COMMISSION OF THE EUROPEAN COMMUNITIES

FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS

First Colloquium
Ispra (Italy), April 29, 1971

EUR 4772 e

FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS

by J. FLAMM and D. VAN VELZEN

Commission of the European Communities
Joint Nuclear Research Centre — Ispra Establishment (Italy)
Technology
Luxembourg, May 1972 — 62 Pages — B.Fr. 85.—

A First Colloquium on Fluidised Bed Coating of Nuclear Fuel Particles towards Industrial Scale Operations was held at the EURATOM Joint Research Center, Ispra, Italy on 29 April, 1971.

Abstracts of the papers presented are given together with the ensuing discussions.

The meeting served to review the subject matter with various organizations active in this field and to sound out spheres of common interest.

A second colloquium on the same topic is scheduled to be held at the Euratom Joint Research Centre, Ispra, Italy on 26-27 October, 1971.

EUR 4772 e

FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS

by J. FLAMM and D. VAN VELZEN

Commission of the European Communities
Joint Nuclear Research Centre — Ispra Establishment (Italy)
Technology
Luxembourg, May 1972 — 62 Pages — B.Fr. 85.—

A First Colloquium on Fluidised Bed Coating of Nuclear Fuel Particles towards Industrial Scale Operations was held at the EURATOM Joint Research Center, Ispra, Italy on 29 April, 1971.

Abstracts of the papers presented are given together with the ensuing discussions.

The meeting served to review the subject matter with various organizations active in this field and to sound out spheres of common interest.

A second colloquium on the same topic is scheduled to be held at the Euratom Joint Research Centre, Ispra, Italy on 26-27 October, 1971.

EUR 4772 e

FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS

by J. FLAMM and D. VAN VELZEN

Commission of the European Communities
Joint Nuclear Research Centre — Ispra Establishment (Italy)
Technology
Luxembourg, May 1972 — 62 Pages — B.Fr. 85.—

A First Colloquium on Fluidised Bed Coating of Nuclear Fuel Particles towards Industrial Scale Operations was held at the EURATOM Joint Research Center, Ispra, Italy on 29 April, 1971.

Abstracts of the papers presented are given together with the ensuing discussions.

The meeting served to review the subject matter with various organizations active in this field and to sound out spheres of common interest.

A second colloquium on the same topic is scheduled to be held at the Euratom Joint Research Centre, Ispra, Italy on 26-27 October, 1971.

COMMISSION OF THE EUROPEAN COMMUNITIES

Proceedings
of the First Colloquium on

FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS

Ispra (Italy), April 29, 1971

prepared and edited by

J. FLAMM and D. Van VELZEN

Joint Nuclear Research Centre
Ispra Establishment - Italy
Technology Division

Published by the
Directorate -General Dissemination of Information
Centre for Information and Documentation - CID

Luxembourg, August 1972

EUR 4772 e

NOTICE

The Commission of the European Communities is not responsible for the use which could be made of the information presented in this document.

TABLE OF CONTENTS *)

*) Manuscript received on January 5, 1972

FOREWORD

Under contract with the Dragon Project CCR-EURATOM, Ispra commenced by mid 1970 room-temperature model work on fluidized bed coating systems.

It was clear from the beginning that the coating step is a major cost determining factor in HTR fuel manufacture and that there is still a need for further serious investigations. Efforts in this field are presently widely spread and it is considered paramount to arrive at some point of coordination. Although there has been some mutual exchange of information in the past between individual European groups, active in the field, the system used was thought to be fairly incoherent particularly when viewed in the light of development potentials towards industrial scale operations. In the latter context present individual efforts (running in parallel and to some extent in duplication) are considered essentially too small to exhaust the problems of the coating process.
It is here that some thought should be given to streamlining a common programme on principle problem areas without infringing on profit-orientation of private manufacturers.

Many problems are unsolved in optimization of equipment and processes, and the efforts required are of considerable scale. Funds available, as almost anywhere in research and development today, are very limited and cannot justify the dispersion of activities.

The close cooperation between CCR-EURATOM, Ispra and the Dragon Project in this field as well as the supranational status of these two organizations, provided an excellent launching pad to call on the various organizations for a concerted review of the subject matter and for sounding out spheres of common interest.

The meeting had to be postponed twice for internal and external reasons; but finally, the interest shown in this colloquium was both surprising and gratifying to its organizers. Both the number attending and the number of presentations offered exceeded original expectations.

Whatever the success of this meeting may have been, thanks are due to the persons who participated in its organization and conduct.
Special thanks are due to S. Finzi, for his encouragement and support in preparing as well as chairing the meeting, C. Vivante for initiating contacts with the participants, M.S.T. Price and R. Lefevre for their contributions in programme planning and A. Faraoni and H. Langenkamp for their help in the smooth conduct of the meeting.

Finally, the interest of a group of participants in CCR-EURATOM, Ispra, experimental activities must be mentioned, which were subject of discussions the day following the colloquium. Here it was agreed to organize a second colloquium on this subject to be held at CCR-EURATOM, Ispra on 26-27 Oct. 1971.

FIRST COLLOQUIUM ON FLUIDIZED BED COATING OF NUCLEAR FUEL PARTICLES TOWARDS INDUSTRIAL SCALE OPERATIONS.

(Thursday, 29 April, 1971 - CCR - EURATOM, ISPRA - Italy)

LIST OF PARTICIPANTS

Name	Company Affiliation	Country
AERTS L.	Belgonucléaire	B
TRAUWAERT E.M.	"	B
DECAMPS F.	C.E.N. - MOL	B
VAN GEEL J.	" "	B
WALLROTH C.F.	K.F.A. - JÜLICH	D
VYGEN P.	NUKEM	D
BASCHWITZ R.	CERCA - Romans	F
MORIN C.	" "	F
SOULHIER	C.E.A. - Grenoble	F
THOMAS J.P.	S.I.C.N.	F
FACCHINI A.	Agip Nucleare - Milan	I
MORCELLI G.	" " "	I
HIBBERT N.S.	UKAEA, RFL, Springfields	GB
DE BACCI M.	Dragon Project	
LEFEVRE R.	" "	
PRICE M.S.T.	" "	
TYTGAT D.	Euratom - Brussels	
VIVANTE C.	" "	
LUNGAGNANI	" Petten	
SOUILLART C.	" "	

Name	Company Affiliation
ANGELINI A.	Euratom - Ispra
BOTTELIER P.	" "
ERBEN E.	" "
FARAONI A.	" "
FLAMM J.	" "
FINZI S.	" "
GRASS G. (part-time)	" "
LANGENKAMP H.	" "
LOOMAN B.	" "
MUSSO G.	" "
NOBEL L. (part-time)	" "
VAN VELZEN D.	" "
VOLTA G.	" "

SUMMARY REPORT

1. Opening remarks by chairman - S. Finzi

Gentlemen,

I would like to welcome you here to Ispra for an informal meeting on Fluidized Bed Coating of Nuclear Fuel Particles towards Industrial Scale Operations.

You are in a better position than I, to judge, if there is anybody missing from the "coating family" in Europe. Looking at the attendance list we can certainly assume that some real interest has been roused for all parties active in the field. This being the first gathering on this topic, we can only hope the trouble you took in coming here will be made worthwhile.

To-day's programme of presentations best reflects the variety of activities in the various establishments, and indicates at the same time that after some ten years of development efforts, there is still a need for model research in support of industrial scale operations. Therefore, it is hardly necessary to stress that present-day coating practice has only reached the stage of sketchy analysis with indicative views on future trends in and guides for development.

I presume that all of you working in this field have had an exchange of views at one time or another time; but under the circumstances it was thought that a meeting like this one today would serve, in the first instance, to report, condense and digest preponderant problems observed by the various investigators and manufacturers. To my own knowledge a regular exchange of expert views on a scale like this does not exist as yet in the field.

Therefore, today's meeting may well serve to crystallize the participants' opinion on the necessity and benefits of such regular information exchange.

As you will have noted, today's programme has been arranged to give a situation report highlighting economical aspects and development efforts in the first two papers. We will then turn to a more general technical assessment followed by three presentations on supporting experimental work. Time for discussion during the morning is necessarily limited. It has therefore been decided to give combined discussion time on the first two and again on the following four papers. The first presentation of the afternoon session blends in with the experimental work, leading on to two presentations on industrial aspects. The following general discussion should then also provide some time for any unanswered questions from the morning session.

2. The economics and future development of fluidized bed coating of nuclear fuel particles (M.S.T. Price)

For the prismatic HTR comprising fuel pins in moderator blocks, a recent Dragon Project assessment of fabrication costs can be summarized as:

Graphite fuel pins and moderator blocks - material and machining	52%	(approximately equally divided between pins and blocks)
Interest charges on fissile material hold-up and losses	11%	
Conversion from UF6 to finished fuel element, overheads, interest on non-fissile stocks, R and D, contingency	37%	

Although it is correct to point out the large contribution of graphite to the fabrication cost, it must also be observed that the general level of HTR fabrication costs is too high.

This note examines one particular topic, the cost of coating and from an analysis of a standard cycle, derived from Dragon experience, attempts to deduce the routes by which economies can be achieved. Most of these affect the direction taken by the forward development programme.

Reference coated particle

The reference coated particle was defined as follows:

Kernel

mean diameter	800 μm
composition	UO_2
kernel density	80% dense (i.e. 8.77 g/cm^3

enrichment in U-235 6%

Coating

Layer No.	Layer description	Layer thickness (μ m)	Layer density (g/cm^3)
1	Porous buffer	25	1.1
2	Seal	30	1.6
3	Inner higher density isotropic pyrocarbon	30	1.8
4	Silicon carbide	35	3.2
5	Outer high density isotropic pyrocarbon	35	1.8

Other input data comprise gas costs, maintenance times, utilities other than gases, salaries and labour content and a number of assumptions on capital, amortization and maintenance charges.
The cost of one coating furnace (152 mm diameter, single nozzle system) has been estimated as £ 15,000 - uninstalled, but equipped with all necessary automation devices.

Using these economic assumptions, the cost of the unit operation of coating can be estimated to be about 6.900 £/teU, the make-up of this figure being given in Table 1. It must be emphasized that these are raw figures which do not take into account costs arising from scrap recycle, quality control, changing rooms, overheads, profit, etc.. Nevertheless, such a "standard" cost is useful as a means of illustrating the ways in which the cost of coating could be reduced.

TABLE 1

The cost of coating with the reference coating

I t e m	Cost (£/teU)		%	
Utilities				
- gases	1107		16.1	
- electricity	912		13.2	
- water	162		2.4	
		2181		31.7
Salaries		745		10.8
General expenses		1306		19.0
Amortization and interest charges				
- for equipment	1901		27.6	
- for buildings	251		3.6	
- interest on working capital and insurance	53		0.8	
		2205		32.0
Maintenance charges		447		6.05
Total specific cost of coating		6884		100.0

COST OF COATING WITH THE REFERENCE COATING = 6884 £/teU for a 100 te U/y throughput

We can view the gases used for deposition of the various coating layers as the only "quasi inevitable" costs (520 £/teU or less than 8% of the overall cost). Deducting the total gas cost from the overall standard cost we can then allocate all the other costs directly in proportion to the time involved for each operation. Adding back the non-deposition gas costs, as appropriate for each operational stage, we abtain the cost allocation given in Table 2.

TABLE 2

Cost allocation by operations for coating with the reference coating.

Operation	Time involved (minutes)	Fluidizing gas costs (non-deposition gases only) (£/teU)	Overall cost allocation (£/teU)
Deposition of layer			
1	10.8	14.1	148
2	41.4	67.6	580
3	38.4	48.8	524
4	141.7	319.2	2075
5	51.8	65.5	707
Temperature adjustment between layers	18.0	25.3	248
Heating up and equilibration	10.0	16.4	140
Cooling and unloading	125.0	29.9	1578
Maintenance	29.4	nil	364
	466.5	586.8	6364
Gases for layer deposition			520
Total			6884

Two items contribute more than half to the cost of coating - the deposition of the silicon carbide layer and the time taken to cool the furnace for unloading at the end of the batch deposition process.

The high cost of the silicon carbide layer is a consequence of the restriction on the concentration of silane vapour to 3% by volume.

There are two obvious questions:

a) is a silicon carbide layer a technical requirement?
b) can satisfactory silicon carbide coatings be laid down in large scale furnaces with silane vapour concentrations appreciably greater than 3%?

The time interval during cooling of the finished coated batch down to a temperature suitable for handling is completely dead time. A great saving (~14%) would come from hot unloading at 1000°C because the cycle time could be reduced by ~ 65 minutes.

The rate of deposition of coatings from methane is slow, in the range 0.5 - 2 μm/minute. A molecule with a greater number of carbon atoms can potentially increase the deposition rate.

Huschka and Schmutz pointed out the economic advantages of using propylene instead of methane. The main advantage lies in the greatly increased rate of deposition which reduces the specific labour content and increases coating furnace productivity. In addition, a lower deposition temperature can be employed, leading to reduced energy and cooling water costs, as well as increasing the life of the graphite components. The lower deposition temperature also enables the seal layer to be deleted.

A combination of replacement of methane by propylene and replacement of the silicon carbide layer by a coat based on propylene would, using very pessimistic assumptions, lead to at least a 63% reduction in the cost of coating. With hot unloading superimposed, the reduction would have been more than 70%.

Additionally, one of the most normal avenues for economy is to scale up to larger batch sizes since this will considerably reduce the specific labour cost and lead to reductions in other areas.

This gives a reason for using a multinozzle system to permit a larger batch size for given sphericity and coating thickness variation criteria.

It is suggested that the main developments will be dictated by economic considerations as part of a drive to reduce the cost of HTR fuel fabrication. Topics will include:

a) development of furnaces with larger batch sizes particularly to reduce the specific labour cost;
b) development of automatic control of furnace temperature and gas flows. Present experience shows that a small but finite number of runs fail due to operator error;
c) investigation of silicon carbide deposition conditions to:

   A - increase the deposition rate
   B - increase the average and thereby increase the minimum fracture strength of the silicon carbide layer

d) investigation of pyrocarbon deposition conditions and fluidization to increase the deposition rate e.g. by the use of propylene and to achieve the smallest standard deviation of coating thickness;

e) investigation of methods of increasing the flow rate used in coating with hydrocarbons and methyltrichlorosilane e.g. by using a multinozzle arrangement and by employing model and tracer studies;

f) investigation of hot unloading of coated particle batches to increase furnace productivity;

g) investigation of factors affecting the residual hydrogen content in pyrocarbon coatings deposited at lower temperatures (e.g. when using propylene) including the effects of post-coating heat treatment;

h) development of peripheral equipment e.g. for slot sieving and shape separation of finished coated particles;

i) maximization of process controls and minimization of quality control, e.g. adjustement of the coating conditions in relation to kernel properties, control of the increase in mass on coating, etc...

3. Using cost studies of the coating process as a guide for development scientists and engineers (R. Lefevre)

We have been presented with a breakdown of the cost of fuel particle coating and the areas in which the more important cost reductions can be obtained have been put forward. It is now the work of the Delelopment Scientists and Engineers to make "the dreams come through".

It is then necessary to put down a priority of work and to do so the following four points must be taken into account:

a) the cost saving that can be achieved by tackling a particular problem;
b) the estimated effort required to achieve the target improvement, this more particularly within the context of the total effort available for development within the organization where the work is performed;
c) the time required to prove that what has been achieved yields a satisfactory product;
d) the interaction between one subject and another.

If the two first points are self-explanatory the two last points are worth commenting upon.

The time required to prove that a new system works can vary enormously depending on the subject. Proving a hot unloading system requires some design effort, and the testing only requires a few experiments whereas an improvement related to design of a coated particle must be proven by long and expensive irradiation experiments. Consequently, the time available between now and the freeze of the design of the fuel element and its manufacturing plant must be taken into account in the choice of the subjects that will be tackled.

The definition of a satisfactory product also needs commenting on. There has been, up to now, a tendency in the field on HTR fuel to dismiss anything which is less than perfect. We must depart from this attitude which leads to very high costs and not only search for "the best at the lowest price" but also go further and aim at "the lowest cost for a product which is good enough".

Commenting on the fourth point which is the interaction between one subject and another, it is not only obvious that there are interactions within the coating field (e.g. between the design of a hot unloading system and a multi-nozzle furnace), but also between the successive fields involved in the manufacture of the final product: the fuel element. A similar interrelationship of course exists between the fuel element design and the fuel manufactured.

Assumptions

The cost study, presented by Mr. Price, is based upon a number of assumptions, as a "reasonably optimized production plant".

The first assumption made consists in considering that the design of the particle will be the Dragon Reference Particle, nominally an 800 μm diameter kernel with 20% porosity and a succession of five coating layers having a total thickness of 155 μm including 35 μm of silicon carbide. It is clear that a change to a different type of particle can drastically alter the overall cost of the fuel. For instance an increase of the coating thicknesses will result not only in a higher manufacturing cost per tonne of uranium but also lead to a lower heavy metal density in the reactor and thus to a general cost increase

A capacity of 100 te U/year is assumed for the production plant. A smaller unit will necessarily lead to a higher cost, at least of the final product, due to a proportionally higher investment particularly relative to auxiliaries and testing equipment.

A fully automatic 6" diameter column furnace with a single gas injection nozzle has been chosen. Its cost with all auxiliary equipment was estimated at £ 15,000. No such furnace exists at the Dragon Project where the larger size available is 5" diameter.

A large effort is necessary to prove the suitability of such a furnace and to find the appropriate deposition conditions.

As to the automation of the furnace a fully designed project exists but all work concerning its realization has been stopped. The penalty for not working in this area in time for the design of a production plant is twofold. The labour content of the cost of coating is put at 10.8% assuming one process worker for two automatic furnaces. If the furnaces are manually operated one needs two process workers per furnace corresponding to a 32.4% penalty on the present cost estimates. Furthermore it must be realized that the rate of mistakes for an operator varies from 1 to 5% according to his skill. This will lead to a large rejection rate which again increases the cost.

A reasonable amount of effort is required to meet the assumptions but it is worth mentioning that in each area there is a strong chance of reaching the target if the adequate development effort is made available and that none of them includes a long proving period.

Possible cost savings

It must be realized that the most important savings possible are not in the coating field but in the graphite components (52% of the overall cost of fabrication approximately equally divided between pins and blocks). Cheaper manufacturing methods or designs and the possibility of re-using some of the components must be investigated.

A saving of about 14% of the coating costs can be achieved by a hot unloading system. The design of such a system exists within the Dragon Project but is up to now untried. The effort necessary to complete the work is very small. The time to prove the system is short and it becomes therefore one of the first priorities in the list of the projects to be tackled. There are two problems associated with the hot unloading. There is no benefit in having hot unloading if there is no hot loading system. This is currently achieved by gravity but this means also that the UO_2 kernels must be strong enough to withstand the thermal shock when they are loaded. The second associated problem is relative to the soot control.

The cost study suggests that a large saving ($\sim$30 % with the assumptions employed) can be achieved if propylene is used as coating deposition agent instead of methane. The savings arise not only from reduction of the deposition time and power but also from the fact that the low deposition temperature enables the sealing layer to be deleted.

The experience on propylene coatings laid down at reasonable concentrations lies mainly outside the Dragon Project but it is evident that work must be performed in this field as a matter of urgency in view of the large saving possible and of the long lead time associated with a change in coating source.

The proportionally very high cost of deposition of silicon carbide clearly indicates that a large effort must be concentrated in this area. The first question in wheter the presence of this layer is a necessity. Its deletion from the specification of the coated particle would obviously achieve a very important cost reduction but it is evident that other factors must be taken into account.

The main avenue for development work is to find a method to increase the concentration of silane during deposition to reduce the time to achieve the necessary thickness. Two routes for work are open. The first approach postulates that the scientifically near perfect silicon carbide that is obtained with low concentrations is a necessity.

In this case new deposition methods must be investigated. The second approach consists of questioning the necessity and even the suitability of this very perfect silicon carbide.

There is an urgent need to prove which quality of silicon carbide is the best and also to find what quality is "good enough" so that the specification can be relaxed and thus the manufacturing cost reduced.

To reduce the capital investment and the labour cost there is an advantage is scaling-up the equipment presently used. The effort necessary to evaluate a batch of coated particles is independent of the size of this batch. Economies can thus also be foreseen in that.

Increasing the size of the coating column automatically implies that a multi-nozzle system must be used.

It is thus wise to go through a period of model study in cold conditions where at least some of the phenomena can be investigated.

The results obtained will provide an excellent guide for further work in large coating furnaces and by this reduce the number of very expensive experiments necessary to commission the system.

Conclusion

A tentative list of priorities has been made, keeping in mind all above mentioned arguments. This priority list is given in Table 1.

TABLE 1

Development work in the coating field

Subject	approx. factor on cost	Priorities of subjects If adequate funds and trained staff is made available and if time before freeze of design is: 1 year	2 years	3 years
6" diam. furnace	1.5	2	3	-
Automatic control system	1.4	1	1	1
Elimination of carbon monoxide	1.1	6	7	-
One-stage coating	1.25	5	6	-
Particle handling		3	4	-

Table 1 contd.

Hot unloading and soot control	0.85	4	5	5
Propylene coating	0.7	-	-	2
Silicone carbide	0.7 or less	-	2	3
Scale-up	S.G.	-	-	4

This table does not take into account a possible change in the specification of the Dragon reference particle.

4. Discussion on items 2 and 3

Hibbert: i. Has the use of cheaper fluidizing gases, such as N2, been considered and what is the effect on coating economy?

ii. You are trying to eliminate CO from the coating process, whereas in our current line of thinking there is some advantage in introducing CO for various reasons, particularly to apply buffer and sealing layers at temperatures as high as possible to minimize effects of differential thermal expansion.

iii. Intermediate unloading seems to introduce in our opinion problems and is not practiced in our laboratoires.

iv. While you propose to depart from a sinlge nozzle system for beds larger than 6 in. I.D. we are operating at 6.5 in.I.D. successfully with such a single-nozzle.
In the past we have been spear-heading multi-inlet systems for coating operations.

v. I would make a plea to give priority to the development of a large coating unit rather than automatic process-control particularly for low-enrichment fuels.

Price: You can see from page 14 of the paper that if you debit all the cost of argon you gain 238 pounds per tonne U. The use of nitrogen has been considered but the paper attempted to highlight the main avenues for economy.

Lefevre : The need for use of CO is not quite clear, and present costs as well as its toxicity make it rather an unwanted gas. Intermediate unloading is only exercised at the Dragon Project in order to minimize rejects due to uncertainties in varying kernel quality. The use of single injector for large beds is still in doubt and anyway multi-nozzle systems have to be considered, the optimum spacing still being open to discussion.

The automatic furnace should still retain first priority as such a system can be realized with relatively small efforts, and it increases the confidence in the results obtained from expensive and long-term irradiation experiments as well as from development work.

Flamm : Putting priorities aside, development work on scale-up (facing still a large number of uncertainties) should be commenced at once.

Vygen : Hot-unloading is already practiced at NUKEM and heating and cooling cycles are optimized in the furnace itself.

Lefevre : We have a proposal for gravity-unloading from the bottom (against common practice of suction-unloading from the top) and a design exists.

Vygen, Flamm : Problems of maintenance and clean-up still have to be solved.

5. Principles and practice in fluidized bed coating of nuclear fuel particles - J. Flamm

The task of process scale-up is to provide equipment and process specifications which suit actual demand and give operational flexibility, where possible. The question is "how" and "how far" can or should scale-up go, and, last not least, where will expenditure on develpment outweigh benefits.

The main problem areas - apart from auxiliary circuits and operations - are still the coating process and the coating furnace itself.

Upon analysis of available process data it can be seen that very few data are digestible or digested in a form suitable for the designer. This is not surprising, considering the number of interacting parameters, namely hydrodynamics with superimposed gas injection effects, heat and mass transfer in a system with extremely fast phase interchange and reaction kinetics and their combined effect of coating structure and physical properties. Therefore, stepwise analysis of characteristic process parameters is essential, provided analysis methods become highly refined and laws for recombination of these parameters are developed. A review of the state of the art, interspaced with experimental evidence, is attempted below.

Bulk effects in the fuidized state (i.e. both minimum and maximum gas flows at incipient fluidization and elutriation, resp) have been analyzed from earlier work at the Dragon Project and found in agreement with theory on hydrodynamic grounds. Particularly the upper limit of operation (incipient elutriation), can now be predicted by assessing the fluctuation ratio (r) of the bed level as an exponential function of the fluidization index (N) (adjusted to operating temperature, gas phase composition, particle density, etc..) and a parameter (t) being an empirical function of the particle diameter, i.e.

$$r = e^{t(N-1)}$$

In brief, beds operated at fluctuation ratios in excess of 1.6 showed violent bed movement and beds operated in excess of 1.9 showed strong elutriation. Operational conditions varied over a wide range (i.e. bed diam. 5.5 - 12.7 cm, particle diam. 500-1100 μm. particle densities 3-10 g/cm^3, operating temperature 1400-2150°C).

It must be stressed that the knowledge of these features is important to bracket physical limits of operation for design purposes, but beyond that point there is no resemblance with parameters governing the coating process.

We are only at the beginning of describing the problem of heat and mass transfer and a qualitative review has been given.

Similarly temperature gradients have been observed upstream and downstream of the gas injector, but a critical quantitative alalysis of these effects is still to be made. Mr. Souillart's work is going in that direction and we hope that CCR model work will spearhead improved injector designs.

Reactant decomposition and deposition kinetics are in my opinion the least quantitatively analysed parameters of the entire process. A multitude of investigations may have led to some general conclusions, but no scale-up orientated data have been produced so far. Work of basic nature is projected by Mr. Souillart and CCR model work emanates some tentative formulations of criteria.

At this stage of analytical progress only qualitative illustrations could be given in order to classify areas where further work is needed.

As an example, the effect of reactant throughput (CH_4) on coating properties (PyC density) has been demonstrated for various diameter beds, having similar single gas inlets. For scale-up this indicates the effectiveness limit of a single jet and gives a first lead for the geometrical arrangement of a multi-jet injection system.

The interactions between hydrodynamic forces and heat and mass transfer, as well as deposition kinetics in a high temperature coater could be demonstrated by comparing coating structures obtained in a series of experiments, where charge sizes and gas flows had been increased proportionally. The increase in fluidization index or fluctuation ratio, the higher introduced momentum, i.e. higher nozzle exit velocities, and hence, a shift in deposition zone and temperature profile are responsible to degrees still to be defined. Here again our model work is directed to an analysis.

Finally there is still a field wide open to speculation. This is the effect of decomposition - deposition retarding or catalysing diluents.

Turning to practice and design of coating equipment a brief review of systems in use has been given.

Systems of conventional geometry and heating, i.e. graphite resistance heating, may be doomed on scale-up because of difficulties in accommodating the required resistance in sturdy heaters, the enhanced problem of radiation shielding and transient or unsteady state heat flow over increasing distances. Some remedy may be expected by a "direct resistance heated" bed, a prototype of which has been thermally tested at CCR - Ispra and power savings over the conventional design are estimated at around 50%. A report on the experimental work is in press. Some ideas for scale-up have been shown in two reference designs, i.e. for a 200 mm I.D. and a 300 mm I.D. unit with overlapping scale-down margins to 75 and 150 mm I.D., resp.

Further, single or three-phase operation is feasible. The upper operational limit is estimated at around 40 Kg of standard 800 μm UO2 fuel, but without experimental verification, this is a frivolous degree of extrapolation.

Despite largely increased thermal efficiency, distances for heat transport even in this type of bed still increase with increasing bed diameter. This may best be demonstrated by comparing characteristic path lenghts for heat transfer, derived from the ratio of bed volume and the wall surface or heat source area in contact with this bed. Based on these considerations an elongated "trough type" bed may be the answer. Construction is feasible and can overcome several drawbacks of the other systems, but horizontal servicing will request new engineering endeavours.

Some feasible parameters have been chosen to demonstrate the capacity of such a system. Taking a target charge weight of 50 kg UO2 kernels, 90% T.D. 800 μm Ø, 150 μm total coating thickness and allowing a settled bed height (H_{sf}) after completion of coating of only 25 cm, only a bed length (L) of approximately 80 cm will be needed, which is well realizable.

A properly detailed design may well allow for bed lengths in excess of 1 m and if higher settled beds can be allowed the capacity of such a system could easily reach the 100 Kg mark.

With respect to refinements two points may be mentioned for process simplification:

1) to develop single-flow injectors in order to reduce the unavoidable congestion in multi-point injection systems and
2) to concentrate development efforts on a single family of gases, i.e. H_2, C_2H_2, CH_4 and CH_3SiCl_3, in order to further reduce gas metering and to simplify gas recycle.

Further, digestion of theory in experiments with feedback from quality control is in need of attention.

In order to achieve these goals, a full sized pilot plant-instrumented for vital parameter studies - would be needed; in the first place to provide basic information in the many areas mentioned above and secondly to verify the model work proceeding in the various laboratoires under proper operating conditions. A stream-lined programme may well require a quarter of a million dollars.

6. Model research at KFA - Julich - C.F. Wallroth

In the Institute of Reactor Materials at KFA Julich (Head of Department Prof. B. Liebmann), there are various activities in the field of fluidized bed coating. Detailed work is carried out in the chemical department under the administration of Prof. H. Nickel as follows:

1)- E. Gyarmati:

- a - standard coating for irradiation tests
- b - propylene coatings for the experimental testing of the Scott-Prados-Walther models
- c - systematic SiC parameters research comprising preparation of specimens for irradiation

2)- R. Haange:

gaschromatic analysis of the decomposition of hydrocarbon-gases and localization of the decomposition zone in the fluidized bed.

3)- D. Böcker:

mass spectrometric measurements on pyrocarbon to determine hydrogen and pyrocarbon content related to coating temperatures and various coating gases.

4)- E. Wallura:

investigation of the uranium transport mechanism in pyrocarbon layers by fluorimetric uranium analysis on discriminated parts of the layers removed in 10 microns steps.

5)- B. Püher:

research on advanced ceramic coating materials.

6)- C.F. Wallroth:

research models of conical fluidized beds using Petunia seeds.

7)- B. Kalthoff:

research models of conical fluidized beds using magnesium spheres.

I shall be solely concerned with model bed studies. The idea behind these model studies is:

1. to simulate particle movement of the hot coating furnaces at room temperature in transparent equipment,
2. to analyse the influence of the various parameters,
3. to optimize the particle movement, to yield uniform layers at minimum gas costs and
4. to attempt to translate these optimized conditions from smaller bed diameters to industrial scale operation units.

One dimensionless expression mainly describes the physical similarity. It is a combination of the well-known Reynolds and Newton numbers and may be called Berànek number (after a Czecho-Slovakia scientist)

$$Be = \frac{Re}{Ne} = \frac{v_g^3 \cdot \rho_f^2}{g \cdot \eta \cdot (\rho_p - \rho_f)}$$

v_g = free falling velocities of particles
ρ = density (index p = particle, index f = fluid)
g = acceleration due to gravity
η = dynamic viscosity

As a result of the high temperature in the coating furnace the gas density is very low and the viscosity is very high, resulting in Be- values in the range of 1 to 100. If the same particles were used for the model work, then Be-values will result which are two or three orders of magnitude out of range. Physical similarity only exists at the same values in the model bed and the coating bed. This will be done by applying suitable gases of particular low density (like helium or hydrogen) and suitable particles of also lower density. To this end Petunia seeds (density 1.1 g. cm^{-3}) were chosen.

The following results were obtained:

1. Variation of the gas distribution between the central and the ring nozzle produces the three states of fluidization called "spouting", "bubbling", and "slugging".

2. For the interpretation of these observations a critical ratio of the central gas flow Q_z over the total gas flow Q for the transition from one movement form to another is defined and this ratio is plotted against the empty tube velocity w.

   Curves of same central velocity form a hyperbolic function:

$$(Q_c/Q)_{Q_c} = \text{const} \qquad = \frac{w_c}{w}$$

   where w_c is the empty tube velocity of the centre gas flow. With increasing gas velocity the influence of the particle charge on the form of bed movement decreases.

3. Tests with varied nozzle geometry demonstrated how the exit velocities of the nozzles govern the mode of particle movement.

4. Tests with different cone angles proved that the critical flow ratio for the transition from spouting to bubbling and the particle velocity near the wall is decreasing

exponentially with increasing cone angles.

5. Tests with different bed diameters have shown that reasonable fluidizing conditions are obtained in a 1-inch diam. fluidized bed with a relatively high particle concentration spread over the entire gas bubble volume. In the bigger bed geometries the particle concentration in the gas bubbles reduces greatly and with the increased inhomogeneity of the fluidized bed the overall conditions may result in non-uniform coatings.

6. Coating runs with different particle movement have brought the best results in the case of bubbling, the worst for slugging with respect to the sphericity of the coated particles and the homogeneity of the layers.

   It is very difficult to find an undisturbed bubble formation, because slugging is the dominant form of movement. Therefore, one of the KFA activities is to find a bed and nozzle geometry which presents a good and steady bubble production and bed movement over a wide range of the flow ratios.

   Since it is without any doubt that there is a great influence of the particle movement on the shape of the coating layers, it is essential to know all the parameters which influence the particle movement.

   The model tests demonstrate that the fluidizing conditions in conical fluidized beds are governed by the velocity, the model state, the charge size, the bed and nozzle geometries.

   From the work done so far we formulated a function of the critical gas flow ratio to describe the transition from one fluidized bed state into another. This function comprises all the necessary parameters and reads as follows:

$$Q_c = \text{const.} \quad (Q_c/Q)_{crit.} = f(w, Be, G, A_c/A_b, A_c/A_r, \alpha, d_p/d_b, d_b)$$

where:

Q = gas flow
w = empty tube velocity
Be = Beranek number
G = particle charge
A = cross section
α = cone angle
d = diameter

subscripts:

c = centre nozzle
b = bed
r = ring nozzle
p = particle

To solve this mathematical function considerably more model research work is required.

7. CCR - EURATOM - Ispra model work relevant to scale-up

D. Van Velzen

Introduction

The basic principle underlying the CCR model work is the fact that it should be directly useful for scale-up. Room temperature model work should narrow the field for the very expensive high temperature development work and thus build a cost-saving factor.

In order to define the criteria to judge the model experiments, the chemical reaction system of the coating process is described as:

"A very fast gas phase reaction, depositing the reaction products on a moving solid, the whole process taking place in a non-isothermal reactor".

The main parameters determining the efficiency and the yield of the process are thus:

1) the gas residence time profile in the reaction zone
2) the solid transport through the reaction zone
3) the gas temperature profile of the reaction zone

Needed as basic tools for the model work are therefore: methods to quantify these three parameters, or anyhow two of them, the gas residence time distribution and the solids transport.

The first step of model work should be: investigation of the gas and solids flow patterns in models of reactors actually in use to establish a basis for comparison with new geometries.

Experimental

The experimental work, done so far, has been concentrated on the gas flow pattern in the 125 mm. I.D. bed of Dragon geometry.
Air was always used as the fluidizing gas and four different solids and three gas inlet nozzles have been used.

The solids were:

	sieve-fraction (μm)	density (gcm^{-3})
Zirconium dioxide	590 - 850	4.4
Glass ballottini	890 - 1220	2.6
Glass ballottini	745 - 890	2.6
Permutit ballottini	590 - 850	1.6

The nozzles were all two flow nozzles of the following dimensions:

Central bore diameter	Annulus
2.5 mm	3.8/4.3 mm
4.0 mm	5.3/5.8 mm
8.0 mm	10/11 mm

With this experimental set-up always stable spouting beds were obtained. The experiments consisted of the determination of the dynamic pressure exerted by the upward flowing gas in the spout at well defined positions in the bed.
The measurements were carried out with a movable Prandtl-tube arrangement.

Results were presented as the dynamic pressure profile of the bed.

The vertical component of the gas velocity was calculated from the dynamic pressure and from the point velocities. he total gas quantity flowing upwards in the spout was obtained by numerical integration of the equation

$$V_x = \int_0^{r_s} \pi \cdot rdr \quad v_{xr}$$

This quantity is better expressed as the transport ratio, Vx/Vo, that is the ratio of the total gas flow in the spout to the injected gas flow. It appeared that the transport ratio lies between 0.6 and 1.5, depending on the axial distance from the nozzle, the gas flow and the type of nozzle used. Generally speaking, it can be deduced that practically all gas injected by the nozzle is traveling upwards in the spout - together with a sensible quantity of gas swept along from the rest of the bed.

The observation of the influence of the total gas flow and the nozzle geometry (i.e. the nozzle exit velocity) lead to the hypothesis that the most important quantity governing the behaviour of a spouting bed is the quantity of momentum introduced per unit time by the fluidizing gas.

This quantity is expressed as $M = \dfrac{Q_1 v_1 + Q_2 v_2}{g}$

where Q = the quantity of gas $(g.s^{-1})$
v = the exit gas velocity $(cm.s^{-1})$
g = acceleration of gravity $(cm.s^{-2})$

and is thus expressed in grammes and is equal to the maximum force exerted on the particles. The hypothesis has been checked by plotting the average gas velocity in the spout at a distance x

$(v_x = \dfrac{V_x}{\pi r_s^2})$ against the introduced momentum

on a log/log scale.

It appears that the average upward gas velocity in the spout is clearly dependent upon M, irrespective of particle properties, and that the slopes of the curves are dependent on x, the axial distance. By regression analysis the relation between velocity, momentum and axial distance has been determined.

$$v_x = \frac{16.1}{x + 4.76} \cdot M^{(0.63 - 0.018x)}$$

Strictly speaking, this relation is only valid for the 125 mm bed with air as a fluidization medium, but it is likely to be used also for other gases - even at high temperature - because the momentum is the only variable, and this is not influenced by gas properties.

As a demonstration the velocity profile for an actual coating run (High Density Coating, 1900°C) has been calculated by the above relation and the results expressed as residence time of the gases in the spout. It appears that the residence times in the spout are extremely short, i.e. 2.7 milliseconds at 5 cm from the nozzle and 7 milliseconds at 10 cm above the nozzle.

Assuming a hypothetical axial temperature profile (based on actual measurements carried out at the Dragon Project), the methane decomposition rates can easily be calculated using kinetic data from the literature. It appears that presumably the whole reaction is taking place between 2 to 5 cm from the nozzle in spite of the very short residence times which are to be expected.

Future activities

The developed method is considered a valuable tool for the evaluation of gas flow in the reaction zone, but still needed is a method for quantification of solids transport in this zone.

At the time also this variable can be described quantitatively, it is possible to investigate the influence of bed diameter, bed geometry, nozzle geometry, etc....., thus sensibly contributing to scale-up work by room temperature model experiments.

8. Pyrocarbon deposition with respect to gas preheating and inlet effects - C. Souillart

CCR Petten works on two aspects of pyrocarbon deposition

1 - Fluidized bed deposition
2 - Static deposition

In fluidized bed deposition we investigated the influence of particle properties, including density and diameter, with reference to the state of fluidization. Experiments with tungsten and vitreous carbon (density, 19 and 1.5 g cm^{-3}, resp.) showed the strong influence of the state of fluidization on coating structure and properties. Care has to be taken that the actual coating process in both cases takes place at the same temperature.

This temperature can be influenced by the position of the column within the heater (Dragon bed), and the degree of preheating of the entering gases in the nozzle. It has been shown that preheating of the gas has a crucial effect on coating properties. Experimentally, preheating was achieved by extension of the section between water-cooled support and nozzle exit and, in a second stage, it is foreseen to insert an additional resistance heater into this zone. Having the control of gas inlet temperature, the influence of the particle diameter of the coating properties has been shown clearly.

For instance, B.A.F. increases with increasing particle diameter.

Coated particles produced under various conditions (state of fluidization, rate of deposition, etc.) showed crushing behaviour dependent on the presence of certain layer structures. When these structures are absent, measured crushing strengths increase considerably.

This again stressed the importance of the state of fluidization as well as the adjustement to the rate of deposition per unit bed surface area.

In contribution to scale-up, an idea for a continous coating unit in a multi-column system has been presented for reflection.

A special system has been developed and built in Petten for static deposition experiments. Gas injection can be arranged in three directions at variable distance from the substrate.

The system should be suitable to study the intermittent stages a particle sees on its path through the fluidized bed. The equipment is very versatile; allows for rotation of the substrate and thus simulation of local hydrodynamic conditions in the actual fluidized bed.

Preliminary experiments have shown that there exists a temperature range for specific types of deposits and that small changes in deposition temperature may cause crucial variations in deposit structure and physical properties. It is advisable (if possible) to choose real coating conditions outside these critical temperature zones.

9. Discussion on items 5 to 8

Price: How can similarity rules be applied in so complex a system. Conditions vary axially, radially and as a function of time and similarity rules can in my opinion only be applied to average values.

Wallroth: My work was only concerned with particle motion and, to reduce the great number of variables, we have to accept average conditions as a basis for comparison of model and high temperature work. Agreed, this is only the first step in model work, the work done by Mr. Van Velzen presents the logical follow-up, and further work will be needed on heat & mass transfer, etc.

Price: Taking into account that this line of research is just beginning to get started, it may be mentioned that there is ample opportunity for other parties to join in funding this work. This could lead to an enhanced development being of mutual benefits to all concerned in the coating field and last not least to overall cost reduction.

Baschwitz: Can one, under operating conditions, determine the state of fluidization (bubbling, slugging, spouting) while the bed cannot be seen through the soot.

Wallroth: There is no direct method unless you assess the noise emitted from the furnace; and observe the pressure pattern at the exhaust side of the furnace. Otherwise intermittent checking under dummy conditions must be employed.

Flamm: Is polygonality of the coated particle the only critical parameter or should not physical properties be considered at the same time?

Wallroth: Certainly other parameters have to be investigated, but polygonality was chosen as criterion for this work.

Price: Is there a relationship between polygonality and the coating thickness variation between individual particles, which can be related to the state of fluidization?

Wallroth: There is a direct relationship between polygonality and coating thickness variation as the calculation of the polygonality was carried out on the measured maximum and minimum thicknesses of the outer layer (of a PyC-SiC-PyC coating).

Van Velzen: Upon discussing energy requirements, I should like to make a general remark. If the momentum per unit time introduced by the gas injector is too small to obtain stable spouting conditions, the spout collapses and a bubbling bed develops. If, on the other hand, too much energy is introduced, the upward solids transport in the spout exceeds the supply. This causes irregularities in the solids flow and causes a slugging bed. This hypothesis has been sustained by calculations carried out on Wallroth's work.

Flamm: A bubbling bed may be desirable, but coating economy requires maximum possible reactant throughput, which is apparently, for the injector system used, unfavourable to bubbling conditions (i.e. increased outer nozzle flows are needed to sustain the bubbling state).

Further, strong bubbling or slugging may lead to soot inclusions in the coating, which, by experience, have been found a continuous rings of soot.

Price: Bubbles seem to form at some point moderately above the nozzle, at which decomposition of the reactant is already far advanced and considering possible short-circuiting of particles above that region this could lead to a wider coating thickness variation, a parameter which cannot be overlooked in product quality.

Hibbert: We have been for some time extremely keen on bubbling fluidization for coating scale-up but under these conditions we were not able to make particles with the required coating properties. That's why we have doubts on the usefulness of the bubbling regime for coating scale-up.

Flamm: The nozzle exit velocity seems to be an important parameter, not only with respect to bed movement and coating properties, but also with respect to deposits on nozzle and bed parts. Model work should also be considered in this field.

<u>Souillart</u>: These effects should certainly not be overlooked, and research should (in my opinion) be directed towards a design which allows the coating to be formed at a controllable temperature. Further, ways have to be found to characterize the reaction zone.

10. Experimental studies and calculations on fluidized bed models suitable for coating of U, Pu dioxide kernels - F. Decamps

Fluidized bed model work at CEN-Mol was initially concerned with fluoride reprocessing techniques forming part of a contract between Euratom and the Belgian Authorities. Typically closed circuit techniques were to be employed and simultaneous fluidization of dissimilar solids as well as their transport was to be achieved. Gas solids contact and heat transfer were parameters of dominant interest. Measuring techniques for heat transfer (cross sensor technique - PhD Thesis, approx. August, 1971), gas residence time distribution (tracer injection) and a model for solids transport were developed. Application of this type of reactor to coating of nuclear fuel particles is presently being investigated.

The system consists of a transport device (riser tube) submerged in the bed of particles. At the lower end the riser is fitted with a bell shaped cap. A secondary flow of gas into this cap controls the solids transport. A mathematical model for solids transport in this system has been developed and will be published in one of the next issues of "Powder Technology". Solids transport can be calculated according to the following equation:

$$G_s = \frac{\Delta p}{(u_g - \alpha u_r) + \dfrac{\alpha^2 g L}{(u_g - \alpha u_r)}} \quad \left[\text{Kg m}^{-2} \text{ sec}^{-1} \right]$$

Δp is the total available pressure drop, and α and u_r (for non-spherical particles) are the characteristic parameters of the solid.

Gas residence time distribution measurements form a further part of the CEN programme. In a 2 inch I.D. column a tracer (He, approx. 3 to 6.3% of total flow) is injected axially approximately 40-45 cm above the gas inlet. Samples are taken at two levels downstream of the point of injection and analysis via a computer programme is in progress.

Further, a series of gas distributor systems and their effect on flows for minimum fluidization is being investigated. These systems comprise perforated plates, rigid meshes, conical distributors, rectangular and round bubble caps, etc. Quantification of the relative effects is still outstanding.

Basic knowledge gained on fluidization and gas-solids contact so far is thought to be a useful guide for other applications, such as coating. Appropriate modifications of the riser type fluidized bed are proposed in that sense.

Slab-type reactors (proposal to call this system in future LINEAR REACTOR, a term used by N.S. Hibbert), safe from the criticality point of view and of interest to our original work, may be investigated with our present tracer methods.

<u>The Chairman invited to a brief discussion at this point</u>:

<u>Van Velzen</u>: Was continuous or pulsed tracer injection used and which type of model has been employed for analysis?

<u>Lefevre</u>: Has a single shot or cycling pulse technique been used?

Decamps: The method used was a continuous injection with a response time of the cell of approximately half a second. The model for analysis represents a two-phase model in which the downward flow of gas is assumed to take place outside the bubble phase.

11. Problems in coating on industrial scale - P. Vygen

NUKEM has experience in coating of 200 to 800 microns diam. kernels. As most of the work is related to the THTR reactor our activities are mainly concerned with 400 to 600 microns diameter particles and our upper limit of operation is set by criticality limitations. Hence, our present ceiling in batch size is around the 5 kg mark and further scale-up is outside the scope of our current programme.

Properties of high temperature coatings were analysed. Porous coatings, derived from C_2H_2, as often observed, show variations in coating thickness from particle to particle. Measured on production batches the standard deviation was 9 microns for a mean coating thickness of 50 microns. This is due to a number of parameters but mainly to the high rate of deposition (of the order of 400 microns per hour). After porous coating the particles are almost perfectly spherical. Even small deviations in kernel sphericity are levelled out. This is not the case with the sealing and high density layers deposited from CH_4, leading to polygonality, depending on coated geometry and state of fluidization. Here, polygonality as well as coating thickness variation from particle to particle enter the standard deviation. Further, properties vary across the coating thickness (e.g. BAF).

Although model work will give a lead, production people have to carry on and make a better product. Three coater geometries have, therefore, been examined with respect to coating thickness variations, i.e.:

a) standard single inlet system,
b) modified single inlet system, and
c) three-cone and three-nozzle system.

The following results were obtained:

	system		
	a	b	c
<u>Porous coating - C2H2</u>			
mean coating thickness (microns)	50	50	50
standard deviation	9	8	5,5
<u>Sealing + HDI - CH4</u>			
mean coating thickness (microns)	120	120	120
standard deviation	15	10.5	10.8

Anisotropy, in the coating of one and the same particle, for instance, has been shown very sensitive to poligonality, i.e. at points of maximum thickness anisotropy shows a minimum.

The use of propylene has not only economic advantages but also renders a more uniform coating with respect to anisotropy. Another advantage is the low heat of decomposition. A comparison of high and low temperature coatings is best given below, clearly showing the advantages in the use of propylene:

Coating agent	CH_4	C_3H_6
Temperature (°C)	2000-2100	1300-1350
Deposition rate (µm/h)	35-45	150-200
Coating density (g/cm^3)	1.90	1.9-2.0
Crystallite size (Å)	90-140	20-40
Anisotropy (BAF)	1.15	1.05

Compared on cost basis, methane coating set at 100%, prolylene coatings would be at a favourable 33%, mainly due to large savings in time and personnel.

Irradiation experiments are in progress and show encouraging results, but this product is still not generally accepted.

12. Some comments on the manufacture of coated particle fuel for prismatic fuel elements - N.S. Hibbert

At the reactor fuel element laboratories (RFL) of the UKAEA a programme of work towards the scale-up of coating processes has been accomplished by a team of the laboratories and the British Nuclear Fuel Company (BNFL).

Tonne quantities of nominally 800 microns diam. kernels, having composite coatings of up to 190 microns, have been made for Zero-Power Experiments. Kernels are being made by the RFL powder agglomeration route, i.e. 75-85% T.D. following a carbon burn-out process and 90-95% T.D. a straight U02 route. Coating is carried out in units of up to 165 mm I.D. The plant capacity is of the order of several te/year. Work is backed by a sizeable characterization team.

Along with the work on standard coatings from acetylene, methane and methyltrichlorosilane, detailed studies on coatings from acetylene, propylene, butane, etc. are carried out.

On scale-up the main target is to retain coating properties achievable in small units. For instance, polygonality is one of the main problems in scale-up and compaction experiments have shown that the ratio of particle breakage is of the order of 100 comparing spherical and angular coated particles. For this reason care is being taken to achieve uniform coated particle properties from particle to particle as well as from batch to batch, and to remove misshapen particles by a centrifugal shape separation technique.

The effect of the choice of the kernel diameter on the economics of the process cannot be neglected. There have been voices to reduce to reference kernel diameter from presently 800 microns to 600-500 microns. Based on general parameters governing the production route, it has been shown that the manufacturing costs will increase by a factor of about 2.5 for both the kernel and coated particle fabrication. For a plant already installed this means a reduction in out-put of about 60%.

13. Discussion on items 10 to 12 and general discussion

Hibbert: The idea of an elongated coater geometry (here called "trough-type" bed, in our terminology "linear bed") has been pursued in our laboratories to the construction of a demonstration unit. Low-temperature equipment is almost complete, but experimental coating work had to be postponed for priority reasons. The system is kept as a fall back position for real large scale operations.
Further, the use of H2 throughout, i.e. instead of other diluents, seems to have an additional negative effect on the thermal economy of the process.

Flamm: The use of H2 as diluent has mainly been thrown in to open the debate on off-gas treatment, a field which is usually treated in stepmotherly fashion, especially concerning economics. The effect of additional hydrogen on the coating depends entirely on the gas inlet geometry and should help to increase reactant concentration, and, thus, throughput.

Price: Recycle of gases, particularly such derived from decomposed hydrocarbons, may even lead to a cost penalty in case hydrocarbon residues cannot be removed properly.

Lefevre: Along this line safety regulations have been treated in a stepmotherly fashion as well.

Price: Low-temperature coatings (C3H6) are known to retain higher amounts of hydrogen and I would like to ask Mr. Vygen if any post-coating heat treatment is considered necessary or is the normal heat treatment of compacts sufficient for hydrogen removal.

Vygen: Hydrogen retention has so far not been investigated in detail. Emission of hydrogen has been observed and a heat treatment after coating is considered to be necessary.

A general discussion developed on the pros and cons for low and high temperature coatings but those present were not in a position to draw decisive conclusions.

Lefevre: Directly heated coating columns have to fulfill two functions, i.e. to provide the coating chamber as well as part of the heater. Normally the coating column has to be replaced more often than the heater, which seems to be one disadvantage. Further, electrical contacts have to be remade after each disassembly, which seems to me to add some operational uncertainties. Along the conventional route of heating there is still the graphite cloth type resistance heater (a design of which exists) which keeps these two functions separate.

Flamm: The directly heated system renders a very compact design, large power savings and in addition heat generation at the point of consumption. Naturally cooling periods are increased due to the two thermal barriers.

Baschwitz: I would like to add a remark with respect to a fast cooling system. We have developed a design with granular thermal insulation which can be fluidized during cooling down periods. The system still has to be proven.

General discussion

Vivante: Summarizing today's presentations, discussions and undertones to the meeting areas of outstanding work comprise the following topics:

i determination of optimum or maximum bed size for a low enrichment fuel

ii the use of single- or multi-nozzle systems and the most favourable state of fluidization

iii the type (s) of hydrocarbons to be used (propylene seems to be very promising) and can silicon carbide be eliminated from the coating

iv is one stage coating acceptable or should intermittent clean-up (i.e. shape separation and/or leaching) be employed

v feasibility of isothermal coating

vi is elimination of carbon monoxide from the coating process feasible.

A point not stressed in this meeting is the need for an irradiation programme for coated particles manufactured in industrial installations.

Price: Part of the Dragon irradiation programme should be geared to satisfy the latter request.

Vygen: Some irradiation tests on production batches are already under way and a joint programme of NUKEM and KFA exists but an enlargement on this programme is certainly desirable. Also the A.V.R. reactor can be considered as an irradiation experiment of industrially made particles.

Price: It is a disadvantage that exact irradiation conditions in the A.V.R. are not known, and this is important for extrapolating the results to other types of reactor.

14. Summary by Chairman

The Chairman invited the participants to give a survey of the relative manpower contributing to this type of work in Europe.

Although incomplete for any type of analysis provisional figures given for the deployment of staff by the various organizations indicate the overall effort in this field. As programmes and targets vary grossly from organization to organization and contributions to research, development and manufacture could not clearly be separated at this meeting, a figure of about 80-100 persons (irrespective of qualifications) may be taken as indicative for total efforts in Europe. Product evaluation and quality control- instrinsically linked to development in this field - may well more than double the above number.

Finzi: Having this estimate and a global view of the various activities it seems useful to me to take a further step towards collaboration and possibly forming a working party in this field. I would appreciate comments on this proposal from the more industrial organizations.

Price: Collaboration is naturally the prime object of the Dragon Project. We see here again the typical European approach of working in isolation and duplication, launching a project but covering only the first 10% of the route, and, so allowing the United States an easy conquest of the European market.

Vygen: We in industry certainly do not have the opportunity to do the fundamental work and in this respect a collaboration is certainly necessary. The results of any fundamental work should be communicated to industry.

<u>Finzi:</u> It is very necessary for research centers to obtain their guide lines and input data from the industrial groups, in order to keep their programmes in line with up-to-date problems. On the other side, it is common knowledge that industry does not always make optimal use of research data, simply because their existence is not known to them. Therefore, the idea of closer contacts has a certain bearing.

<u>Vivante:</u> In this context it will be advisable to have another meeting on some well defined subject areas.

<u>Wallroth:</u> Collaboration between KFA and Ispra in the field of model work seems very well possible and of mutual interest.

<u>Van Velzen:</u> I certainly agree with that and there is ample scope for work on well tuned programmes. In addition it would be favourable to our work not to have only directions from the Dragon Project but also from the manufacturers.

<u>Flamm:</u> A similar need exists for component development and this is usually hampered by the non-availability of the expensive basic furnace unit or the "skeleton". The idea of launching a common project may be vented here.

<u>Finzi:</u> But does such a proposal not infringe on commercial interest?

Lefevre: There is still ample scope for common development before infringing on commercial interests and such development work will help to improve the validity and representativeness of results obtained from research oriented irradiation programmes.

Baschwitz: This meeting has shown that there is common interest in these topics and in the work done by the Fuel Production Development Group of the Dragon Project, and by Euratom under Dragon contract. We should, therefore, support them; where common work is carried out may be debatable.

Finzi: Finally summarizing, there seems to be considerable scope for continuing this discussion and I propose to organize another meeting of this kind later in the year. Organizationally preparations may best be made as for this meeting. The programme should then be more directed to specific topics and proposals for mutually beneficial programmes.
I wish to thank you for your encouraging interest in these affairs and I hope you will carry the spirit of the meeting home to your parent organizations.

NOTICE TO THE READER

All scientific and technical reports published by the Commission of the European Communities are announced in the monthly periodical **"euro-abstracts"**. For subscription (1 year: B.Fr. 1025) or free specimen copies please write to:

Sales Office for Official Publications
of the European Communities
P.O. Box 1003
Luxembourg 1
(Grand-Duchy of Luxembourg)

SALES OFFICES

The Office for Official Publications sells all documents published by the Commission of the European Communities at the addresses listed below, at the price given on cover. When ordering, specify clearly the exact reference and the title of the document.

GREAT BRITAIN AND THE COMMONWEALTH

H.M. Stationery Office
P.O. Box 569
London S.E. 1

UNITED STATES OF AMERICA

European Community Information Service
2100 M Street, N.W.
Suite 707
Washington, D.C. 20 037

BELGIUM

Moniteur belge — Belgisch Staatsblad
Rue de Louvain 40-42 — Leuvenseweg 40-42
1000 Bruxelles — 1000 Brussel — Tel. 12 00 26
CCP 50-80 — Postgiro 50-80

Agency:
Librairie européenne — Europese Boekhandel
Rue de la Loi 244 — Wetstraat 244
1040 Bruxelles — 1040 Brussel

GRAND DUCHY OF LUXEMBOURG

Office for official publications
of the European Communities
Case postale 1003 — Luxembourg 1
and 29, rue Aldringen, Library
Tel. 4 79 41 — CCP 191-90
Compte courant bancaire: BIL 8-109/6003/200

FRANCE

Service de vente en France des publications
des Communautés européennes
26, rue Desaix
75 Paris-15ᵉ — Tel. (1) 306.5100
CCP Paris 23-96

GERMANY (FR)

Verlag Bundesanzeiger
5 Köln 1 — Postfach 108 006
Tel. (0221) 21 03 48
Telex: Anzeiger Bonn 08 882 595
Postscheckkonto 834 00 Köln

ITALY

Libreria dello Stato
Piazza G. Verdi 10
00198 Roma — Tel. (6) 85 09
CCP 1/2640

Agencies:
00187 Roma — Via del Tritone 61/A e 61/B
00187 Roma — Via XX Settembre (Palazzo Ministero delle finanze)
20121 Milano — Galleria Vittorio Emanuele 3
80121 Napoli — Via Chiaia 5
50129 Firenze — Via Cavour 46/R
16121 Genova — Via XII Ottobre 172
40125 Bologna — Strada Maggiore 23/A

NETHERLANDS

Staatsdrukkerij- en uitgeverijbedrijf
Christoffel Plantijnstraat
's-Gravenhage — Tel. (070) 81 45 11
Giro 425 300

IRELAND

Stationery Office
Beggar's Bush
Dublin 4

SWITZERLAND

Librairie Payot
6, rue Grenus
1211 Genève
CCP 12-236 Genève

SWEDEN

Librairie C.E. Fritze
2, Fredsgatan
Stockholm 16
Post Giro 193, Bank Giro 73/4015

SPAIN

Libreria Mundi-Prensa
Castello, 37
Madrid 1

OTHER COUNTRIES

Office for official publications
of the European Communities
Case postale 1003 — Luxembourg 1
Tel. 4 79 41 — CCP 191-90
Compte courant bancaire: BIL 8-109/6003/200

Price: BFr. 85.–

OFFICE FOR OFFICIAL PUBLICATIONS OF THE EUROPEAN COMMUNITIES
Case postale 1003 — Luxembourg 1 5924

CDNA04772ENC