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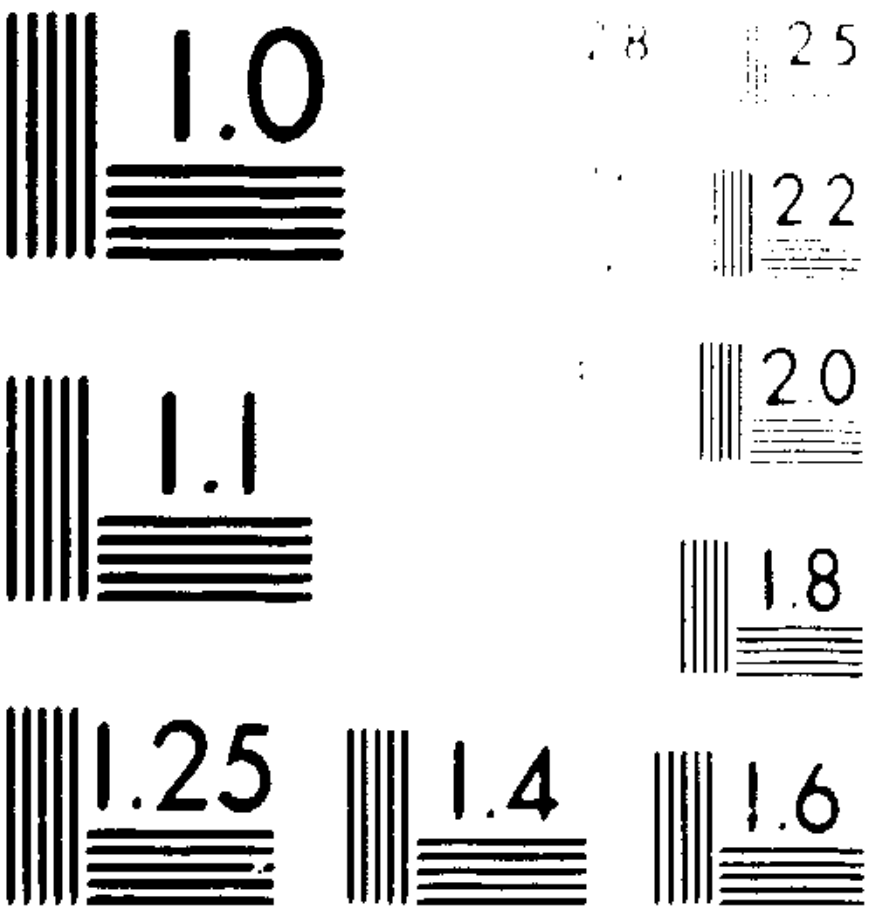
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TECHNIQUES OF DIRECT COAL LIQUEFACTION

DP/CPR/80/048

CHINA

Trip Report on Evaluation Mission to Beijing, People's Republic of China

24-29 June 1982

by M.D. SCHLESINGER, UNIDO Consultant

EXECUTIVE SUMMARY

The purpose of my visit to the Central Coal Mining Research Institute (CCMRI) was to review progress and observe the status of research performed under Project DP/CPR/80/048 - Techniques of Direct Coal Liquefaction. Besides the above activity I reviewed the technical plans for continuing the programme with possible further support from UNDP/UNIDO.

All of the equipment for the project, purchased with UNDP funds, was received and is in operation. Purchase arrangements were made through UNIDO.

The coal liquefaction building has been completed, personnel occupy the offices and technicians are preparing the plant areas. It is anticipated that the 5 kg. per hour and the 20 kg. per hour (slurry rate) units will be delivered this summer. Operations should be underway around the end of 1982. Experimental work up to now has been with autoclaves. About 200 experiments were completed and candidate coals were selected for further testing. Autoclave testing will continue for solvent evaluation, catalyst testing and kinetics studies.

A comprehensive plan of research has been developed for the steps to be taken from the laboratory scale to larger systems.

Introduction

China has a vast supply of coal of considerable variety as to geological age and composition. Of particular interest to this project is the large tonnage of coal that has a high content of sulfur and ash. High sulfur coals are particularly undesirable because the products of combustion pollute the atmosphere and can cause crop damage, health problems and water pollution. By hydrogenating the coal, a clean fuel can be produced with industrially useful elemental sulfur recovered as a by-product.

A research programme began in 1979 by the Chinese Government and since 1980, supported by UNDP/UNIDO, has progressed satisfactorily. New laboratory space is being outfitted and the research programme that precedes larger scale development has been formulated.

Review of the Project

About two years have elapsed since the beginning of the coal liquefaction project in China. In that time a number of important items of equipment were received and placed into operation. Each of the nine items were seen in the laboratory space provided for them. Specific items were:

Item	Date received
1. Viscometer	March 1981
2. Sulfur Analyzer	Sept. 1981
3. Carbon, Hydrogen, Nitrogen Analyzer	Oct. 1981
4. Balance	Aug. 1981
5. Autoclaves (2 AE)	March 1982
6. Rotary Evaporators (2)	April 1981
7. Refrigerated Centrifuge	Feb. 1982
8. Nuclear Magnetic Resonance Unit	Dec. 1981
9. Differential Thermal Analysis Unit	May 1982

During the laboratory visit the items purchased by the Chinese Government were pointed out. Most impressive was the three storey building that contains offices, laboratories and pilot plant areas. The building was only recently occupied and much of the equipment was installed on a temporary basis - but it was in operation.

Some equipment purchased by the Government has not yet been received - in particular a small hydrogenation unit (continuous process development unit) purchased from Xytel Corp. and scheduled for delivery in August 1982. A larger hydrogenation pilot plant will be received from Japan. Foundations for the heavy items are already in place.

Research Accomplished

During the past two years about 200 autoclave experiments were completed using 24 different coals and eight lignites. At present the preliminary evaluation of the coal types is advanced to the point where candidate coals can be selected.

High conversions could be obtained with all coals and is generally a function of the hydrogen to carbon ratio and the reactive maceral content. Younger coals and lignites give higher conversions but the oil yields are lower because of the water and oxygen present in the starting material, i.e. the carbon content of younger coals is lower.

China has large reserves of high sulfur coals and they cannot be used directly without causing serious pollution problems. Thus, it was a primary objective in this programme to utilize these coals in an acceptable manner. Early experiments in the autoclave demonstrated conversions of over 92% with good sulfur and nitrogen reduction.

Another research segment involves the selection of the solvent used for the extraction. Presently, four oils are being evaluated, tetralin, two anthracene oils and a wash oil. These are known solvents but modification can increase their effectiveness. The analysis of these product oils can be used to designate the critical concentration of hydroaromatic components to promote hydrogen transfer to the solubilized coal. It is proposed to use infrared analysis for this purpose.

Mineral matter is also important in the solvent refined coal process, SRC II. The ash in some coals can be catalytically active and by recycling some with the fresh feed, higher conversions can be achieved.

Originally it was thought that a heavy fuel oil produced by the SRC I process would be the preferred product. This material is usually solid at room temperature and not very stable at elevated temperatures. Presently however the thinking is to use the SRC II system wherein the coal products are hydrogenated further and a lighter product, more amenable to catalytic upgrading is formed.

Process Development Units

One process development Unit purchased with Government funds will process about 5 kg/hr of slurry (one third coal). It was built by Xytel Corp., located near Chicago, Il. USA, and a group from the Institute checked the unit before it was shipped to Beijing. Receipt is expected in August 1982 and experiments should begin around the end of the year if everything goes according to schedule.

The second process development unit is being provided by the Japanese who have a co-operative agreement with the Chinese Government. This larger unit will process about 20 kg/hr of slurry that contains one third coal. The equipment will be assembled in the high-bay section of the building with a separate room for control and another for the hydrogen compressor. Ceiling height in the high bay is about 8 m. Bases for mounting the vessels and pumps have been cast in place.

Hydrogen and nitrogen will be supplied from three banks of 15 cylinders each. Gases will be purchased locally and will be costly. In the present configuration there is no hydrogen recycle nor is there a large gas holder to store additional supplies. Because of the limited amount of hydrogen available it is planned to operate the unit for only eight hours at a time, then shut down. This method of operation is not advisable for obtaining process data; the system will not have time to reach equilibrium and meaningful material balances may not be achieved.

Project Document Outputs

The project document calls for five outputs. All of them addressed and mostly completed.

1. A report was submitted on the evaluation of eight lignites that can be used for liquefaction.

2. In the same report as above, the evaluation of tetralin as a solvent was presented.

3. An evaluation was to be made of 5 or 6 solvents other than tetralin and their influence on conversion. A report is completed on four solvents but it has not yet been delivered to UNDP/UNIDO.

4. Semi-annual and annual reports were submitted to UNDP/UNIDO as required.

5. The technical staff was to be increased from 40 to 50. Actually the staff was increased to 60.

Impact of Training

Fellowship training has been very useful to those who spent time at the Pittsburgh Energy Technology Centre. Visits were made to other facilities too so that a good picture was obtained of developments in the United States. In this way some prespective could be obtained as to the development of a coal liquefaction programme for China. Four areas of work were decided upon - First, a plan for the research programme; second, establishment of the procedures to be used; third, that autoclaves would carry the bulk of the basic research load; and fourth, that the main products would be a light oil and a middle oil. These can be later processed catalytically to stable, clean fuels and chemicals, particularly the more useful aromatics.

Other significant guidance obtained by the fellowships for the research at CCMRI were:

1. The use of viscosity measurements as an index of operability.

2. Testing of Chinese coals at the standard conditions used at PETC.

3. Solids separation under laboratory conditions. A similar apparatus will be used at CCMRI.

4. Solvent separation of product oils. This is a well developed procedure using aromatic and aliphatic solvents to separate oils, asphaltenes and the insoluble residue.

5. The 5 kg per hour system purchased from Xytel was altered with 13 changes suggested by the experiences at PETC where a similar unit exists. Operability and safety will be improved.

6. Environmental and safety aspects of plant operation are appreciated and better protection will be provided to personnel responsible for operation of the equipment.

7. Samples of coal tested in autoclaves at PETC and CCMRI will be checked for consistency of results. Data are not yet complete but comparability has been demonstrated.

Fellowships supported by the Government

There has been a major effort to train the technical personnel both at home and abroad. About one third of these people are or have had some form of specialized training. Foreign fellowships are at universities in Ohio, Kentucky and Chicago (Argonne National Laboratory).

At home, the technical staff has been increased from 39 to 60 on the Coal Liquefaction Project. This higher number does not include personnel working on coal and petrographic analyses which apply to the overall research programme at the Institute (CCMRI). Of the 60 professionals, 5 are senior engineers and these are some recent graduates who are attending technical courses and short courses in English and German. By the time the two liquefaction continuous process development units are installed, all of the personnel will have a sound basis on which to build both technical and literature experience.

Tsinghua University

The Chemical Engineering department at Tsinghua University has established a microautoclave system and a thermogravimetric apparatus for the evaluation of hydrogenation catalysts. These catalysts can have a wide range of composition and depends on many factors. The more important ones such as iron in the form of red mud and molybdenum are in good supply in China. These must be evaluated with the coals selected for potential use in a coal liquefaction system.

The Research and Development Programme

A well planned programme has been formulated to carry forward the work from the laboratory to commercial application. There are several parts to the concept. Basic research will help to select the coals that will be utilized. Many coals and coal like materials are amenable to hydrogenation but the ones of most interest in this project are those containing high concentrations of sulfur and ash. The second research project concerns the formation of a stable slurry that can be transported through pumps and pipes to the reactor without separation of the solid and liquid phases. It is also desirable to achieve the highest concentration of coal in the oil, preferably around 50 per cent.

Solvent preparation is also an important step because the hydrogenation reaction takes place through the transfer of hydrogen to the coal. Workable processes depend on control of this hydrogen factor.

The last item consists of catalyst evaluation, a significant component of most chemical processes. In the first stage of liquefaction the coal conversion can be affected by added catalyst or the catalytic effect of ash associated with the coal. In later stages other catalysts can be used for hydroprocessing and the formation of specific end products.

Combination of the four research programmes provides an evaluation of the system and methods to achieve desired results.

A second part of the evaluation scheme involves the procedures used which apply the knowledge gained from the basic research scheme. The initial work, already underway, is the autoclave study. Here, a lot of semiquantitative information can be obtained on a small scale concerning conversion and yields on a particular sample with a particular oil and at a particular set of operating conditions.

The next step involved the processing of the coal in a dynamic system and at constant operating conditions, generally free of the constraints of the autoclave. From the process development units samples can be obtained from periods when the system inputs and outputs are at equilibrium. Analysis of the feed and product streams can then be used to define the system within the limits of equipment size. Ultimately, the demonstration size equipment provides the material and economic information.

A final step is upgrading the primary products to those of industrial use. This process is generally done catalytically so that the final products can be separated into fractions that meet the specifications for particular fuels and chemicals.

As the large scale development proceeds it is important to keep the smaller scale systems in operation. Problems are sure to develop as the systems become larger and these are more easily solved on a small scale. Typical problems involve material failure, catalyst deactivation and product distribution.

Future plans have been established within the Government's programme. Sometime between 1985 and 1990 a process will be selected for commercialization. Besides the development schemes described previously, support activities will include work in microautoclaves, modeling, hydrodynamics, reactor design, materials selection and other related studies. A broad spectrum of disciplines must be available for the development and ultimate commercialization of a process.

Co-operation with Other Countries

A broad research programme is being pursued to bring the process development to an advanced stage within a relatively short time. As stated elsewhere the Government has committed itself to a plan whereby coal will be a major source of direct and indirect energy. Agreements have been reported here and in the technical literature between China and organizations in Japan and the United States. In the development stage is also a potential co-operative agreement with Germany.

The important aspect of the above inter-governmental agreements is that all of the major processes under development around the world are part of the programme in China. Within the broad framework are single-stage, two-stage and indirect systems.

Conclusion

The programme as outlined in the project document for DP/CPR/80/048 was essentially completed. National objectives have been established and the procedures needed to approach the objectives were established. Attainment of the goals will be enhanced by the international agreements for larger scale development which will, in turn, be dependent on the studies achieved with the UNDP/UNIDO project in China.

Recommendations

Progress to date has demonstrated the ability of CCMRI, with the support it received from the Government and UNDP/UNIDO to carry out the research programme. Therefore, I recommend that the work continue as planned with the flexibility to shift emphasis if a particular process is selected for large scale application.

In the separation and analysis of coal liquefaction products, the techniques of gas chromatograph/mass spectrometer combination should be employed to enhance the value of the data obtained from the continuous process development units. Without the application of this technique of structural analysis, the full value will not be derived from the Government's investments in the continuous process development units. It is therefore recommended that the liquefaction laboratory of CCMRI should purchase a gas chromatograph/mass spectrometer combination, which is a vital piece of instrument for this type of work, as well as acquire expertise and experience in this technique through inviting a foreign consultant and through training abroad.

Fellowships and study tours have made significant contributions to the programme and these should also continue. Consultants, too, should visit the laboratory at intervals, especially during the early phase of operation so that discussions can be held that bear on any problems that arise or can be foreseen.

Progress of the programme should be reviewed every 12 to 18 months.

Personnel of CCMRI Contacted

25 June 1982

Fan Weitang	Director, CCMRI
Wang Yinren	Director, Beijing Research Institute of Coal Chemistry
Ovang Yuan	Head, Office of Liquefaction and Gasification
Pang Weijin	Engineer, Translator
Dai Hewu	Deputy Director
Lei Xiang Qin	Translator

Other members of the staff joined the meeting for short periods of time.