

The Belgian Program on the management of the bituminised radioactive waste.

09:00 Welcome

09:05 Context and global objectives of the exchange meeting (J.P. Boyazis, ONDRAF/NIRAS)

General considerations:

09:20 Origin, types, bituminisation processes and inventory of the bituminised radioactive waste produced in Belgium (C. Cosemans, ONDRAF/NIRAS and A. Meeus, BELGOPROCESS)

Short and middle-term management of bituminised radioactive waste

09:50 Behaviour and evolution of bituminised radioactive waste during interim storage (G. Van Zaelen *et al.*, ONDRAF/NIRAS)

10:30 Coffee break

Long term evolution of the Eurobitum bituminised waste

10:45 Handling of Eurobitum bituminised waste in safety assessment and implication for the R&D program (P. De Preter *et al.*, ONDRAF/NIRAS)

11:15 Evolution of the characteristics of the Eurobitum matrix (F. Rorif *et al.*, SCK•CEN)

11:40 Impact of the radiolytic and chemical degradation products on the sorption of plutonium and americium in undisturbed Boom Clay (E. Valcke *et al.*, SCK•CEN)

12:00 Lunch

13:15 Processes related to the uptake of water and release of NaNO_3 by Eurobitum bituminised waste (E. Valcke *et al.*, SCK•CEN)

14:15 Gas generation and potential impact on repository performance (D. Mallants *et al.*, SCK•CEN)

14:40 Research into possible alternative solutions for the long term management of bituminised radioactive waste (N. Impens, SCK•CEN and F. van den Bergh, SHELL)

15:20 Coffee break

15:40 Discussion with a focus on the acceptability of bituminised waste for disposal and compatibility of this waste with the Boom Clay host formation

16:45 Closure drink



ONDRAF/NIRAS



**STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ÉTUDE DE L'ÉNERGIE NUCLÉAIRE**



ESV EURIDICE GIE

The Belgian Program on the Management of Bituminised Radioactive Waste

Exchange Meeting

26th October 2004

Club House SCK•CEN, Mol

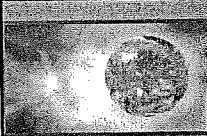


Bituminisation processes at Belgoprocess

André Meeus & Paul Luycx
Waste Processing Department of
Belgoprocess, Dessel, Belgium



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Bituminisation processes at Belgoprocess

- **Batch process**
Mummie
Bituminisation for low-level
water purification sludges
- **Continuous process**
Eurobitum
Bituminisation of
decontamination solutions



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Research into possible alternative solutions for the long term management of bituminised radioactive waste

N. Impens

SCK•CEN

Exchange meeting about bituminized radioactive waste 1

26/10/2004

Just in case...

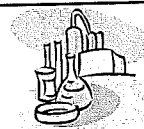
- Belgian Reference Concept = geological disposal
- If phenomenological studies would show that this would be unsafe, another solution is needed
- Therefore, we study bituminised waste re-treatment

Which bituminised waste first?

- We first look at wastes originating from ...
 - nuclear fuel decladding slurries and/or waste concentrates from the nuclear fuel cycle
- ... containing
 - High amounts of NaNO_3
 - BaSO_4 and/or CaSO_4
 - Radiotoxic and chemotoxic species

The retreatment process should be applicable to other homogeneous bituminised wastes

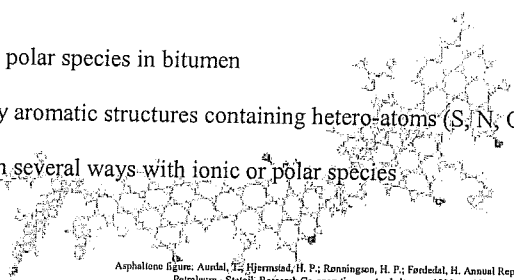
Possible solutions



- Geological disposal
- Chemical re-treatment: a combination of existing technologies
 - Petroleum technology
 - Nitrate destruction
- Plasma re-treatment

Bitumen = maltenes + asphaltenes

- **Maltenes are**
 - saturates, aromates and resin-like fractions of bitumen
 - The more saturated a molecule, the more sensitive it is to radiolysis
- **Asphaltenes are**
 - the largest, most polar species in bitumen
 - complex, mainly aromatic structures containing hetero-atoms (S, N, O)
 - able to interact in several ways with ionic or polar species



Asphaltene figure: Aurdal, T.; Hjermstad, H. P.; Rønning, H. P.; Færevik, H. Annual Report Saga Petroleum - Statoil Research Co-operation on Asphaltene, 1998, pp 100.

Our approach is to separate unstable fractions from radioactive waste



- **Separate maltenes**
 - Most sensitive towards radiolysis
- **Separate water soluble species**
 - Mostly NaNO_3 but also soluble chemotoxic species

This results into concentration of the radioisotopes into the asphaltene phase. Result is a graphite-like solid.

Our two step separation looks promising!

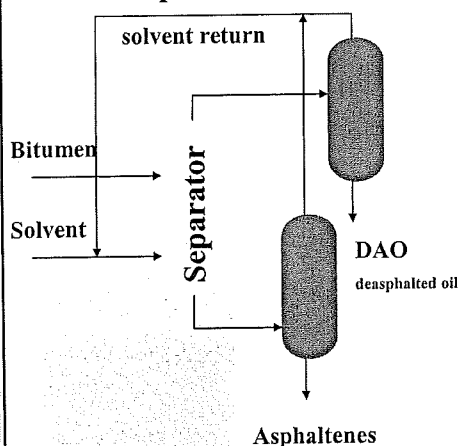
- 1. Separation of maltenes through adapted solvent deasphalting process**
- 2. Separation of water soluble matter from the precipitate**

STEP 1

Maltene separation through adapted solvent deasphalting process

Solvent deasphalting - general

Simplified flow scheme



- Commercially applied in hundreds of applications

➢ Lubricating oil manufacture

Example: Shell Pernis refinery, 1500 t/d

➢ Production of conversion feedstock

Example: Texas City refinery, 6500 t/d

➢ Tarsands upgrading

Ex: Alberta Oil Sands Project, 20.000 t/d

- Suppliers: KBR (ROSE process), UOP (Demex), Foster Wheeler

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Solvent deasphalting – process parameters

- **Solvent type** (lower MW hydrocarbons produce cleaner DAO but in lower yield)
- **Bitumen feedstock quality** (a.o. asphaltene concentration)
- **Solvent to bitumen ratio** (higher S/B ratios give better separation, at a higher cost)
- **Temperature** (higher T gives less DAO but at better quality)
- **Separator design** (various types exist, e.g. packed column, rotating discs, tilted plates)

Solvent deasphalting-advantages

- Proven technology, easy to operate
- Robust: range of options for optimisation and product quality control
- Hardware relatively cheap (low capital expenditure);
 NB: operating costs determined by energy costs
- Theoretical framework available (solubility theory, thermodynamics) – modelling relatively straightforward.

Solvent deasphalting - application to radio active waste

- Much smaller unit required – may be batch process instead of continuous and makes solvent recycle unnecessary. DAO/solvent stream may be used as afterburner fuel for nuclear incinerator
- Different solvent for safety reasons (tests were done with C11/C12 mixture).
- Work-up of asphaltene stream (with radio-active material) is different. Product is expected to be a hard solid at ambient temperature and require high temperatures for pumping.

→ Further development work required

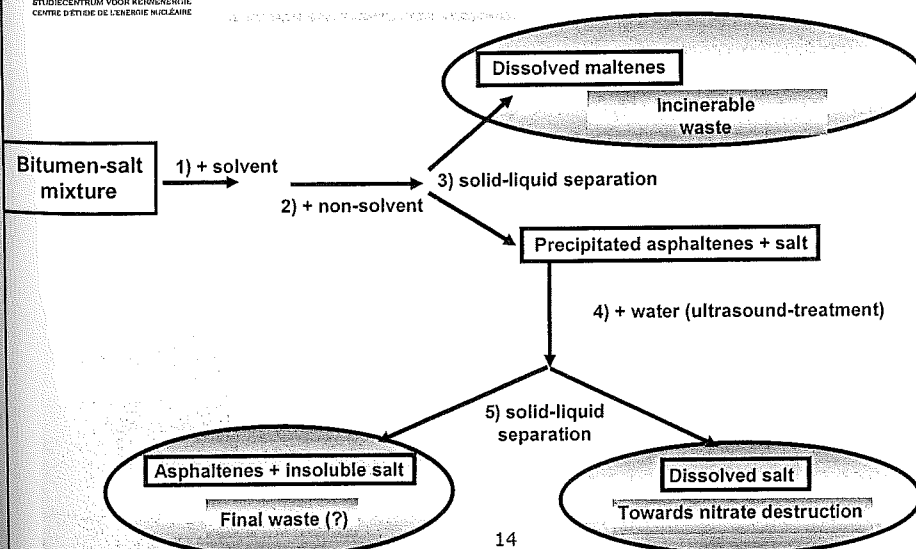
STEP 1
Maltene separation through “adapted” solvent
deasphalting

STEP 2
Removal of water soluble salts

HOW?

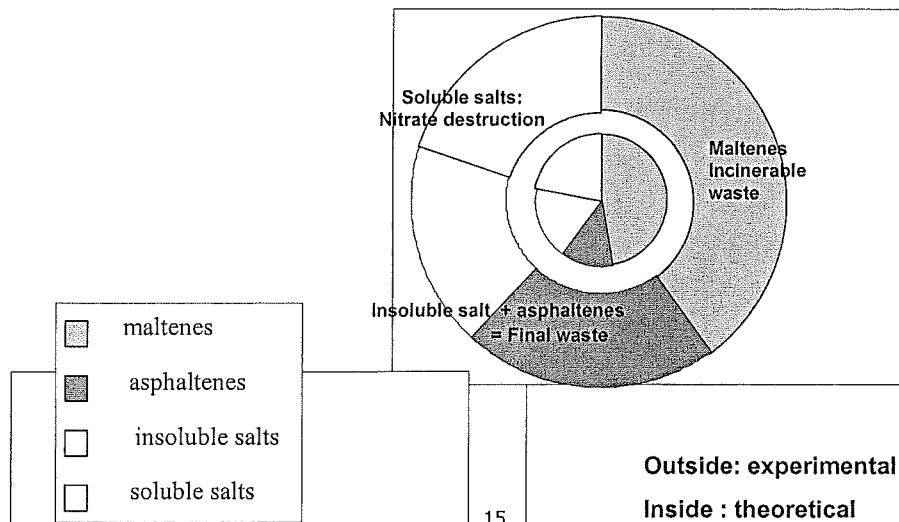
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Separation into 3 fractions



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Resulting waste fractions



Results of maltene separation *

- “Ash free” incinerable fraction obtained
 - Detection limit for fraction of minerals going to the maltene fraction = $1 \cdot 10^{-5}$
 - We want to improve the detection limit using radioisotopes
- Other batches should be checked based on inventory. We need inventory.

*N. Impens et al., Energy and Fuels, submitted

Step 2: Separation of soluble salts

- If pure water is used, the composition of the secondary aqueous waste is mostly determined by the quality of the Neutralisation Self Solidification Process applied at Eurochemic
- Optimisation of aqueous secondary waste purity not tested
- Nitrate destruction (towards N_2) not tested

Chemical re-treatment TO DO (1)

- Optimise the dissolution of water soluble salts
 - Radioisotopes should remain in solid phase!
- Test the precipitation on several bitumen-salt mixtures representing the waste which could be considered for re-treatment
 - We need inventory
- Set up the precipitation tests using neutron activated bitumen salt mixtures or real samples
 - We can follow up the precipitate formation using gamma spectrometry

Chemical re-treatment TO DO (2)

- Secondary waste reduction
 - Maltene incineration tests (BP?)
 - Nitrate destruction research
- For the process engineers: From lab scale tests towards process conditions

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Chemical re-treatment: TO DO (3)

- Applicability towards other types of bituminised waste

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...Or should we look at **plasma** treatment?

- Plasma treatment of nuclear waste is considered as a versatile tool
- A literature and theoretical study has been started very recently

Conclusions (1)

- A chemical re-treatment method for bituminised waste is studied. It is partially based on a well-demonstrated deasphalting process, but adapted for safety reasons
- The aim of the re-treatment is to stabilise the waste by separation of species sensitive towards radiolysis and separation of water soluble (mostly nitrate) salts
- Up till now, the method shows promising results for 1 studied bitumen-salt batch

Conclusions (2)

- Now that we have set up a method to test the re-treatment, we can proceed using radioactive test material to improve the detection limit
- Tests should be extended towards other bitumen-salt batches and other bituminised waste types
- We need the waste inventory to select the most appropriate test batches to cover the existing range of bituminised waste

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Conclusions (3)

- Last but not least: A thorough study on plasma treatment should be considered too.

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Acknowledgements

- Frans van den Berg, Shell Global Solutions – Amsterdam
- Wamibox Research Group
Jo Van Laer, Fons Fonteyne, Stefan Dekelver, Liliane Vos, Robert Vandevoorde
- Swat Janssens: Graphical assistance

Behaviour and evolution of bituminised radioactive waste during interim storage

**Exchange Meeting October, 26th 2004
Mol, Belgium**

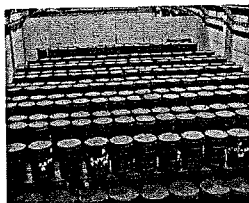
G. Van Zaelen (NIRAS), A. De Goeysse (NIRAS), B. Ghys (Belgoprocess)



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Exchange Meeting Bituminised Waste

26/10/2004



- Requirements for interim storage
- Storage facilities used for bituminised conditioned waste
- Physical inventory
- Behaviour and evolution
- Conclusions



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Exchange Meeting Bituminised Waste

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Requirements for interim storage (1/3)

- Mechanical stability requires containment vessel
 - To maintain integrity and quality during storage
- Remote vertical stacking of drums
 - Avoid possible flow of bitumen
 - No horizontal storage of drums



Requirements for interim storage (2/3)

- Reduce swelling in the bituminised waste
 - Natural shrinkage after cooling
 - Allows space for swelling
- Consider reduction of extent to filling of drums
- Avoid water uptake
 - E.g. thin layer of pure bitumen on top of bituminised waste



Requirements for interim storage (3/3)

- Reduce swelling in the bituminised waste
 - Presence of radiolytic gases requires 2 actions
 - Drums must not be closed tightly
 - Build up of Hydrogen in building or container needs to be limited
- Shielding provisions as result of the low self-shielding of the bitumen
- Retrievable storage concepts



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Storage facilities used for bituminised conditioned waste (1/8)

Storage buildings

BUILDING	WASTE CATEGORY	DOSE RATE (contact)	CAPACITY
127 bunker 1-3	B	> 5 mSv/h and < 2 Sv/h	3 x 1100 m ³
127 bunker 4	A and B	> 5 mSv/h and < 2 Sv/h	1350 m ³
151	A and B	< 5 mSv/h	14000 m ³
270M	A and B	< 5 mSv/h	2000 m ³

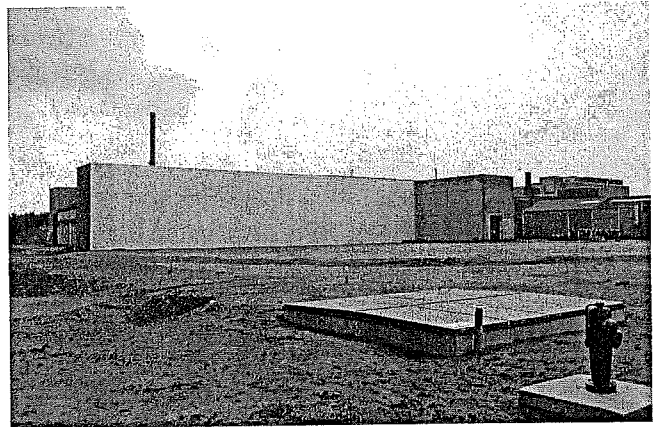


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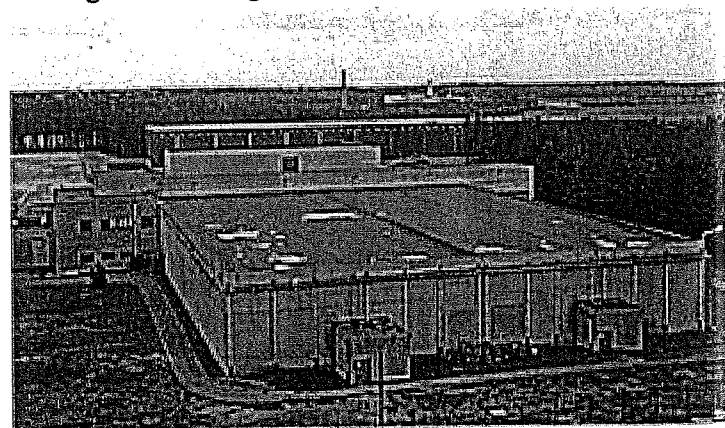
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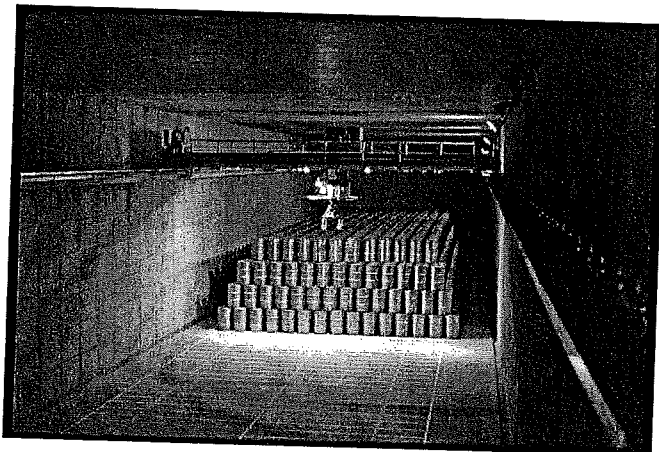
Storage facilities used for bituminised
conditioned waste (2/8)
Storage building 127



Storage facilities used for bituminised
conditioned waste (3/8)
Storage building 127



Storage facilities used for bituminised
conditioned waste (4/8)
Storage building 127

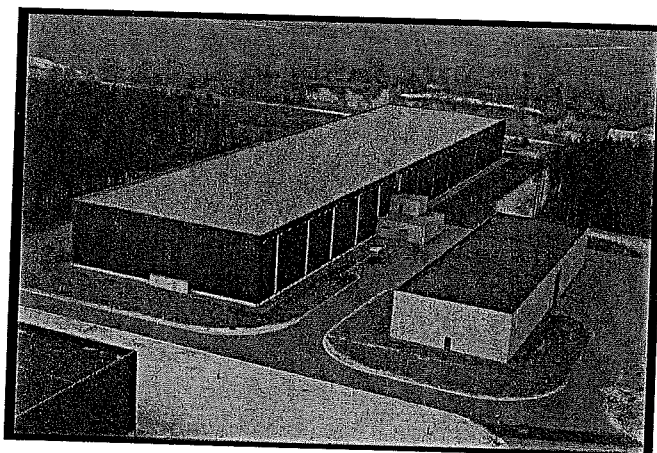


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Storage facilities used for bituminised
conditioned waste (5/8)
Storage building 151



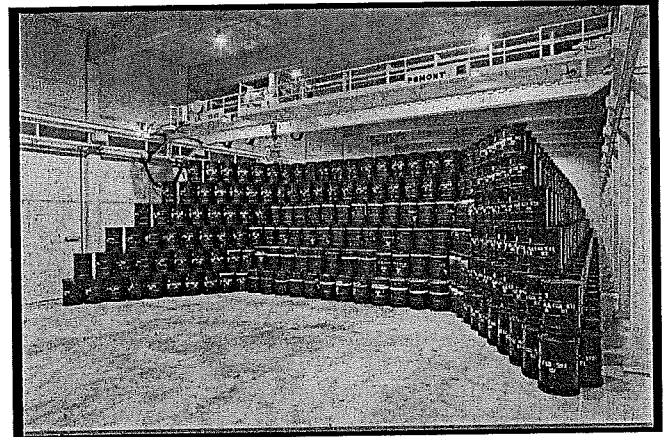
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Storage facilities used for bituminised conditioned waste (6/8)

Storage building 151



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Storage facilities used for bituminised conditioned waste (7/8)

Storage building 151



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Storage facilities used for bituminised conditioned waste (8/8)

Storage building 270M



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Physical inventory (1/6)

Waste productions - Terminology

- Historical Waste
 - Waste productions < February 10, 1999
 - Passive Waste
 - Waste productions < January 1, 1989
- Current Waste
 - Waste productions > February 10, 1999



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Physical inventory (2/6)

- Conditioning processes
 - EUROBITUM (homogeneous bituminised waste)
 - MUMMIE (homogeneous bituminised waste)
 - 'Alpha Chamber' (heterogeneous bituminised waste)
- Storage Bitumen Category A and B
 - Building 127
 - Building 151
 - Building 270M

Physical inventory (3/6) EUROBITUM

Storage
building 127
bunkers 1, 2 and 3

Category : B

Year	Drums	Type of package
1978	499	220 l
1979	1845	220 l
1980	2840	220 l
1981	2809	220 l
1982	1926	220 l
1983	1839	220 l
1984	293	220 l
1985	48	220 l
1986	151	220 l
1987	43	220 l
1988	64	220 l
1989	166	220 l
1990	146	220 l
1991	116	220 l
1994-1995	16	220 l
1997-1998	743	220 l
TOTAL	13544	drums

Physical inventory (4/6) MUMMIE

Storage
buildings 127, 151
and 270M

Category : A or B

Year	Drums	Type of package
1982	462	220 l
1983	241	220 l
1984	88	220 l
1984	63	400 l
1985	73	400 l
1986	39	400 l
1987	4	400 l
1989	56	400 l
1990	184	400 l
1991	164	400 l
1992	180	400 l
1993	134	400 l
1994	91	400 l
1995	128	400 l
1996	138	400 l
1997	120	400 l
1998	95	400 l
1999	95	400 l
2000	55	400 l
2004	39	400 l
TOTAL	2449	drums



Physical inventory (5/6) Heterogeneous bituminised waste

Storage
buildings 151
and 270M

Category : A or B

Year	Drums	Type of package
1975	14	400 l
1983	624	220 l
1984	322	220 l
1985	384	220 l
1986	180	220 l
1986	569	400 l
1987	131	220 l
1988	21	220 l
TOTAL	2245	drums



Physical Inventory (6/6)

- Building 127 bunker 1
 - 5028 drums EUROBITUM
- Building 127 bunker 2
 - 5006 drums EUROBITUM
- Building 127 bunker 3
 - 3510 drums EUROBITUM
- Building 127 bunker 4
 - 70 drums MUMMIE
- Building 151
 - 1634 drums MUMMIE
 - 2026 drums heterogeneous bituminised waste
- Building 270M (Frisomat)
 - 745 drums MUMMIE
 - 219 drums heterogeneous bituminised waste
- **TOTAL : 18238 drums of bituminised conditioned waste**



Behaviour and evolution (1/7) Follow up condition of stored drums

- Building 127
 - During project optimisation of the dose in bunker 3 visual inspection by camera
 - Visual Inspection Program : → planning
- Building 151
 - Periodical inspection of drums and building
 - Visual Inspection Program : → planning
- Building 270M
 - 6-monthly inspection of drums



Behaviour and evolution (2/7) Visual Inspection Program

- Inspection of all stored drums
 - Started in 2003
 - Objective : to classify drums based on acceptance criteria
- Planning:

Year	2004	2005	2006	2007	2008	2009	2010	2011	2012
Building									
•151									
•127 bunker 4									
•127 bunker 1									
•127 bunker 2									
•127 bunker 3									

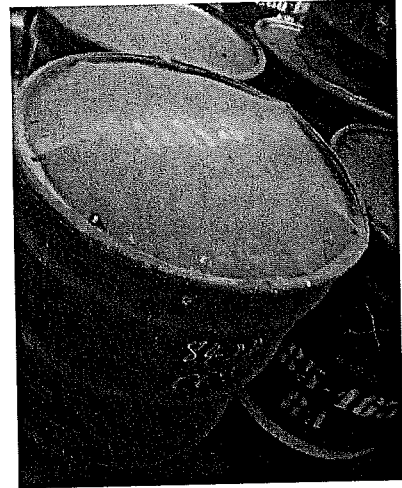


Behaviour and evolution (3/7)

- Drums EUROBITUM
 - No problems observed
- Drums MUMMIE
 - No problems detected
- Drums heterogeneous bituminised waste
 - **Swelling of drums**



Behaviour and evolution (4/7) Heterogeneous bituminised waste



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Behaviour and evolution (5/7) Heterogeneous bituminised waste



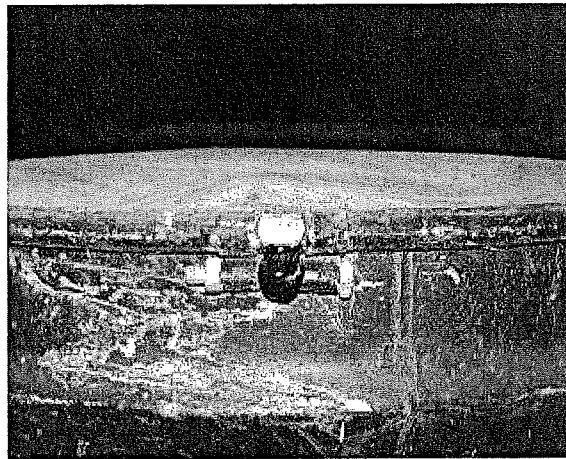
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Behaviour and evolution (6/7)

Heterogeneous bituminised waste



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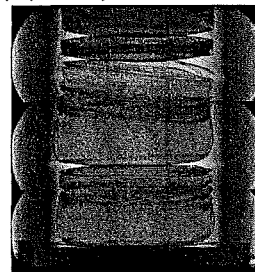
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Behaviour and evolution (7/7)

Project 'Determination of the origin of the swelling of drums for heterogeneous bituminised waste'

- Objective : inventory of waste (chemical and radiological contents, origin of waste) with regard to the swelling of drums
- Assessment (origin of swelling) (CEA)
 - Localisation of gas
→ TOMOGRAPHY
 - Evaluation of risk for non-swollen drums
- Definition of scenarios for reconditioning of drums



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Conclusions

- Behaviour and evolution of bituminised waste
 - Homogeneous waste (EUROBITUM and MUMMIE)
 - No problems observed so far with regard to conditioning process
 - Heterogeneous waste ('Alpha Chamber')
 - Swelling of drums → determination of cause
- Follow up
 - Periodical inspection of drums
 - Visual Inspection Program



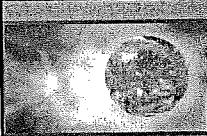


Bituminisation processes at Belgoprocess

André Meeus & Paul Luycx
Waste Processing Department of
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Bituminisation processes at Belgoprocess

- **Batch process**
Mummie
Bituminisation for low-level
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- **Continuous process**
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Bituminisation of
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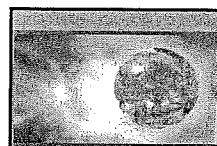
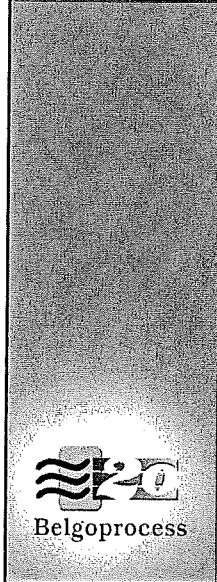
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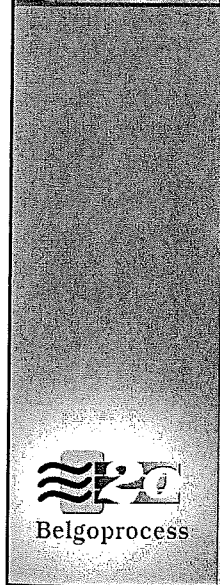
	Origin of low-level sludges
	• Effluents mainly produced by nuclear energy research and radioactive waste treatment • Water purification by co-precipitation and flocculation, followed by decantation ~ purified water is discharged to river ~ sludge is conditioned in mummie

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	Process description (1/3)
	• Transfer of sludge to 20 m³ buffer tank ~ homogenisation, sampling, analysis eventually chemical pre-treatment • Transfer of sludge to 1.2 m³ tank for a freeze (to -15°C) thaw cycle • Transfer of sludge to 1 m³ feed tank ~ further homogenisation, sampling determination of dry substance content

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	Process description (2/3)
	• Filtration of sludge in filter ~ solids content increased to 40 % • Transfer of dried waste to bitumen bath through hopper and horizontal double screw

	Process description (3/3)
	• Bituminisation in 400 l tank by mixing with blown bitumen at 205 – 215 °C • Pouring of end product (40 wt% waste solids) into 400 l drums in several steps

	Operational experience
	• Since 1989 • 202 t dry substance • 747 t end product • 592 m³ end product

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	Eurobitum plant
	• Constructed in 1970s to process Eurochemic's ~ chemical decladding wastes ~ other MLW from reprocessing • Conditioning of medium level liquid wastes • In operation since 1978

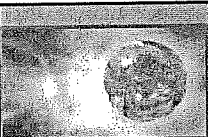
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	Bituminisation process (1)
 Belgoprocess	• Elaborate characterisation of feed product • Specific batchwise chemical pre-treatment ~ aim: ☐ insolubilise long-lived radionuclides ☐ eliminate corrosive compounds ☐ eliminate ammonium ☐ good ratio liquid-dry solids 9

	Bituminisation process (2)
 Belgoprocess	~ achieved by: ☐ sequential precipitation ☐ ammonium boil off 10

	Bituminisation process (3)
 Belgoprocess	• Incorporation into blown bitumen using a four screw extruder-evaporator ~ continuous operation ~ end product: salts: bitumen ratio of 40:60 ~ temperature just below 200 °C to exclude risk of exothermic reaction to ensure efficient evaporation

	Bituminisation process (4)
 Belgoprocess	• Extruder ~ nominal evaporation rate of 80 l/h jacket heated by saturated steam at 20 bars ~ 4 feed lines: waste slurry, molten bitumen; water and organic solvent ~ water vapours routed to condensers through 3 domes allowing visual checking

	Bituminisation process
 Belgoprocess	• Drum filling station ~ protruding outlet of extruder ~ revolving table with 6 positions ~ level detection in drum during filling ~ conveyors and carriage on rails for drum transport ~ end product packed in 220 l drums ▫ 1st step: up to 70 % of drum volume ▫ 2nd step: up to 90 % of drum volume

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	Bituminisation process
 Belgoprocess	~ telemanipulator and overhead crane ~ external contamination check ~ measurement of radioactivity contents of drums • Extruder operation and drum manipulations remotely controlled

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Operational experience

- 32.000 hours of active operations
- 2955 m³ of end product



Belgoprocess

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Historical process

- Heterogeneous
Bituminisation of low level solid or
solidified wastes

Aim: sea dumping



Belgoprocess

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	Origin of wastes
 Belgoprocess	■ Belgonucleaire ■ FBFC ■ Eurochemic ■ SCK-CEN ■ Doel - Tihange

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	Waste forms
 Belgoprocess	■ Solidified liquids ■ Precompacted incinerator ashes ■ Precompacted filters ■ Precompacted non-burnable waste ■ Non-burnable waste

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Process

■ Pretreatment

- ~ Incineration
- ~ Compaction
- ~ Solidification of liquids
- ~ Cutting

■ Putting solid waste in 220 l-drum (50 % in volume)

■ Pouring hot bitumen on waste in 2 steps

■ Reconditioning of 220 l-drums



Belgoprocess

Handling of bituminised waste in safety assessment and implications for the R&D program

Exchange meeting 26 October 2004

P. De Preter – NIRAS/ONDRAF



26:10/2004

General aims of disposal (1/2)

1. Prevent as long as possible or needed any dispersion
 - No release
 - Decay within the system or part of the system
 - Total impact reduced
2. Slow down as much as possible or needed releases to the environment
 - Retardation within the system → decay → total impact reduced
 - Physical consequence of retardation → spreading of releases → total impact spread in time



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General aims of disposal (2/2)

3. Prevention of direct access to the waste
 - Prevention of direct exposure
 - Avoidance of system disturbance and waste denudation
 - By men
 - By natural phenomena



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Long-term safety functions (1/4)

- **Physical containment (C)**
 - Prevention of contact water – waste → no dispersion of radionuclides
 - Effective containment = decay within first barrier = total impact diminishes
 - At least effective during transient (mainly thermal) phase of disposal system (100s – 1000s y for vitrified waste and spent fuel)
 - Two sub-functions
 - **Water tightness (C1)** = impermeable barrier
 - **Limit the water influx (C2)** = slow and limited water infiltration



ONDRAF/NIRAS

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Long-term safety functions (2/4)

- **Delay and spread the release (R)**

- After partial or total containment failure : limit and slow down radionuclide migration / dispersion within the disposal system
- Decay within disposal system = total impact diminishes
- Eventual releases in the biosphere spread in time = impact per unit of time (generations !) diminishes
- Function depending on
 - Type of radionuclide
 - Transport characteristics of components



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Long-term safety functions (3/4)

- **Delay and spread the release (R) – cont.**

Two sub-functions

- **Slow release (R1)** = spreads in time radionuclide releases from waste matrix into near field (resistance to leaching, co-precipitation and secondary phases, ...)
- **Diffusion et retention (R2)** = slow migration within the disposal system.

This delays and spreads in time radionuclide releases into the environment

(retention includes both sorption and precipitation)



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Long-term safety functions (4/4)

- **Limitation of access**

- Prevention of direct access to the waste
- Avoidance of human intrusion
- Resilience of the system in case of intrusion
- Natural events (denudation of the waste)
- Examples of possible contributing factors
 - Depth and "impenetrable" barriers
 - Absence of natural resources
 - Institutional control period
 - Land use restrictions
 - Memory and markers



Role of the environment

- **Dilution and dispersion (D)**

- Dilute and disperse the radionuclide fluxes that eventually leave the disposal system
- Water bearing strata and biosphere (rivers, lakes, water wells, ...)
- Not to be optimised, but effects radiological impact (linear effect of dilution factor)
- Not a primary site selection criterion



Possible states of the safety functions

- In the successive time frames of the life time of the system different functions contribute to the safety of the system
- Various possible states of a function
 - **Effective**
 - **Latent**: function that becomes partially or totally active only when other safety functions do no longer achieve the expected performances
 - **Reserve** safety function: function that, at present, is not sufficiently well characterised to be fully relied upon in the safety case, but that contributes to the confidence in the safety



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Bituminised waste in the disposal system

- Range of different types of bituminised waste (see previous presentations)
- Safety considerations in this presentation valid for all types
- Eurobitum waste presents most challenging issues
 - Physico-chemical characteristics
 - Total volume
 - Total activities



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Bituminised waste in the disposal system

1. Direct contribution from bituminised waste form to the safety functions
2. Effect (negative) of bituminised waste form on the safety functions of other components



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Direct contributions to the safety functions (1/2)

Physical containment

- No thermal phase → no required initial containment (water tightness – C1) of bituminised waste
- Boom Clay will limit water inflow (C2 function) in a sealed repository - no role for waste forms in general



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Direct contributions to the safety functions (2/2)

Delay and spread the release

- Slow release from waste form (matrix) – R1
 - Bituminous matrices will slowly release radionuclides
 - Release rates depending on evolving matrix properties (aging phenomena as a result of radiolysis a.o.)
 - Instantaneous release from waste form considered in assessments for Category B waste
 - So, R1 to be considered reserve safety function



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Effects on the safety functions (1/5)

Delay and spread the release

- Diffusion and retention (R2) function of near field (EBS) and far field (host rock)
 - R2 of host rock dominates R2 of EBS
 - Bituminous waste form can negatively affect R2 of host rock:
 - Mechanical disturbance due to pressure build up by water uptake or by gas production – affecting the performance of the Boom Clay as a migration barrier
 - Chemical disturbance by migrating chemical substances from the waste form (heavy metals, NaNO_3 – 750 ton in $\approx 3000 \text{ m}^3$ of waste)



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Effects on the safety functions (2/5)

- Mechanical disturbance by swelling/water uptake
 - Swelling rate controlled by rate of water uptake and production of gases (radiolysis)
 - Does swelling rate causes rapid or slow disturbances ?
 - How will it affect the barrier function of a plastic clay ?
 - How many meter (cm) of Boom Clay can we loose ? – knowing that a few meters is not problematic (see excavation disturbed zone)
 - Swelling phenomena affected by matrix aging



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Effects on the safety functions (3/5)

- Mechanical disturbance by gas production
 - Mainly anaerobic corrosion (Stainless steel and Cr-carbon steel)
 - Biodegradation and radiolysis contribution relatively uncertain, but probably of minor importance = general conclusions all waste types
 - Gas production situation comparable to Category A waste disposal in Boom Clay (see presentation D. Mallants)



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Effects on the safety functions (4/5)

- Chemical disturbance – NaNO_3 migration
 - 750 ton of NaNO_3 in $\approx 3000 \text{ m}^3$ waste
 - Can this quantity of NaNO_3 significantly change the performance (migration properties) of the clay ?
 - Ionic strength effects
 - Redox effects
 - Sorption competition effects
 - How many meters (cm's) of Boom Clay can we loose ?



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Effects on the safety functions (5/5)

- Chemical disturbance - formation of complexing agents
 - Chemical and radiolytic degradation $\rightarrow$ formation of complexing organic molecules (oxalate, acetate, formate, ...): international evidences for unimportance of effects
 - Biodegradation: extremely slow process in closed disposal system + non-complexing end products
 - Organic molecules present in Boom Clay
 - Significant effects not to be expected



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In conclusion

- Safety function approach allows systematic analysis of the question
- Key processes from disposal system safety evaluation = effect on performance of the natural clay barrier
 - Mechanical pressure build up by water uptake (rates influenced by aging phenomena)
 - Chemical disturbance by NaNO_3 plume



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Inventory of Bituminised Radioactive Waste

Exchange Meeting 26th October 2004
Mol, Belgium

Christian Cosemans
Hughes Van Humbeeck (presentation)



Overview

- Objectives of an inventory
- Classification of the conditioned radioactive waste
- Classification of the Bituminised Waste Products (BWP)
- Quantities of BWP
- Chemical content
- Radiological content
- Conclusions



Inventory of Radioactive Waste

Objectives of an Inventory

- Inventory = legal obligation for ONDRAF/NIRAS (Royal Decree 12/12/1997) to
 - Establish a repertory of all radioactive **substances** present on the Belgian territory (existing waste included)
 - Determine the potential nuclear liabilities (further waste have to be evaluated)
- Also required to obtain a global view on all radioactive **waste** that already exists and will exist on Belgian territory in order to :
 - Plan the interim storage needs (capacities of the storage buildings)
 - Provide input data for final disposal program

→ Information needed : quantities, chemical and radiological contents



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Inventory of Radioactive Waste

On the waste classification system

A : Surface disposal possible
B and C : Only geological disposal

	Low Level	Intermediate Level	High Level
Short Lived (30 years or less)	A	A	C
Long Lived (more than 30 years)	B	B	C



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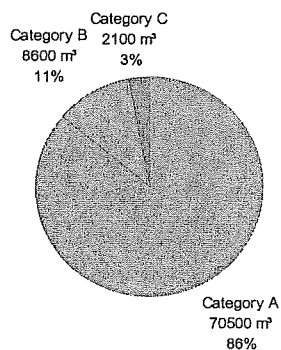
4

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Inventory of Radioactive Waste

Quantities

Total Inventory = 81.200 m³
(produced and to be produced)



Inventory of Radioactive Waste

Classification (1/2)

Bituminised Waste = Cat A as well as Cat B

- **HETEROGENEOUS BWP** (produced in the 80's)
 - Mainly waste produced by the former SCK Waste Department
 - Chemical and radiological characterisation : preliminary actions are running (detailed examination of the existing information and preliminary experimental program)
→ category A and B
- **MUMMIE BWP**
 - Waste produced by the former SCK•CEN Waste Department (< 1989) and Belgoprocess (> 1989) : sludges from treatment of low level aqueous effluents produced by R&D activities (mainly SCK•CEN)
 - Chemical and radiological characterisation : in progress (Belgoprocess production)
→ Category A and B



Inventory of Radioactive Waste

Classification (2/2)

Bituminised Waste = Cat A as well as Cat B

■ EUROBITUM BWP

- Mainly low and middle active level waste long lived produced during reprocessing of spent fuel – decontamination solution (EUROCHEMIC)
- Chemical and radiological characterisation well known
→ belong to B category

■ SYNATOM BWP (not yet present)

- Abroad reprocessing of spent fuel at COGEMA
- Chemical and radiological : some information already available
→ belong to B category



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Inventory of Radioactive Waste

Quantities (1/2)

Bituminised Waste = Cat A as well as Cat B

BWP	Types of package	Category A	Category B
		[# packages]	[# packages]
HETEROGENEOUS + MUMMIE BWP	220 l drum	(598)	(1.35)
	400 l drum	(2.038)	(2.7)
EUROBITUM BWP	220 l drum		13.5
	400 l drum		
COGEMA BWP	220 l drum		480 (900)
	400 l drum		

EUROBITUM and HETEROGENEOUS BWP : all drums already produced
MUMMIE : further campaign planned
COGEMA : not yet present in Belgium



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Inventory of Radioactive Waste

Quantities (2/2)

Bituminised Waste = Cat A as well as Cat B

BWP	Types of package	Category A	Category B
		[m³]	[m³]
HETEROGENEOUS + MUMMIE BWP	220 l drum	(132)	(307)
	400 l drum	(815)	(1.085)
EUROBITUM BWP	220 l drum		2980
	400 l drum		/
COGEMA BWP	220 l drum		106 (198 ?)
	400 l drum		/



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Inventory of Radioactive Waste

Chemical composition (1/3)

MUMMIE BWP (BELGOPROCESS production)

Mainly sludge and ... bitumen

Components	kg/drum	%
H2O	0	(< 0,2%)
Sludge	291	46%
Matrix Mexphalt 2030 (hard)	280	44%
Package	61	10%
Total (kg)	632	



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Inventory of Radioactive Waste

Chemical composition (2/3)

EUROBITUM BWP

Mainly salts and ... bitumen

Components	kg/drum	%
H2O	3	1%
NaNO3	63	27%
CaSO4	12	5%
Matrix Mexphalt 85/40 (hard)	129	55%
Package	19	8%
others	7	3%
Total (kg)	233	



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Inventory of Radioactive Waste

Chemical composition (3/3)

COGEMA BWP

Mainly salts and ... bitumen

Components	kg/drum	%
NaNO3	28	13%
BaSO4	29	14%
Matrix Mexphalt M80/100 (soft)	128	61%
Package	20	10%
Others	6	3%
Total (kg)	211	



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Inventory of Radioactive Waste

Radiological content (1/2)

Bituminised waste from Cat B (1/2)

Isotope	COGEMA [Bq/Cello]	EUROBIT [Bq/Cello]
H-3	3,57E+09	3,39E+07
C-14		2,79E+07
Cl-36		1,65E+01
Mn-54		9,33E+08
Fe-55		7,19E+11
Co-60	3,59E+10	5,91E+11
Ni-59		1,45E+09
Ni-63		2,16E+11
Se-79		5,33E+08
Kr-85		7,62E+10
Sr-90	2,42E+11	8,43E+11
Zr-93		4,65E+07
Nb-94		2,86E+08
Tc-99	3,57E+07	1,77E+08
Ru-106	1,07E+12	1,94E+10

Isotope	COGEMA [Bq/Cello]	EUROBIT [Bq/Cello]
Tc-99	3,57E+07	1,77E+08
Ru-106	1,07E+12	1,94E+10
Pd-107		9,65E+05
Ag-108m		2,22E+02
Ag-110m		3,54E+06
Sn-126		8,91E+06
Sb-125	8,00E+10	2,16E+10
I-129	3,57E+07	3,83E+05
Cs-134	8,90E+10	4,66E+10
Cs-135		6,11E+06
Cs-137	2,50E+11	1,07E+12
Ce-144	3,59E+11	1,38E+10
Pm-147		3,40E+11
Sm-151		8,29E+09



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Inventory of Radioactive Waste

Radiological content (2/2)

Bituminised waste from Cat B

Isotope	COGEMA [Bq/Cello]	EUROBIT [Bq/Cello]
Eu-154	3,21E+10	1,96E+10
Eu-155		1,37E+10
Ra-226		9,19E+00
Th-229		4,48E-01
Th-230		4,08E+03
Th-232		2,04E-03
Pa-231		3,79E+02
U-233		3,22E+02
U-234		4,41E+07
U-235	3,86E+02	1,23E+06
U-236		4,28E+06
U-238	3,86E+03	1,22E+07
Np-237		2,65E+06
Pu-238	9,45E+09	7,08E+09

Isotope	COGEMA [Bq/Cello]	EUROBIT [Bq/Cello]
Pu-239	1,09E+09	8,80E+09
Pu-240	1,52E+09	6,69E+09
Pu-241	5,88E+11	9,84E+11
Pu-242		6,48E+06
Pu-244		2,11E-01
Am-241	5,99E+09	1,81E+10
Am-242m		6,46E+07
Am-243		2,22E+07
Cm-242	1,69E+08	5,35E+07
Cm-243		1,19E+07
Cm-244	1,19E+09	5,37E+08
Cm-245		2,33E+04
α Act	1,94E+10	4,14E+10
β/γ Act	2,75E+12	4,99E+12



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Conclusions

- SYNATOM BWP
 - Chemical and radiological composition : qualification of the process and acceptance of the waste have still to be done (some information about characterisation already available)
- Following BWP are well characterised :
 - EUROBITUM BWP
 - o 55 % of the total volume of bituminised waste
 - o > 95 % of the total alpha activity of bituminised waste
 - ... But < 10 % of the total activity produced by the reprocessing of spent fuel from EUROCHEMIC
 - (> 90% present the vitrified PAMELA waste)
 - MUMMIE BWP (BELGOPROCESS production)
- For other BWP : characterisation in progress
 - HETEROGENEOUS BWP
 - MUMMIE BWP (former SCK WASTE DEPARTMENT)



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