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COAL GASIFICATION DP/IND/80/004 INDIA

Technical Report * Mission 25 January to 4 February 1986

Prepared for the Government of India by the United Nations Industrial Development Organization, acting as executing agency for United Nations Development Programme

> Based on the work of K.H. van Heek, Chief Technical Adviser

United Nations Industrial Development Organization Vienna

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Summary

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From January 26 to February 1, 1986 I have visited the Regional Research Laboratory (RRL) in Hyderabad, India, to act as Chief Technical Adviser of the UNIDO funded coal gasification project. The main activities of my visit had been:

- Review of the pilot plant work and the results from test runs made.
- Review of the supporting laboratory work on gasification kinetics and gas conversion as well as reactor modelling.
- Discussion on further programme of the pilot plant and the supporting work.
- Review of the results of training provided for the RRL scientists and of experts visits.
- Assistance in preparation of the Tripartite meeting, discussion of the terminal report and attendance of the Tripartite meeting.

The pilot plant is fully operational and the UNDP provided analytical and electronic instruments are in use. On the whole 7 trial runs have been performed since January 1984, 3 of them in 1985 and 1986 using different Indian coals. Also quenching tests have been made to determine material profiles in the gasifier. Moreover residence time distribution studies for the solids were carried out.

The data gathered from the different runs have been evaluated carefully and in-depth. They show the different gasification characteristics of the coals gasified and have been used to establish material and energy balances of the gasifier. All the results have been presented to me in well arranged tables and diagrams. Thus they are easily accessable whenever they are required in a future design and scale-up work.

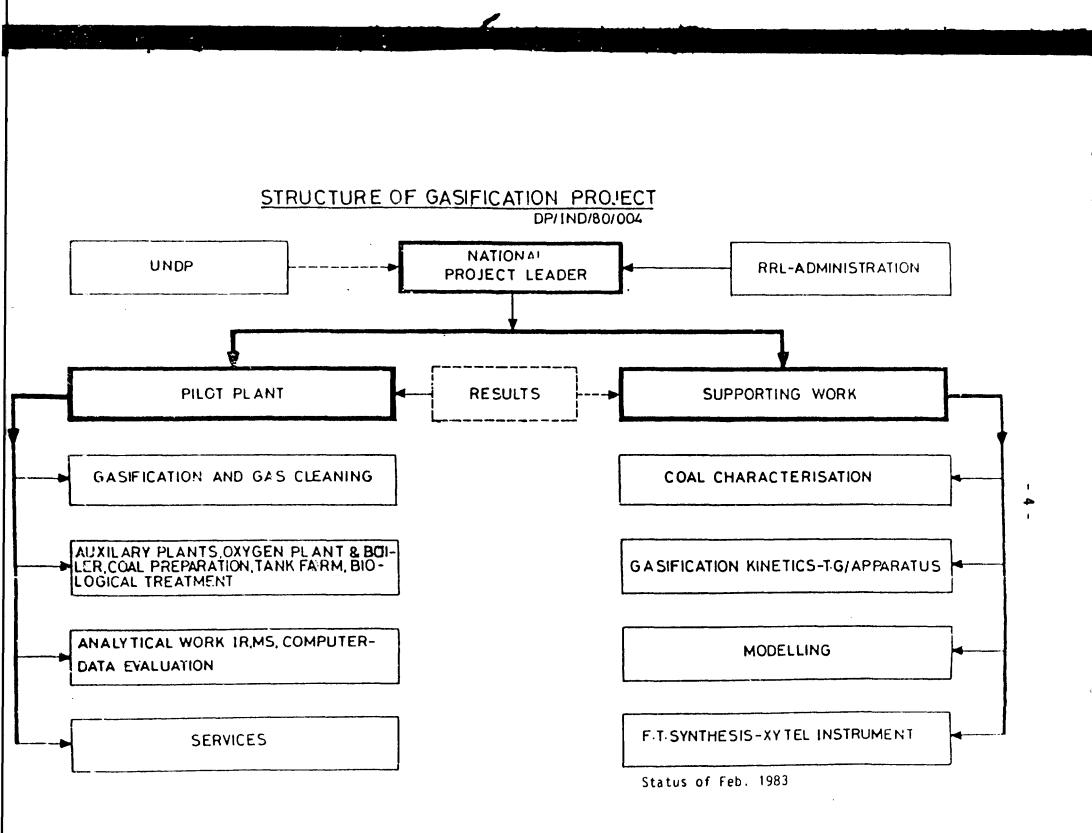
Concerning the future work of the pilot plant it is proposed to test 4 other solid feedstocks from India. This programme could last the next 1 1/2 to 2 years, thereby giving a good overview over the potential candidate Indian coals. A long term programme for the pilot plant could be the demonstration of coal gasification combined with synthesis processes for chemicals and motor fuels. The experimental programme of the plant, enlarged by the gas cleaning and synthesis unit, would include tests of purification processes for raw gases, tests of synthesis reactors and relevant catalysts, analysis and further processing tests for the liquid products and further possibilities for testing coals.

As to the supporting work the studies on gasification kinetics using the thermogravimetric analysis (TGA) have reached a fully satisfactory status. Work is in progress to characterise the reactivity of char prepared from Indian coals (as used in the pilot plant) and to establish kinetic data needed for reactor modelling. The XYTEL datalyst screening unit has been in use in 1985 to test Zeolite catalysts with various active compounds as promotors in 10 gscale for about 1000 hours. Throughout the last year also modelling work had been progressing. In the meantime data predicting the behaviour of the pilot plant gasifier are available using simple and more extended models on the basis of the gasification characteristics of Indian coals as measured in the TGA apparatus. As to the matching of the model predictions with the experiments it can be stated that the simple model shows already the trends quite well whereas the more sophisticated one describes the course the fixed carbonand the particle size profiles in the gasifier satisfactorily.

The experts and training programme of the last year concentrated fully on the improvement of gasification kinetics measurements and the use of the results in reactor modelling. By the training of Mr. M.M. Mallikarjunan at Bergbau-Forschung and Berkeley and by the visit of Dr. Mühlen to RR' the Indian colleagues are in meantime fully aquainted with the experimental technique, the data evaluation and the theoretical background of gasification kinetics as well as the performance of model calculations. In this sense the choices made for the expert's visit and the training have been a good success.

At the end of UNDP's support for the project it can be stated that at the Regional Research Laboratory there exists a group of scientists and engineers, well experienced both in pilot plant work and supporting laboratory and theoretical studies in the field of pressurized fixed bed coal gasification. Thereby the help of the UNDP in financing sophisticated instruments, visits of experts and training has been used very successfully by the Indian colleagues and the project has reached fully satisfactory status and can operate on itself. It forms a sound basis for further development of the Indian coal as a source of energy and chemical feedstock and for possible testing programmes of coal from other countries.

I wish to express my sincere thanks to the national project director, Dr. Vaidyeswaran, and all colleagues of RRL, especially T.S.R. Anjaneyulu, K.S. Rao and M.M. Mallikarjunan, for the open information and the free discussion on status, problems and organisation of the project and the fruitful cooperation whenever changes or improvements of the ongoing and future work had to be considered during the last 4 years. Again the organisation of my visit was excellent and the friendly atmosphere, the kind hospitality and manifold assistance during my stay are highly appreciated.



Time table and activities of the mission

- January/February 1986 -

Date	Day	Action
25/26	Sat/Sun	Travel to Hyderabad
27	Mon	morning: - Visit to the Pilot Plant (Dr. Vaidyeswaran, T.S.R. Anjaneyulu, K.S. Rao)
		afternoon: - Overview and first discussion of pilot plant results (K.S. Rao)
		- Preparation of Tripartite Meeting. discussion of the final report (Dr. Vaidyeswaran, T.S.R. Anjaneyulu, K.S. Rao)
28	Tue	<pre>morning: - Tripartite Meeting including visit to the pilot plant along with the members of the Committee (Not included in this report, as detailed minutes will be circulated by UNIDO)</pre>
		afternoon: - Meeting with the group working on gasification kinetics (TGA) and reactor modelling (M.M Mallikarjunan, K.S. Prasad)
29	Wed	<pre>morning: - Continuation of discussion on TGA experiments and reac- tor modelling Review of expert's visit on TGA (Dr. Mühlen) Review of training received (M.M. Mallikarjunan, K.S. Prased)</pre>
		afternoon: - Detailed review of Pilot plant results (K.S. Rao, T.S.R. Anjaneyulu, M.S. Sulraha Manyam, S. Narayana Reddy, T. Krishnudu)
30	Thu	<pre>morning: Meeting with the group working on gas conversion (M. Janardana Rao, A. Satyanarayana, K.V.L.N. Prakash, G. Muralidhar, K. Venkateswarlo, (Mrs.) V.Durgakumari)</pre>
		 Detailed review of pilot plant results (continued) (T.S.R. Anjaneyulu, K.S. Rao)

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Date	Day	Action
30	Thu	afternoon: - Meeting with managers of BHEL Discussion on future work and possible collaboration (N.N. Rao, Gen. Manager, N.N. Ramakrishna, Dr. Rana)
31	Fri	<pre>morning: Final review of TGA and modelling work (M.M. Mallikarjunan, K.S. Prasad)</pre>
		- General discussion of coal research and future work of Pilot Plant (Dr. Vaidyeswaran)
		afternoon: - Final review of pilot plant results (K.S. Rao)
Feb. 1/2	Sat/ Sun	Travel to Essen
3/4	Mon/ Tue	Writing of the report

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1. Technical status of pilot plant

1.1 General

The technical status of the plant is nearly the same as it was on my last visit in Feb. 1985. In the meantime the biological effluent treatment is completed and reaching for commissioning. The plant has been fully operational the whole last year. Of course there have been minor maintenance problems.

1.2 Control and instrumentation, including UNDP-provided instruments

As described in previous reports, the pilot plant is sufficiently equipped and the overwhelming part of the instruments in use are obviously operating well. For a better and more efficient gas analysis UNDP has provided a gas chromatograph (GC), two sets of Infrared analysers (IR) - for the raw gas and the gas achieved after cleaning - and a mass spectrometer (MS), the latter combined with a computer for the calculation of gas concentrations from the mass spectra and for collecting and evaluation of process data such as relevant temperatures, pressures and flows.

Out of the two sets of IR-analysers available in the plant only raw gas can be monitored for the following streams: CO, CO_2 , CH_4 , H_2 . With regard to the O_2 -analysers, the cell life is only 2 months, because the CO_2 content is too high, a fact which was overlooked by the suppliers. Thus the replacement cells are used already and the module is not in function anymore. The H_2S monitor is operational. The reason, why not all IR detectors are in use is, that some of the detectors installed are found to be defective from the beginning, as has been reported to UNIDO and the supplier. Similar is the case for the spare parts supplied by the firm. On the whole the performance of these units is not satisfactory.

The MS-computer combination worked well at the technical status as reported earlier. The needed data could be collected. Especially the data on gas analysis are used to supplement the information collected by the Orsat and Janak instruments. The problem connected with frequent calibration, as discussed in earlier reports, is practically solved for the next 1 or 2 years, as the consumption of calibrating gas is fairly low. The development of additional software for a more detailed evaluation of the plant using the Minc computer is in progress, however, for the time being the outputs of the computer are at the same status as last year.

With respect to a better evaluation of the process and future work the following additional instrument facilities are planned by the group:

- Measurements of various streams of water going to the system. Relevant meters were not provided in the plant earlier. This additional evaluation will enable to establish a more improved material balance;
- Temperature profiling of the gas generator to establish a better data base for modelling as has been suggested by several UNDP experts;
- Instruments for air/steam runs, especially steam and air flow recorders.

2. Operation and trials performed

2.1 Runs and tests performed

From 18.01.1984 to 30.01.1986 trials runs have been carried out as listed in the following:

Trial Run No	Duration	Coal	Gasification agent pressure
1	18.01 21.01.1984	Ramagundam	air steam 5 to 10 bar
2	02.03 30.03.1984	Ramagundam	air steam 50 to 10 bar
3	16.07 24.07.1984	Ramagundam	air steam 5 to 10 bar
4	20.08 04.09.1984	Ramagundam	oxygen steam 10, 15, 20, 21 bar
5	06.02 18.02.1985	Monuguru	oxygen steam 10, 15, 20, 24 bar
6	08.08 19.08.1985	Hindustan Lalpet	oxygen steam 10, 15, 20, 24 bar
			air steam 10 bar
7	22.01 29.01.1986	Samla	oxygen steam 10, 15, 20 bar

The performance of the run 1 to 5 was described in my last report. Run 6 was done with a coal from Wordha Valley coal field which has high moisture (appr. 12 %), high volatiles (appr. 42 % dmf) and relatively low ash content (16 %). The Samla coal is low ash (15 %) and high volatile (42 % dmf) and weakly caking (caking index: < 5). With this coal the first step was made to study the behaviour of caking coal. The coal was processed without distributor and the run successfully completed. The data are under evaluation. Besides these runs the following special tests have been made:

- Quenching experiments following the runs 4 and 5 and recently following run 7: The procedure is described in my last report, except that in run 7 the quenching had been carried out by a flow of nitrogen, whereas the former quenching was done by locking the gasifier and allowing it to cool down by the jacket water. By nitrogen cooling the quenching time goes down from 10 to 3 days. Moreover the ritrogen suppresses the heterogeneous reactions totally.
- During March 1985 residence time distribution (RTD) studies were carried out. Three tests were performed during the gasification of Monuguru coal at 20 bar under full operating conditions. In these experiments about 120 kg of mullite pebbles 6 - 25 mm size have been charged in between the coal charging and their appearance in the ash has been monitored. Resistance to temperature of the pebbles was tested before in the laboratory, size and density suited that of the coal bed.

2.2 Data evaluation and assessment

The runs 4 and 5 as well as the special tests mentioned before have been completely evaluated, whereby the informations achieved can be classified as follows:

- evaluation of gasification characteristics of the coals,
- material and energy balances,
- internal profiles of the gasifier,
- residence time distribution of the solids in the gasifier.

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As to the gasification characteristics the steam oxygen ratio was optimized taking into consideration for each coal the CO_2 level in the output gas, the texture of the ash and the outlet temperature. Then the specific consumption of steam, oxygen and coal per unit of raw gas were precisely evaluated. Also the yield of tar, oil and gas liquor have been determined. Thereby each coal has shown its own different gasification-characteristics. All these values form the basis for the design of commercial processes for given coals.

Material balances have been established from the data collected during stable periris of plant operation. These include elemental balances for C, H and O for the gasifier. In the absence of measuring instruments of flow of water to the jacket, as mentioned earlier, this amount of water was taken as the difference between the output and input to the gasifier.

The energy balance has been calculated from the material balance and operational conditions (temperatures, pressure) established in the plant. The balance between input and output energy is in good agreement. The limitations are the unknown effects of errors at different stages of collection of information, which I recommended to estimate to the extent possible. Also so far the losses by radiation and coal dust were not taken into account. As to the efficiencies it is advised that calculation of cold gas efficiency of the gasifier will be sufficient.

As results of the quenching experiments the internal profiles of the gasifier concerning total carbon, fixed carbon, hydrogen, volatile matter, particle size and bulk density have been determined. These give information on the extent of different zones (drying, devolatilization, gasification, combustion) and the degree of consumption of carbon in these zones during its flow through the gasifier. They show that the main coal conversion takes place in two sections: in the upper part by pyrolysis, whereby the extent of the zone is between 20 and 40 cm and in the lower part by gasification and compustion. The data have been of great use in the checking of gasifier modelling predictions as will be mentioned in chapter 4.3.

The residence time distribution experiments have shown that there is hardly a backmixing of solids in the gasifier. All three tests conducted have confirmed a plug flow behaviour. The average residence time was about six hours after which the tracing pebbles appeared with a half width time of 30 minutes.

All experimental results have been presented to me in well arranged tables and diagrams and - as the balances are concerned - summarized in process flow sheets. Thus they are easily accessible whenever they are required in a future design and scale-up work. From all my discussions I can confirm, that all calculations have been done very carefully and can be checked at any time in required. To sum up: the data achieved form a sound basis for further design work in the field of fixed bed gasification and are nowhere published in literature even not for other coals.

3. Future work of the pilot plant

In the short time programme it is proposed to test other coals and solid feedstock from India. Among these are:

- coal from Singrauli (U.P.),
- lignite briquettes from Neyvely (T.N.),
- semicoke from Singareni Collieries (A.P.),
- one more coal to be identified.

These runs will also include air-steam gasification to generate information required for combined cycle electricity generation. If all these coals are tested in the same way as above, then a good overview over the potential candidate Indian coals will be available.

During these tests, the measurement of temperature profiles inside the gasifier will also be carried out as mentioned earlier.

It has been discussed also, what could be the long term programme for the pilot plant. Among other possibilities the following proposal seems to be worth to be taken into detailed consideration:

Enlargement of the existing facility by a synthesis unit for chemicals and middle distillates, which can be used for kerosine substitutes. Such an exten-

sion of the plant offers advantages such as

- test of gas purification processes for raw gases from the gasification of Indian feedstocks with respect to the adjacent synthesis step,
- test of synthesis reactors and relevant catalysts,
- analysis and further processing of the liquid products with sufficient amounts, as the plant output would be in the order of 100 200 kg liquid products/day,
- further possibility for testing coals.

These possibilities must be discussed, taking into account the future requirements of the Indian energy and chemical industry. Also it should be taken into consideration that by this the present know-how of RRL-Hyderabad both in the field of gasification and catalyst development can be used furtheron. Also the knowledge of other Indian institutes already dealing with catalyst development could be pooled up.

4. Supporting work

4.1 Kinetics of gasification (TGA)

By the training of Mr. M.M. Mallikarjunan at Bergbau-Forschung in October 1985 and the visit of Dr. Mühlen to the laboratory as UNIDO expert (see chapter 5) in November 1985 the TGA work has reached a fully satisfactory status. This concerns both the performance of the experiments and the kinetic evaluation of the data using the computer.

Since then about 10 runs with steam and 20 runs with CO_2 have been performed to study the kinetics of gasification of Monuguru char prepared in the Gray King assay (800 °C, 1 h) at different pressure levels (1 - 40 bar). The results show already, that the Indian coals might behave differently from the German ones, which could be ascribable to the high content of ash, which is intensively intergrown with the organic matter. Work is in process with the aim to establish the Langmuir-Hinshelwood type of kinetics needed for modelling, which takes into account the influence of the product gases. This has to be done stepwise from pure gases (H_2O , CO_2 , H_2) to mixtures. The recommendation is, to perform these experiments not only with the Gray King model char but also with the chars from the quenching experiments, which may show a markedly higher reactivity.

On the whole, the Indian colleagues are in the meantime fully acquainted with the experimental technique, the mode of data evaluation and the theoretical background of gasification kinetics. Thus this group forms a good basis for gasifier modelling, which is needed for the design and assessment of gasification processes. Thereby it should be mentioned that the data collected are very useful also for others than fixed bed gasifiers e.g. fluidized bed reactors.

4.2 Gas conversion (XYTEL-instrument)

The main aim of the supporting work on the conversion of gases as they are produced by gasification is to manufacture olefins as precursors for polymers. Moreover, especially during the last year, also the aspect of producing hydrocarbons as motor fuel has been taken into account.

The instruments provided by UNDP funds - XYTEL testing unit and GC - have been fitted into the ongoing stional programme, which consists of

- preparation of catalysts,
- screening tests in "minireactors" (g-scale),
- tests in larger scale (Xytel),
- analysis of the products achieved (GC).

The catalysts studied are

- Zeolite based catalysts, ion exchanged Fe, Fe-Mn, Fe-Cu with differen promotors,
- other Iron-based catalysts.

The work on the XYTEL apparatus concentrated in 1985 on tests in 10 gr-scale of Zeolite catalysts with various active compounds as promotors. Thereby especially the physical conditions were changed. The tests includes results on selectivity and life studies. For these investigations the XYTEL instrument was in use for about 1000 hrs and showed reliable performance. - Only the graphic display is still not working, a failure, which is there since last year as mentioned in my last report.

4.3 Gasifier modelling

As it has been mentioned in my last report, the modelling group generally is working on 3 models. These are

- 1) one-dimensional, neglecting the radial effects,
- 2) two-dimensional, including also the heat flux from the gasifier axis to the jacket,

as been introduced by Prof. Denn, who visited RRL twice as an UNDP expert and

3) one-dimensional heterogenous,

as explained by Prof. Klose, who also visited RRL as an expert on the project. In the heterogeneous model the temperatures between the solids and the gases are regarded to be different, whereas in the homogeneous cases 1 and 2 they are asumed to be equal.

The programme concerning 1 is operational on the RRL computer, the programme for version 2 is now being installed at RRL computer. However, M.M. Mallikarjunan has performed about 12 runs on the computer at Berkeley, where he was staying during a training mission in October 1985.

Following the discussions and recommendations (f my last visit, the input data have been improved. Especially the kinetics of the gasification of Monuguru char - as processed in the pilot plant - have teen used. These were measured during the stay of M.M. Mallikarjunan at Bergbau-Forschung in October 1985. Also the experimental results from the quenching experiments have been used for adjustments in the programme, especially regarding the combustion zone and for comparison with the predictions of the model, thereby showing its degree of validity.

As to the results: Prediction for the gasifier of the RRL pilot plant have been made with the two versions concerning the following items:

- gas composition at the outlet,
- profiles of C, ash, volatile matter (for comparison with quenching experiments),
- particle size profile,
- temperature profile (to be compared later with the measurements planned; see chapter 3),
- simulation of the gasifier for various input conditions e.g. C/O_2 ratio, O_2 /steam ratio, inlet temperature.

The data base had been the feed conditions of the pilot plant of run 5, Monuguru coal and the kinetics as said before.

For the matching of the model predictions with quenching experiment data, outlet gas composition and unconverted carbon in the ash, the following conclusions can be drawn:

- model 1 shows the trends already quite well,
- model 2 matches the fixed carbon- and particle size profiles definitely better than version 1.

For further work it can be recommended:

- improvement of kinetic data taking into account the inhibiting effects of product gases and introducing the Langmuir kinetics,
- concentration of work on version 2, especially to have it installed on the RRL computer as soon as possible.

The status of the more complicated version 3 is almost the same as last year. I recommend that it could be taken up later, as for the time being version 2 seems to be satisfactory.

On the whole, I am very content with the progress which has been made in this field. It proves also that it has been a good decision to have Prof. Denn as expert and to send M.M. Mallikarjunan twice to receive training in this field.

5. Experts and training programme

From Nov. 4 to Nov. 30 Dr. H.-J. Mühlen from Bergbau-Forschung, W. Germany was sent as an experi on gasification kinetics to the project. He had been chosen mainly for the reason that Bergbau-Forschung is using the same experimental and evaluation technique. His visit can be regarded as very successful, as - as mentioned in chapter 4.1 - after that the RRL scientists working on the TGA have been fully instructed and capable to perform the work.

Mr. M.M. Mallikarjunan had been on a study tour in October 1985. Thereby he visited first Bergbau-Forschung and got more acquainted with the TGA technique uncluding all evaluation procedures. He had taken part in practical experiments on Monuguru char, he had brought with him, the results of these have been used already in modelling (see chapter 4.3). The last two weeks of his mission he has spent with Prof. Denn's group at Berkeley, Cal. USA. Thereby it has been of utmost importance for the future steps in modelling that he could perform runs with version 2 (see chapter 4.3).