

炭催化富甲烷气重整制合成气技术产业化推广应用的重大装备研制和关键技术研发简介

太原理工大学



项目概述

- 我省有丰富的焦炉煤气资源。焦炉煤气或热解煤气（ CH_4 : 23%—27%, CO_2 : 3%—10%）制宝贵合成气的**关键和难点是将其中的甲烷和二氧化碳重整转化** $\text{CH}_4 + \text{CO}_2 \rightarrow \text{CO} + \text{H}_2$ 。 CH_4 - CO_2 重整制合成气研究被公认为全球化学和能源研究领域最具挑战性的研究方向之一。张永发教授自主创新开发了**炭催化富甲烷气重整制合成气技术产业化技术**，其意义在于：
 - 实现了焦炉煤气原子水平科学利用
 - 实现了二氧化碳减排



任务来源

- 国家973计划
- 国家重大项目
- 国家自然科学基金
- 山西省自然科学基金
- 山西高校科技研究开发项目

张永发 张
孙亚玲 李

你们
炉煤气制
省高等学
等奖。

山西省科学技术奖 证书

为表彰山西省科学技术奖获得者，
特颁发此证书。

项目名称：炭催化 $\text{CH}_4\text{-CO}_2$ 重整及焦炉煤气制合成气
化学化工基础研究

获奖类别：自然科学类

奖励等级：二等

获奖者：张永发 张国杰 肖 睿 孙亚玲 李香兰



证书号：2012-Z-2-002

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十月

技术被国内外学术界认可的情况

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E-mail a

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Wei-Hsin Ch

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Blast furnace

1. Introdu

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Options for the
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Maria T. Johansson
Division of Energy Systems, Dep

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Excess energy
CO₂ emissions

1. Introduction

The threat of rising er
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Production and characterization of *Lemna minor* bio-char
and its catalytic application for biogas reforming

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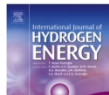
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1. Introduction

Due to ever growing concerns over depleting fossil fuel
resources and their negative ecological impact, the develop-
ment of environmentally benign and efficient processes for
converting biomass energy to clean transportation fuels,
chemicals and other value-added materials has received
a worldwide attention. The advantages of using biomass are
three-fold: it is a distributed, abundant and carbon-neutral
resource. However, the use of terrestrial biomass for energy
and fuel production is frequently queried by the relatively
low solar energy conversion efficiency of plants and potential

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Investigation of H₂O and CO₂ Reforming and Partial Oxidation of
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Mixtures of carbon and Ni/Al₂O₃ as catalysts for the microwave-assisted CO₂

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An evaluation of hydrogen production from the perspective
of using blast furnace gas and coke oven gas as feedstocks

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Blast furnace gas (BFG) and coke
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Hydrogen production
Partial oxidation
Water gas shift reaction
CO₂ reduction
Ironmaking process

ABSTRACT

Blast furnace (BF) is a large scale reactor for producing hot metal where coke and coal are
consumed as reducing agent and fuel, respectively. As a result, a large amount of CO₂ is
liberated into the atmosphere. The blast furnace gas (BFG) and coke oven gas (COG) from
the ironmaking process can be used for H₂ production in association with carbon capture
and storage (CCS), thereby reducing CO₂ emissions. In this study thermodynamic analyses
are performed to evaluate the feasibility of H₂ production from BFG and COG. Through the
water gas shift reaction (WGS) of BFG, almost all CO contained in BFG can be converted for
H₂ production if the steam/CO (S/C) ratio is no less than unity and the temperature is at
200 °C, regardless of whether CO₂ is captured or not. The maximum H₂ production from
WGS is around 0.21 Nm³ (Nm³ STP)^{−1}. Regarding H₂ production from COG, a two-stage
reaction of partial oxidation (POX) followed by WGS is carried out. It is found the
proper conditions for syngas formation from the POX of COG is at the oxygen/fuel (O/F)
ratio of 0.5 and the temperature range of 1000–1750 °C where the maximum syngas yield is
2.83 mol (mol hydrocarbon)^{−1}. When WGS is subsequently applied, the maximum H₂
production from the two-stage reaction can reach 0.83 Nm³ (Nm³ STP)^{−1}.
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1. Introduction

In recent years, on account of global warming stemming from
a huge amount of greenhouse gas emissions, the reduction of
carbon dioxide formation has been considered as a vital
countermeasure to lessen the greenhouse effect in the atmo-
sphere. It is known that blast furnaces (BFs) are a remarkable
source of carbon dioxide emissions, as a result of using coke as
a raw material to reduce iron oxides [1]. In addition to coke, coal
has also been extensively consumed in BFs for supplying heat
through pulverized coal injection (PCI) [2–4]. However, coal
combustion will cause a number of serious problems of air

pollution. At the same time, the consumptions of coal and coke
will liberate a large amount of greenhouse gases, especially for
carbon dioxide. In fact, according to statistics [6], the steel
industry accounts for 5–7% of total anthropogenic CO₂ emis-
sions in which around 70% of CO₂ emissions come from iron
production in BFs. Therefore, if a substantial cut of CO₂ emis-
sions from BFs is implemented, it will give a significant
contribution in abating atmospheric greenhouse effect.

When coke and pulverized coal are transported into BFs for
producing hot metal, the reduction processes of iron oxides
can be divided into two different ways, one is the indirect
reduction (IR) [7,8] and the other the direct reduction (DR) [9].

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rhombic materials and an
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Energy Fuels 2005,

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K. Proc. Combust. Inst. 2007, 31, 1983–1990.
S., Xie, R. Korean J. Chem. Eng. 2007, 24, 688–

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技术被国内外学术界认可的情况

◆ 被美国西肯塔基大学和西班牙教授认为是生产合成气的理想方法

“甲烷部分氧化需要纯氧，这样不仅增加了投资而且增加了甲烷部分氧化操作费用。那么怎样才能得到现代工业需要的理想合成气那？联合煤气化及甲烷重整是一个理想的好方法，该方法可以通过调节入口气成分来得到合适的氢炭比（1.5-2）。”



Mixtures of carbon and Ni/Al₂O₃ as catalysts for the microwave-assisted CO₂ reforming of CH₄
B. Fidalgo, A. Arenillas, J.A. Menéndez*

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Microwave heating

ABSTRACT

In this work, the microwave-assisted CO₂ reforming of CH₄ over mixtures of carbonaceous materials and an Al₂O₃ tab prepared (Ni/Al₂O₃) was studied. Ni/Al₂O₃ is not heated by microwave radiation, and for this reason, microwave receptors, such as carbonaceous materials, must be mixed with this catalyst. In order to evaluate the role of the carbonaceous component of the blend, two different carbonaceous materials were used: an activated carbon, PVS, and a metallurgical coke, CO. The carbonaceous component acted not only as microwave receptor but also as catalyst and, consequently, it influenced the catalytic activity of the mixture. PVS + Ni/Al₂O₃ was found to be a better catalyst than CO + Ni/Al₂O₃, since PVS on its own showed a better catalytic activity than CO. Ni/Al₂O₃, which consists of an impregnated directly onto the microwave receptor, was also evaluated as a catalyst. It was found that the catalytic activity of the mixture PVS + Ni/Al₂O₃ was better than that of Ni/Al₂O₃. Finally, the influence of the heating time on the catalytic activity of PVS + Ni/Al₂O₃ was studied. Conclusions over PVS + Ni/Al₂O₃ and microwave heating were found to be similar to conventional over Ni/Al₂O₃ and conventional heating.

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1. Introduction

In recent years, the CO₂ reforming of CH₄ or dry reforming (reaction 1), has emerged as a promising alternative to steam reforming for the production of synthesis gas (H₂ + CO) and its resulting byproducts, such as hydrogen [1,2]. The dry reforming of methane is generally favored by the environmental and they try to increase carbon CO₂ emissions. Moreover, it produces a syngas with a low ratio of H₂/CO (stoichiometric ratio of 1), which is suitable for the synthesis of higher hydrocarbons and subsequent derivatives [3–5].



However, the CO₂ reforming of methane has not yet been industrially implemented since there are no commercial catalysts that can operate without undergoing deactivation due to carbon deposition. Several metal-based catalysts have been tested as catalysts for the dry reforming reaction. Noble metals like Pt, Ru, or Rh have shown the best catalytic activity and the lowest susceptibility to coke formation, although Ni is usually preferred due to its availability and low cost [1,3,6]. Several materials have been evaluated as supports, including inorganic oxides [2–4] and carbon materials [6–10]. Carbon materials have also been used as catalysts in their own in methane reforming [6,11,12]. However, Ni

Al₂O₃ has been found to be one of the catalysts that have given the best results [2–5].

Apart from the search for new catalysts, changes to the process are being investigated in order to achieve high and steady conversions. The novel process of microwave-assisted CO₂ reforming of CH₄ using carbon-based catalysts has been widely studied in previous works by this group [6,12,13–15]. Good CO₂ and CH₄ conversions were obtained by combining microwave heating with carbon catalysts. It was found that microwave heating improves heterogeneous catalytic reactions, such as the dry reforming of CH₄, and as the rate of reaction increases, the residence time decreases, selectivity is improved and higher yields are achieved [12,15–19]. It was also observed that carbon materials show a good catalytic activity in dry reforming, both when used alone [6,12,13,16] and as supports for metal catalysts [6–10]. Besides, they are low cost, resistant towards sulfur and other potentially harmful impurities in the feedstock and good microwave absorbers, which make them suitable not only for being used as catalysts but also as microwave receptors [6,14,19].

In the best of our knowledge, the combination of Ni-based catalyst and microwave heating has not yet been investigated. For this reason, the main objective of the present work is to study the catalytic activity of Ni/Al₂O₃ in the microwave-assisted dry reforming of CH₄ by using carbon materials as supports, including inorganic oxides [2–4] and carbon materials [6–10]. Carbon materials have also been used as catalysts in their own in methane reforming [6,11,12]. However, Ni

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“炭材料被成功用于甲烷和二氧化碳重整制合成气”



Dry reforming of coke oven gases over activated carbon to produce syngas for methanol synthesis
J.M. Bermúdez, B. Fidalgo, A. Arenillas, J.A. Menéndez*

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ABSTRACT

The dry reforming of coke oven gases (COG) over an activated carbon as a catalyst has been studied in order to produce a syngas suitable for methanol synthesis. The primary aim of this work was to study the influence of the high amount of hydrogen present in the COG on the process of dry reforming, as well as the influence of other operating conditions, such as temperature and volumetric hourly space velocity (VHSV). It was found that the reverse water gas shift (RWGS) reaction takes place due to the hydrogen present in the COG, and that its influence on the process increases as the temperature decreases. This situation may give rise to the consumption of the hydrogen present in the COG, and the consequent formation of a syngas which is inappropriate for the synthesis of methanol. This reaction can be avoided by working at high temperatures (above 1000 °C) in order to produce a syngas that is suitable for methanol synthesis. It was also found that the RWGS reaction is favored by an increase in the VHSV. In addition, the active carbon PVS was proven to be an adequate catalyst for the production of syngas from COG.

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1. Introduction

In recent years, the CO₂ reforming of CH₄ or dry reforming (reaction 1), has emerged as a promising alternative to steam reforming for the production of synthesis gas (H₂ + CO) and its resulting byproducts, such as hydrogen [1,2]. The dry reforming of methane is generally favored by the environmental and they try to increase carbon CO₂ emissions. Moreover, it produces a syngas with a low ratio of H₂/CO (stoichiometric ratio of 1), which is suitable for the synthesis of higher hydrocarbons and subsequent derivatives [3–5].

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Investigation of H₂O and CO₂ Reforming and Partial Oxidation of Methane: Catalytic Effects of Coal Char and Coal Ash

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The reforming and partial oxidation was studied to evaluate the catalytic effects of coal chars and coal ash on methane (CH₄) conversion, sum selectivity (the sum of H₂ and CO), and ratio selectivity (the ratio of H₂ to CO) in a fixed-bed reactor. The kinetics study presented the possibility of CH₄ reforming and partial oxidation with a favorable H₂/CO ratio, greater than 5. The higher H₂/CO ratio in CH₄ reforming and partial oxidation process can reduce the consumption of CH₄ needed to adjust the H₂/CO ratio during combined methane reforming and partial oxidation. Coal ashes failed to be good candidates of catalysts on CH₄ reforming and partial oxidation because of their very low specific surface area available for catalytic reactions. The coal chars presented very promising catalytic performance on CH₄ reforming and partial oxidation because of their high specific surface area. In this study, no other constituents in coal fly ash or special surface properties were correlated with the enhanced methane-conversion efficiency. It seems that the specific surface area is the most important variable in controlling methane-conversion efficiency.

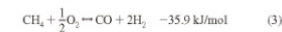
1. Introduction

It is well-known that many chemical products are produced through syngas (H₂ and CO). The production of syngas has great importance in the chemical industry because it is the main material for methanol synthesis, Fischer–Tropsch (F–T) synthesis, and dimethyl ether (DME).^{1,2} Natural gas is an important resource for syngas production. With an insufficient supply and the rising price of petroleum, great importance has been attached to the research and development of natural gas reforming. Coal gasification is also a promising resource for syngas production because the carbon in coal can react with H₂O to produce CO and H₂. Therefore, natural gas reforming and coal gasification are two primary resources for the production of syngas and may become the new source of the modern chemical industries in the future instead of petroleum.

The downstream synthesis of different chemical products requires syngas with different H₂/CO ratios. The H₂/CO ratio of syngas usually depends upon the H/C ratio of raw materials and the reaction routes of the syngas production. The desired H₂/CO ratios for methanol synthesis and F–T synthesis of different chemical products are usually 1.5–2.² Presently, syngas is mainly produced by H₂O reforming of methane. However, syngas produced from H₂O methane reforming has a H₂/CO ratio between 3 and 4 higher than what is needed for the downstream synthesis processes and thus requires further adjustment to be used in methanol synthesis and F–T synthesis. Syngas produced from CO₂ methane reforming and steam

gasification of coal cannot also be directly used in methanol synthesis or F–T synthesis because the H₂/CO ratio is close to 1. Methane partial oxidation needs to be carefully controlled to obtain an available yield of H₂ and CO, although this reaction can produce syngas with a H₂/CO ratio close to 2. At the same time, H₂ partial oxidation requires pure O₂, and thus increases investment and operation costs of CH₄ partial oxidation. How can we get a desired syngas to meet the demand of the modern chemical industry? Combined methane reforming and coal gasification is expected to easily produce syngas with the desired H₂/CO ratio of 1.5–2 by changing feed composition.^{3,4}

With the use of H₂O and CO₂, methane can be reformed to produce H₂ and CO according to the following reactions shown in eqs 1 and 2. These two reactions are so-called methane reforming. With a supply of lower stoichiometric coefficients of oxygen, methane can be partially oxidized to produce H₂ and CO according to the following reaction, which is shown in eq 3. Carbon monoxide can further react with an excessive supply of H₂O to produce more H₂. This reaction is called the water–gas shift reaction, as shown in eq 4. Carbon deposit is one of major problems during methane reforming and partial gasification. The possible carbon deposit reaction is shown in eq 5.



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(1) Bai, Z. Q.; Chen, H. K.; Li, W.; Li, B. Q. *Int. J. Hydrogen Energy* 2006, 31, 899–905.

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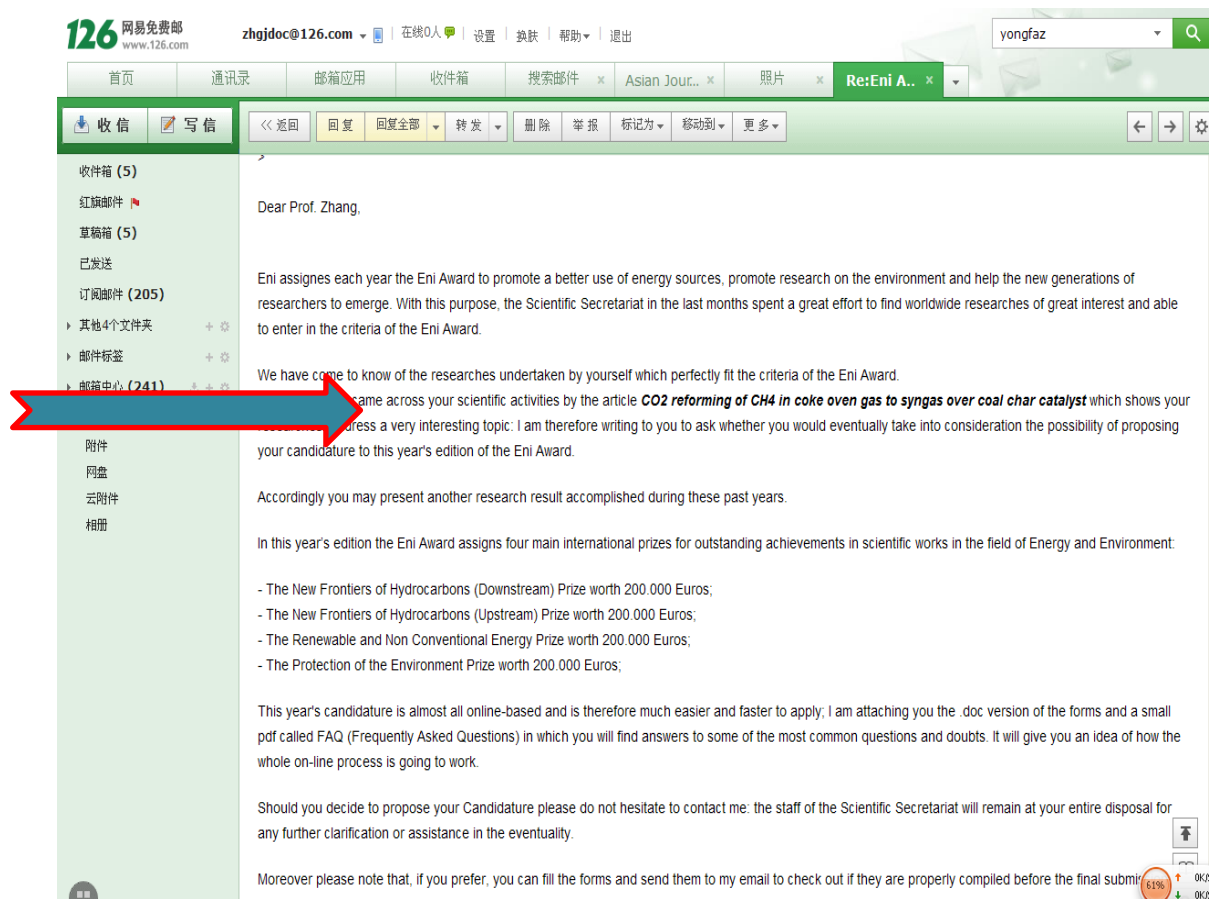
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技术被国内外学术界认可的情况

◆ Eni Award奖秘书在来信中指出我们的学术思想和观点很有创新性

“我们知道你们所做的研究这完全符合埃尼奖标准。特别是，通过你们发表的“煤焦催化CO₂重整焦炉煤气CH₄制合成气”我们认为你们的研究十分具有新颖性和创造性。因此，我写信给你，问你是否愿意作为今年Eni Award候选人，参加今年的埃尼奖评选。”





技术转化程度

- 本技术研究获得和申请国家发明专利**7**项。该技术已于**2012**年**3**月**24**日在成功临汾同世达煤化工集团开车，使这一创新性技术向产业化迈进了关键性的一大步。
- 同时该技术受到榆次神龙焦化厂、宁夏宝丰集团、山西襄汾焦化集团、山东青岛青岛泰能燃气集团有限公司和山西发鑫集团的高度关注。



试车方案制定



开启输气阀门开车成功



技术转化程度



输送原料的管路系统



混合和控制系统



技术转化程度



甲烷二氧化碳重整反应器 ($\phi 2.6\text{m}$)



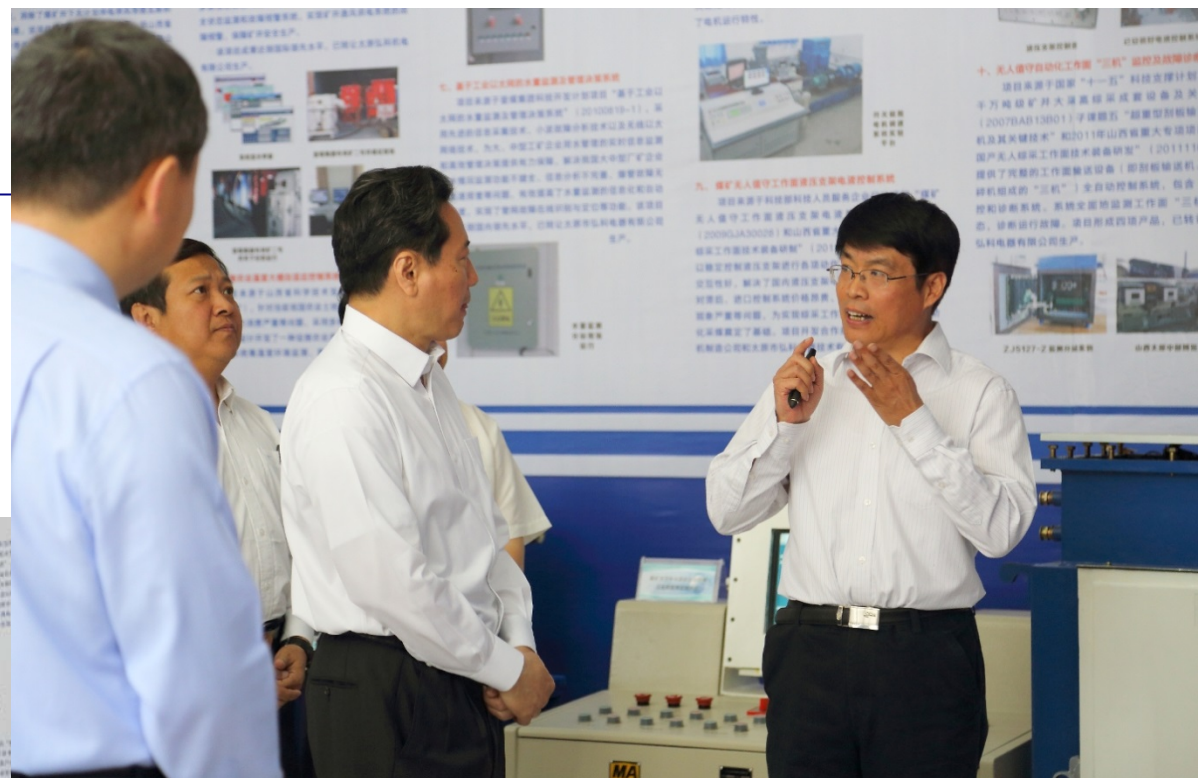
技术转化程度





技术转化程度

● 李小鹏省长调研



Key Laboratory of Coal Science and Technology of
Shanxi Province and Ministry of Education



经济和社会效益

该研究部分成果在同世达煤化工集团（年产焦炭100万吨）对 $5.2 \times 10^7 \text{Nm}^3/\text{年}$ 的富甲烷焦炉气进行转化处理，可增加二甲醚产量4.3万吨，企业增加产值**1.7亿元/年**，**同时减少CO₂排放77880吨/年**，经济和环境效益显著。总体上该技术已实施出现良好效果。本项技术高效地将焦炉煤气转化成合成气，其在我省乃至全国都有广阔的推广市场。

