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CATALYSIS AND CLEAN ENVIRONMENT: WHAT DO THESE TWO HAVE IN COMMON?

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ABSTRACT

The theme of the 1990s, "Continual Expansion through Sustainable Development," is the outcome of interdisciplinary studies undertaken in the preceding two decades to arrest ongoing global environmental deterioration. A critical component of various international efforts to formulate and implement environmentally-sound technologies is the planned involvement of developing countries and countries with economies in-transition which are poised for explosive growth well into the twenty-first century but do not have policies or financial resources to simultaneously manage their environment. Though CO_2-producing, technologies utilizing relatively abundant and inexpensive carbon-based feedstocks will still play a major role on the scene. Related to this, the aim of the ongoing effort at Brookhaven National Laboratory is to develop technologies for enhancing natural gas and coal usage through liquid phase catalysis. A successful application of this concept in the synthesis of oxygenates is described. The potential impact of this activity on methanol-energy, non-petroleum feedstocks development, H_2-energy, CO_2-recycle, and several other areas in relation to CO_2-mitigation, is also discussed.

INTRODUCTION

Since petroleum became a major energy source in the 1900s, the world economy has been dictated by the price of oil. To gain independence from this energy source, a concerted effort was dedicated in the US after the first oil embargo in 1973. In the following years, several potential feedstocks were recognized. But the overall economics of these alternatives were tied to the price of oil which is dictated by politics. In early 1980s, with a glut of oil in the world market, any incentive to utilize feedstocks other than petroleum faded, and several promising technology developments were abandoned by major corporations. It is only in the past few years that revival of this activity is being noticed. The thrust behind this effort is based on **environmental concerns**. In the US, severe pollution problems in several major cities (Los Angeles, Denver, to name a few) prompted the US Congress to pass, in 1990, Amendment to the Clean Air Act. This Act specifically mandates increased utilization of oxygenated fuels and natural gas and decreased use of petroleum.

The trend towards clean environment is growing world-wide. To meet this goal, the utilization of natural gas, a clean-burning gaseous fuel, is being expanded. For example, in Japan where raw materials are limited and the people/land ratio is relatively high, a constant effort is being made to stay at the forefront of technology development to meet the energy needs with minimum environmental impact. An impressive lead taken by the Japanese government is to implement methanol/oxygenates (recent projects funded by MITI through Mitsubishi and Mitsui) and natural gas (one of the biggest LNG projects undertaken in Australia a few years ago). These examples demonstrate Japan's national commitment to introduce state-of-the-art and environmentally-sound technologies to meet energy demands in the twenty-first century. To this effect, several end uses of desired raw materials are being developed.

It is important to mention that utilities, transportation, steel manufacturing, and other industries together emit > 70% of the total CO_2 generated. Therefore, a wise selection and implementation of technologies affecting these targeted areas is of prime interest.

ENERGY STRATEGIES--ROLE OF DEVELOPED COUNTRIES

Though adverse effects of industrialization on environment are now well documented, it is hard to imagine giving up all modern day comforts. It is clear that energy usage will expand as world population continues to grow [1]. In this scenario, it is critical that CO_2-mitigation strategies must involve developing countries which are poised for explosive growth well into the next century but do not have policies or resources to effectively manage their environment. With a theme, **"Continual Expansion through Sustainable Development,"** several international efforts are under way. Discussed below are basic elements of two such efforts.

US Initiative

Due to availability of large scientific talent and resources, a large percentage of novel technologies is still developed in developed countries with the US taking a lead. Earlier this year, in response to management of global environment, the US initiated the ADEPT (Assisting Deployment of Energy Practices and Technologies) program through International Technology Cooperation Branch of the Department of Energy. This program comprises six components (Need Studies, Technical Information, Institution Building, Training, Technology and Practices Adaptation, and Technology and Practices Demonstration) and targets developing countries and countries with economies in-transition to promote wise selection and implementation of energy technologies.

Japanese Initiative

To integrate energy policy with global environmental issues of the 1990s, the Industrial Technology Inquiry Commission recently issued an Interim Report [2]. The following goals are set forth:

- Fundamentals of developing energy and technology that will make continual expansion possible.
- Implement "Greenhouse Effect Prevention Action Plan."
- Provide support to neighboring developing countries in energy/environmental areas.
- Implement "Earth Regeneration Plan."

The outcome of this report is formulation of "The New Sunshine Project." A critical aspect of this project addresses "Development of Broad Area Energy Utilization Network System--Basic Concept of Eco-energy City" [2,3]. The ultimate aim is to reduce overall energy needs by 6% and CO_2 by 9% in Japan. The heart of this topic is storage/transport/utilization of low grade industrial waste heat (< 200°C). To implement this program, two innovative technologies are sought:

- Low temperature liquid phase methanol synthesis/decomposition.
- A workable metal hydride system for reversible H_2 storage.

PRIMARY ENERGY OPTIONS

There are several non-petroleum (non-nuclear) energy sources which are of interest:

- Solar
- Wind
- Geothermal

- Natural Gas
- Coal
- Biomass
- H_2O/H_2

Solar and wind are the most favored but at present, these cannot be utilized on a major scale. Water (from oceans) is abundant but an economical H_2-production process needs to be developed. Natural gas and coal are the most abundant and inexpensive available feedstocks. **A sensible exploitation of these C-based resources is desirable.**

CHEMICAL SYSTEMS FOR SPECIFIC APPLICATIONS--A CASE STUDY

Since Japan meets most of its energy needs with imported feedstocks, there is a pressing need to address environmental issues. A recent study [4] was dedicated to consider six coupled chemical systems for transportation of clean fuels.

1. $H_2O \rightarrow H_2 + \frac{1}{2}O_2$
   $H_2 + \frac{1}{2}O_2 \rightarrow H_2O$

2. $3H_2 + CO_2 \rightarrow CH_3OH + H_2O$
   $CH_3OH + H_2O \rightarrow 3H_2 + CO_2$

3. $2H_2 + CO \rightarrow CH_3OH$
   $CH_3OH \rightarrow 2H_2 + CO$

4. $2H_2 + HCOOCH_3 \rightarrow 2CH_3OH$
   $2CH_3OH \rightarrow 2H_2 + HCOOCH_3$

5. $3H_2 + N_2 \rightarrow 2NH_3$
   $2NH_3 \rightarrow N_2 + 3H_2$

6. $3H_2 + C_6H_6 \rightarrow C_6H_{12}$
   $C_6H_{12} \rightarrow 3H_2 + C_6H_6$

For Case 1, H_2 was assumed to be generated via water electrolysis. For electric power generation, the cost calculation followed the order: $H_2O/H_2 > CH_3OH/CO_2 > CH_3OH/HCOOCH_3 > C_6H_{12}/C_6H_6 > NH_3/H_2 > CH_3OH/CO$, with the lowest and the highest cost differing by a factor of two (assuming 4,000 MW power plant and fuel transportation distance of 5,000 km). Since systems containing large quantities of ammonia (Case 5) and benzene (Case 6) may have a problem gaining environmental acceptance, a preference for methanol-energy (Systems 2, 3, and 4) is clearly demonstrated.

It is important to review general criteria used to favor non-petroleum feedstocks for environmental applications. Table 1 lists energy densities [5] and CO_2 emission (per fuel heating value) [4] for selective fuels. The emissions decrease in the order: coal > gasoline > methanol > CH_4 > H_2. H_2 is assumed to be generated through water electrolysis [6,7] and methane is in the form of LNG. The picture changes when H_2 is produced from fossil fuels and the total process (mining → combustion) is taken into account [4]: under this scenario, CO_2 emissions follow: H_2 ($\geq$ 1.0) > methanol (0.8) $\geq$ LNG (0.65 - 0.85). From the energy density point of view, H_2 and CH_4 appear favorites but technical problems associated with storage need to be resolved [8]. Therefore, liquid methanol is of immense interest.

ROLE OF CATALYSIS

Considering that "Catalysis" accounts for > $2 trillion worldwide product stream, it is not surprising that this topic is a subject of several recent reviews [9,10,11,12]. But of interest here is application of catalysis to environmental problems. It is envisioned that CO_2-mitigation could be achieved through more energy-efficient processes. For example, in a recent catalysis meeting, of the 10 critical areas identified for future development, **3 involve methanol and other oxygenates** [13]. These are:

1. Direct methane conversion to methanol using low temperature liquid phase catalysts.

$$CH_4 \rightarrow CH_3OH \qquad (1)$$

2. Since > 40% of all manufactured organic chemicals utilize ethylene feedstock, oxidative coupling of methanol to ethylene is highly desirable.

$$2CH_3OH \rightarrow C_2H_4 \qquad (2)$$

3. The Anti-Markovnikov addition of H_2 and NH_3 to unsymmetrical olefins to selectively produce primary alcohols and primary amines respectively.

$$C = C \xrightarrow{H_2O/NH_3} ROH/RNH_2 \qquad (3)$$

Obviously, catalysis is poised to play an important role in development of energy technologies.

ONGOING BNL CATALYSIS EFFORT

Given the criteria set forth in diverse applications, it is convincing that methanol and other oxygenates will play an important role in the application of clean-energy technologies. It is, therefore, critical that more energy-efficient technologies to synthesize methanol and other oxygenates be developed. Coincidently, a need to develop such catalysts was realized several years ago at BNL. Since Natural Gas and Coal are the most abundant and inexpensive carbon-based non-petroleum feedstocks available, a wise development of these resources is of interest.

Since economical processes for direct conversions of coal (direct liquefaction) and natural gas (direct methane conversion) are yet to be targeted for commercial applications, the BNL effort is dedicated to synthesize oxygenates via the "Indirect Route," i.e., through synthesis gas. The stoichiometry of synthesis gas, produced from these two sources, is of interest:

$$\text{Coal} \xrightarrow{\text{Gasification}} CO + xH_2 + CO_2 \ (x < 1) \qquad (4)$$

$$CH_4 \xrightarrow{\text{POX}} CO + 2H_2 \qquad (5)$$

$$CH_4 \xrightarrow{\text{Steam Reforming}} CO + 3H_2 \qquad (6)$$

Thus, the H_2/CO ratio varies from < 1 for coal-derived syngas to 3 for syngas from steam-reforming of natural gas. In order to maximize C utilization, the BNL effort is dedicated to a One-Step synthesis of identified products of commercial interest without complications of other possible secondary reactions that normally accompany during synthesis of these products. Thus, the Catalyst-By-Design

(CBD) approach for synthesis of methanol and higher oxygenates is ongoing under the "BNL Catalyst Development" program. The central theme of this effort is to achieve:

- ideal syngas source/product match
- favorable thermodynamic parameters
- high activity
- enhanced product selectivity
- liquid phase performance
- one-step synthesis
- low temperature and pressure operation
- better process economics

It is noteworthy that this "Homogeneous Catalysis" approach is finding a niche in the commercial domain of heterogeneous catalysis [14,15,16].

LLTMeOH Technology

The methanol synthesis reaction is commercially catalyzed by a Cu/ZnO heterogeneous catalyst [17].

$$CO + 2H_2 \overset{Cat.}{\leftrightarrow} CH_3OH \qquad \Delta H^{\circ}_{298} = -21.6 \text{ kcal.mol}^{-1} \tag{7}$$

The following disadvantages:

- large capital investment
- < 20% conversion per pass requiring gas recycle

render the present technology unsuitable for several desirable applications which could potentially expand the role of methanol.

A successful application of the aforementioned BNL concept utilized thermodynamically favorable parameters to design a series of catalysts which synthesize methanol in liquid phase with > 95% selectivity and > 90% per pass conversion with balanced gas. This process requires no recycle and the catalyst is inert to CH_4 and N_2. Partial oxidation of natural gas with air is envisioned making the overall process of methanol synthesis from natural gas economically attractive. Summarized below are salient features of the BNL LLTMeOH technology:

- The liquid phase catalyst employed is inexpensive and easy to prepare.
- Liquid medium provides better heat control.
- Methanol (product) is the main solvent used in the process.
- The system operates efficiently between 50° to 150°C at pressure as low as 0.5 MPa.
- A > 90% per pass conversion is routinely achieved.
- The process is highly selective (> 95%) to methanol with methyl formate generated as a byproduct.
- At low levels (< 0.1%), the catalyst is tolerant to sulfur.
- Since the catalyst is inert to N_2, syngas manufacture with air (instead of O_2) is envisioned which eliminates the need for an O_2-separation plant.
- An EPRI-sponsored study indicates that at $1/MMBTU natural gas, methanol could be produced at ~ $0.20/gallon.

The LLTMeOH technology has been patented [18,19] and is the subject of several publications and presentations [20,21,22].

A comparison of operating conditions of methanol process (BNL LLTMeOH versus commercial) is shown in Table 2. Initially, the importance of BNL technology for the following applications was realized:

- Coupling with the Integrated Gasification Combined Cycle (IGCC) System which requires a Once-Through-Methanol (OTM) process.
- Remote natural gas transport where a skid-mounted unit will be economical.

Table 3 compares energy delivery efficiency of BNL LLTMeOH with transported LNG. Given the advantages associated with the BNL approach, several potential environmental applications of methanol synthesis via this technology are being identified.

LLTMF Technology

As emphasized earlier, one of the goals of BNL approach is to maximize energy efficiency. Realizing that,

- Syngas derived from natural gas is ideal for methanol synthesis ($H_2/CO = 2$ via POX).
- Syngas from coal is always CO-rich ($H_2/CO < 1$).
- Alcohols, higher than methanol, normally produce H_2O as a byproduct.

Methyl formate (b.pt. = 31.5°C; m.pt. = -100°C; F.pt. = -19°C) was identified as a product of interest due to the following reasons:

- Storable liquid
- Miscible with alcohols
- Potential as a fuel if produced cheaply. Presently, this product is a specialty chemical.
- Higher CO utilization ($CO/H_2 = 1$)
- Can be easily decomposed to methanol
- IGCC application

The commercial synthesis of methyl formate is carried out in two steps via methanol (equations (7) and (8):

$$CH_3OH + CO \rightarrow CH_3O_2CH \qquad (8)$$

Recognizing that methyl formate was produced as a byproduct during methanol synthesis via the LLTMeOH process, the BNL methanol catalyst was successfully modified to develop a new liquid phase catalyst for synthesis of this C2 oxygenated product [23].

$$2CO + 2H_2 \rightarrow CH_3O_2CH \qquad (9)$$

This patented process [24] has these advantages:

- One-step synthesis
- Low temperature (50° - 200°C) and low pressure (0.5 - 5 MPa) operation.
- High yields with methanol as a byproduct
- Initial estimate indicates that methyl formate is produced at about the price of methanol.

This potentially inexpensive route is expected to promote utilization of methyl formate as a desirable feedstock for several downstream processes [25]. This BNL LLTMF technology is expected to find distinct applications as environmental concerns continue to grow.

SUMMARY

It is beyond doubt that the 1990s will be the "Decade of the Environment" and several state-of-the-art technologies will be implemented during this period. Due to economics and their sheer abundance, natural gas (near term) and coal (long term) will still play a major role in energy supply. But applications of these carbon-based materials will be dictated by their relevance to environmentally-sound technologies. In this respect, catalysis will play a key role.

This paper describes advantages of new technologies, LLTMeOH for methanol synthesis and LLTMF for methyl formate synthesis, based on the liquid-phase concept and developed at BNL. Several diverse emerging applications of these materials are reviewed. A few noteworthy are:

Methanol

- Fuel (M100/M85)
- Fuel-additive
- Feedstock (MTBE, and other downstream chemical processes) coupled with IGCC system
- Waste heat utilization
- Liquid energy carrier for transport of H_2 (12.5 wt% H_2).

Methyl Formate

- Several applications are very similar to methanol.
- A C2 feedstock

Obviously, new applications are emerging for which these technologies were not originally intended. Now, the challenge is to customize technologies for specific applications. To this effect, participation in planned international efforts to implement CO_2-mitigation technologies will ensure that the ultimate goal of a "Clean Earth" is realized.

THE FUTURE

Looking into the future, it is not unrealistic to assume that if the presently targeted global CO_2-stabilization is not achieved, even further CO_2-emission reduction may be implemented through a Carbon Tax proposed in the US (replaced with a BTU Tax which is replaced with a 4.3¢/gallon Gasoline Tax, as of today!) and elsewhere [26]. This scenario may provide incentive to reduce CO_2 at the emitting source. At BNL, CO_2-recycle through catalytic conversion is being considered:

$$CO_2 \xrightarrow{\text{catalyst}} \text{Oxygenates} \qquad (10)$$

This reaction is endothermic and coupling with H_2 is an option to make the overall process exothermic. This and several other activities, ongoing at BNL, are:

- The CO_2-recycle has specific applications to stationary CO_2-emitting sources. But transport of cheap H_2 to site is required for oxygenates synthesis.
- Technologies related to methanol ($MeOH/H_2$ cycle) as a liquid energy carrier for H_2 storage are desirable.
- Improvement in kinetics of the Mg/MgH_2 system (7.6 wt% H_2) for development as an energy carrier for H_2.
- A vision of flexible oxygenates synthesis (for specific environmental applications) skid-mounted (for economical transport of feedstocks located in remote areas) plant is to be realized. The

key to this strategy is development of oxygenates synthesis technologies with closely-related chemistry that will allow such integration.

- Incorporation of waste utilization technologies in the overall scheme.

These and other related technologies, being developed at BNL, will ensure that BNL plays a role in addressing global environmental problems now and in the future.

NOMENCLATURE

ADEPT	Assisting Development of Energy Practices and Technologies
BNL	Brookhaven National Laboratory
CBD	Catalyst-By-Design
EPRI	Electric Power Research Institute
IGCC	Integrated Gasification Combined Cycle
LLTMeOH	Liquid Low Temperature Methanol
LLTMF	Liquid Low Temperature Methyl Formate
LNG	Liquefied Natural Gas
MITI	Ministry of International Trade and Industry (Japan)
OTM	Once-Through-Methanol
POX	Partial Oxidation
US DOE	United States Department of Energy

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TABLE 1. ENERGY DENSITY AND RELATIVE CO_2-EMISSION OF SELECTIVE FUELS[1]

Fuel	Energy Density $\times 10^{-3}$ kJ.Kg^{-1}	CO_2 Emission from Combustion[2]
Coal	29.9	1.0
Gasoline	44.5	0.7
Methanol	21.6	0.65
CH_4	48	0.55[3]
H_2	125	0.0

[1]Data Sources: References [4] and [5].
[2]Relative Ratio
[3]LNG

TABLE 2. A COMPARISON OF METHANOL SYNTHESIS PROCESS CONDITIONS [20]

	BNL LLTMeOH	Conventional[1]
Reactor temperature, °C	110	265
Reactor pressure, psia	150	750
Equilibrium CO conversion, %	94	61
Operating CO conversion, %	90	16
Volume of gas recycle, mols CO/mol prod.	0.11	5.25
Reactor feed, mols CO/mol prod.	1.11	6.26
Overhead gas cooling duty, Btu/mol prod.	4,100	71,000
Separator temperature, for 95% product recovery, °F	163	77

[1]Supp. E. Hydrocarbon Processing, March 1984 (pp. 71-75); July 1984 (pp. 340-345)

TABLE 3. ENERGY DELIVERY EFFICIENCY [20]

	Billion Btu/Day (Higher Heating Value)		
	BNL LLTMeOH	Methanol Conventional	LNG Mixed Refrigerant
Energy Delivered			
Fuel	323	323	323
Electricity	27.5	----	----
Total	350.5	323	323
Energy Consumed			
Process Feed	476.6	487.5	397.3
Process Fuel	16.3	32.8	included
Total	492.9	520.3	397.3
Delivery Efficiency %	**71**	**62**	**81**

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