the catalytic upgrading of renewable feedstocks into gasoline-range hydrocarbons (C5–C12) has emerged as a strategically important pathway toward low-carbon mobility [6].

Among renewable carbon intermediates, bioethanol occupies a uniquely advantageous position [7]. It is already produced at an industrial scale from sugarcane, corn, and increasingly from lignocellulosic biomass and waste streams [8]. The global fermentation infrastructure for ethanol production is mature, geographically distributed, and

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continuously expanding [9]. Moreover, ethanol is liquid at ambient conditions, readily transportable, and can be synthesized from a broad spectrum of carbohydrate-rich feedstocks, including agricultural residues and municipal organic waste [10]. These attributes render ethanol not merely a fuel additive but a versatile platform molecule for sustainable carbon chemistry.

Despite its widespread use in gasoline blending [11], ethanol exhibits physicochemical limitations that restrict its direct deployment as a complete substitute for gasoline [12]. Its high oxygen content reduces volumetric energy density relative to hydrocarbons, leading to lower mileage per unit volume [13]. Ethanol is hygroscopic and can induce phase separation in fuel systems [14, 15], posing challenges for storage and pipeline transport. In addition, its vapor pressure and combustion characteristics impose blending constraints within existing engine architectures [16]. These limitations motivate the development of catalytic strategies to transform ethanol into fully deoxygenated hydrocarbons compatible with current fuel infrastructure [17, 18].

Ethanol conversion to gasoline-range biofuels offers a compelling solution by integrating renewable carbon into established refinery logistics. Unlike blending strategies that partially displace fossil gasoline [19], catalytic upgrading enables the production of drop-in hydrocarbons—paraffins, iso-paraffins, olefins, naphthenes, and aromatics—within the conventional C5–C12 boiling range. Such hydrocarbons match the energy density, volatility profile, and combustion properties of petroleum-derived gasoline [20]. Importantly, this approach leverages existing distribution networks, storage facilities, and engine technologies, thereby reducing the need for systemic infrastructure modifications.

From a chemical perspective, ethanol upgrading represents a highly complex reaction network involving dehydration, oligomerization, hydrogen transfer, cyclization, aromatization, and cracking reactions [21]. The catalytic conversion of oxygenates to hydrocarbons has been extensively studied in related processes such as methanol-to-hydrocarbons and methanol-to-gasoline technologies [22]. However, ethanol introduces distinct mechanistic nuances. Its molecular structure, possessing both a hydroxyl group and a carbon–carbon bond, enables parallel pathways including direct dehydration to ethylene, bimolecular dehydration to diethyl ether, and subsequent carbon–carbon coupling reactions [23]. Understanding and controlling these intertwined pathways remain central scientific challenges.

The thermodynamic and kinetic features of ethanol conversion further underscore its complexity [24]. Dehydration reactions are exothermic and proceed readily over Brønsted acid sites, yet downstream oligomerization and aromatization steps require careful control of acidity strength, pore topology, and diffusion properties. Excessive acidity can accelerate undesired cracking and coke formation, while insufficient acidity suppresses carbon chain growth. Water, generated in situ during dehydration, competes with ethanol for active sites and influences catalyst stability [25]. Consequently, rational catalyst design must balance activity, selectivity, hydrothermal stability, and resistance to deactivation.

Beyond catalyst chemistry, process considerations play a decisive role in determining overall feasibility. Ethanol upgrading is highly exothermic, necessitating efficient heat management strategies to prevent thermal runaway and catalyst degradation [26]. Reactor configuration—whether fixed-bed, fluidized-bed, or circulating riser—affects mass transfer, temperature gradients, and catalyst lifetime. Integration with upstream fermentation and downstream separation units influences energy efficiency and carbon utilization [27].

In recent years, advances in hierarchical zeolite synthesis, metal-modified bifunctional catalysts, operando spectroscopy, and microkinetic modeling have substantially deepened mechanistic understanding [11, 28, 29]. Tailoring pore architecture to mitigate diffusion limitations, incorporating dehydrogenation metals to enhance aromatization, and tuning acid site density to optimize product distribution have collectively improved performance. Simultaneously, emerging concepts such as tandem catalysis and process intensification aim to increase carbon efficiency and suppress coke formation [30, 31].

Against this backdrop, ethanol conversion to gasoline-range biofuels stands at the intersection of catalysis science, reaction engineering, and sustainable energy systems. It embodies a broader paradigm shift: transforming renewable oxygenates into infrastructure-compatible hydrocarbons through precise control of catalytic reaction networks. This review aims to systematically examine the mechanistic foundations, catalyst design strategies, structure–activity relationships, process engineering considerations, and future directions of ethanol upgrading technologies. By integrating insights from fundamental catalysis with considerations of scalability and sustainability, we seek to clarify

the opportunities and remaining challenges in constructing efficient, durable, and economically viable ethanol-to-gasoline processes.

2 Guerbet Pathways

The Guerbet reaction represents one of the most atom-economical and strategically attractive pathways for upgrading ethanol into higher alcohols and, ultimately, gasoline-range hydrocarbons [32]. First reported by Marcel Guerbet in the late nineteenth century, the reaction involves the self-condensation of primary alcohols to yield β-alkylated alcohols with doubled carbon number. In the case of ethanol, the Guerbet reaction converts C2 alcohol into n-butanol (C4) [30, 32], which can subsequently undergo dehydration, oligomerization, hydrogenation, and cyclization to produce C5–C12 hydrocarbons suitable for gasoline blending [33, 34]. Compared to direct ethanol-to-hydrocarbon routes over strong acids, the Guerbet pathway offers improved carbon efficiency, moderated coke formation, and enhanced flexibility in downstream processing [23].

At the molecular level, the Guerbet reaction proceeds via a redox-neutral hydrogen autotransfer cycle [35]. The first step involves metal-catalyzed dehydrogenation of ethanol to acetaldehyde [36], generating surface hydrogen species. This is followed by base-catalyzed aldol condensation between two acetaldehyde molecules to form 3-hydroxybutanal [37], which subsequently undergoes dehydration to crotonaldehyde. In the final step, crotonaldehyde is hydrogenated by the previously released hydrogen to yield n-butanol. The overall stoichiometry, $2\ C_2H_5OH \rightarrow C_4H_9OH + H_2O$, highlights the absence of net hydrogen consumption. The hydrogen released during dehydrogenation is internally recycled during hydrogenation, a feature that enhances sustainability and reduces external hydrogen demand [38, 39]. This intrinsic hydrogen economy is one of the most compelling advantages of the Guerbet pathway.

The catalytic requirements of the Guerbet reaction are inherently bifunctional [40]. Efficient ethanol dehydrogenation demands metallic sites capable of reversible hydrogen abstraction [41], while aldol condensation requires basic or amphoteric sites to facilitate enolate formation [42]. Therefore, catalyst systems must provide intimate proximity between metal and base functionalities [43]. Copper-based catalysts have been extensively investigated due to their moderate dehydrogenation activity and high selectivity toward acetaldehyde without excessive C–C cleavage [44]. Nickel exhibits higher intrinsic activity but can promote undesirable hydrogenolysis and reforming reactions [43]. Noble metals such as ruthenium or palladium enhance hydrogen transfer kinetics but introduce economic constraints for large-scale fuel production [45].

The basic component is equally critical in determining selectivity and turnover frequency. Solid bases such as MgO [42], hydrotalcite-derived Mg–Al mixed oxides, and alkali-modified metal oxides provide the necessary sites for acetaldehyde condensation [46]. The strength and distribution of basic sites must be carefully tuned [47]. Weak basicity limits enolate formation and reduces C–C coupling rates, whereas excessively strong basicity accelerates side reactions, including ester formation via Tishchenko pathways or over-condensation leading to heavy oligomers. Optimal performance arises from moderate basicity that promotes selective aldol condensation while minimizing competing pathways [48].

Active site proximity strongly influences overall efficiency. If metal and base sites are spatially separated, intermediate acetaldehyde molecules may desorb before condensation, or hydrogen species may diffuse inefficiently. Consequently, modern catalyst design emphasizes interfacial engineering [49], such as dispersing metal nanoparticles directly onto basic oxide matrices or constructing core–shell architectures that maximize metal–base contact [41, 50]. Strong metal–support interactions stabilize small metal particles, suppress sintering, and facilitate hydrogen spillover, thereby enhancing hydrogen recycling during the autotransfer cycle.

Selectivity control remains a significant mechanistic challenge because ethanol can undergo multiple parallel reactions under Guerbet conditions [51]. Acidic impurities or residual acid sites may catalyze dehydration to ethylene [52], diverting carbon away from higher alcohol formation. At elevated temperatures, ethanol may undergo steam reforming or dehydrogenation beyond acetaldehyde, generating CO, CO_2, and hydrogen [53]. Additionally, crotonaldehyde intermediates may polymerize if hydrogenation is insufficient, leading to carbonaceous deposits. Thus, reaction temperature, typically in the range of 200–300 ℃, must be optimized to balance dehydrogenation kinetics and condensation rates while minimizing side reactions [21]. Maintaining an appropriate hydrogen balance within the catalyst bed is crucial; insufficient hydrogenation activity promotes unsaturated intermediate accumulation, whereas excessive hydrogen partial pressure can suppress dehydrogenation equilibrium [54–56].

Water management represents another key factor in Guerbet chemistry [57]. Water is generated during the aldol condensation–dehydration sequence and can adsorb strongly on basic sites, reducing catalytic activity. Accumulated water may also influence metal dispersion and promote sintering under hydrothermal conditions [58]. Strategies such as operating under continuous flow, reducing pressure, or integrating selective water removal technologies can shift equilibrium toward higher alcohol formation and enhance long-term stability.

The Guerbet reaction serves as an intermediate platform rather than a final fuel production step. n-Butanol produced from ethanol can be dehydrated to butenes over acidic catalysts [59], followed by oligomerization and hydrogenation to produce iso-paraffins and branched hydrocarbons within the gasoline boiling range [60]. Alternatively, tandem catalytic systems can integrate Guerbet coupling and downstream hydrocarbon synthesis in a cascade configuration. Compared with direct ethanol-to-aromatic pathways [61], this stepwise strategy allows greater control over branching and suppresses rapid aromatic condensation that leads to coke formation. The resulting hydrocarbon mixture can be tailored to optimize octane number, volatility, and combustion characteristics.

Process intensification is essential for translating Guerbet chemistry from laboratory studies to industrial application [31]. Tandem reactor designs allow independent optimization of metal–base and acid-catalyzed steps, preventing mutual neutralization of active sites [28, 62]. Structured catalysts and monolithic reactors improve heat transfer, mitigating localized hot spots that may accelerate deactivation. Continuous flow operation enhances product removal and limits over-condensation [63], while membrane-assisted water separation can shift reaction equilibria toward higher alcohol yields [64, 65]. Emerging electrified or plasma-assisted catalytic systems offer the potential to lower effective activation barriers and improve energy efficiency by decoupling surface activation from bulk thermal conditions [39, 66].

Catalyst stability remains a central issue in long-term operation [67]. Deactivation mechanisms include carbon deposition from aldol polymerization, metal particle sintering, and poisoning of basic sites by water. Designing catalysts with robust oxide frameworks, controlled basicity, and strong metal anchoring can significantly extend lifetime [22]. Hydrotalcite-derived mixed oxides exhibit structural resilience under hydrothermal conditions [68], while copper-based catalysts benefit from moderate redox stability [44]. Periodic regeneration protocols may further enhance durability in industrial settings [69].

Mechanistic understanding has advanced substantially through operando spectroscopy and computational modeling [70]. Infrared studies reveal surface-bound acetaldehyde and crotonaldehyde intermediates, confirming the hydrogen-borrowing cycle [71]. Isotopic labeling experiments demonstrate internal hydrogen recycling, while density functional theory calculations identify aldol condensation as a key rate-determining step sensitive to base strength and coordination environment [72]. Microkinetic modeling highlights the importance of balancing dehydrogenation and hydrogenation rates to prevent intermediate accumulation and deactivation. These mechanistic insights provide a rational basis for catalyst optimization.

From a sustainability perspective, the Guerbet pathway offers superior carbon utilization relative to direct dehydration–aromatization routes [44]. Its redox-neutral character minimizes external hydrogen demand, and when coupled with bioethanol derived from lignocellulosic biomass, the overall lifecycle greenhouse gas emissions can be substantially reduced. Economically, the viability of Guerbet-based ethanol upgrading depends on catalyst cost, durability, and process integration efficiency. Non-noble metal systems, particularly Ni-based catalysts supported on mixed oxides, provide a promising balance between performance and scalability.

3 Ethylene Oligomerization Pathways

Ethylene oligomerization constitutes one of the most direct and industrially relevant pathways for converting ethanol-derived intermediates into gasoline-range hydrocarbons [73]. In integrated ethanol upgrading schemes, ethanol is first dehydrated to ethylene [74–77], after which carbon chain growth proceeds via oligomerization to generate C4+ olefins [78, 79]. These higher olefins can subsequently undergo hydrogenation, isomerization, cyclization, and aromatization to produce paraffinic, iso-paraffinic, naphthenic, and aromatic hydrocarbons within the C5–C12 boiling range [80]. Compared with the Guerbet route, which relies on redox-neutral alcohol coupling, the ethylene oligomerization pathway emphasizes acid-catalyzed carbon–carbon bond formation through carbocation chemistry [81]. Its mechanistic versatility and compatibility with established petrochemical processes render it a cornerstone of

ethanol-to-gasoline technologies.

The initial step in this pathway is ethanol dehydration to ethylene, typically catalyzed by Brønsted acid sites over solid acids such as HZSM-5 [82]. The dehydration reaction is rapid and exothermic, producing ethylene and water. Once formed, ethylene enters a complex network of oligomerization reactions that proceed via protonation to form surface-bound ethyl carbenium ions [83, 84]. These reactive intermediates undergo chain growth through successive insertion of additional ethylene molecules, generating higher olefinic species [73]. The fundamental mechanistic motif involves the formation and rearrangement of carbenium ions within confined catalytic environments, where acidity strength and pore architecture strongly influence reaction trajectories [85].

Two primary mechanistic frameworks describe ethylene oligomerization over heterogeneous catalysts: the classical carbocation mechanism [86] and the metallacycle mechanism [87]. In solid acid systems such as zeolites, oligomerization typically follows the carbocation route. Ethylene is protonated at a Brønsted acid site to form an ethyl cation, which subsequently reacts with another ethylene molecule to generate a butyl cation. Repetition of this sequence leads to C6, C8, and higher olefinic chains [88]. Termination occurs via β-hydride elimination, producing linear or branched olefins and regenerating the acid site. The balance between propagation and termination steps governs the carbon number distribution of products.

In contrast, homogeneous or supported transition metal catalysts, particularly those based on nickel or chromium complexes, can promote ethylene oligomerization through metallacyclic intermediates [89]. In such systems, ethylene coordinates to a metal center, inserts into a metal–carbon bond, and forms a metallacyclopentane intermediate [90, 91]. Subsequent β-hydride elimination releases linear α-olefins. While metallacycle-based catalysis is widely used industrially for producing linear α-olefins, its application in integrated ethanol upgrading remains less common due to sensitivity to water and oxygenates. Nevertheless, hybrid catalytic strategies combining metal-mediated oligomerization with acid-catalyzed downstream transformations are gaining attention [88].

Within zeolitic frameworks, pore topology exerts a profound influence on oligomerization pathways. Medium-pore zeolites such as ZSM-5 favor the formation of C6–C10 hydrocarbons due to shape selectivity and diffusion constraints [92]. The 10-membered ring channels restrict excessive chain growth and limit the formation of bulky polyaromatic species, thereby suppressing coke deposition relative to large-pore materials. In contrast, larger pore zeolites such as H-Beta and HY allow formation of heavier oligomers and polycyclic aromatics [93, 94], often at the expense of stability. Therefore, rational selection of pore architecture enables tuning of product distribution toward the gasoline range [95].

The hydrocarbon pool concept, originally developed for oxygenate-to-hydrocarbon processes, also contributes to understanding ethylene oligomerization in confined zeolite environments. Once initial C4+ species are formed, they can act as co-catalytic centers, undergoing methylation, cracking, and hydrogen transfer reactions that interconvert olefins and aromatics [85, 95]. This dynamic network leads to a quasi-equilibrated distribution of hydrocarbons governed by thermodynamic and kinetic constraints. In this context, oligomerization is not a simple linear chain growth process but part of a complex autocatalytic cycle involving olefin and aromatic intermediates.

Selectivity toward gasoline-range hydrocarbons depends strongly on acidity strength and density [96]. High acid site concentration accelerates oligomerization but may also promote excessive cracking and hydrogen transfer, increasing light paraffin formation and coke deposition. Conversely, insufficient acidity limits chain growth and results in predominantly C4 products. Optimal catalysts exhibit moderate Brønsted acidity that supports controlled propagation without rapid deactivation [88]. Adjusting the Si/Al ratio in zeolites provides a practical means of tuning acid density and strength, thereby modulating product selectivity [97].

Reaction temperature further influences oligomerization pathways [95]. Lower temperatures favor longer-chain olefins by suppressing β-scission and cracking, whereas higher temperatures increase aromatic formation and coke deposition. Typically, temperatures in the range of 250–400 ℃ are employed in ethanol-derived ethylene oligomerization systems. Water, generated during ethanol dehydration, competes with ethylene for acid sites and may attenuate activity. Designing hydrophobic zeolite surfaces or hierarchical structures that facilitate water desorption can mitigate these inhibitory effects.

Branching and isomerization are critical determinants of fuel quality [60]. Linear olefins generated during oligomerization can undergo skeletal isomerization via hydride shift and methyl shift reactions within the zeolite framework. Branched iso-olefins exhibit higher octane numbers after

hydrogenation, enhancing gasoline performance. The interplay between oligomerization and isomerization thus enables tuning of fuel properties. Confinement within medium-pore zeolites promotes the selective formation of mono-branched structures while limiting multi-branched or bulky species that could lead to deactivation [98].

Downstream hydrogenation of oligomerized olefins yields paraffinic hydrocarbons compatible with gasoline blending [99]. Integrating bifunctional metal–acid catalysts allows simultaneous oligomerization and hydrogenation, reducing olefin content and stabilizing products. Metals such as nickel or platinum provide hydrogenation functionality [100, 101], while acidic supports drive oligomerization. The spatial arrangement of metal and acid sites influences hydrogen transfer efficiency and suppression of coke formation.

Catalyst deactivation in ethylene oligomerization primarily arises from coke deposition formed through polyaromatic condensation [102, 103]. Prolonged residence of bulky intermediates within micropores accelerates deactivation. Hierarchical zeolites incorporating mesoporous channels improve mass transfer, facilitating the removal of heavy intermediates and extending catalyst lifetime. Additionally, phosphorus modification or controlled dealumination can moderate acidity and reduce coking propensity.

Process intensification strategies aim to enhance efficiency and scalability. Fluidized-bed reactors improve heat management for highly exothermic oligomerization reactions, while circulating catalyst systems enable continuous regeneration [104, 105]. Integration of ethanol dehydration and ethylene oligomerization within a single reactor reduces capital cost but requires careful thermal control. Alternatively, staged reactor configurations allow independent optimization of dehydration and oligomerization conditions, improving overall carbon efficiency.

Mechanistic insights from operando spectroscopy and isotopic labeling studies reveal rapid interconversion between olefinic species and confirm the dynamic nature of carbenium ion chemistry within zeolite pores [106]. Computational studies indicate that propagation barriers depend strongly on pore confinement and acid strength, while β-scission activation energies determine chain length distribution [107]. These insights support rational catalyst design aimed at maximizing gasoline-range selectivity.

From a sustainability perspective, the ethylene oligomerization pathway leverages mature petrochemical knowledge while enabling renewable carbon incorporation via ethanol. When integrated with bioethanol derived from lignocellulosic biomass, this route can produce drop-in hydrocarbons with reduced lifecycle carbon intensity. Its compatibility with existing refinery infrastructure enhances feasibility relative to entirely novel fuel systems.

4 Conclusion and Outlook

The catalytic conversion of ethanol to gasoline-range biofuels represents a strategically significant pathway for integrating renewable carbon into existing fuel infrastructures. Across the mechanistic landscape discussed in this review, two dominant upgrading paradigms emerge: the Guerbet-mediated carbon chain growth strategy and ethylene oligomerization networks. Each pathway offers distinct advantages and limitations in terms of carbon efficiency, catalyst stability, process complexity, and product distribution control. Collectively, they demonstrate that ethanol is not merely a blending component but a versatile platform molecule capable of being transformed into fully deoxygenated, infrastructure-compatible hydrocarbons.

Direct dehydration–oligomerization routes over acidic zeolites provide high activity and process simplicity, leveraging well-established oxygenate-to-hydrocarbon chemistry. However, strong acidity and confined microporous environments often accelerate coke formation and limit catalyst lifetime. The Guerbet pathway, by contrast, introduces controlled C–C coupling through hydrogen-borrowing chemistry, improving carbon utilization and moderating aromatic overproduction. Meanwhile, ethylene oligomerization offers a petrochemically mature and tunable strategy for constructing gasoline-range olefins and paraffins with desirable branching characteristics. The future of ethanol upgrading will likely depend on rational integration of these approaches rather than exclusive reliance on a single pathway.

Looking forward, several scientific and technological priorities will shape progress in this field. First, catalyst design must move toward atomic-level precision in balancing acid, base, and metal functionalities. Engineering interfacial active sites with optimized proximity can enhance hydrogen transfer, suppress side reactions, and extend catalyst durability. Second, hierarchical and defect-engineered porous materials are essential to overcome diffusion limitations and mitigate coke deposition under hydrothermal conditions. Third, operando characterization combined with microkinetic modeling should continue to elucidate reaction networks under realistic operating environments, enabling predictive catalyst optimization.

From a process engineering perspective, intensified reactor configurations—such as tandem beds, circulating systems, and electrified catalytic reactors—offer opportunities to improve thermal management and energy efficiency. Integration with lignocellulosic ethanol production and biorefinery concepts will be crucial for achieving favorable life-cycle greenhouse gas performance. Additionally, techno-economic analyses must guide the selection of scalable catalyst systems based on earth-abundant metals and robust supports.

Ultimately, ethanol-to-gasoline technologies embody a broader transition toward renewable hydrocarbon manufacturing. By merging advances in heterogeneous catalysis, materials science, and systems engineering, it is possible to transform bioethanol from a blending additive into a cornerstone of sustainable fuel production. Continued interdisciplinary innovation will determine whether this pathway can achieve the selectivity, stability, and economic viability required for large-scale deployment in a decarbonized energy future.

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