

## Fuel surface measurement on spent fuel using BET-technique

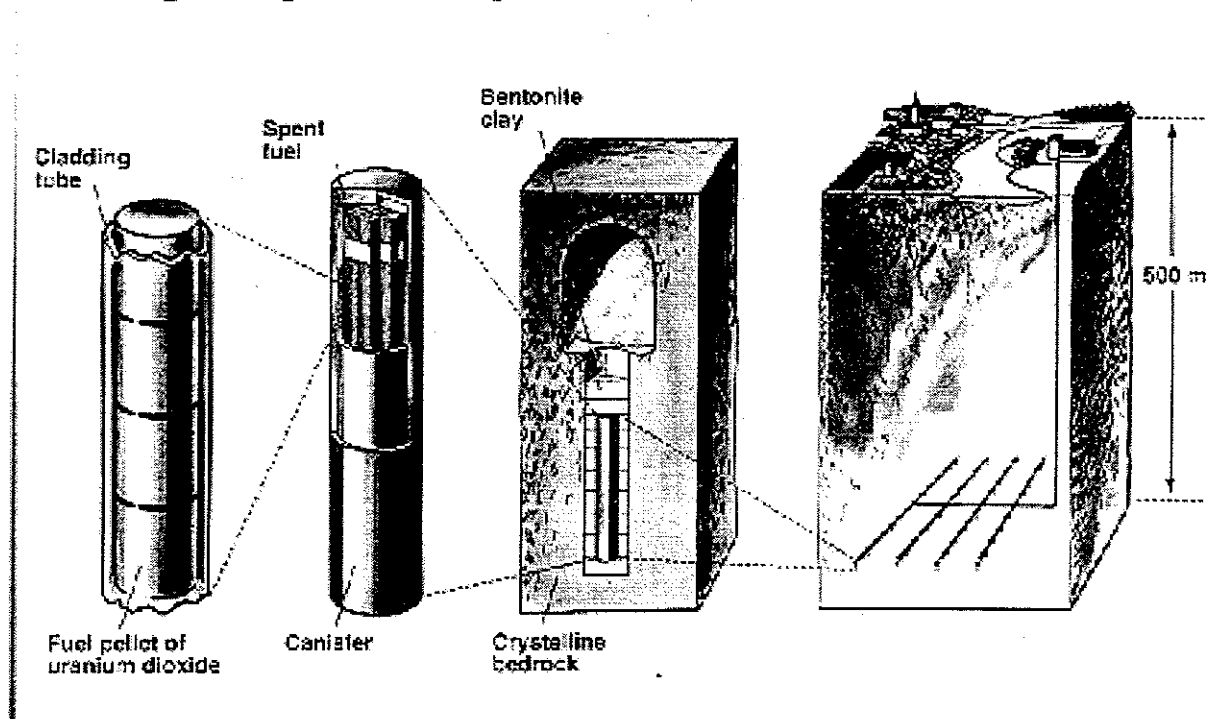
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### Abstract

It has been decided not to reprocess but to store spent fuel in Sweden. For this reason research has been going on for a long time in Studsvik evaluating the release of fission products and actinides from such fuel. This has been performed as leaching tests in ground water. It was found that the leaching rate ought to be normalised to the fuel surface. The method chosen to measure the surface area of spent fuel was BET (Brunauer, Emmet and Teller – ISO/DIS 9277). The basis for this method is measurement of the adsorption of a gas on the fuel surface. In-cell equipment was built for this purpose. It was found that the amount of surface area to be measured was at the limit of resolution for the instrument. It was thus decided to use krypton instead of the more commonly used nitrogen for the measurements although this gives more experimental difficulties. A further concern with measurement on spent fuel is the decay heat. Extensive tests were performed both before and after installing in the cell. Finally measurements were performed on fresh fuel, on PWR and BWR fuel with a burnup of about 40 MWd/kgU and on fuel with a burnup of 70 MWd/kgU.

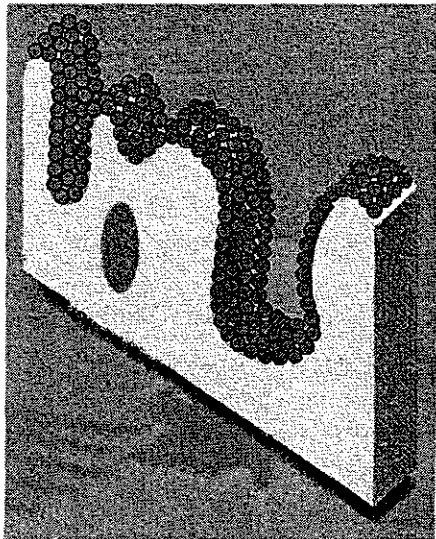
It has been decided not to reprocess, but to store spent fuel in Sweden. This is briefly the Swedish model for the final repository for spent nuclear fuel. The spent fuel will be encapsulated in copper capsules, embedded in dense bentonite clay, 500 meters down in the bedrock.

## Deep repository for spent nuclear fuel



In order to obtain experimental data for the modelling and performance assessment of the repository, spent fuel has been exposed to simulated ground water under different conditions. Research has been going on for twenty years in Studsvik evaluating the release of fission products and actinides from such fuel. This has been performed as leaching tests in ground water. If the ground water is in contact with the fuel, the leach rate can be related to the fuel inventory. But it was found that the leaching rate ought to be normalised to the fuel surface area.

The method chosen to measure the surface area of spent fuel was the BET-method. It has been named after the initial letters of the three authors of the method - Brunauer, Emmet and Teller. The principle of this method is the determination of the quantity of adsorbed gas needed to completely cover the surface of a specimen with a single layer of gas molecules. The method is standardized according to ISO/DIS 9277.



Surface detected by the BET  
(adsorption) method, developed by  
Brunauer, Emmet and Teller.

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In order to carry out in-cell measurements with the BET-method we met some problems.

One of the problems was the commercially available instruments, that we judged would be difficult to be adapted to in-cell use. For example almost all electric components and electronics would be removed from the instrument and be placed on the outside of the cell due to the radiation. Such instruments would also be hard to handle practically with the manipulators in the cell. They often had small knobs and keys. The specimen holder was also difficult to handle, specially when the fuel was charged, and difficult to reach.



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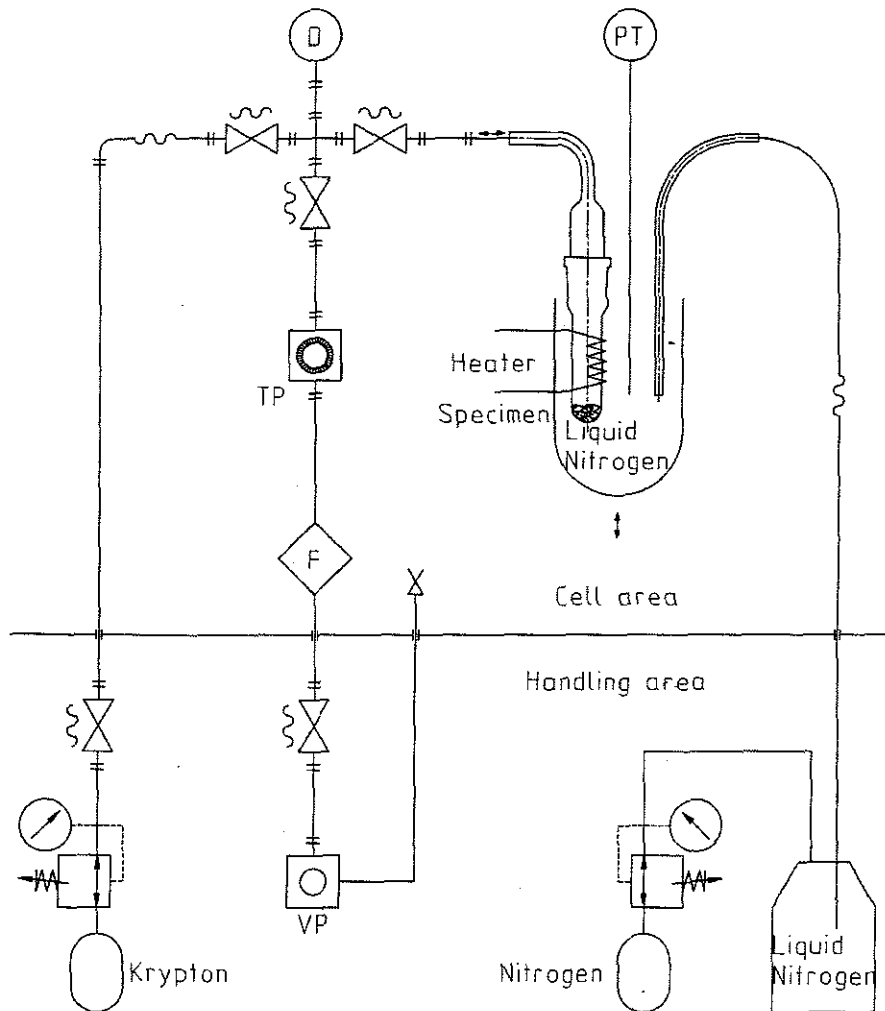
Commercial  
instrument

A department for powder technology had earlier existed in Studsvik and they had experience of BET-measurements. Ideas and experience from their own-design instrument was used when we constructed, tested and adapted to cell our BET-equipment.

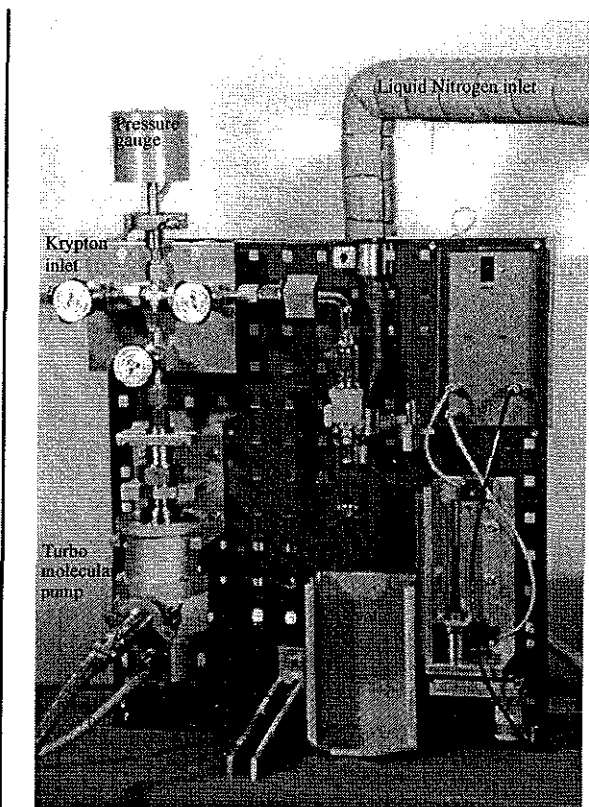
This is a flow chart of our BET-equipment. When we constructed it, we chose to have as little as possible of the equipment in the cell. Practically all electric components and electronics were placed outside the cell. The cell equipment was connected to the equipment outside the cell through specially made plugs in the cell wall. There were for example plugs for liquid nitrogen, Krypton gas and turbomolecular pump controls.

It had to be a rugged construction which was easy to handle with the manipulators.

The figure below shows the specimen tube made of glass which is heatable for degassing of the sample. This is a movable thermos flask for the liquid Nitrogen. Its temperature is checked and the level is controlled by a couple of Pt-100 probes. There are valves to control the Krypton gas, which we chose instead of Nitrogen to improve the sensitivity of the instrument, which is fed to the sample this way. A pressure gauge is mounted on top of the instrument to measure the pressure of the Krypton gas, before inlet to the sample, and after adsorption on the sample. To evacuate the instrument a turbomolecular pump (TP) is used, followed by a filter and finally a backing pump.



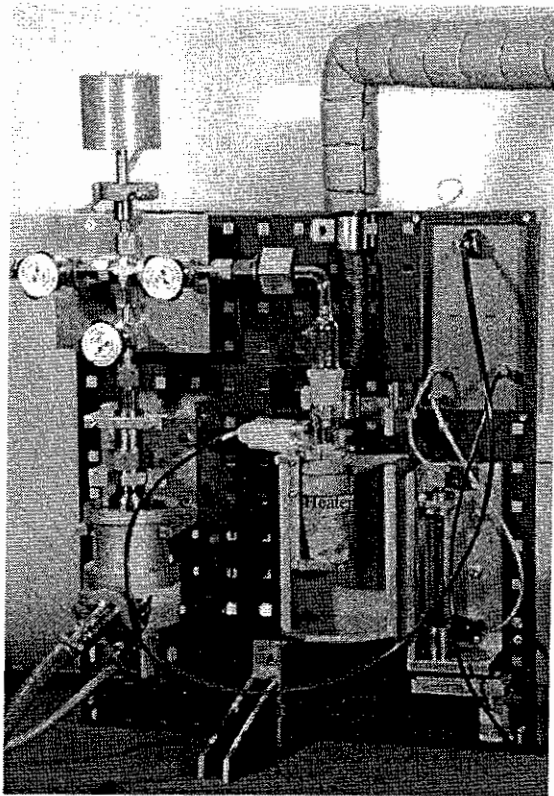
The equipment looks like this in the initial setup. One can see the different parts of the instrument, for example the valves, the specimen tube, the liquid Nitrogen inlet and the elevator.



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Initial  
Setup

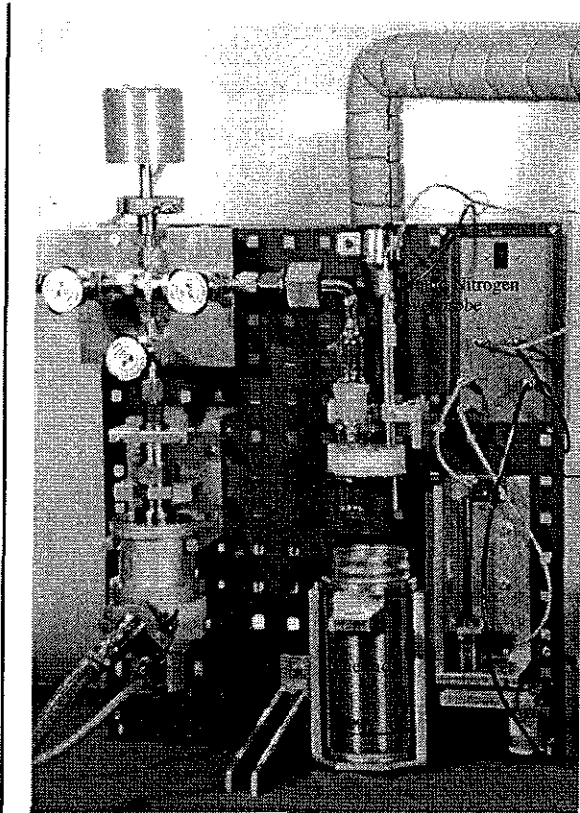
In the degassing setup a circular heater is placed around the sample, as shown.



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Degassing  
Setup

In this setup the instrument is ready for measuring, but for explanation the thermos flask is in its lowest position. One can see the probes for liquid Nitrogen level and temperature control. There is an insulation layer for the liquid Nitrogen.

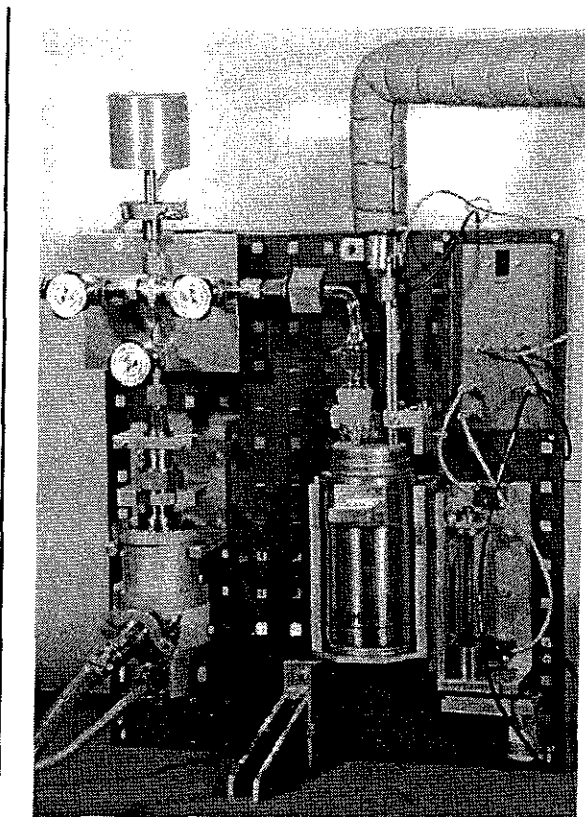


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Pre-measuring  
Setup



This is the measuring setup with the elevator in the top position.



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Measuring  
Setup

It was found that the amount of surface to be measured was at the limit for the instrument. A further concern with measurement on spent fuel is the decay heat. Extensive tests were performed both before and after installation in the cell.

Finally measurements were performed on fresh fuel, on PWR and BWR fuel with a burnup of about 40 MWd/kgU and on fuel with a burnup of 70 MWd/kgU.

The measured values were all in the range 50-300 cm<sup>2</sup> per gram.

This corresponds to a surface to volume ratio of 500-3000 cm<sup>-1</sup>.

The average intercept length of separated particles is equivalent to  $4x(S/V)^{-1}$ , so that a surface area of 500-3000 cm<sup>-1</sup> corresponds to an average intercept particle size of 13-80 μm. This should be compared to the average intercept length grain size of 5-10 μm, and the fuel fragment size of >1 mm (>98 % by weight).

The measurement reproducibility was generally well within ±20 %.

## Measurements

- Unirradiated and irradiated fuel (40-70 MWd/kgU)
- Measured specific area 50-300 cm<sup>2</sup>/g  
 $S/V = 500-3000 \text{ (cm)}^{-1}$
- Mean intercept length of particles =  $4x(S/V)^{-1}$   
So:  $500-3000 \text{ (cm)}^{-1} = 13-80 \text{ μm}$
- Compare to
  - Fragment diameter >1 mm (>1000 μm)
  - Mean intercept length grain size 5-10 μm
- Reproducibility ±20 %

One of the few problems that occurred with the equipment was that an electrical component in the turbomolecular pump didn't resist the radiation. This was solved by shielding the pump with lead blocks.

The comprehensive sample preparation, for example: cutting of the rod, grinding of the fuel, sieving the fuel and weighing, means that every test cycle is time consuming. Pumping with leak tests, degassing of the sample, cooling of the sample and measuring, takes about forty eight hours.