Key Technologies and Demonstration Postprint for Clean and Efficient Cascade Utilization of Low-Rank Coal

Authors: Shanxi Institute of Coal Chemistry, Chinese Academy of Sciences

Date: 2017-02-08T00:00:00+00:00

Abstract

In the global energy structure, coal has consistently occupied a significant position. The World Energy Council' s *World Energy Vision: Energy Scenarios for 2050* indicates that by 2050, fossil fuels will remain the predominant energy source, with coal continuing to play an extremely important role. China' s energy situation is particularly severe, characterized by scarce oil and natural gas resources but relatively abundant coal reserves; coal accounts for nearly 70% of the nation' s primary energy structure, holding a dominant position. With the development of new energy sources such as nuclear, wind, and solar power, the proportion of coal will gradually decline, but its foundational status will not undergo fundamental changes for a considerable period in the future.

Full Text

Preamble

Strategic Priority Research Programs (Category A) of the Chinese Academy of Sciences
ChinaXiv Partner Journal
Key Technologies and Demonstration for Clean and Efficient Cascade Utilization of Low-Rank Coal

1. Project Background and Significance

Coal has long occupied a crucial position in the global energy structure. According to the World Energy Council' s "World Energy Vision: Energy Perspectives for 2050," fossil fuels will remain the dominant energy source through 2050, with coal continuing to play an extremely important role. China' s energy situation is particularly severe, with scarce oil and natural gas resources but relatively abundant coal reserves. Coal accounts for nearly 70% of China' s primary energy mix, establishing its dominant position. While the proportion of coal will gradually decline with the development of nuclear, wind, and solar energy, its foundational status will not fundamentally change for a considerable period.

Low-rank coal (lignite/sub-bituminous coal), which represents over 55% of China' s proven coal reserves (1,020 billion tons), has a low degree of coalification. The volatile matter contained within is equivalent to 100 billion tons of oil and gas resources. However, due to its high moisture content, direct combustion or gasification of low-rank coal is inefficient, and existing technologies cannot fully utilize its resource value, resulting in enormous waste of coal resources. Based on the composition and structural characteristics of low-rank coal, developing a clean and efficient cascade utilization technology system to achieve its rational and optimized utilization is of great significance.

[Figure 1: see original paper]

2. Progress Achieved

In response to these challenges, the Chinese Academy of Sciences launched the Strategic Priority Research Program "Key Technologies and Demonstration for Clean and Efficient Cascade Utilization of Low-Rank Coal" in February 2012. Based on the composition and structural characteristics of low-rank coal, the program proposed an overall solution for clean and efficient cascade utilization, with efficient pyrolysis as the leading process to extract existing oil and gas resources from coal. The remaining semi-coke can then be used for power generation or converted into fuels and chemicals through gasification, forming three technical routes: "Pyrolysis—Oil/Gas Upgrading—Semi-coke Combustion—Power Generation," "Pyrolysis—Gasification—Synthesis," and "Pyrolysis—Gasification—Fischer-Tropsch Synthesis—Co-processing of Oil Products."

After nearly five years of dedicated effort, the program has achieved remarkable success in scientific objectives, platform construction, and talent development, meeting its predetermined targets. Many key technologies have reached demonstration or industrialization readiness, with some already being demonstrated in collaboration with enterprises, laying a solid foundation for industrial promotion. Specific achievements include:

A 240 ton/day solid heat carrier pulverized coal low-temperature pyrolysis unit and a 10,000 ton/year low-rank coal hydrogenation pyrolysis pilot plant have achieved stable operation at full load. A 25,000 standard cubic meter/hour industrial demonstration test for integrated dry removal of multiple pollutants from flue gas has been successful. A 10,000 cubic meter/hour CO reforming unit for syngas production, a 30,000 ton/year acetic acid hydrogenation to ethanol plant, and a 50,000 ton/year cobalt-based Fischer-Tropsch synthesis technology unit have all been successfully commissioned. Catalysts developed for methanol-to-fuel conversion have been applied in a 200,000 ton/year industrial demonstration. Additionally, industrial demonstrations for 350 MW supercritical circulating fluidized bed combustion, kiloton-scale multi-stage gasification technology, 50,000 ton/year methanol synthesis of polyether oxygenates, and 100,000 ton coal-based dimethyl ether carbonylation to methyl acetate and ethanol are under construction and expected to be completed and operational by 2018.

2.1 Scientific and Technological Objectives

Pyrolysis can directly extract volatile matter from coal to produce oil and gas, with the resulting semi-coke serving as a high-quality clean fuel for combustion or gasification. This is a graded conversion process that leverages coal's inherent composition and structure to produce oil, gas, and chemicals. As the leading technology in the coal program, pyrolysis focuses on breakthroughs in key technologies for coal pyrolysis to prepare oil and gas. In low-temperature pyrolysis, through innovative solid material heating, material circulation control, and pyrolyzer coke discharge methods, the program successfully controlled energy and material exchange between the solid heat carrier combustion bed and pyrolysis bed, achieving organic coupling of the dual beds. In November 2015, a 240 ton/day solid heat carrier coal low-temperature pyrolysis pilot plant achieved stable operation at full load [Figure 2: see original paper], marking the first time in China that a 100,000 ton/year scale circulating fluidized bed pulverized coal pyrolysis unit has reached full-load operation, laying a solid foundation for subsequent semi-coke combustion and gasification objectives. The program also overcame technical challenges in gas heat carrier pyrolysis gasification engineering, as well as multiple key technologies including large-rate solid-solid mixing for solid heat carriers, solid level control under high-temperature and high-dust conditions, pyrolysis-combustion coupling, and high-temperature gas-solid separation suitable for pyrolysis oil and gas. A 3,000 ton/year solid heat carrier coal pyrolysis pilot plant and a 100,000 ton/year solid heat carrier pyrolysis-gasification industrial demonstration have been constructed, with the latter already in normal operation.

In hydrogenation pyrolysis, a mild-condition hydrogenation liquefaction process was developed based on coal type characteristics. The program solved the problem of difficult regulation of liquefaction product structure distribution through "solvent cycle self-balancing." By the end of 2015, a 10,000 ton/year Xinjiang Hami coal hydrogenation pyrolysis unit in Ordos, Inner Mongolia achieved 128 hours of continuous stable operation [Figure 3: see original paper], collecting complete process data and samples. The 2 million ton/year Xinjiang Hami coal graded comprehensive utilization project prepared based on these results has been included in the National Energy Administration' s "13th Five-Year Plan" key demonstration reserve projects for efficient and clean coal utilization, with system energy efficiency reaching 55-56%, opening a viable path for clean utilization of Xinjiang Hami coal.

Combustion represents an important utilization pathway for coal pyrolysis semi-coke. Targeting efficient power generation and gas pollutant control, and combining steam cycle power generation with semi-coke and mixed fuel combustion and post-combustion flue gas pollutant control, the program conducted research on supercritical circulating fluidized bed combustion power generation and coal-fired flue gas pollutant removal and purification technologies. The program completed a 2 MW pilot test of fine semi-coke preheating combustion characteristics and NOx emission characteristics, developed a 20 MW-grade preheating burner, and commissioned a high-temperature semi-coke transport pilot plant, obtaining pneumatic solid valve high-temperature semi-coke transport characteristics. The program broke through core technologies for large-scale circulating fluidized bed combustion uniformity, designed a new polygonal furnace type reaching domestic leading levels, and secured a technical demonstration project for a 350 MW supercritical circulating fluidized bed boiler. The developed integrated dry removal technology for multiple flue gas pollutants has achieved continuous stable operation in a 25,000 standard cubic meter/hour industrial demonstration test unit [Figure 4: see original paper], with all indicators meeting "ultra-low emission" standards and results superior to publicly reported domestic and international levels.

Gasification is the leading technology for synthesis in the coal program, aiming to efficiently and cleanly gasify semi-coke produced from pyrolysis based on existing pressurized fluidized bed pulverized coal gasification technology and CFB combustion technology, and to address key issues in large-scale and engineering of pressurized coal gasifiers. Major progress includes optimization of a 100 ton/day multi-stage bed pilot plant, conducting 22 tests with cumulative operation exceeding 400 hours, achieving system stability under 2.0-2.8 MPa pressure, reaching international advanced level process indicators, verifying core equipment performance, and completing process design packages and preliminary engineering design for kiloton-scale multi-stage fluidized bed gasification technology. The program also completed combustion tests of gasification fine ash in a 40 ton/day industrial test unit, achieving stable combustion with combustion efficiency reaching 99.18%, providing support for engineering of gasification fine ash CFB re-burning technology.

Synthesis is an important pathway for high-value utilization of coal. As is well known, bulk chemicals such as olefins and aromatics, as well as liquid fuels like gasoline and diesel, are important cornerstones of national economic development. Traditional production methods mainly involve refining petroleum or its derivatives using various processes. A new viable strategic approach is to partially replace petroleum resources using China' s relatively abundant coal resources for further synthesis into various bulk chemicals or fuels. The coal program focuses on developing technologies for producing bulk chemicals and fuels from syngas and methanol. Currently, the core catalytic technology for "coal-to-syngas-to-C2 oxygenates and hydrogenation to ethanol technology" is at a world-leading level in all key indicators. In April 2016, a 30,000 ton/year acetic acid hydrogenation to ethanol industrial unit was successfully started up in one attempt [Figure 5: see original paper], producing ethanol with purity above 99.9%, exceeding national industrial ethanol standards. The "cobalt-based

Fischer-Tropsch fixed-bed synthesis technology and demonstration," completed in collaboration with Lu' an Group, represents China' s first 50,000 ton/year industrial demonstration unit, which has been in trial operation. The catalyst, evaluated by BP and other international energy companies (under rated conditions), outperforms existing commercial catalysts and reaches international advanced levels.

Technologies for producing chemicals and fuels from methanol are significant for promoting methanol industry structural adjustment and upgrading, improving the ecological environment, and driving regional economic development. The "methanol-to-propylene key technology" has completed large-scale cold fluidization tests and 300 ton/year hot tests, obtaining data required for preparing industrial plant process packages, representing a new generation of methanol-to-propylene technology. The "methanol synthesis of polyether oxygenates" mainly produces DMM3-8 (polyoxymethylene dimethyl ethers) from methanol through trioxymethylene under ionic liquid catalysts. The program conducted hundred-ton engineering scale-up and system integration, achieving continuous operation and process optimization with conversion rates $>$90%, DMM3-8 selectivity $>$50%, and methanol consumption $<$1.5 tons per ton of DMM3-8 product. A 50,000 ton/year industrial demonstration plant is currently under construction. The catalyst developed for "methanol-to-fuel key technology and industrial demonstration" has been successfully applied in Yunnan' s 200,000 ton/year MTG (methanol-to-gasoline) industrial demonstration test. Meanwhile, a fixed-bed adiabatic reactor developed jointly with enterprises to achieve one-step conversion of methanol to high-quality gasoline is unprecedented internationally. The program also achieved breakthroughs in catalyst activity and stability for "coal-based dimethyl ether carbonylation to methyl acetate and ethanol," determined the reaction process route, completed pilot tests, and is constructing a 100,000 ton/year demonstration plant with Yanchang Petroleum. Currently, there is no industrial-scale application research on this technology domestically or internationally, and the technical level is world-leading.

In CO conversion and utilization, the program collaborated with Lu' an Group to construct the world' s first 10,000-50,000 cubic meter/hour CO methane reforming unit, which was successfully ignited and operated in July 2015. The 50,000 cubic meter/hour reforming complete technology process package has been approved, laying an important foundation for technology commercialization and promotion.

Furthermore, process simulation has provided strong support for optimization of pyrolysis, gasification, combustion, and synthesis unit technologies.

2.2 Platform Construction

During program implementation, the Institute of Process Engineering of the Chinese Academy of Sciences and Pingmei Group jointly established the Key Laboratory of Efficient and Clean Coking Technology of the National Energy Administration. The Shanxi Coal Chemistry Institute of the Chinese Academy of Sciences and Lu' an Group jointly established the National Coal-based Synthesis Engineering Technology Research Center of the Ministry of Science and Technology. The Shanxi Coal Chemistry Institute also established the ICC-Shell Coal Chemistry Joint Laboratory with Shell. The program also constructed pilot platforms for gas heat carrier pyrolysis, solid heat carrier pyrolysis, light component hydrogenation of pyrolysis oil, circulating fluidized bed combustion, oxy-fuel combustion, multi-stage bed gasification, CFB combustion of gasification ash and fine ash, transport bed gasification, methanol-to-olefins, methanol synthesis of polyether oxygenates, methanol-to-gasoline, syngas complete methanation to natural gas, CO reforming, as well as process simulation scale-up and system simulation integration platforms. These platforms have greatly enhanced independent innovation capabilities and laid a solid foundation for successful program completion and subsequent technology development and optimization.

2.3 Talent Team Building

The program assembled approximately 500 outstanding team members from the Coal Chemistry Institute, Dalian Institute of Chemical Physics, Institute of Engineering Thermophysics, Institute of Process Engineering, Shanghai Advanced Research Institute, Lanzhou Institute of Chemical Physics, Fujian Institute of Research on the Structure of Matter, and other institutions in fields including pyrolysis, gasification, catalysis, combustion, chemical engineering, consulting, and simulation. The program further optimized and enriched the research talent team, particularly providing training and advancement opportunities for young researchers while cultivating outstanding graduate students. According to preliminary statistics, two individuals were elected as academicians, two were selected for the National "Ten Thousand Talents Program," two received National Science Fund for Distinguished Young Scholars, 57 received National Science Fund for Excellent Young Scholars, two received Zhou Guangzhao Applied Science Awards, one was named among the Top Ten Scientific and Technological Innovation Figures at the 2013 CCTV Science and Technology Ceremony, 25 were promoted from associate senior to senior professional titles, and 87 were promoted from intermediate to associate senior professional titles. The program trained 435 graduate students, with 32 receiving funding from the National Natural Science Foundation and 16 receiving funding from the Ministry of Science and Technology. This has assembled and stabilized a high-level professional team in the field of efficient low-rank coal utilization in China.

3. Originality

Through implementation of this program, fundamental chemical and chemical engineering theories have been developed for low-rank coal pyrolysis, semi-coke/coal combustion, semi-coke/coal gasification, C1 directional conversion, and CO capture and storage. The program has revealed the intrinsic relationships between low-temperature pyrolysis, hydrogenation liquefaction, and residue gasification characteristics and the structure and chemical composition of low-rank coal. Key factors affecting directional conversion of syngas and methanol have been identified, catalytic mechanisms and coking behavior have been elucidated, and catalytic theory has been advanced. The program has also revealed the influence of material combustion rate and distribution on heat transfer uniformity and supercritical parameters in ultra-large furnace chambers and their matching relationship with thermal power during fluidized bed combustion.

A series of key technologies have been developed and breakthroughs achieved, including low-temperature pyrolysis with gas/solid heat carriers, low-rank coal hydrogenation pyrolysis, integrated dry removal of multiple pollutants from flue gas, CO reforming to syngas, acetic acid hydrogenation to ethanol, cobalt-based Fischer-Tropsch fixed-bed synthesis, 350 MW supercritical circulating fluidized bed combustion power generation, kiloton-scale multi-stage bed gasification, methanol synthesis of polyether oxygenates, and coal-based dimethyl ether carbonylation to methyl acetate and ethanol. Corresponding pilot or demonstration plants have been constructed and operated.

4. Significance for Industry

Many key technologies developed in the program in pyrolysis, combustion, gasification, and synthesis have reached demonstration or industrialization readiness, with some already being demonstrated in collaboration with enterprises. The successful operation of industrial demonstrations particularly signifies basic maturity of processes and technologies, which is of great significance for technological innovation and advancement in China' s clean coal utilization field and has laid a good foundation for industrial promotion.

During program implementation, approximately 10 billion RMB in investment from large and medium-sized enterprises has been stimulated, with an estimated 40 billion RMB investment expected during the "13th Five-Year Plan" period. This provides important technical guarantees for enterprise transformation, industrial upgrading, and clean and efficient coal utilization in China, while cultivating new momentum and expanding new space for economic and social development.

5. Recommendations for Future Deployment in Disciplines, Industrial Advancement, and Talent Cultivation

Coal' s position in China' s energy structure is unquestionable, and clean and efficient coal utilization represents the main direction of China' s energy development. National leaders attach great importance to clean coal utilization, proposing at the 6th meeting of the Central Financial and Economic Affairs Leading Group to "vigorously promote clean and efficient coal utilization." Relevant industrial policy documents have explicitly identified "promoting efficient and clean coal utilization" as a primary task, and enterprises have unprecedented emphasis on technological innovation. We must seize opportunities, overcome difficulties, focus on program priorities, achieve predetermined goals, and promote technological progress in coal power and coal chemical industries to drive industrial upgrading.

The "Key Technologies and Demonstration for Clean and Efficient Cascade Utilization of Low-Rank Coal" is a Category A Strategic Priority Research Program oriented toward major national needs, targeting major scientific outputs and industrial applications. Currently, individual project applications and promotions face numerous difficulties and challenges, and cannot form integrated advanced technologies throughout the coal chemical industry chain, resulting in insufficient social impact and economic benefits. It is recommended to actively sign comprehensive strategic cooperation agreements with leading enterprises in China' s coal chemical industry or major coal-producing provinces, establish incubation parks, and concentrate on scale-up, demonstration, and integrated application of various coal chemical technologies developed in the program to accelerate application and promotion of mature technologies, which will certainly generate good economic benefits and significant social impact.

(Host Institution: Shanxi Institute of Coal Chemistry, Chinese Academy of Sciences)

Coal is China' s most important primary energy source, with low-rank coal accounting for more than half of China' s proven coal reserves. Clearly, there is an urgent need to develop key technologies for clean and efficient utilization of low-rank coal. In response to this major national demand, the Chinese Academy of Sciences timely launched the Strategic Priority Research Program "Key Technologies and Demonstration for Clean and Efficient Cascade Utilization of Low-Rank Coal" in February 2012.

Over the past five years, with Researcher Wang Jianguo, Director of the Shanxi Institute of Coal Chemistry, serving as chief commander, the program has assembled advantageous research teams from the Chinese Academy of Sciences. Based on the composition and structural characteristics of low-rank coal, the program has carried out full-chain key technological innovation and industrial demonstration, achieving major breakthroughs in low-rank coal pyrolysis, oil/gas upgrading, gasification, semi-coke fuel, and synthesis of oil products and chemicals, forming innovative technologies with independent intellectual property rights in China. Taking the synthesis of liquid fuels and chemicals, which China has more contact with, as examples, the program has developed world-leading technologies including coal (via syngas via C2 oxygenates and hydrogenation) to ethanol technology, cobalt-based Fischer-Tropsch fixed-bed synthesis technology, and a new generation of methanol-to-propylene technology. The program has constructed the world' s first hundred-ton industrial demonstration unit for methanol synthesis of polyether oxygenate fuel additives, achieving long-term continuous operation. Breaking international precedent, the program creatively developed a fixed-bed adiabatic reactor to achieve one-step conversion of methanol to high-quality gasoline. By creating novel catalysts and overcoming constraints in catalyst activity and stability, the program has developed world-leading technology for coal-based dimethyl ether carbonylation to methyl acetate and ethanol, completed pilot tests, and will soon achieve industrialization first in the world. The program has also completed construction of the world's first large-scale CO methane reforming to syngas unit, which has achieved long-term stable operation.

These major breakthroughs will form a low-rank coal comprehensive utilization technology characterized by high efficiency, low pollution, low emissions, and high value, greatly promoting industrial development of clean and efficient low-rank coal utilization and driving the adjustment and upgrading of China's coal industry structure. The promotion and application of these technologies will greatly alleviate China's energy and environmental pressures, generate significant economic and social benefits, and enable China to occupy an internationally leading position in the new round of energy competition.

Wang Fuchen is a professor and doctoral supervisor at East China University of Science and Technology, a Changjiang Scholar, chief scientist of the National "973" Program, a national candidate of the "New Century Talents Project," leader of the Ministry of Education's "Changjiang Scholar and Innovation Team Development Plan" innovation team, and an outstanding talent in the Ministry of Education's New Century Excellent Talents Program and Shanghai's Outstanding Academic Leader. He has long been engaged in applied basic research and engineering development of coal gasification and gaseous hydrocarbon conversion processes, and is one of the main inventors of multi-nozzle coal water slurry gasification technology. He has published over 250 papers, authorized more than 70 patents, published 2 monographs, received 1 second prize of the National Science and Technology Progress Award, and 5 first prizes of provincial and ministerial science and technology progress awards.

Coal is China's foundational energy source and important raw material, and clean and efficient coal utilization is of great significance. The Strategic Priority Research Program "Key Technologies and Demonstration for Clean and Efficient Cascade Utilization of Low-Rank Coal" focuses on major national needs and urgent industrial development requirements. Targeting China's coal resource endowment, the program has proposed an overall solution for clean and efficient cascade utilization of low-rank coal, aiming to form three innovative technical routes: "Pyrolysis–Oil/Gas Upgrading–Semi-coke Combustion–Power Generation," "Pyrolysis–Gasification–Synthesis," and "Pyrolysis–Gasification–Fischer-Tropsch Synthesis–Co-processing of Oil Products." Through five years of collaborative innovation among the Academy's advantageous units, important breakthroughs have been achieved in basic research and common key technologies.

The research results have advanced the fundamental chemical and chemical engineering theories for low-rank coal pyrolysis, gasification, and C1 directional conversion, deepening understanding of the intrinsic relationships between low-temperature pyrolysis, hydrogenation liquefaction, and the structure and chemical composition of low-rank coal. The program has revealed and elucidated catalytic mechanisms and coking behavior in directional conversion processes of syngas and methanol, developing new catalytic theories. It has revealed the influence of material combustion rate and distribution on heat transfer uniformity and supercritical parameters in ultra-large fluidized bed furnace chambers and their matching relationship with thermal power. Key process technologies have been developed, including solid heat carrier pulverized coal low-temperature pyrolysis, low-rank coal hydrogenation pyrolysis, integrated dry removal of multiple pollutants from flue gas, CO reforming to syngas, acetic acid hydrogenation to ethanol, cobalt-based Fischer-Tropsch fixed-bed synthesis, 350 MW supercritical circulating fluidized bed combustion, kiloton-scale multi-stage bed gasification, methanol synthesis of polyether oxygenates, and coal-based dimethyl ether carbonylation to methyl acetate and ethanol, with corresponding pilot or demonstration plants constructed and operated.

These research results have comprehensively elevated China's basic research level in low-rank coal utilization, laid a technical foundation for clean and efficient low-rank coal utilization, and opened new directions for promoting transformation and upgrading of the modern coal chemical industry.

Liu Haichao is a professor and doctoral supervisor at Peking University, a Changjiang Scholar Distinguished Professor, recipient of the National Science Fund for Distinguished Young Scholars in 2008, the 4th "China Catalysis Youth Award" in 2012, and the first "Min Enze Energy Chemical Outstanding Contribution Award" in 2013. He has long been engaged in heterogeneous catalysis and fundamental energy chemistry research, understanding catalyst structure-activity relationships at the atomic/molecular level, studying catalytic reaction kinetics and mechanisms, and designing and constructing efficient catalysts for important catalytic processes oriented toward efficient utilization of fossil energy and renewable biomass energy.

Note: Figure translations are in progress. See original paper for figures.

Source: ChinaXiv —Machine translation. Verify with original.