



EUROPEAN  
COMMISSION

European  
Research Area

# Carbon-14 Source Term

## CAST



## Workshop 1 Proceedings (D7.11)

**Authors:**

**G. Buckau, D. Bottomley, E.A.C. Neeft**

Date of issue of this report: 14/11/2016

**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project**

Dissemination Level		
PU	Public	PU
RE	Restricted to the partners of the CAST project	
CO	Confidential, only for specific distribution list defined on this document	



EUROPEAN  
COMMISSION

European  
Research Area



## ***CAST – Project Overview***

The CAST project (CARbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focuses on the release of carbon-14 as dissolved and gaseous species from irradiated metals (steels, Zircalloys), irradiated graphite and from ion-exchange materials.

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of release of carbon-14 from the corrosion of irradiated steels and Zircalloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:

<http://www.projectcast.eu>

*CAST*  
*1<sup>st</sup> CAST Project Workshop Proceedings (D7.11)*

CAST		
Work Package: 7	CAST Document no. :	Document type: R
Task: 7.4	CAST-2016-D7.11	R = report, O = other
Issued by: JRC		Document status: Final
Internal no. :		Draft/Review/Final

Document title
Workshop 1 Proceedings

## **Executive Summary**

The 1<sup>st</sup> Workshop of the CAST project was held 5 - 6 October at the COVRA facilities, Vlissingen, Netherlands. The different actors involved in generating C-14 containing waste, managing the waste through to disposal, and regulating the activities responded to invitations to participate.

Presentations were given showing the status of project work and scientific-technical achievements, but also how the project work falls in the wider context. A series of topics were identified where R&D is still required in order to obtain the desired confidence in safety for geological disposal of the concerned waste materials. This is also relevant for the question to which extent confidence in safety for disposal in less ambitious facilities than deep geological ones can be justified, with or without pre-treatment of the waste material.

Two workshops are envisaged in CAST for participants with an interest in the research executed in CAST, but who can also contribute to the confidence in national safety assessments. The research is evaluated from different perspectives in order to specify this contribution. The scientific progress is already evaluated by the CAST Advisory Group and results obtained in CAST have been and will be presented at several scientific fora. For an implementation of the new understanding developed in CAST, stakeholders with a responsibility in the management of radioactive waste are envisaged. These stakeholders are regulators, waste management organisations and waste generators.

Performance indicators of the involvement of (type of) actors have been set in the CAST dissemination plan (D7.3). The involvement of regulators was a success: more than 50% of the countries with an organisation participating in CAST had send 1 or 2 persons from their national regulatory body to the workshop. The involvement of waste generators was less successful: 9 waste management organisations participate in CAST and was considered achievable to have at least 9 waste generators participating at the workshop. Three waste generators attended the workshop of which 2 are also waste management organisations for the types of waste investigated in CAST.

All EU countries contribute to the funding of EU research programs. But not every EU country has the same benefit of the executed research. Mainly Western EU waste management organisations - that are responsible for an end point management of the types of waste investigated in CAST - participate in CAST. Most Eastern EU waste management organisations with this responsibility participated in the workshop or contributed to the workshop proceedings.

The waste management organisations were requested to provide their radiological characterisation of the types of waste investigated in CAST for their participation in the workshop. As carbon-14 during its decay does not emit gammas, the radionuclide cannot be detected with usual monitoring (detection of gammas) like cobalt-60. Gamma-emitting radionuclides present in the waste investigated in CAST usually have a shorter half-life than carbon-14. The methodologies to determine the carbon-14 concentration in the waste appeared to vary a lot within the EU and in some EU countries carbon-14 has not yet been identified to be present for the types of waste investigated in CAST. The sharing of the methodologies and discussing them with the regulators present in the workshop is expected to upgrade the radiological characterisation and thereby increase the confidence of the carbon-14 concentration as input for the safety assessment but also the confidence in clearance of the neutron irradiated graphite and steel. Parts of these two types of dismantling waste may after some period be clarified as conventional waste while the ‘true’ carbon-14 concentration is larger than its clearance level as set in the latest council directive laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation.

The overall outcome of the workshop was that the different actors could exchange views and experiences, the state-of-knowledge was presented and discussed, and potential topics for consideration at the second workshop were identified.

List of Contents

Executive Summary ..... i

List of Contents ..... iii

1 Background and Objectives of WS1 ..... 1

1.1 Selected observations .....2

1.1.1 C-14 Sources Term.....2

1.1.2 C-14 Migration .....3

1.1.3 Biosphere .....3

1.1.4 Up-scaling, representativity and relevance .....3

1.1.5 Involvement actors .....4

2 The second CAST Workshop .....5

Annex .....I

## **1 Background and Objectives of WS1**

The overall objectives of working on the C-14 containing waste is to provide evidence for the:

1. safety of handling, treating, storing and eventually disposing the original or modified waste, and
2. possibility to justify using less costly management, and in particular less costly disposal routes.

The particular problems with C-14 containing waste is that the half-life does not allow disposal to rely on institutional control, that C-14 in the waste cannot easily be detected, the chemical form and thus the mobility of C-14 containing species released from the waste are difficult to assess, and that C-14 contributes to dose, especially if entering the carbon cycle of the biosphere. A final problem is that C-14 is generated over different routes, and the amounts and chemical form in waste components frequently rely on modelling under different assumptions, and to a lesser extent on a solid base of analytical data. Consequently, there is a great need to bring the overall understanding to a higher level of maturity. In order for the higher level of maturity to result in improved safety or less costly waste management, interaction and information exchange between the different actors involved is required, including regulators. The C-14 containing waste investigated in CAST are irradiated metals (steel and Zircaloy), irradiated graphite and spent ion exchange resins. Irradiated Zircaloy and spent ion exchange resins is usually operational waste. The largest volumes of irradiated steel and irradiated graphite usually appear at dismantling of nuclear reactors.

With this background, the 1<sup>st</sup> Workshop of the CAST project aimed at:

- a. Learning from the present waste management practices for the waste investigated in CAST in the European Union, Japan, Switzerland and Ukraine (organisations from these non EU countries also participate in CAST),
- b. Prepare the second workshop in which (traceable) information can be shared to characterise the waste to calculate disposal of the (processed) waste investigated in CAST;
- c. Presenting national safety assessments as examples of calculating the safety of disposal of the waste investigated in CAST but also
- d. Involving waste generators, waste management organizations and regulators in a discussion on what the status is, and where future work should go

The outcome is documented in the annex in the form of the Agenda, the presentations given, and the outcome of the Wrap-up and Closure. Some aspects are briefly discussed below.

## 1.1 Selected observations

Below some selected observations are briefly discussed. The aim is not to provide a summary of the information provided at the Workshop, nor the outcome of discussions or the state-of-knowledge in the field. The CAST work-programme during the remaining time does not allow for major changes, thus focus remains on ensuring implementation of the programme as agreed upon. Within this context, the selected observations aim at discussing some key questions, giving attention to topics that can be discussed at the final workshop, as well as preparing for documenting pending information and knowledge that could be dealt with in a follow-up activity.

### 1.1.1 C-14 Sources Term

The production routes of C-14 from nuclear reactors are<sup>1</sup>:

Parent Isotope	Natural abundance (%)	Cross section for thermal neutron capture	Reaction
<sup>14</sup> N	99.634	1.81	<sup>14</sup> N(n,p) <sup>14</sup> C
<sup>13</sup> C	1.103	0.0009	<sup>13</sup> C(n,γ) <sup>14</sup> C
<sup>17</sup> O	0.0383	0.235	<sup>17</sup> O(n,α) <sup>14</sup> C

C-14 containing material is thus generated from reactor operation, in particular:

1. Graphite from graphite moderated reactors,
2. Ion-Exchangers for keeping the water clean,
3. Zircaloy hulls and cladding, and
4. Steels.

The experimental programme in the CAST project considers all four types.

The C-14 content is estimated based on neutron activation calculations. These calculations are subject to assumptions. In the case of C-14 from nitrogen activation, the uncertainty about the nitrogen content of source material adds to the assumptive character of C-14 content

---

<sup>1</sup> <https://en.wikipedia.org/wiki/Carbon-14>

estimates. C-14 content is also estimated through scaling from other rather quantifiable radionuclides, such as Co-60. Comparison with measurement shows the limited applicability of such scaling. Some additional uncertainties are:

- Distribution of pre-cursors in the source material,
- Distribution of C-14 in the source material,
- Chemical form of C-14 in the source materials,
- Release mechanisms, and
- Chemical form as released from the source material

### 1.1.2 C-14 Migration

Once C-14 species are released from the source term, their migration will depend on their chemical form along the migration path, in combination with the physical, chemical and hydrological properties of the migration path. Sufficient confidence in chemical retention requires plausible evidence for speciation of the C-14 compounds. This is subject to considerable uncertainty, in particular as the chemical form may change along the migration path. Measurements of the migration of C-14 species is outside the scope of CAST.

### 1.1.3 Biosphere

The chemical form of C-14 compounds entering into the biosphere is essential for determining the metabolism and eventually dose delivered. There was no clear outcome concerning the role of the present project in this context because biosphere processes are outside the scope of CAST.

### 1.1.4 Up-scaling, representativity and relevance

Up-scaling from the lab-system to the real system scale is associated with transfer across scale, but also with the question of representativity of the lab-systems investigated with respect to physico-chemical conditions in the lab and the representativity of the sampling material for the actual overall system. The lack of relevance thus can be the result deficiencies in the investigated systems, including failing to identify important parts of the real system. Finally, the relevance of investigations may also lack identification of processes. Examples could be microbial processes providing for retention by incorporation into secondary phases,



conversion of C-14 compounds to more/less mobile ones, or impact on physico-chemical conditions, including formation of important ligands.

### 1.1.5 Involvement actors

All waste management organisations in the European Union, Switzerland, Japan and Ukraine have been invited to participate in the first CAST workshop. For EU countries without nuclear power plants, there may be less interest to participate in this workshop as the waste investigated in CAST mainly originates from these plants and not research reactors. Based on the expressed interest from these organisations, efforts could have been made to invite regulators from these countries. All regulators in the European Union with (past, future) nuclear power plants, Switzerland, Japan and Ukraine had been invited. Eight budgets were reserved from the EC funding for CAST to reimburse the travel costs and subsistence to attend the workshops if sufficient funding was not available. All eight appeared to be necessary. In some EU countries, the responsibility of the management of the types of waste investigated in CAST is the same organisation that generates the waste. The involvement of waste generators in the first workshop appeared to be difficult despite (several) efforts made by the waste management organisation and some research organisations participating in CAST.

In the dissemination plan (D7.3), the performance indicators concerning the involvement of actors have been set to assess the success of the workshop:

1. Workshop was judged successful if one expert from each national regulatory body of 50% of countries participating in CAST participates in the workshop
  - 15 countries (12 EU countries) participate in the workshop and 8 countries had send a representative of their national regulatory body to the first workshop. Involvement of regulators was successful.
2. Workshop was judged successful if more than 9 waste generators/producers participate in the workshop since 9 waste management organisations participate in CAST
  - 3 waste generators participated; 2 of them are also waste management organisations. Involvement of waste generators / producers was not successful.

## **2 The second CAST Workshop**

Based on observations from the present workshop, and suggestions made during concluding discussion, topics can be brought forward for consideration for the second workshop (cf. also Annex, Presentation No. 23):

- a) Discuss the status concerning the different source materials, an integrated descriptions of C-14 routes, from the source material to the biosphere metabolism, including modelling and comparison with measurements of the nitrogen precursor, as well as quantification, speciation and migration behaviour of C-14 compounds along the transport path.
- b) Consider inviting analytical expertise from other areas in order to identify potential future speciation developments, including relevance and representativity of lab measurements and modelling.
- c) Discuss the transfer of knowledge into the Safety Case, including the scientific-technical basis subject to the concerned R&D, as well as coupling with general topics when implementing scientific-technical knowledge from present types of investigations into the real-scale Safety Case.
- d) Provide training on those aspects where the state-of-knowledge is sufficiently mature.
- e) In view of the way ahead: How mature is the scientific-technical basis and how does it feed into the Safety Case? What are the key remaining needs for knowledge as identified from the Safety Case? How could such key remaining needs be met by joint R&D activities?

As indicated in the invitation for the first workshop, the waste generators are intended to give a contribution to the second workshop. The confidence in the radiological characterisation of the waste highly depends on the information that can be provided by waste generators / producers. The aim is to have a participation of at least 9 waste generators / producers in the second workshop. At the CAST General Assembly Meeting held in October 2016 in Switzerland it has also been decided to held the second workshop in Lithuania.

## **Annex**

The Annex consist of:

1. Attendance 1<sup>st</sup> CAST workshop
2. 1<sup>st</sup> CAST Workshop Agenda
3. Presentations 1 – 22
4. Wrap-Up and Closure

<i>Name</i>	<i>Country</i>	<i>Organisation</i>
Mayia Mateeva	Bulgaria	BNRA
Andrei Turnea	Romania	CNCAN
Olli Nummi	Finland	Fortum
Péter Molnár	Hungary	PURAM
Mindaugas Pranaitis	Lithuania	RATA
Jaap Hart	the Netherlands	NRG
Ewoud Verhoef	the Netherlands	COVRA
Erika Neeft	the Netherlands	COVRA
Penka Avramova	Bulgaria	DPRAO
Simon Norris	United Kingdom	RWM
Colin Campbell	United Kingdom	Environment agency
Sophia Necib	France	ANDRA
Jan Wieman	the Netherlands	EPZ
Jürgen Hansmann	Switzerland	ENSI
Nikitas Diomidis	Switzerland	NAGRA
Manuel Capouet	Belgium	ONDRAF/NIRAS
Pierre De Cannière	Belgium	FANC
Frédéric Bernier	Belgium	FANC
Dalia Grigaliūnienė	Lithuania	LEI
Tomofumi Sakuragi	Japan	RWMC
Sabine Dörr	Germany	DBE Tech
Miguel Cuñado	Spain	ENRESA
Thierry Louis	the Netherlands	ANVS
Stelian Orasanu	Romania	ANDR
Gunnar Buckeau	Germany	JRC
Dmitry Lukin	Czech Republic	SURAO

Attendance regulators:

In orange regulatory organisations, in green support for regulator



Participation waste management organisations,

In orange attendance workshop, in yellow participation limited to proceedings of the workshop



Attendance waste generators / producers



Attendance waste generators / producers









**5 October 2016**

No.	Time	Presentation	Organisation	Presenter
Coffee and tea				
1	10:00-10:15	Introduction of the waste investigated in CAST & Audience of this workshop	COVRA	Erika Neeft
2	10:15-10:25	Dutch contribution		
3	10:25-10:35	Lithuanian contribution	RATA	Mindaugas Pranaitis
4	10:35-10:45	English contribution	RWM	Simon Norris
5	10:45-10:55	German contribution	DBE Tech	Sabine Dörr
6	10:55-11:05	Belgian contribution	ONDRAF/NIRAS	Manuel Capouet
7	11:05-11:15	Hungarian contribution	PURAM	Péter Molnár
8	11:15-11:25	French contribution	ANDRA	Sophia Necib
9	11:25-11:35	Czech contribution	SURAO	Dmitry Lukin
10	11:35-11:45	Romanian contribution	ANDR	Stelian Orasanu
11	11:45-11:55	Japanese contribution	RWMC	Tomofumi Sakuragi
12	11:55-12:05	Bulgarian contribution	DPRAO	Penka Avramova
13	12:05-12:15	Spanish contribution	ENRESA	Miguel Cuñado
Lunch				
	13:00-13:30	3D Movie	COVRA	
	13:30-14:30	Visit storage facility LILW	Group regulators & TSO	Ewoud Verhoef
		Irradiated steel Spent Ion exchange resins	Group WMO & waste generators	Erika Neeft
Coffee and tea				
14 / 15	14:50-16:00	Finnish contribution Example assessment spent ion exchange resins	Fortum	Olli Nummi
16	16:00-16:15	Dutch regulator	ANVS	Thierry Louis
17	16:15-16:30	Dutch waste generator	EPZ	Jan Wieman
18	16:30-16:45	Dutch WMO	COVRA	Ewoud Verhoef



## 6 October

Time		Presentation	Organisation	Presenter
Coffee and tea				
19	10:00-11:00	Lithuanian example assessment irradiated graphite	LEI	Dalia Grigaliuniene
20	11:00-12:00	Dutch example assessment irradiated Zircaloy	NRG	Jaap Hart
Lunch				
	13:00-14:00	Visit storage facility Irradiated Zircaloy	Group regulators & TSO	Ewoud Verhoef
			Group WMO & waste generators	Erika Neeft
Coffee and tea				
21	14:20-15:30	Swiss contribution <hr/> Example assessment irradiated steel	NAGRA	Nikitas Diomidis
22	15:30-16:30	Wrap-up and closure	JRC	Gunnar Buckau



Presentation No. 1

1

# Carbon-14 Source Term CAST

Name: **Erika Neeft**

Organisation: **COVRA**

Date: **5 October 2016**



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project



## Introduction of the waste investigated in CAST & Audience of this workshop

- Audience
- Generation of carbon-14
- Generation of waste in types of reactors
- Determination carbon-14 content
  - Speciation in carbon-14 release from waste
  - Radiation protection
- Audience - outlook

3



## Audience



ENSREG: EU-countries with operational nuclear power plants in orange  
16 EU countries + future waste in Poland, waste from past in Italy  
Also invited: Switzerland, Japan and Ukraine because participation in CAST

4

# Audience



NAGRA: Irradiated steel  
 ANDRA: Irradiated Zircaloy  
 RWM: Irradiated graphite  
 ONDRAF/NIRAS: Safety case relevance  
 COVRA: Dissemination  
*CEA: Spent ion exchange resins*

Agreement with research organisations  
 what type of research is useful

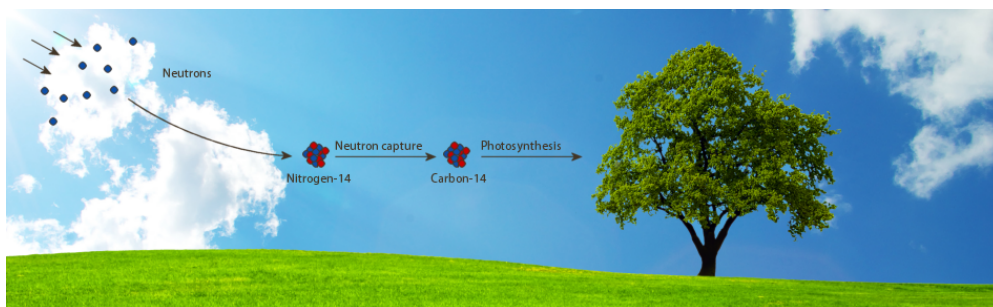
Neeft et al, Deliverable7.3: Dissemination plan; WMO participation in CAST  
 Dissemination is more than publishing documents; aims e.g. prevent 'reinventing the wheel' (limit research load).

what is the background: interest + knowledge

In this workshop: professionals e.g. drafting waste acceptance criteria, characterisation of waste for disposal et cetera

5

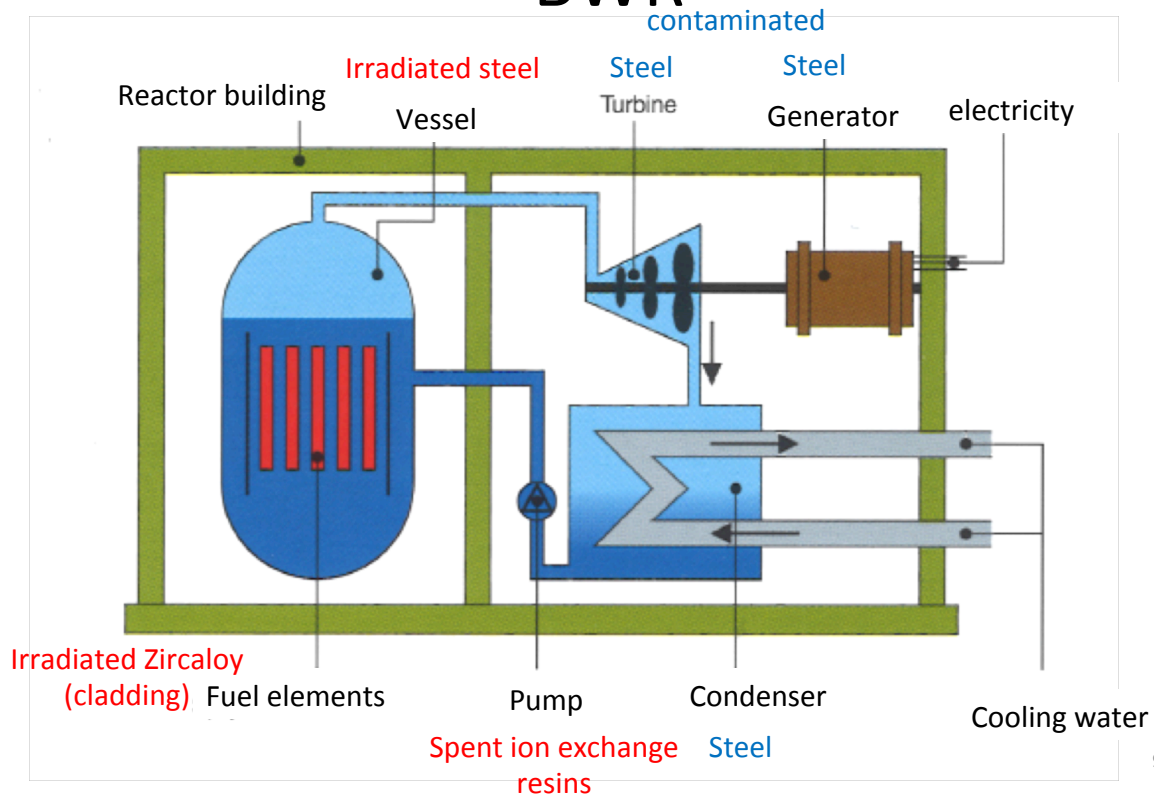
# Generation of carbon-14



6

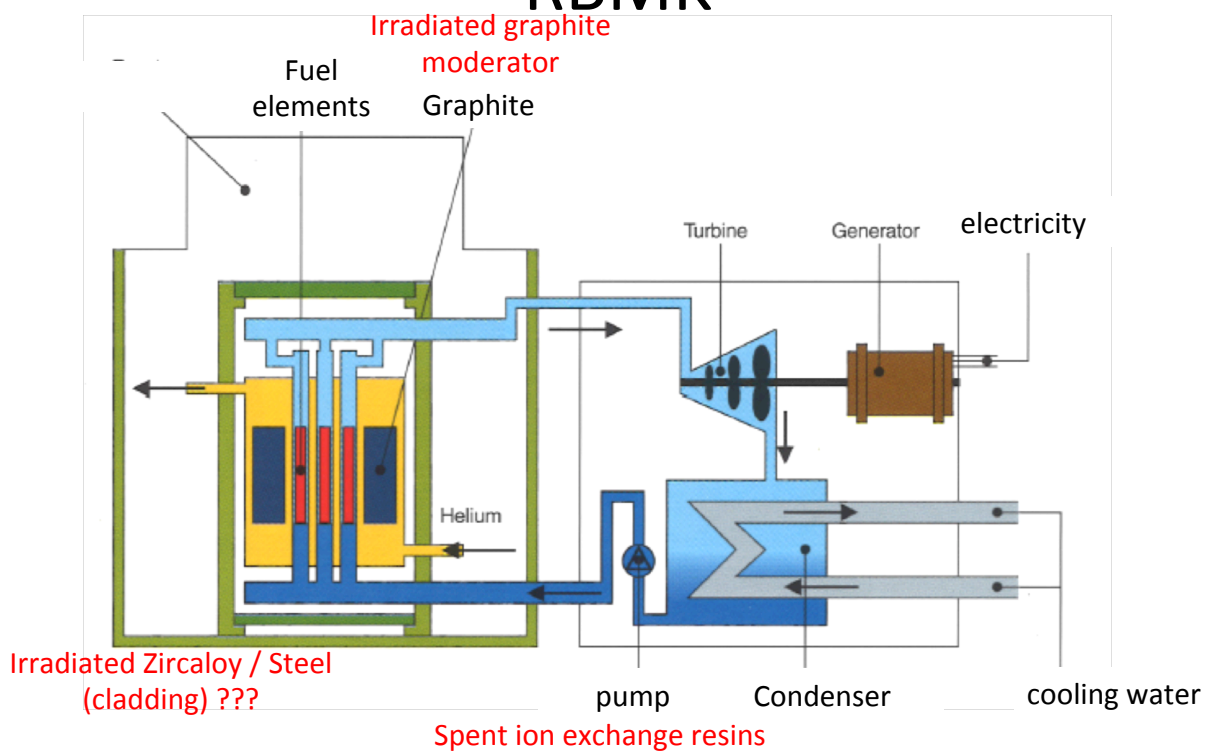


# BWR



9

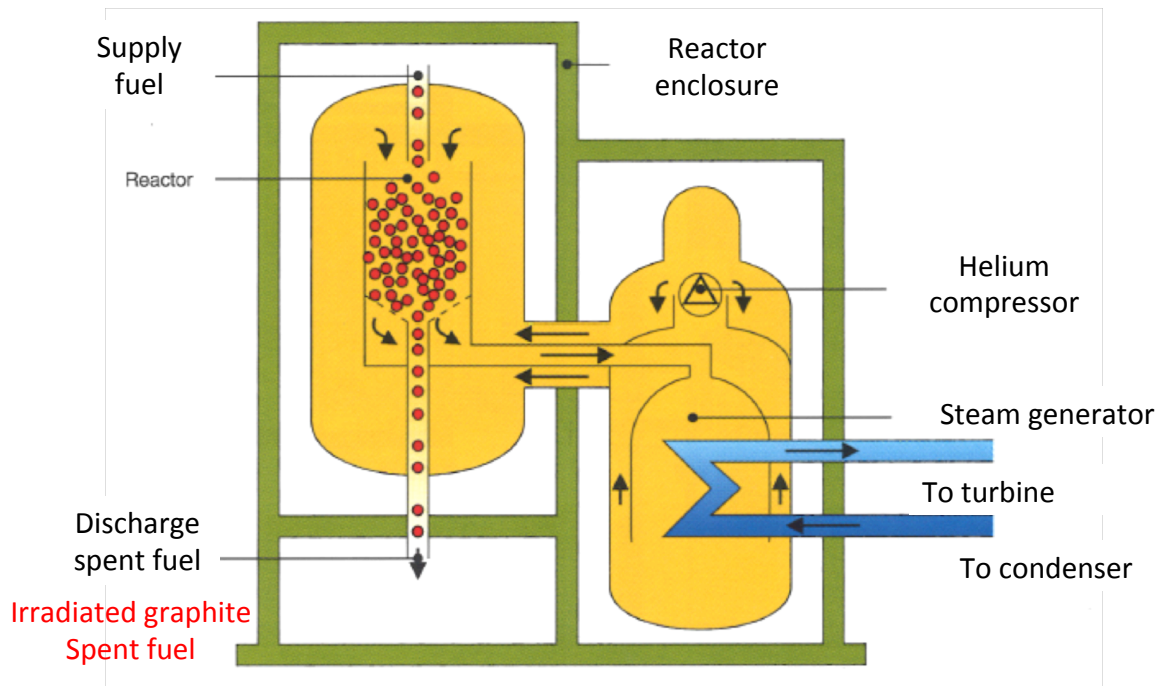
# RBMK



10



# HTR



11

## Determination carbon-14

- DTM radionuclide
  - Destructive analysis
    - Representative samples radwaste
    - Digested in liquid (concentrated acid)
    - Carbon-14 evolving gas needs to be isolated e.g.  $\text{Ba}(\text{OH})_2$
    - Dispersion in scintillating liquid
  - Fission :  $^{135}\text{Cs}$
  - Neutron activation:  $^{36}\text{Cl}$

12

# Determination carbon-14 content

- Neutron activation
  - Chemical details of metals used in reactor (in case of carbon-14 mainly nitrogen impurities)
    - ALARA
      - measured before irradiation
      - (If not possible before irradiation wait as long as acceptable- tour today)
  - Neutron fluence / flux / energy distribution
    - Not closed system, airborne emissions: licence to (annual) discharge carbon-14 similar as tritium
  - Representative sampling
    - ALARA – before waste processing

13

# Speciation carbon-14 release

- $\text{CO}_3^{2-}$  (ionic compound dissolved in ground water)
  - Same or larger migration rate than a carboxylic acid
- $\text{CH}_4$ 
  - As gas dissolved in ground water
  - Gas accumulations?

As dissolved ionic compound smaller migration rate than dissolved as gas ; how much smaller depends on depth, connection between pores, size of pores et cetera

14

# Radiation protection

- Council directive 2013/59/EURATOM
  - Dismantling waste

RN	Activity concentration for exemption clearance [Becquerel per gram solid matter]	Half life [years]	Chemical concentration [ppm per gram iron]	Fission yield $^{235}\text{U}$ thermal [%]	DTM?	Released from waste as
$^{14}\text{C}$	1	5730	0,000024	Not relevant for determination generation	Yes	Gas or ionic?
$^{36}\text{Cl}$	1	301000	0,001269	Same as $^{14}\text{C}$	Yes	Ionic

# Audience outlook

- Sufficient opportunity to ask questions, initiate discussions
  - Regulators
    - WMO outside CAST
      - Waste generators
        - » Research organisations & WMO inside CAST
- WMO contributions
  - 8 minutes
  - 2 minutes questions
- National examples of assessments
  - 45 minutes
  - 15 minutes questions, initiation for discussions

# Audience outlook

- WMO preparations (fast)
  - Snapshots; intend to make selection to optimize your (future) reading load for preventing 'reinventing the wheel' again
    - Characterisation (how Carbon-14 is determined?)
    - (Processed) waste (package) (how carbon-14 is distributed?)
    - If geological disposal : host rock
- Example national assessments (slower)
  - Regulators asked to read *Poskas et al Deliverable 7.7 Overview of achievements on technical results for regulators for Workshop 1* for their preparation
  - WP Leaders in CAST also possible to answer your question

17

# Audience outlook

- Workshop in 2018?
  - WMO
    - 75% EU countries with NPP
      - 87.5% expressed their interest
  - Waste generators
    - Limited to Finland and the Netherlands
      - One of the objectives for the second workshop is not expected to be achieved
  - Regulators
    - 50% EU countries with NPP expressed their interest
    - Possibility special moment in wrap-up and closure inform Gunnar Buckau

18





Presentation No. 2

1

# Carbon-14 Source Term CAST-workshop

Country: **the Netherlands**

Organisation: **COVRA**

Name: **Erika Neeft**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.



# Irradiated Steels



- What are the amounts you expect to have / will be generated?
  - Not (yet) a waste family in the Dutch inventory; aggregated in LILW
    - Carbon-14 not (yet) identified in dismantling waste BWR (60 MW<sub>e</sub>; 28 years) (Siempelkamp); 2045 further dismantling
    - Carbon-14 in PWR (515 MW<sub>e</sub> ; period 60 years); waste not yet owned by COVRA; 2033 dismantling
  - Origin(s) Image(s) of waste (processed) package
    - Most packages Mosaik II-15 EI; not envisaged for disposal
  - How is the carbon-14 content determined / specified?
    - Scaling factors
  - What is the activity of <sup>14</sup>C per waste package / kilogram metal
    - Not investigated in sufficient detail to present an activity
- What is the designated end-point of this waste?
  - If disposed in the Netherlands: deep geological disposal

Previous research programme CORA (1996-2001)  
Dismantling waste (non-heat HLW): 2000 m<sup>3</sup> (COVRA-EIR, 1995)  
In OPERA (2011-2016):  
dismantling waste after storage (100 years; <sup>60</sup>Co decay)



# Irradiated Zircaloy



- What are the amounts you expect to have / will be generated? Only in sufficient detail for carbon-14 available in combination with irradiated Zircaloy –waste (steel a.o.) arising from processing CSD-C: 600 containers: storage volume 117 m<sup>3</sup>
  - Origin(s) irradiated in PWR, processed by AREVA
    - No fuel cladding and fuel grid support from irradiation in BWR; has been exchanged for vitrified waste
  - Image of waste (processed) package (see next slide)
  - How is the carbon-14 content determined / specified?
    - Typical value for 900 MW in technical specification (Dutch PWR 515 MW<sub>e</sub>)
  - What is the activity of <sup>14</sup>C per waste package / kilogram metal? 1.4×10<sup>10</sup> Bq / 528 (393 kg Zr, 19 kg Inconel, 116 kg ss (technological waste) +100 kg = 27 MBq / kilogram waste; 22 MBq / kilogram waste package
- What is the designated end-point of this waste?
  - Deep geological disposal in poorly indurated clay e.g. Boom Clay (for rock salt not yet included in a disposal concept)
    - For cost estimate: each waste container embedded in a 'supercontainer': disposal volume: 4253 m<sup>3</sup>

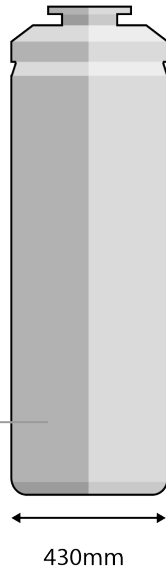


## CSD-c

non-heat generating  
high-level waste

inside diameter 420 mm  
wall thickness 5 mm  
weight loaded: max 850 kg  
empty: 100 kg

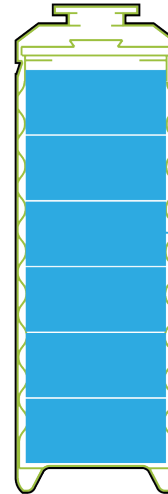
Stainless steel



1335 mm

430mm

cross section



Compacted  
hulls and ends

Typical composition  
393 kg Zircaloy, 19 kg Inconel,  
116 kg waste arising reprocessing



Verhoef et. al, 2016, OPERA-PG-COV023; AREVA, 2001: Specification for compacted waste standard residue from light water reactor



## Spent Ion-exchange resins



- What are the amounts you expect to have / will be generated? **SIER only combined with sludge: 4000 containers : 3927 m<sup>3</sup> (waste family with smallest LILW volume)**
  - Origin(s)
    - PWR, BWR, research reactors
  - Image of waste (processed) package **(see next slide)**
    - PWR, BWR
    - research reactor under investigation
  - How is the carbon-14 content determined / specified?
    - PWR: validated (ANDRA) scaling factor with reported <sup>60</sup>Co activity
      - IAEA:2002 Application of ion exchange processes for the treatment of radioactive waste and management of spent ion exchange series, Technical report series No. 408
    - BWR to be performed
  - What is the activity of <sup>14</sup>C per waste package ~~/ kilogram metal~~
    - **7.2×10<sup>9</sup> Bq / 3000 kg = 2.4 MBq / kilogram waste package**
- What is the designated end-point of this waste?
  - If disposed in the Netherlands: deep geological disposal
    - Storage volume = disposal volume

Verhoef et. al, 2016, OPERA-PG-COV023;



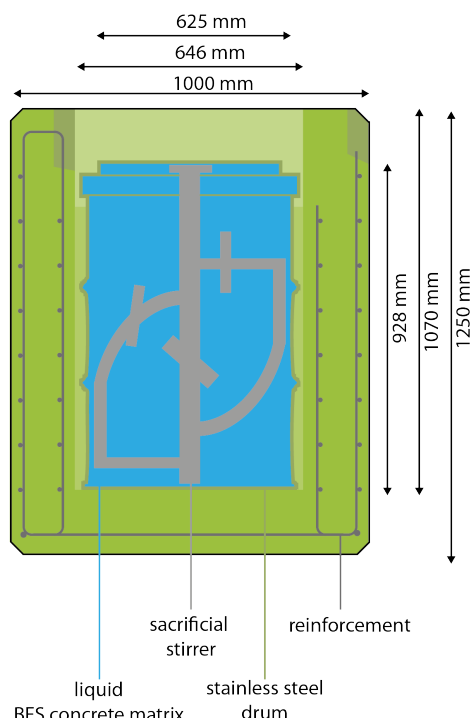


# Sludge & SIER waste container

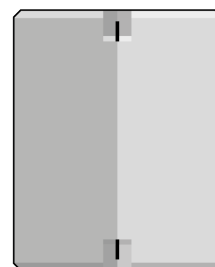
low- and intermediate-level waste



cross section



inside diameter 740 mm  
inside height 940 mm



liquid I: 1,000 l magnetite  
concrete container



## Irradiated Graphite



- What are the amounts you expect to have / will be generated?  
 <<<<<  $54 \times 10^3$  kg (not yet distinguished from other waste categorised as LLW)
- Not (soon) expected to be a separate waste family in the Dutch inventory; aggregated in LILW
  - Origin(s)
    - Research reactor (LFR: type Argonaut; 30 kW<sub>th</sub>)
  - Image of waste (processed) package
  - How is the carbon-14 content determined / specified?
    - Carbon-14 not (yet) identified in Env. Impact Report for decommissioning
  - What is the activity of  $^{14}\text{C}$  per waste package / ~~kilogram metal~~
    - Not (yet) possible
- What is the designated end-point of this waste?
  - If disposed in the Netherlands: deep geological disposal



Presentation No. 3

1

# **CArbon-14 Source Term CAST-workshop**

Country: Lithuania

Organisation: SE Radioactive waste management agency

Name: Mindaugas Pranaitis

This presentation can be used for the Proceedings of the workshop that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**

# Ignalina NPP



## Irradiated Steels



- 3416 tons of irradiated steel alloys in units 1&2 and 949 tons of operational waste



# Irradiated Steels



- 3416 tons of irradiated steel alloys in units 1&2 and 949 tons of operational waste
  - Activated metallic reactor structures



# Irradiated Steels



- 3416 tons of irradiated steel alloys in units 1&2 and 949 tons of operational waste
  - Activated metallic reactor structures
  - Nuclide vectors/scaling factors will be used for activity estimation



# Irradiated Steels



- 3416 tons of irradiated steel alloys in units 1&2 and 949 tons of operational waste
  - Activated metallic reactor structures
  - Nuclide vectors/scaling factors will be used for activity estimation
  - Equipment radiological characterization is not finished yet – all waste are preliminary classified as class E wastes



# Irradiated Steels



- 3416 tons of irradiated steel alloys in units 1&2 and 949 tons of operational waste
  - Activated metallic reactor structures
  - Nuclide vectors/scaling factors will be used for activity estimation
  - Equipment radiological characterization is not finished yet – all waste are preliminary classified as class E wastes
  - Most probable endpoint (class E wastes) – **Deep geological repository**



## Irradiated Zirconium alloys



- Inventory consists of 214 tons of Zr (Nb 2,5%) alloys



## Irradiated Zirconium alloys



- Inventory consists of 214 tons of Zr (Nb 2,5%) alloys
  - Pressure tubes from units 1 & 2



# Irradiated Zirconium alloys



- Inventory consists of 214 tons of Zr (Nb 2,5%) alloys
  - Pressure tubes from units 1 & 2
  - Scaling factors not determined yet



# Irradiated Zirconium alloys



- Inventory consists of 214 tons of Zr (Nb 2,5%) alloys
  - Pressure tubes from units 1 & 2
  - Scaling factors not determined yet
  - Conservative preliminary classification as class E wastes



## Irradiated Zirconium alloys



- Inventory consists of 214 tons of Zr (Nb 2,5%) alloys
  - Pressure tubes from units 1 & 2
  - Scaling factors not determined yet
  - Conservative preliminary classification as class E wastes
  - **Deep geological repository** is considered as an endpoint



## Spent Ion-exchange resins



- Stored spent ion-exchange resins:
  - 500 m<sup>3</sup> of spent ion-exchange resins and perlite mixture stored in tank TW18B01





## Spent Ion-exchange resins



- Stored spent ion-exchange resins:
  - 500 m<sup>3</sup> of spent ion-exchange resins and perlite mixture stored in tank TW18B01
  - 1250 m<sup>3</sup> of spent ion-exchange resins stored in tank TW11B03



## Spent Ion-exchange resins



- Stored spent ion-exchange resins:
  - 500 m<sup>3</sup> of spent ion-exchange resins and perlite mixture stored in tank TW18B01
  - 1250 m<sup>3</sup> of spent ion-exchange resins stored in tank TW11B03





## Spent Ion-exchange resins



- Stored spent ion-exchange resins:
  - 500 m<sup>3</sup> of spent ion-exchange resins and perlite mixture stored in tank TW18B01
  - 1250 m<sup>3</sup> of spent ion-exchange resins stored in tank TW11B03
  - Activity of C-14 determined using scaling factors



## Spent Ion-exchange resins



- Stored spent ion-exchange resins:
  - 500 m<sup>3</sup> of spent ion-exchange resins and perlite mixture stored in tank TW18B01
  - 1250 m<sup>3</sup> of spent ion-exchange resins stored in tank TW11B03
  - Activity of C-14 determined using scaling factors
  - Endpoint – **Near surface repository**



## Spent Ion-exchange resins



- Spent ion-exchange resins resulting from decommissioning
  - 250 m<sup>3</sup> to be cemented and placed to **NSR**



## Spent Ion-exchange resins



- Spent ion-exchange resins resulting from decommissioning
  - 250 m<sup>3</sup> to be cemented and placed to **NSR**
  - 720 m<sup>3</sup> from condensate purification facilities – regenerated, dried and placed to FIBC for radiological characterization –



## Spent Ion-exchange resins



- Spent ion-exchange resins resulting from decommissioning
  - 250 m<sup>3</sup> to be cemented and placed to **NSR**
  - 720 m<sup>3</sup> from condensate purification facilities – regenerated, dried and placed to FIBC for radiological characterization – meets WAC for **LANDFILL** –



## Spent Ion-exchange resins



- Spent ion-exchange resins resulting from decommissioning
  - 250 m<sup>3</sup> to be cemented and placed to **NSR**
  - 720 m<sup>3</sup> from condensate purification facilities – regenerated, dried and placed to FIBC for radiological characterization – meets WAC for **LANDFILL** – placed in ISO or ½ height ISO after radiological characterization



# Irradiated Graphite



- 3843 tons of irradiated graphite, of which:
  - 55 tons of operational waste (graphite sleeves)
  - and 3788 tons of decommissioning waste;



# Irradiated Graphite



- 3843 tons of irradiated graphite, of which:
  - 55 tons of operational waste (graphite sleeves)
  - and 3788 tons of decommissioning waste;





# Irradiated Graphite



- 3843 tons of irradiated graphite, of which:
  - 55 tons of operational waste (graphite sleeves) and 3788 tons of decommissioning waste;
  - GR-280 grade graphite columns and bushings and GR-2-125 grade graphite rings and sleeves (~200 t)



# Irradiated Graphite



- 3843 tons of irradiated graphite:
  - In 7<sup>th</sup> FDP only interim storage of graphite is foreseen, without immobilisation
  - 7<sup>th</sup> FDP foresees placement of graphite sleeves into 200 l drums which would then be placed into containers appropriate for interim storage





# Irradiated Graphite



- 3843 tons of irradiated graphite:
  - In 7<sup>th</sup> FDP only interim storage of graphite is foreseen, without immobilisation
  - 7<sup>th</sup> FDP foresees placement of graphite sleeves into 200 l drums which would then be placed into containers appropriate for interim storage
  - According to 7<sup>th</sup> FDP, graphite columns are to be placed in FIBC and then placed in appropriate containers for interim storage



# Irradiated Graphite



- Radionuclide inventory:
  - Scalling factor has been determined for operational graphite waste
  - Activity of C-14 is in the order of at least  $10^4$  Bq/g in used graphite rings



# Irradiated Graphite



- Possible endpoints:
  - WAC for NSR is not met due to high estimated C-14 content, so NSR is not an option for untreated graphite.



# Irradiated Graphite



- Possible endpoints:
  - WAC for NSR is not met due to high estimated C-14 content, so NSR is not an option for untreated graphite.
  - Most probable endpoint for irradiated graphite – **Deep geological repository**



Thank you for attention!





Presentation No. 4

1

# Carbon-14 Source Term CAST-workshop

Country: **United Kingdom**

Organisation: **Radioactive Waste Management**

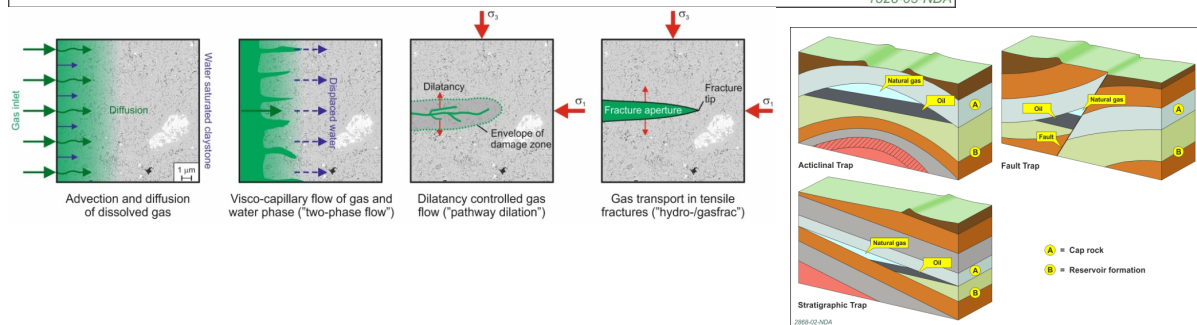
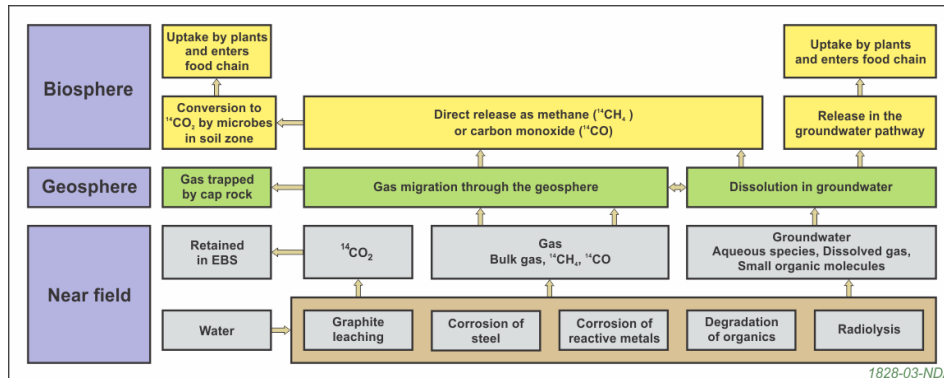
Name: **Simon Norris**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website

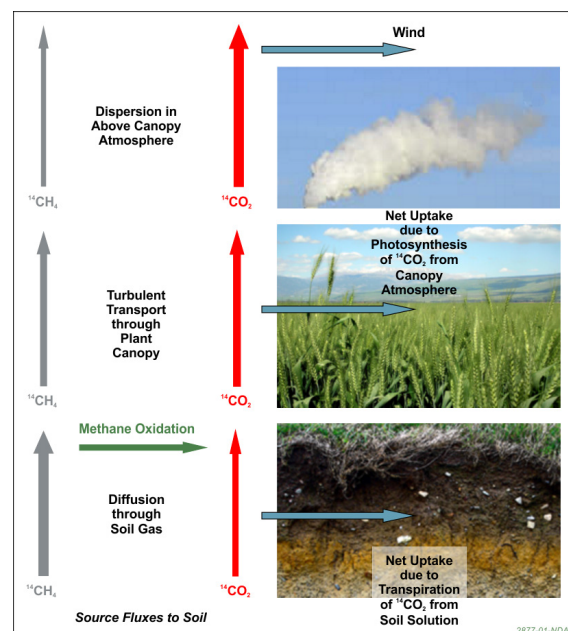
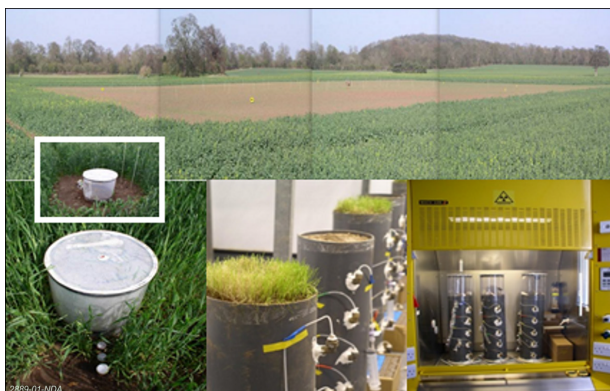


The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.

# Example 'deeper dive' – waste-derived gas



# Example 'deeper dive' - biosphere





# UK C14 Inventory



- RWM 2013 Derived Inventory (DI) for the geological disposal facility (GDF)
  - Based on the 2013 UK Radioactive Waste Inventory (RWI)
  - The DI differs from the 2013 UK RWI by removing wastes not destined for geological disposal and adding wastes to reflect industry best estimates and UK Government policy
  - Leads to an updated carbon-14 inventory for disposal to the UK GDF
- Large number of UK waste streams containing C-14
- For RWM work on C-14 these have been grouped into four categories
  - Graphite
    - in scope of CAST
  - Steels
    - in scope of CAST
  - Reactive metals (uranium, Magnox and aluminium)
    - outside scope of CAST
  - Other wastes (includes Zircaloy, spent ion exchange resins, spent fuel)
    - Zircaloy, SIER in scope of CAST

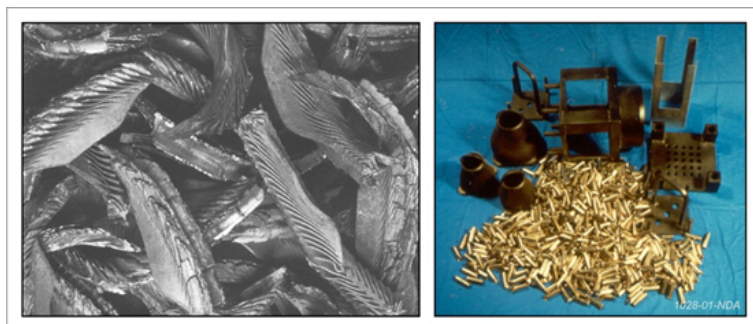


## RWM 2013 DI



- 17,700 TBq of carbon-14
  - Graphite – 6,929 TBq
  - Steels - 7100 TBq
  - Reactive metals - 115 TBq
  - Other wastes - 3579 TBq
    - ILW organics from GE Healthcare (all may not be disposed to the GDF) – 204 TBq
    - Other ILW (includes Zircaloy and SIERS) – 77.1 TBq
    - Spent fuels – 3290 TBq
    - Miscellaneous – 8.2 TBq

# C14 sources



Examples of ILW. (a) The cladding of Magnox fuel elements after size reduction (Magnox swarf); (b) elements of the cladding of PWR fuel elements after size reduction (hulls and ends)



Illustration showing an example of a Magnox fuel element (cladding). The element is typically 1 metre in length

# Irradiated Steels



- 7100 TBq C-14
  - 40% of UK C-14 derived inventory by activity
  - majority is from assumed 16GW(e) new nuclear build (6660 TBq C-14)
  - $\approx 49,000\text{m}^3$  ILW when packaged for disposal
- Seven categories of steel wastes:
  - ILW AGR stainless steel fuel cladding - 29.4 TBq (0.17%)
  - ILW AGR stainless steel fuel assembly components - 38.3 TBq (0.22%)
  - Fuel stringer debris – 99.5 TBq (0.56%)
  - ILW stainless steels from reactor decommissioning - 6770 TBq (38.2%)
  - ILW stainless steel reactor wastes – 7.78 TBq (0.044%)
  - ILW other ferrous metal decommissioning wastes – 126 TBq (0.71%)
  - ILW other ferrous metal reactor wastes – 29.4 TBq (0.17%)
- C-14 inventory from neutron activation calculations
- End point is disposal of packages of conditioned waste to the GDF

Note - % with respect to total UK C14 inventory of 17,700TBq



## Irradiated Zircaloy



- 28 TBq C-14
  - 0.16% of UK C-14 derived inventory by activity
  - $\approx 2,000\text{m}^3$  ILW when packaged for disposal
- Part of 'other wastes'
  - Encapsulated LWR cladding -28.0 TBq (0.16%)
  - FED Zirconium ( $<0.01\text{TBq}$ )
- C-14 inventory from neutron activation calculations
- End point is disposal of packages of conditioned waste to a GDF

Note - % with respect to total UK C14 inventory of 17,700TBq



## Spent Ion-exchange resins



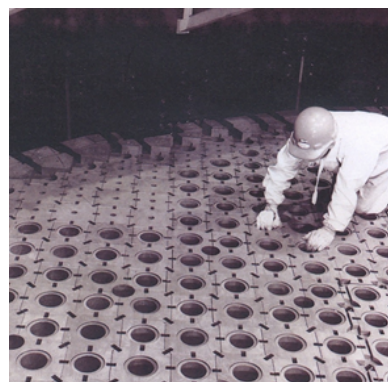
- 9.9TBq C-14
  - 0.06% of UK C-14 derived inventory by activity
  - $\approx 25,000\text{m}^3$  ILW when packaged for disposal
- Part of 'other wastes' in derived inventory
  - CVCS resins and spent resins (ILW) – 2.77TBq (0.016%)
  - ILW submarine IEX resin – 2.22 TBq (0.013%)
  - UILW resins (new build) – 4.93 TBq (0.028%)
- C-14 inventory from theoretical estimates
- End point is disposal of packages of conditioned waste to a GDF

Note - % with respect to total UK C14 inventory of 17,700TBq





# Irradiated Graphite



Oldbury Magnox reactors and core graphite blocks during construction (Photos copyright NDA and Magnox Limited ([www.magnoxsites.com](http://www.magnoxsites.com)))



# Irradiated Graphite

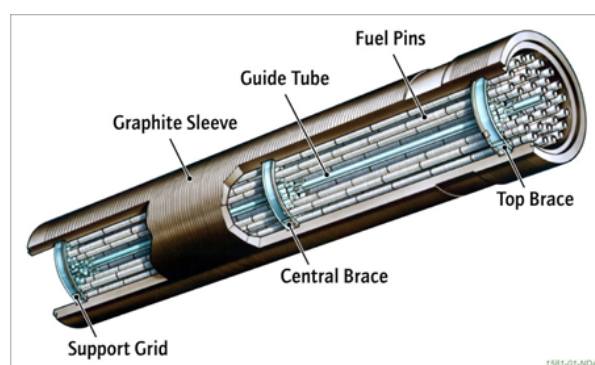


Illustration showing an AGR fuel assembly, together with its graphite sleeve. Note that disposal of spent AGR fuel assumes removal of the graphite before containerisation in disposal containers, consistent with current storage practices at Sellafield. The length of the assembly is about 1 metre



# Irradiated Graphite



- 6929 TBq C-14
  - 39% of UK C-14 derived inventory by activity
  - $\approx 95,000\text{m}^3$  ILW when packaged for disposal
- Four categories of graphite waste:
  - ILW core graphite – 6880 TBq (38.8%)
  - ILW AGR fuel assembly graphite – 45.1 TBq (0.25%)
  - ILW Magnox fuel element graphite – 1.75 TBq (0.01%)
  - LLW core graphite – 2.13 TBq (0.012%)
- C-14 inventory from neutron activation calculations
- End point is disposal of conditioned waste to a GDF

Note - % with respect to total UK C14 inventory of 17,700TBq



## Additional Information



- For more information please see:
  - A.C. Adeogun, Carbon-14 Project Phase 2: Inventory, Amec Foster Wheeler Report AMEC/200047/003 Issue 1 (Pöyry Report /390936/Phase 2 Issue 1), 2016
  - Downloadable from <https://rwm.nda.gov.uk/publication/carbon-14-project-phase-2-inventory/>
- Thank you for your attention







Presentation No. 5

1

# **CArbon-14 Source Term CAST-workshop**

GERMANY

DBE TECHNOLOGY GmbH

Sabine Dörr

This presentation can be used for the Proceedings of the workshop that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**



# Irradiated Steels



- What are the amounts you expect to have / will be generated?
  - Origin: PWR, BWR, VVER
  - Amounts and C-14 activities not known today
- What is the designated end-point of this waste?
  - Konrad repository



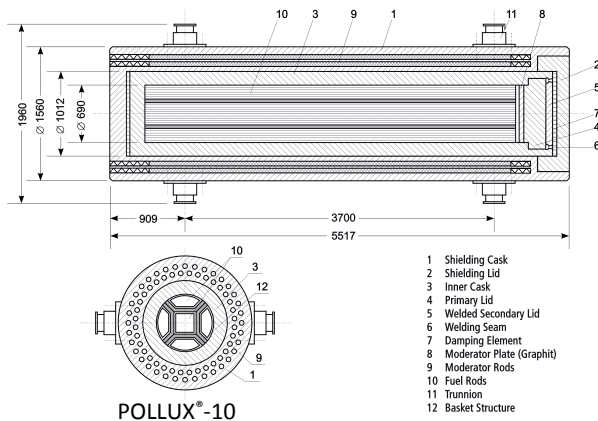
# Irradiated Zircaloy



- What are the amounts you expect to have / will be generated?
  - Origin: Spent nuclear fuel from NPPs, waste from reprocessing of spent nuclear fuel from NPPs
  - C-14 content is determined with OREST calculations (Preliminary Safety Analysis for the Gorleben Site - VSG)

Waste type	Amount		C-14 activity	
	pieces	[tHM)		Total [GBq]
SNF (PWR) UO <sub>2</sub>	12,450	6,415	37.3 GBq/tHM	239,280
SNF (PWR) MOX	1,530	765	23.1 GBq/tHM	17,672
SNF (BWR) UO <sub>2</sub>	14,350	2,465	37.7 GBq/tHM	92,931
SNF (BWR) MOX	1,250	220	21.9 GBq/tHM	4,818
SNF (VVER) UO <sub>2</sub>	5,050	580	10.2 GBq/tHM	5,916
CSD-V	3,729	-	17.9 GBq/CSD-V	66,749
CSD-B	140	-	unkown	unknown
CSD-C	4,104	-	13.8 GBq/CSD-V	56,635

- What is the designated end-point of this waste?
  - HLW repository
  - Typical waste packages for drift disposal is POLLUX®-10
  - Typical waste package for borehole disposal is BSK-3



BSK-3

- What are the amounts you expect to have / will be generated?
  - Origin: PWR, BWR, VVER
  - PWR: ca. 4 - 7 m<sup>3</sup>/a and unit (IAEA TRS408)
  - BWR: ca. 20 m<sup>3</sup>/a and unit (IAEA TRS 408)
  - VVER: ca. 34 m<sup>3</sup>/a and unit (NUKEM)
  - C-14 activity not known today
- What is the designated end-point of this waste?
  - Konrad repository



# Irradiated Graphite



- What are the amounts you expect to have / will be generated?
  - Origin: reflector and/or moderator material in prototype and research reactors
  - Amount and activity is (R&D project CARBONFOREST):

Reactor	Amount [Mg]	C-14 activity	C-14 activity
AKR-2	1.3	unknown	-
AVR	364 <sup>1</sup>	0.53 TBq	1.46 GBq/Mg
AVR	65 <sup>2</sup> / 158 <sup>3</sup>	4.6 TBq / 280 TBq	70.7 GBq/Mg
FMRB	1.5 <sup>4</sup>	1.6 GBq	1.07 GBq/Mg
FRF	7.7	3.0 GBq	0.39 GBq/Mg
FRG-1/-2	11.11	unknown	-
FRJ-1	12.914	unknown	-
FRJ-2	30	unknown	-
FRM	0.25	unknown	-
FRN	27.7	unknown	-
RFR/RRR	0.30	89 MBq	296.67 MBq/Mg
	0.10	920 MBq	9.2 GBq/Mg
THTR-300	551.12	5.0 TBq	9.07 GBq/Mg
	63.02 <sup>5</sup>	0.58 TBq	9.20 GBq/Mg
Others	13	unknown	-

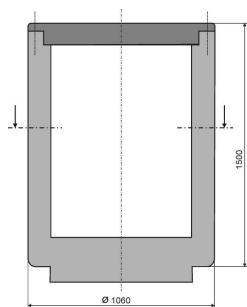
1: Graphite dust and fuel bound in light-weight concrete;  
2: Graphite; 3: Carbon brick;  
4: Packed in one 400-l drum and five 200-l drums;  
5: Graphite and absorber elements



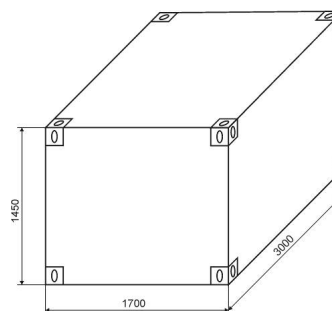
# Irradiated Graphite



- What is the designated end-point of this waste?
  - Konrad repository
  - HLW repository for graphite and carbon brick from AVR
  - Typical waste packages for the Konrad repository are:



Cast iron package Type II



Konrad Container Type IV



Presentation No. 6

1

# CArbon-14 Source Term CAST-workshop

Belgium  
Ondraf/Niras

**VANDOORNE T., MEERT K., BOULANGER D. and CAPOUET M.**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.



# Irradiated Steels



- Stainless steel from UOx& MOx spent fuel (max 55 GWd/tHM – PWR)
  - ~ 11000 assemblies ; Stainless steels : 15-20 kg/ass.
  - A\_C14 = 300 MBq/kg
  - Based on conservative assumption of N=1000 ppm (MCBEND)
  - 12% of total tHM reprocessed => CSD-C
  - Geological disposal (HLW + ILW)

3



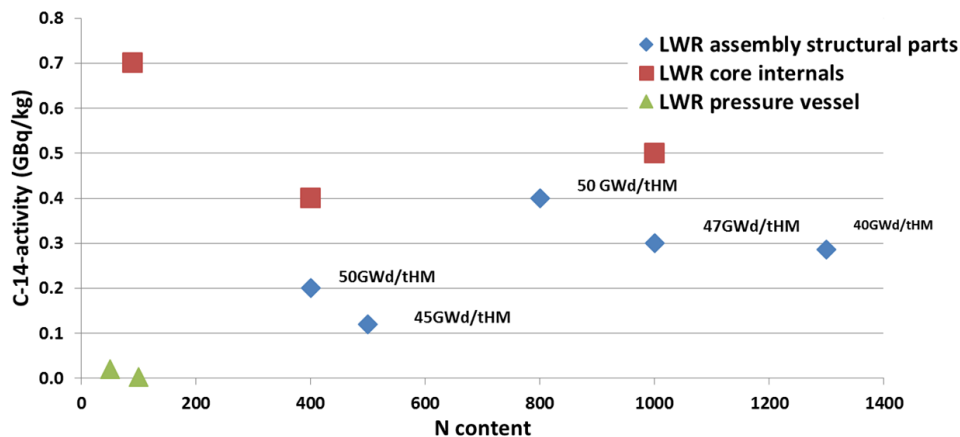
# Irradiated Steels



- Core internals
  - 500 - 600 tons
  - Conditioning: Not decided
  - Geological disposal
  - A\_C14 < 500 MBq/Kg
  - Calculated

4

# Current work in WP6 (D6.2 report)



18

Figure 9:  $^{14}\text{C}$  activity in the steel from different components of light water reactors (LWR).

- $^{14}\text{C}$  activity in the steel from different components of light water reactors (LWR) as provided by different WP6 contributors
- The correlation of  $^{14}\text{C}$  with the nitrogen content is very well reproduced for the structural parts. No clear correlation for core internals and pressure vessels, most likely due to the employed calculation methodology/assumptions.
- **Measurements ?**

5



## Irradiated Zircaloy

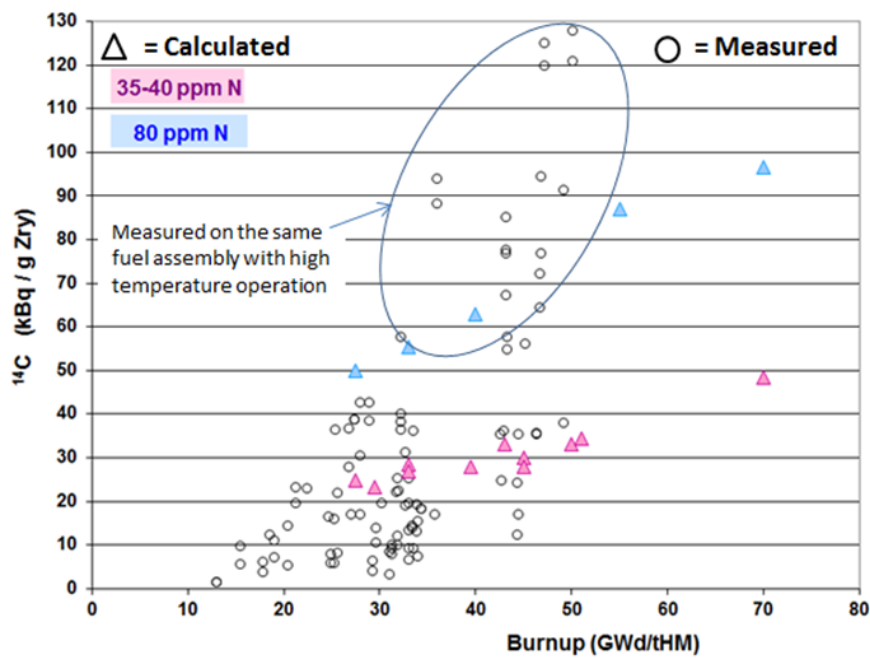


- From UOx spent fuel (max 55 GWd/tHM – PWR)
  - ~ 11000 assemblies
  - Mainly Zr-4, Zirlo and M5
  - ~ 70-150 kg/ass.
  - $A_{\text{C14}} = 80 \text{ MBq/kg}$
  - Based on conservative assumption of  $N=80 \text{ ppm}$  (Spec.)
  - 12% of total tHM reprocessed => CSD-C

6



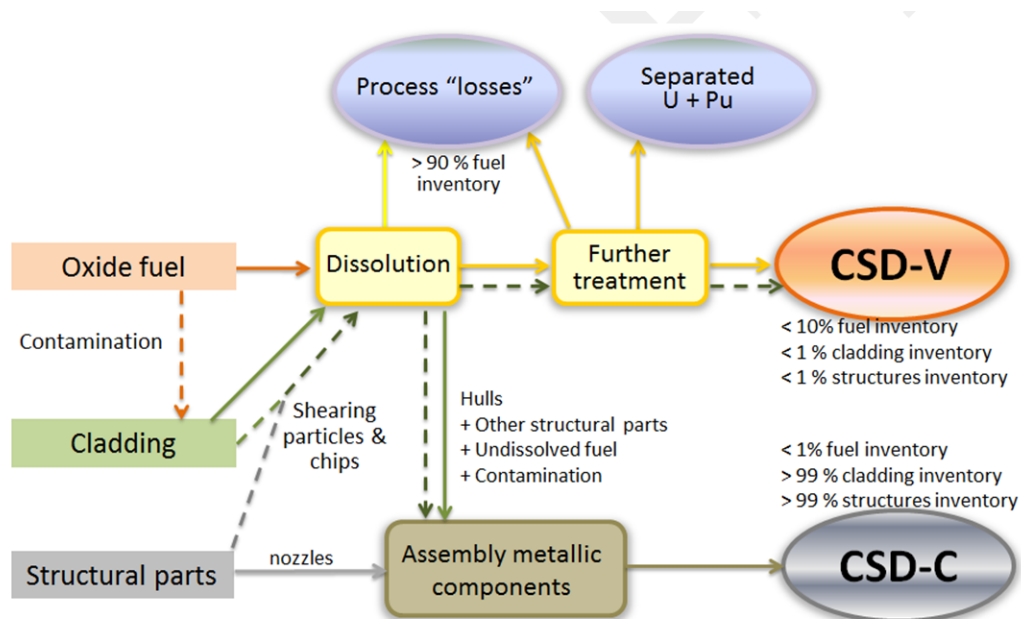
## Current work in WP6 (D6.2 report)



- Based on literature and CAST contributors
- the trendline for the measured data is representative of an initial nitrogen content of 20-30 ppm (as based on the calculations)

7

## Current work in WP6 (D6.2 report)



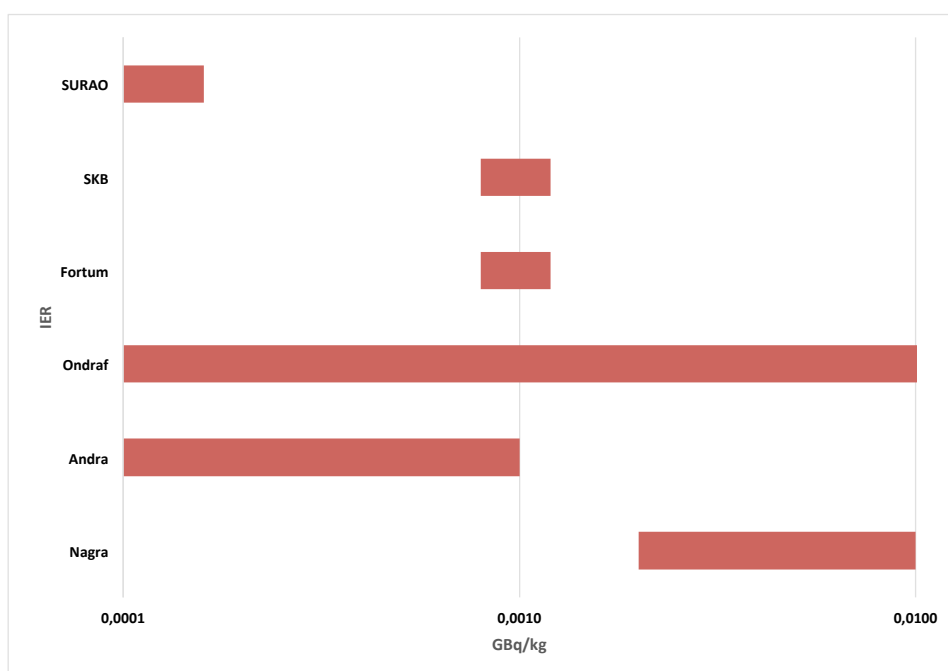
**Figure 8: Waste processing streams. Solid and dash arrows refer to different hypotheses regarding the carry over fraction of  $^{14}\text{C}$  upon spent fuel assemblies reprocessing.**

8

- Average for the different SIER-families range from 0.1 to 25 MBq/kg resin (primary + auxiliary circuits)
- Conditioning: Mostly cementation (previously also polymerization)
- Essentially surface disposal
- Calculated correlation factors

9

## Current work in WP6 (D6.2)



Range of activity depends on several specific factors such as reactor type, resin type as well as storage, handling and conditioning of the resin

10





Presentation No. 7

1

# Carbon-14 Source Term CAST-workshop

Country: **Hungary**

Organisation: **PURAM**

Name: **Péter Molnár**

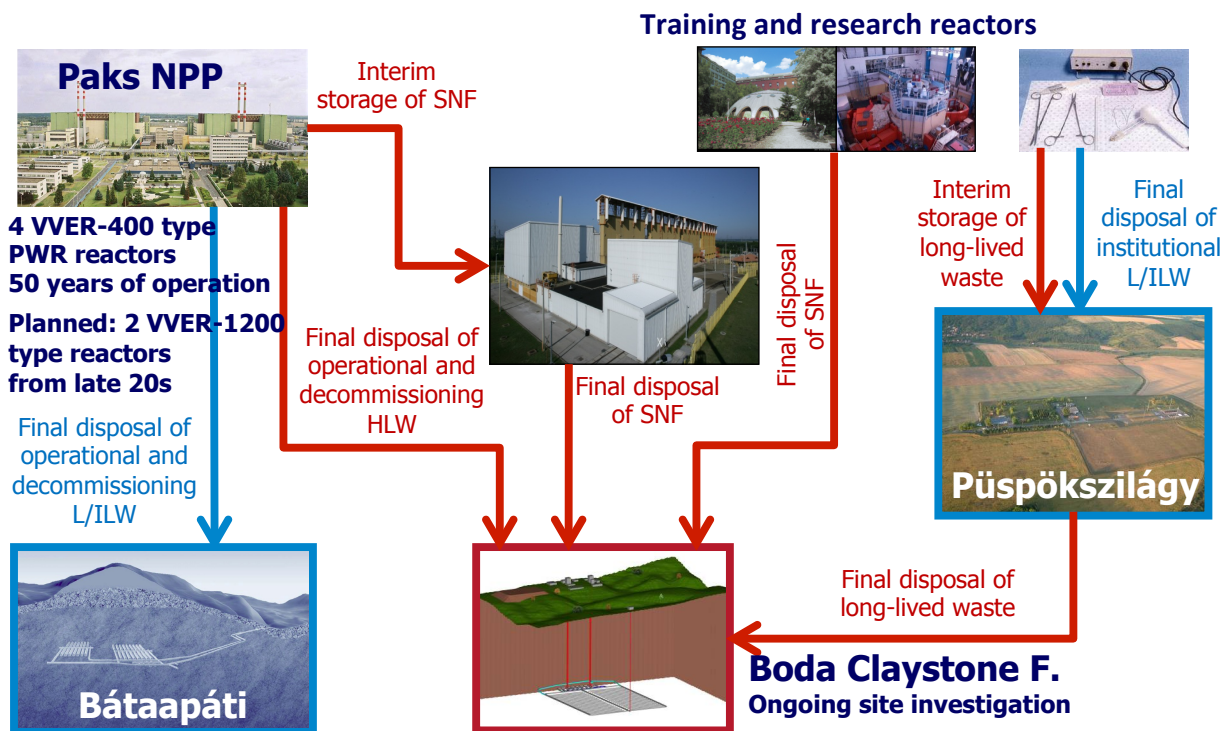
This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.



# Management of RW & SF in Hungary



## Irradiated Steels



- What are the amounts you expect to have / will be generated?  
ca. 1000 t of metal from decommissioning
  - Origin: NPP reactor vessels, stainless steel elements inside vessel and steel reinforcement in concrete around vessel, elements of the primary system
  - Image of waste package: 1.8m<sup>3</sup> steel containers 1.35×1.35×1.05m, max. 6000kg metal in max. 25kg pieces, cemented
  - How is the carbon-14 content determined / specified? Supplier's data service and a few samples for N impurities (0.05% in stainless steel), activation modelling (MCNPX and in-house codes), comparison with international literature data
  - What is the activity of <sup>14</sup>C? typically 1E+08 Bq/kg metal, max. calculated 2E+09 Bq/kg (58% VLLW+LLW, 38% ILW, 4% HLW – the main source of activity is <sup>60</sup>Co)
- What is the designated end-point of this waste?  
Bátaapáti repository for LILW, the planned DGR for HLW



# Irradiated Zircaloy



- What are the amounts you expect to have / will be generated?  
Ca. 17,700 fuel assemblies
  - Origin: ENU fuel structure elements
  - Image of waste (processed) package: reference scenario is direct disposal of SF (KBS-3V concept, copper canisters, 12 assemblies per canister)
  - How is the carbon-14 content determined / specified?  
Supplier's data service (25 ppm  $^{14}\text{N}$  impurity in Zircaloy), activation modelling, international literature
  - What is the activity of  $^{14}\text{C}$ ? Typically  $5\text{E}+05$  Bq/assembly, 35% of it in Zircaloy,  $6\text{E}+06$  Bq/canister (total 1500 canisters)
- What is the designated end-point of this waste?  
Planned DGR for the HLW (reprocessing is still an option)



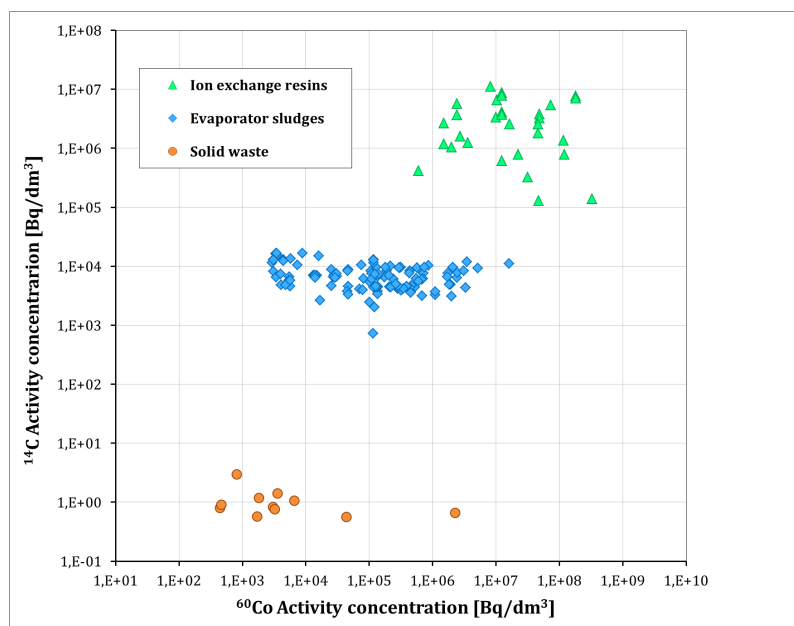
# Spent Ion-exchange resins



- What are the amounts you expect to have / will be generated?  
 $454\text{m}^3$  resin  $\rightarrow$   $1393\text{m}^3$  cemented resin in 757 containers  
 $4250\text{m}^3$  other liquid waste (sludges)  $\rightarrow$   $7225\text{m}^3$  cemented waste as disposed
  - Origin: primary circuit purification system (hydrazine  $\text{N}_2\text{H}_4$  added to the coolant) – resins, and also evaporator sludges from liquid waste
  - Image of waste (processed) package:  $1.8\text{m}^3$  steel containers  $1.35 \times 1.35 \times 1.05\text{m}$  for the cemented waste
  - How is the carbon-14 content determined / specified? Systematic sampling for laboratory analyses (**scaling factor is not applicable!**)
  - What is the activity of  $^{14}\text{C}$ ? Typically  $2\text{E}+06$  Bq/ $\text{dm}^3$  for resins and  $6\text{E}+03$  Bq/ $\text{dm}^3$  for sludges as is,  $7\text{E}+05$  Bq/ $\text{dm}^3$  for cemented resins and  $2\text{E}+04$  Bq/ $\text{dm}^3$  for conditioned and cemented sludges as disposed,  $1\text{E}+09$  Bq/container for cemented resins
- What is the designated end-point of this waste?  
Bátaapáti repository for LILW



# $^{14}\text{C}$ and $^{60}\text{Co}$ activity concentration in different RW



Scaling factor is not applicable!



## Irradiated Graphite



- What are the amounts you expect to have / will be generated?  
only 1.5m<sup>3</sup> waste, 1 container
  - Origin: Training reactor, graphite blocks as reflectors around the reactor and irradiation tunnel
  - Image of waste (processed) package: 1.8m<sup>3</sup> steel containers
  - How is the carbon-14 content determined / specified?  
Activation modelling (MCNPX code)
  - What is the activity of  $^{14}\text{C}$  per waste package?  
9E+04 Bq total activity
- What is the designated end-point of this waste?  
Bátaapáti repository for LILW



Presentation No. 8

1

# CArbon-14 Source Term CAST-workshop

Country: **France**

Organisation: **Andra**

Name: **Sophia Necib, Stéphan Schumacher**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**





# ANDRA

French National Agency for Radioactive Waste



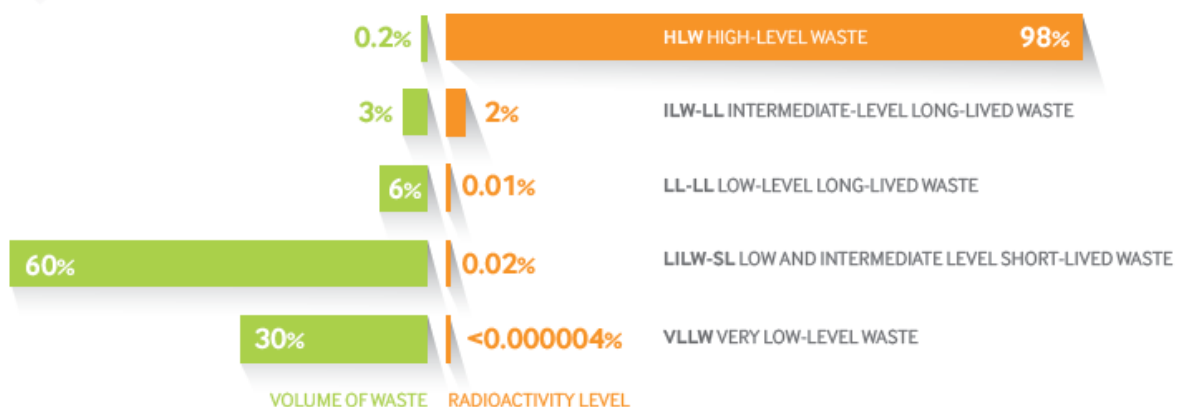
- Andra, the French National Agency for Radioactive Waste Management, was created in 1979 within the French Atomic Energy Commission (CEA)
- Andra was made independent in 1991 by the French law on radwaste management (Law No. 91.1381 of 30 December 1991)
- **Andra's general mission:** To Find, implement and guarantee safe solutions of management for all the French radioactive waste to protect the present and future generations of the risk which this waste present
- Placed under the authority of the ministries of research, ecology, sustainable development and energy
- Independent from waste generators
- Employees ~ 650



<http://www.andra.fr/>

## The five categories of waste

### Breakdown of volume and radioactivity level of radioactive waste





# Irradiated Steels



- What are the amounts you expect to have / will be generated? :
  - Origin(s) : *LWRs (BWRs +PWRs): activated waste*
  - How is the carbon-14 content determined / specified?  
*Modelling (performed by EDF)*
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal?  
 *$\sim 10^{12}$  Bq / waste package considered in the 2016 Cigeo optional safety report (value subject to revision)*
- What is the designated end-point of this waste?  
*Cigéo (deep geological disposal)*



*processed waste package*



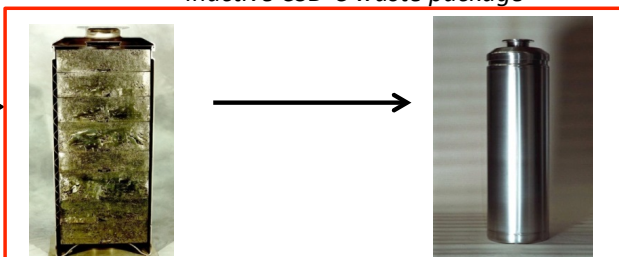
# Irradiated Zircaloy



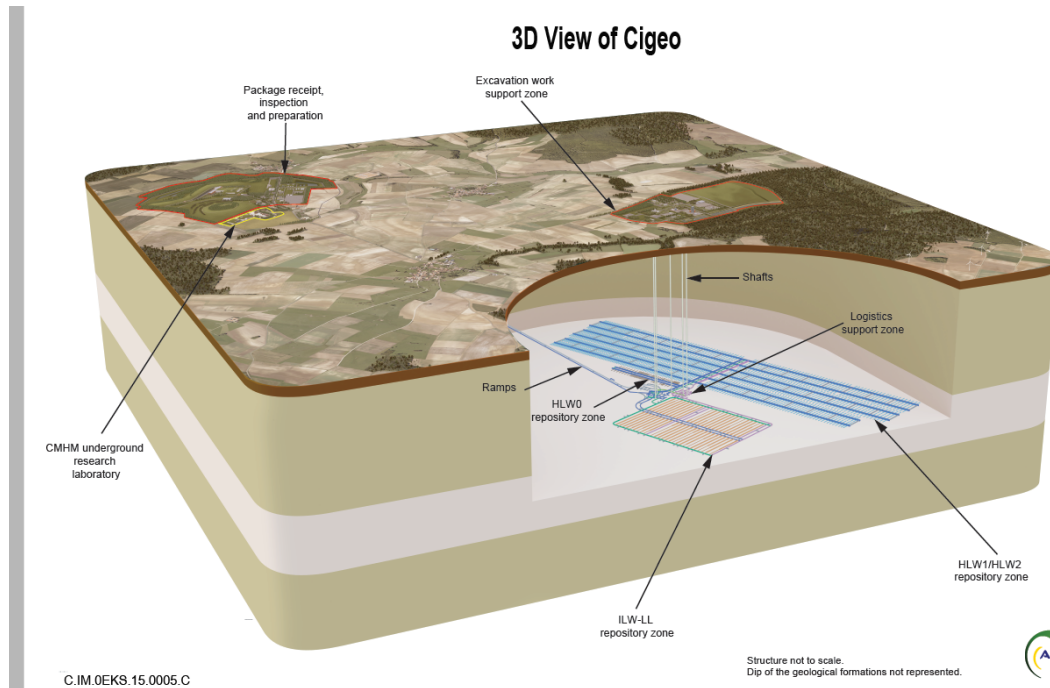
- What are the amounts you expect to have / will be generated?  *$\sim 55,000$  waste packages*
  - Origin(s) *PWRs*
  - Image of waste (processed) package:

*Inactive CSD-C waste package*

*Hulls and end caps*



- How is the carbon-14 content determined / specified?  
*Modelling (CESAR)*
- What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal :  
 *$\sim 20$  GBq/waste package*
- What is the designated end-point of this waste? *Cigéo (deep geological disposal)*



## Spent Ion-exchange resins

- What are the amounts you expect to have / will be generated? *~17,000 waste packages*
- Origin(s) *from the PWR coolant*
  - How is the carbon-14 content determined / specified?  
*By using the scaling factor method based on  $^{60}\text{Co}$  measurement*
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal:  
*Between  $10^3$  and  $10^4$  Bq/g*  
*=> ~1.5 GBq/waste package*  
*(based on 17013 GBq for 11449 waste packages)*
- What is the designated end-point of this waste?  
*CSA (surface disposal in Aube District)*



*Ion exchange resins*

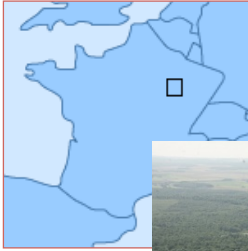


*Cross-section of a French waste-package*



# LLW repository in Aube District

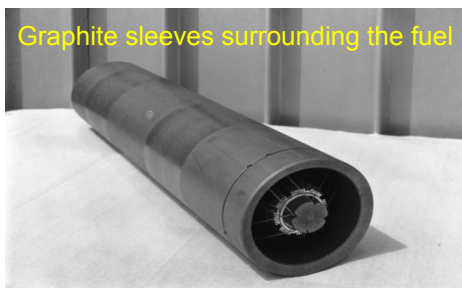
(operating since 1992)



## Irradiated Graphite



- What are the amounts you expect to have / will be generated?  
*8500 waste packages*  
*Approx. 23,000 tons of LL-LLW, i.e. approx. 90,000 m<sup>3</sup> in concrete packages*
  - Origin(s) *UNGG (Uranium Naturel Graphite Gaz) reactor*



- How is the carbon-14 content determined / specified? *It is based on radiochemical measurements and computation*
- What is the activity of <sup>14</sup>C per waste package / kilogram metal ?  
*Between 2.10<sup>5</sup> and 3.10<sup>11</sup> Bq/waste package => 10<sup>4</sup> and 10<sup>5</sup> Bq/g*
- What is the designated end-point of this waste? *Shallow disposal (LL-LLW)*



## To learn more ...





déchets  
radioactifs.com

edité par




[LES DÉCHETS DE LA RADIODACTYLIE](#)

[LES DÉCHETS À LA LOUPE](#)

[LE TOUR DES SOLUTIONS](#)

[LE DÈJÀ DE LA SCIENCE ET DE LA TECHNIQUE](#)



HISTOIRE



ORDONNES



INVENTAIRE



CLASSIFICATION



RÈGLES

[Accueil](#) > [Les déchets à la loupe](#)

## LES DÉCHETS À LA LOUPE

La fabrication des ampoules de la radiodactylie dans de nombreux secteurs est à l'origine de la production de déchets radioactifs qui, pour des raisons techniques ou économiques, ne peuvent être réutilisés ou recyclés. Cables, alternateurs, borniers, plaquettes : la grande majorité des déchets radioactifs sont cependant des déchets classiques. Cependant, étant radioactifs, ils ont la particularité d'être des déchets pouvant présenter un risque pour l'homme et l'environnement. Ce n'est là qu'un exemple des déchets comme des déchets classiques et doivent être pris en charge de manière spécifique.

### LE NOYAU DE L'INFO


» Ou se trouvent les déchets radioactifs ? »




#### HISTOIRE

Comme toute activité humaine, les déchets génèrent des problèmes de radioprotection des déchets, qui, pour certains sont radioactifs.



### NOS PARTENAIRES







[Accessibilité](#)
[Droits](#)
[Politique de confidentialité](#)
[Plan du site](#)
[Contact](#)



Thank you for your attention!





Presentation No. 9



1

# Carbon-14 Source Term CAST-workshop

Country: **Czech Republic**

Organisation: **SURAO**

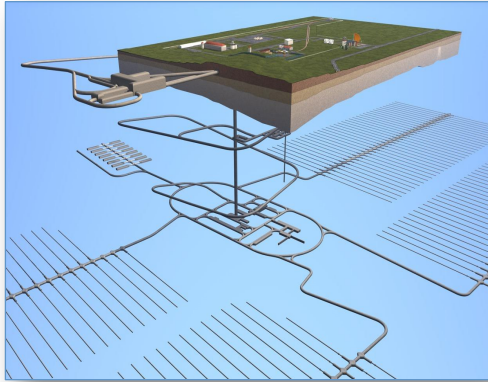
Name: **Dmitry Lukin, Antonín Vokál**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website

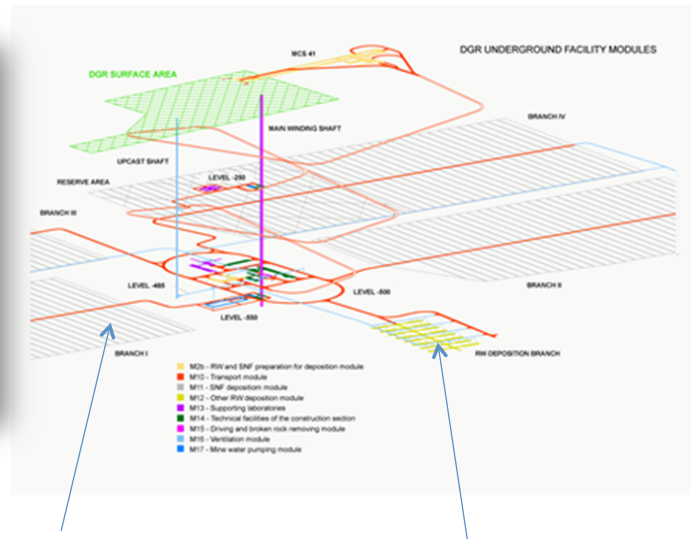


The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.

# Czech Geological Disposal Concept



Visualization of the DGR



SF repository

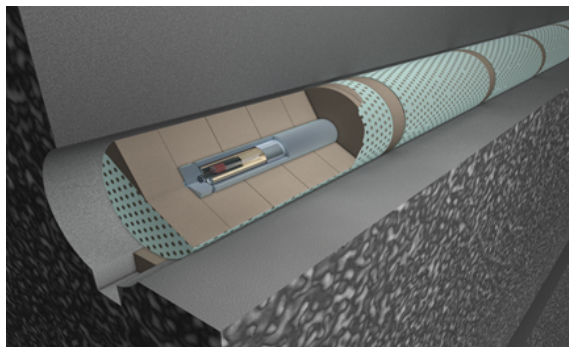
ILW repository



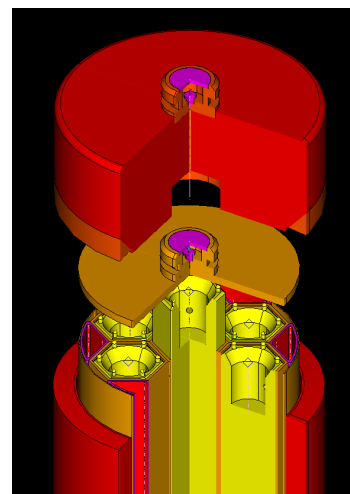
3

# Czech Geological Disposal Concept

## DGR concept for spent fuel assemblies



Visualization of the Czech concept for horizontal boreholes



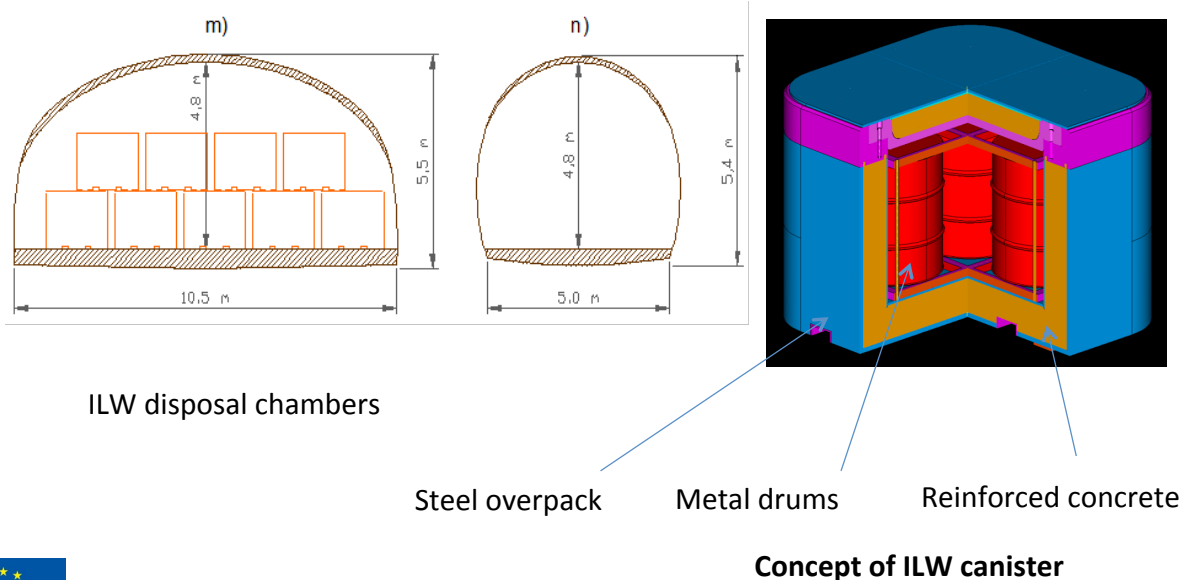
Concept of SF canister



4

# Czech Geological Disposal Concept

## DGR Concept for ILW



5

## Irradiated Steels



- Origin of irradiated steels is mainly decommissioning waste from primary zone of nuclear power plants
- This waste will be disposed in ILW canisters
- carbon-14 content is determined only by calculations
- The activity of  $^{14}\text{C}$  is ca. 77 000 GBq/t (total  $5\text{E}14$  Bq)
- Designated end-point of this waste: deep geological repository





# Irradiated Zircaloy



- The average content of  $^{14}\text{C}$  on one assembly of spent fuel
  - from WWER 440 reactors is  $2.1\text{E}10$  Bq
  - from WWER 1000 reactors  $7.6\text{E}10$  Bq.
  - Total amount of  $^{14}\text{C}$  is  $1\text{E}15$  Bq.
- This waste will be disposed in SF canisters
- The content of carbon-14 was determined by calculations
- The activity of  $^{14}\text{C}$  is about 166-235 GBq/t
  
- Designated end-point of this waste: deep geological repository
- From the safety point of view the  $^{14}\text{C}$  is not so important due to relatively low half time (5600 yr) in comparison to expected lifetime of canisters (100000 yr) for spent fuel assemblies



# Spent Ion-exchange resins



- Spent ion-exchangers from WWER reactors have relatively low concentration  $0.13$  GB/t
- Now there are solidified by geopolymers and disposed of in surface repository at Dukovany
- The concentration of C-14 in spent ion-exchange resins is determined by standard analytical methods in certificated laboratories



# Irradiated Graphite



- In the Czech Republic there is no graphite waste



## $^{14}\text{C}$ Inventory



Waste type	Number of waste units	C-14 activity [GBq/t <sub>hm</sub> ]	Uncertainty
SF from WWER 440 reactors	20 250 SF assemblies	187	High
SF from WWER 1000 reactor	4600 SF assemblies	235	High
SF from WWER 1000 (new type)	8100 SF assemblies	166	High
Decommissioning waste from WWER 440 reactors	1996 waste packages	77	High
Decommissioning waste from WWER 1000 reactors	250 waste packages	148	High
Ion-exchange resins	4200 waste packages	0.13	High





Presentation No. 10

1

# **CARbon-14 Source Term CAST-workshop**

Country: ROMANIA

Organisation: Nuclear Agency and for Radioactive Waste

Names: Stelian ORASANU and Alice Mariana DIMA (ANDR)

Daniela DIACONU (ICN-RATEN), Crina BUCUR (ICN-RATEN)

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**



## Irradiated Steels



- The amounts we expect to be generated: 1,420.288 tonnes core internals.
  - Origins: Cernavoda NPP core internals;
  - Image of waste (processed) package: not available;
  - The carbon-14 content determined / specified: measurements;
  - The activity of  $^{14}\text{C}$  per waste package / kilogram metal is not available.
- The irradiated steels will be disposed in near surface repository or DGR.



## Irradiated Zircaloy



- The amounts we expect to be generated: 92.720 tonnes Zr-2.5%Nb (pressure tubes) and 34.960 Zy 2 (Calandria tubes)
  - Origins: Cernavoda NPP;
  - Image of waste (processed) package: not available;
  - The carbon-14 content determined: for Zr-2.5%Nb ORIGEN simulations;
  - The activity of  $^{14}\text{C}$  per waste kilogram metal: for Zr-2.5%Nb  $1.51\text{E}+09$  Bq and for Zy 2 is not available;
- The irradiated zircaloy will be disposed in DGR



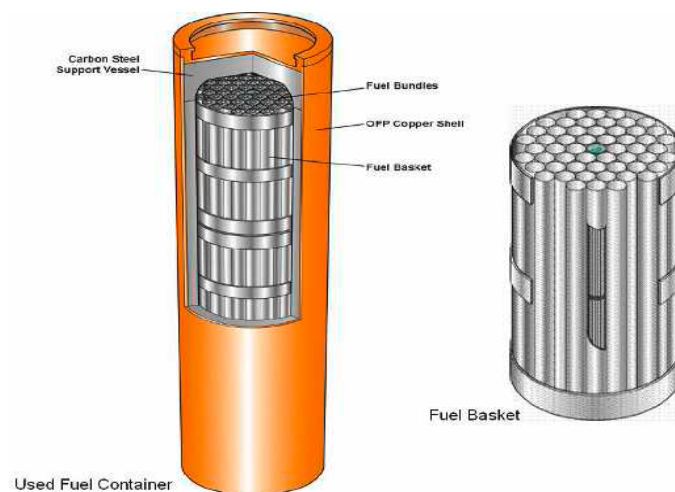
# Irradiated Zircaloy



- The amounts we expect to be generated: ~486500 FB (~9340.8tU)
  - Origins: Cernavoda NPP
  - The carbon-14 content is measured/estimated: ORIGEN simulations;
  - The activity of  $^{14}\text{C}$  per waste kilogram uranium:  $5.7\text{E}+6$  to  $2.03\text{E}+7$  Bq.
- In Romania nuclear spent fuel is waste and will be disposed in DGR



# Irradiated Zircaloy



Disposal canister for CANDU SF



## Spent Ion-exchange resins



- The amounts we expect to be generated: 290 m3.
  - Origin: Cernavoda NPP;
  - Image of waste (processed) package: not available;
  - The carbon-14 content determined: measured on moderator SIERs;
  - The activity of  $^{14}\text{C}$  per waste kilogram resin: up to  $3.45\text{E}+9$  Bq.
- Spent ion-exchange resins will be disposed in DGR



## Irradiated Graphite



- The amounts we expect to have /to be generated: 7.8 tonnes
  - Origins: TRIGA reactor (2.5 tonnes) and VVR-S reactor (5.3 tonnes);
  - Image of waste (processed) package: not available;
  - The carbon-14 content determined by measurement;
  - The activity of  $^{14}\text{C}$  per waste kilogram graphite: ranging from  $1.1\text{E}+7$  to  $1\text{E}+10$  Bq for TRIGA and  $1\text{E}+10$  to  $1\text{E}+11$  Bq for VVR-S.
- The graphite will be disposed in DGR



Presentation No. 11

1

# CArbon-14 Source Term CAST-workshop

Country: **JAPAN**

Organisation: **RWMC** (Radioactive Waste Management funding and research Center)

Name: **Tomofumi Sakuragi**

This presentation ~~can/cannot~~ be used for the Proceedings of the workshop that will be published at CAST website



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.





# Irradiated Steels



- What are the amounts you expect to have / will be generated?
  - End-pieces of fuel assembly (PWR and BWR) after reprocessing of 32,000 tU
  - 1,200 t
  - Four SS canisters packed in a carbon steel package
  - $^{14}\text{C}$  inventory is determined by calculation (ORIGEN)\*
  - $1.2 \times 10^{11}$  Bq/t-SS,  $5.8 \times 10^9$  Bq/canister,  $2.3 \times 10^{10}$  Bq/package (Total  $1.4 \times 10^{14}$  Bq)
- What is the designated end-point of this waste?
  - Geological disposal (24,150 canisters)
  - Remarks: Other steels from operation and decommissioning of NPPs (9,000 t) will be disposed in intermediate depth below 70 meters



\* a typical irradiation condition for PWR (45 GWd/t, Enrichment 4.5%, Specific power 38MW/t)

Federation of Electric Power Companies (FEPC) and Japan Atomic Energy Agency (JAEA), Second Progress Report on Research and Development for TRU Waste Disposal in Japan (2007)



# Irradiated Zircaloy



- What are the amounts you expect to have / will be generated?
  - Fuel cladding (PWR and BWR) after reprocessing of 32,000 tU
  - 10,000 t
  - Four SS canisters packed in a carbon steel package
  - $^{14}\text{C}$  inventory is determined by calculation (ORIGEN)\*
  - $2.3 \times 10^{10}$  Bq/t-Zr,  $9.6 \times 10^9$  Bq/canister,  $3.8 \times 10^{10}$  Bq/package (Total  $2.3 \times 10^{14}$  Bq)
- What is the designated end-point of this waste?
  - Geological disposal (24,150 canisters)
  - Remarks: Zircaloy channel box (5,000 t) will be disposed in intermediate depth below 70 meters



\* a typical irradiation condition for PWR (45 GWd/t, Enrichment 4.5%, Specific power 38MW/t)

Federation of Electric Power Companies (FEPC) and Japan Atomic Energy Agency (JAEA), Second Progress Report on Research and Development for TRU Waste Disposal in Japan (2007)



# Spent Ion-exchange resins



- What are the amounts you expect to have / will be generated?
  - From operation of BWRs (2,553 t) and PWRs (562 t)
  - 1.6 m square package (Carbon steel, 5 cm thick, 647 packages)
  - $^{14}\text{C}$  inventory is determined by calculation (ORIGEN)
  - BWR;  $3.7 \times 10^9$  Bq/t,  $1.9 \times 10^{10}$  Bq/package (Total  $9.4 \times 10^{12}$  Bq)
  - PWR;  $8.3 \times 10^8$  Bq/t,  $3.1 \times 10^9$  Bq/package (Total  $4.7 \times 10^{11}$  Bq)
- What is the designated end-point of this waste?
  - Intermediate depth disposal below 70 meters



<https://www.nsr.go.jp/data/000090254.pdf#search=%27%E4%BD%99%E8%A3%95%E6%B7%B1%E5%BA%A0%E5%87%A6%E5%88%E6%AF%E5%8B%1A%E5%8B%83%E6%A3%84%E7%89%A9%E3%81%AB%E9%96%A2%E3%81%89%E3%82%8B%E5%9F%BA%E6%9C%AC%E3%83%87%E3%83%BC%E3%82%BF%E9%9B%86%27>



# Irradiated Graphite



- What are the amounts you expect to have / will be generated?
  - Moderator and reflector of a GCR\* (1,514 tons)
  - 1.6 m square package (Carbon steel, 5 cm thick, 655 packages)
  - $^{14}\text{C}$  inventory is determined by calculation (ORIGEN)
  - $9.7 \times 10^{10}$  Bq/t,  $2.3 \times 10^{11}$  Bq/package (Total  $1.5 \times 10^{14}$  Bq)
- What is the designated end-point of this waste?
  - Intermediate depth disposal below 70 meters



\* Tokai Power Station (Japan atomic power company)

<https://www.nsr.go.jp/data/000090254.pdf#search=%27%E4%BD%99%E8%A3%95%E6%B7%B1%E5%BA%A0%E5%87%A6%E5%88%E6%AF%E5%8B%1A%E5%8B%83%E6%A3%84%E7%89%A9%E3%81%AB%E9%96%A2%E3%81%89%E3%82%8B%E5%9F%BA%E6%9C%AC%E3%83%87%E3%83%BC%E3%82%BF%E9%9B%86%27>



Presentation No. 12

1

# **CArbon-14 Source Term CAST-workshop**

Country: Bulgaria

Organisation: SERAW

Name: Penka Avramova

This presentation can be used for the Proceedings of the workshop  
that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme  
for research, technological development and demonstration under grant agreement no.  
604779, the CAST project.**



# Irradiated Steels



- The amounts expected are about 1211 tonnes.
  - Origin: Irradiated reactor vessel and reactor internals.
  - Carbon-14 content is determined by calculations using computer codes SCALE, ORIGEN, DOORS.
  - The activity of  $^{14}\text{C}$  depends on the mass share of Carbon in the respective material (stainless steel  $8\text{E-}04$ ; reactor vessel  $1.65\text{E-}03$ ; carbon steel  $1.18\text{E-}02$ )
- The designated end-point of this waste is deep geological disposal.



# Irradiated Zircaloy



- Irradiated Zirconium alloys are not expected.
  - Zirconium alloy, type E110 (99 % Zirconium + 1 % Niobium), is used in the cladding of the fuel assemblies of Russian reactors VVER.
  - According to a framework trade agreement between Bulgaria and Russian federation, Spent Fuel (SF) is shipped for reprocessing to Russia, and after that Bulgaria will receive the resulting HLW.
  - These HLW will not contain Zirconium alloys.



## Spent Ion-exchange resins



- Amounts of spent IER:  $\sim 443 \text{ m}^3$
- IER originate from the water purification systems: (SVO-1) first circuit; (SVO-5) steam generators blow down and (SVO-4) spent fuel pool and emergency feed water tanks.
  - The waste package will be cemented mixture in a barrel.
  - Carbon-14 content will be determined in the frame of decommissioning project 5a: IER Retrieval, Characterization and Conditioning Methodology.



## Spent Ion-exchange resins



- C-14 content is determined by application of scaling factors according to ISO 21238:2007 Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants.  
Radiochemical analyses include Liquid Scintillation Counting (LSC) after chemical treatment of samples.
- The waste package will be cemented mixture in a barrel.
- The end point of this waste is storage in near surface repository.



# Irradiated Graphite



- Reactors VVER do not use graphite moderators and thus SERAW does not have such kind of RAW



Thank you for your attention!



Presentation No. 13

1

# **CArbon-14 Source Term CAST-workshop**

Country: Spain

Organisation: Enresa

Name: Miguel Cuñado

This presentation can be used for the Proceedings of the workshop that will be published at CAST website



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**





# Irradiated Steels



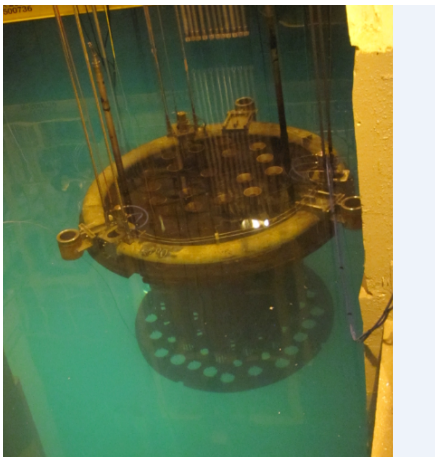
- Expected amount
  - Origin: Structural parts of the fuel assemblies (FA) and internal components of the reactors vessel (main contrib.)
  - Estimated amount:
    - $4.3 \cdot 10^3$  MBq/FA. 14 400 Equivalent Reference FA  $\Rightarrow 6.19 \cdot 10^7$  MBq
    - Internal comp. of one small reactor 150 MWe  $\Rightarrow 1.96 \cdot 10^7$  MBq
    - No precise estimation available of the total (10 reactors)
  - Activity of  $^{14}\text{C}$  per waste package: max.  $1.06 \cdot 10^4$  MBq or  $4.45 \cdot 10^5$  MBq (characterization level 1 or 2), for those to be disposed at El Cabril  $\Rightarrow$  upper internal components
  - No limits on  $^{14}\text{C}$  for High Level Waste



# Irradiated Steels



- Designated end-point of this waste:
  - The L/IW repository at El Cabril.



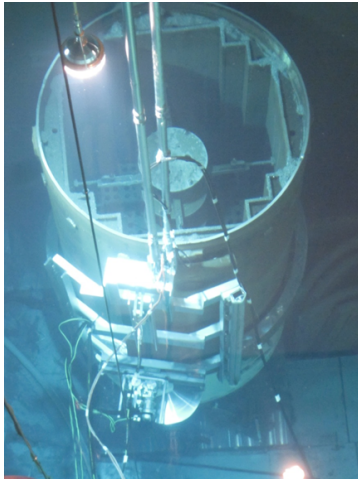
Upper internal components

Conditioning of upper internal components

# Irradiated Steels



- Designated end-point of this waste:
  - Interim Storage and probably geological disposal (L.I.C. and F.A.)



Lower internal components



Storage of lower internal components

# Irradiated Zircaloy



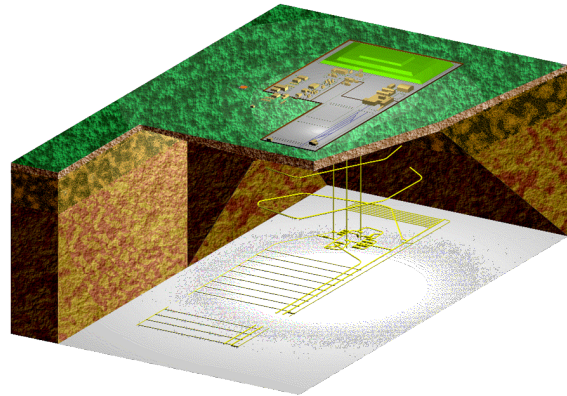
- Expected amount
  - Origin: Sheaths of the fuel assemblies (FA) rods
  - Estimated amount:
    - $8.14 \cdot 10^3$  MBq/FA. 14 400 Equivalent Reference FA  $\Rightarrow 1.17 \cdot 10^8$  MBq
  - Determination: Calculated, based on impurities content
  - No activity limits on  $^{14}\text{C}$  for High Level Waste



# Irradiated Zircaloy



- Designated end-point of this waste:
  - Interim Storage and geological disposal.



# Spent Ion-exchange resins



- Expected amount
  - Origin: Cleaning of liquid systems in NPP's like fuel pool, cooling system, primary circuit.
  - Conditioning is made with cement and some additional water. Hydration liquid is provided by the resin itself. One third of the mass in a standard drum (220 l) corresponds to the resin and two thirds is the cement plus additional water.
  - Activity of  $^{14}\text{C}$  per waste package: max.  $1.06 \cdot 10^4$  MBq or  $4.45 \cdot 10^5$  MBq (characterization level 1 or 2)



# Spent Ion-exchange resins



- Designated end-point of this waste:
  - The L/IW repository at El Cabril.



Conditioned resins



Resine packages ready for disposal



# Irradiated Graphite



- Expected amount
  - Origin: Neutron moderator/reflector of a UNGG (*Uranium Naturel Graphite Gaz*) reactor, cooled by CO<sub>2</sub>.
  - Amount: Graphite sleeves (1000 t), moderator graphite pile (2680 t), plus 8 t from a Material Testing Reactor. Total <sup>14</sup>C activity is 1.65·10<sup>8</sup> MBq
  - Determination: in a sample, similar process like the resins
  - Activity of <sup>14</sup>C per waste package: max. 1.06·10<sup>4</sup> MBq or 4.45·10<sup>5</sup> MBq (characterization level 1 or 2 -- concentration), **BUT:**



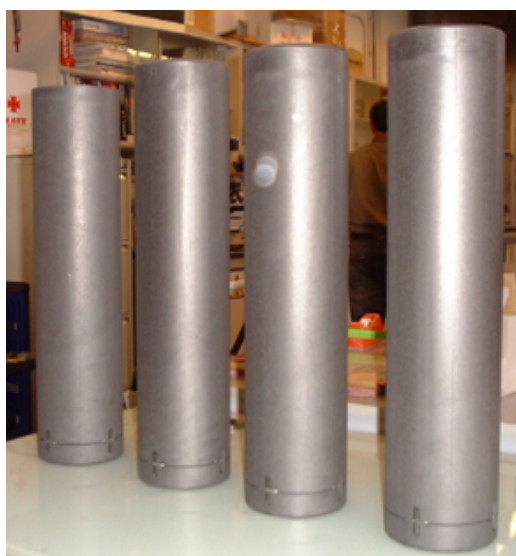
# Irradiated Graphite



- Designated end-point of this waste:
  - Lack of specific acceptance criteria
  - Total graphite activity exceeds eight times the limits of the L/IW repository at El Cabril
  - Selective decontamination is being studied. If successful, graphite could be disposed of at El Cabril, and the high level by-product waste should be sent to an interim storage facility and/or finally to a geological repository.
  - The possibility of impermeabilization of macro porosity would lead to a low  $^{14}\text{C}$  release rate and thus, it could eventually be disposed at El Cabril, provided that the legal limits are modified.
  - Currently is stored inside the reactor building and containers.



# Irradiated Graphite



Virgin graphite sleeves





Presentation No. 14

1

# **CArbon-14 Source Term CAST-workshop**

Country: Finland

Organisation: Fortum

Name: Olli Nummi

This presentation can be used for the Proceedings of the workshop that will be published at CAST website



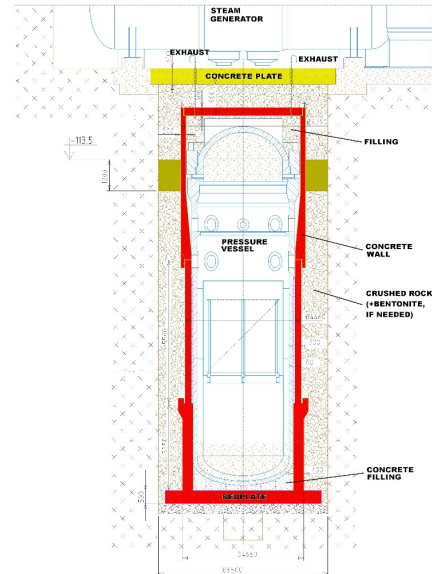
**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**



# Irradiated Steels



- Mainly reactor internals, pressure vessels and other activated components
- Internals packaged inside pressure vessels
- C-14 activity is based activation calculations (MCNP)
  - Internals: 53 800 GBq
  - Pressure vessels: 350 GBq
  - Other waste: 142 GBq



# Irradiated Zircaloy



- Spent nuclear fuel
- Disposal at Olkiluoto by Posiva (KBS-3V concept)
- Not part of Fortum's contribution to CAST

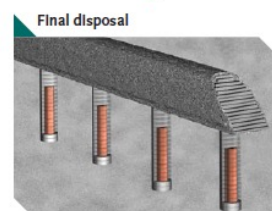


Figure: Posiva



## Spent Ion-exchange resins



- Originates from purification of primary circuit
- Solidified into concrete containers
- C-14 activity based on direct measurements
  - Total activity 500 GBq, on average 0.15-0.25 GBq/container
- Containers disposed inside a concrete basin



## Irradiated Graphite



- No graphite is generated at Loviisa NPP (Not part of Fortum's contribution to CAST)
- Only graphite in Finland originates from a research reactor FiR 1
  - Reflector and thermal column
  - Possible disposal into LILW repository at a Olkiluoto or Loviisa

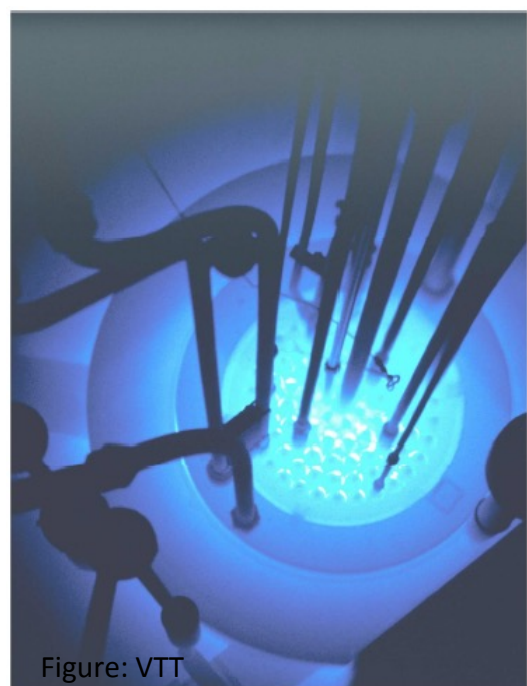


Figure: VTT







Presentation No. 15

1

# **CArbon-14 Source Term CAST**

Name: Olli Nummi  
Organisation: Fortum, Finland  
Date: October 5<sup>th</sup>, 2016



**The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.**



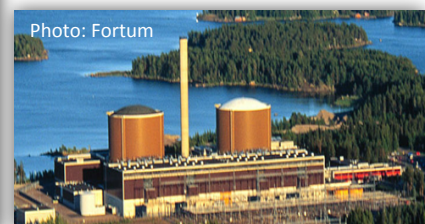
# Outline



- General
- Part I: Solidification and disposal of ion-exchange resins
- Part II: Long term-safety



## Nuclear power in Finland



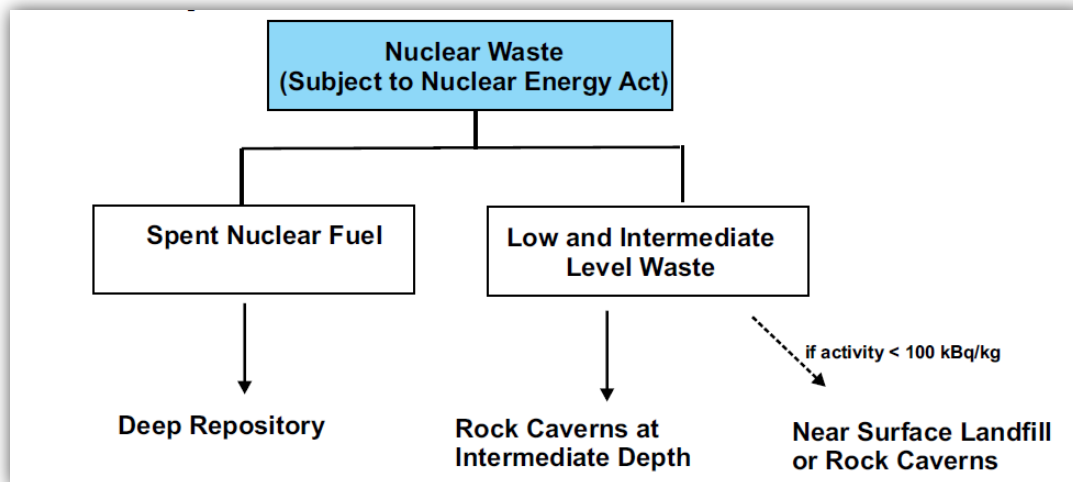


Image: Finnish Radiation and Safety Authority (STUK)

## Part I: Solidification and disposal of ion-exchange resins



# Ion-exchange resins



## Loviisa NPP

- Accumulation: 10-15 m<sup>3</sup>/a
- At storage ~550 m<sup>3</sup>



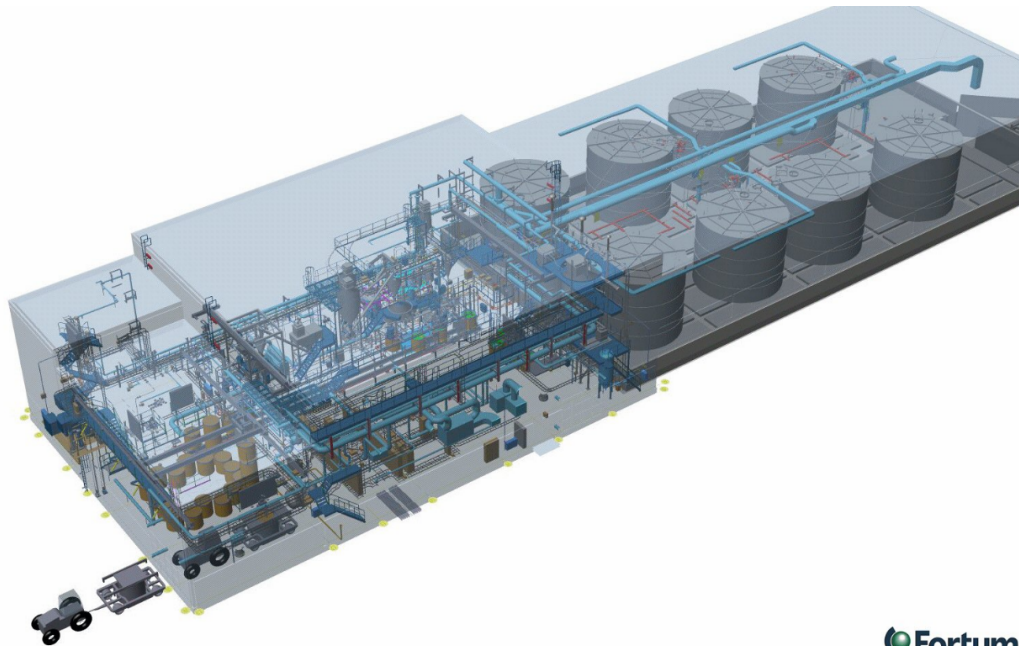
## Loviisa NPP



Solidification plant and liquid waste storage



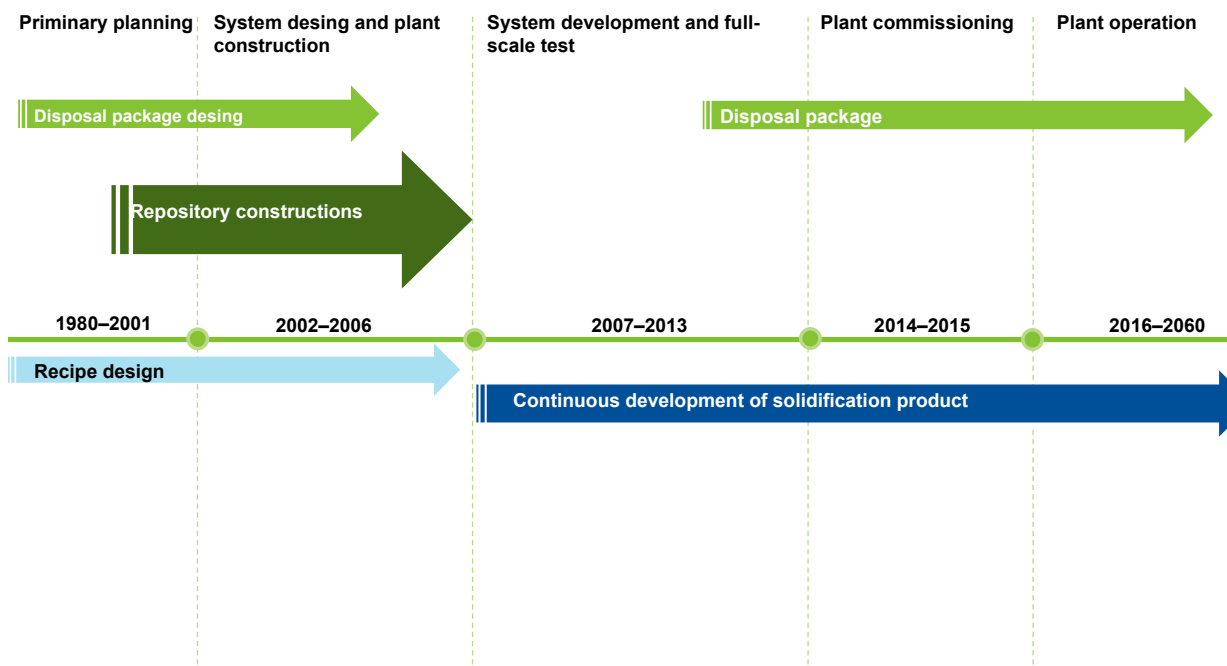
# Solidification plant and liquid waste storage



Fortum



# Solidification plant; design, development and production

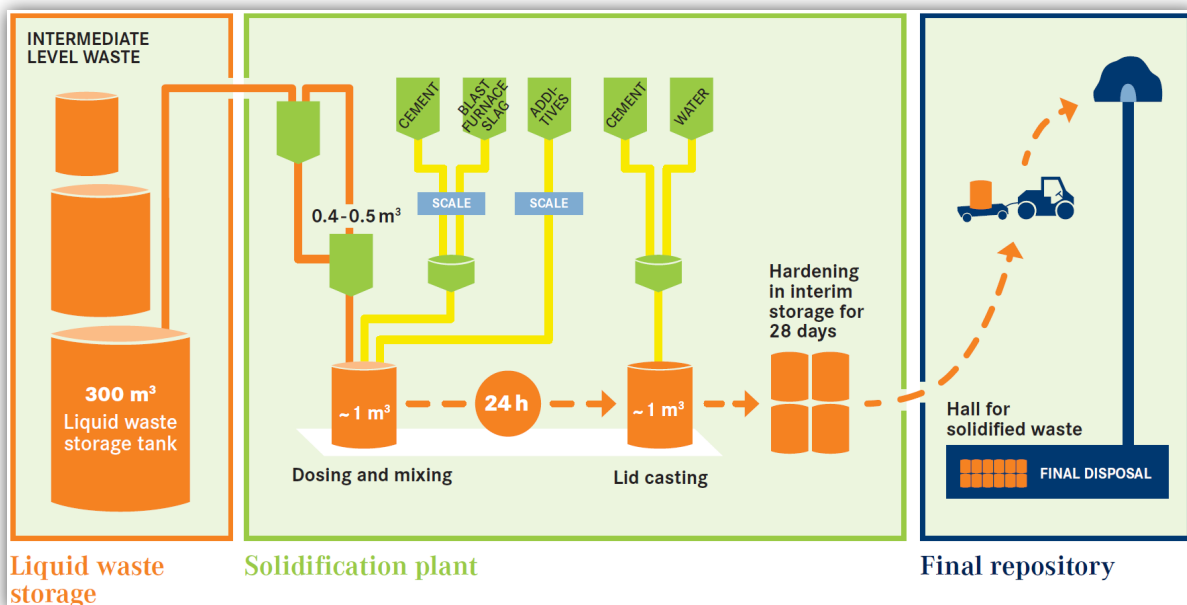




# Waste package



# Solidification process



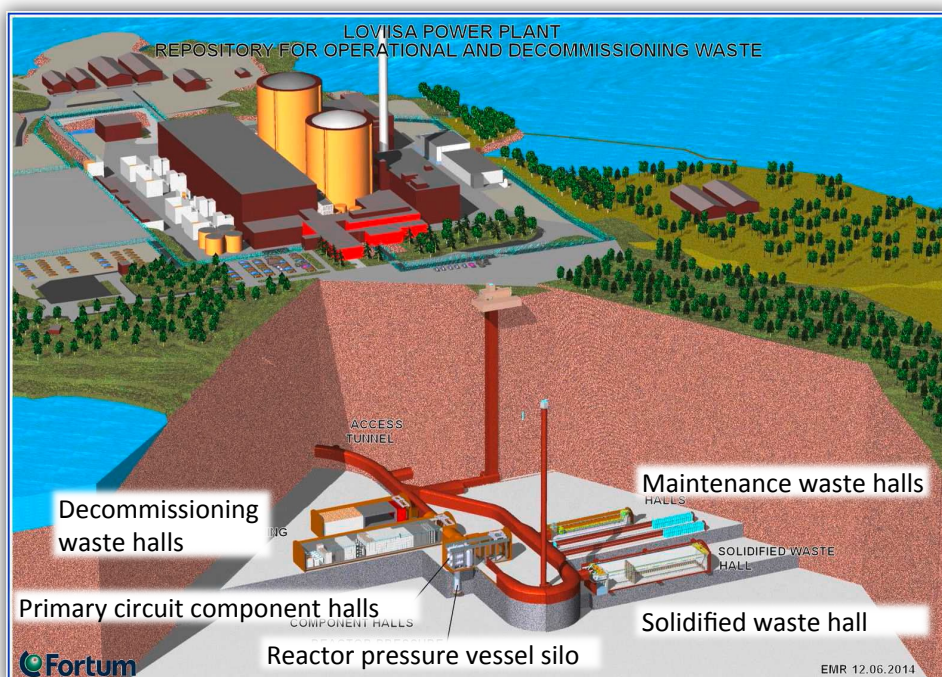




## Solidification process (2)



## Disposal facility







## Hall for solidified waste



## Repository closure



- Hall for solidified waste backfilled
- Concrete plug sealing the hall
- Concrete plugs and backfilling in the access tunnel and shafts



## Part II: Long-term safety



### Radionuclide release



#### Main features and processes

- Groundwater flow
- Diffusion
- Sorption
- Concrete/waste matrix degradation
- (Gaseous transport)



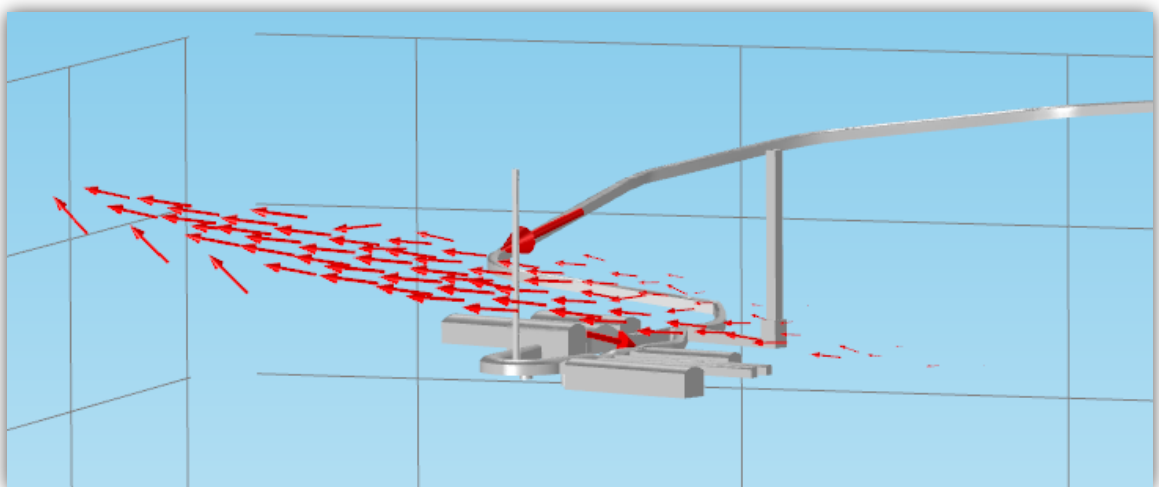
# Safety functions



- Waste and waste package
  - Limit the release
- Concrete basin
  - Limit the groundwater flow
  - Protect the waste package
- Backfill
  - Protect the basin
- Bedrock and closure
  - Limit the groundwater flow and prevent intrusion

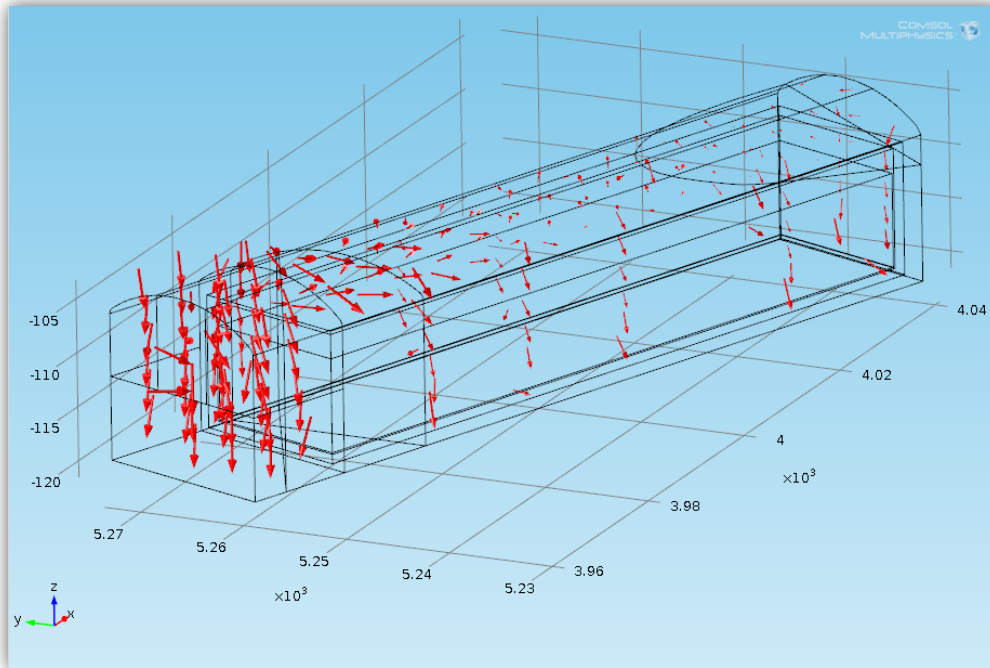


# Groundwater flow

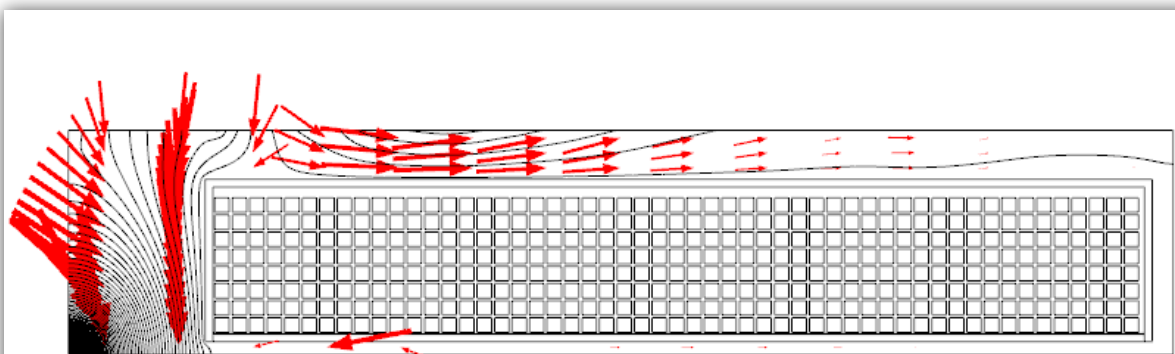




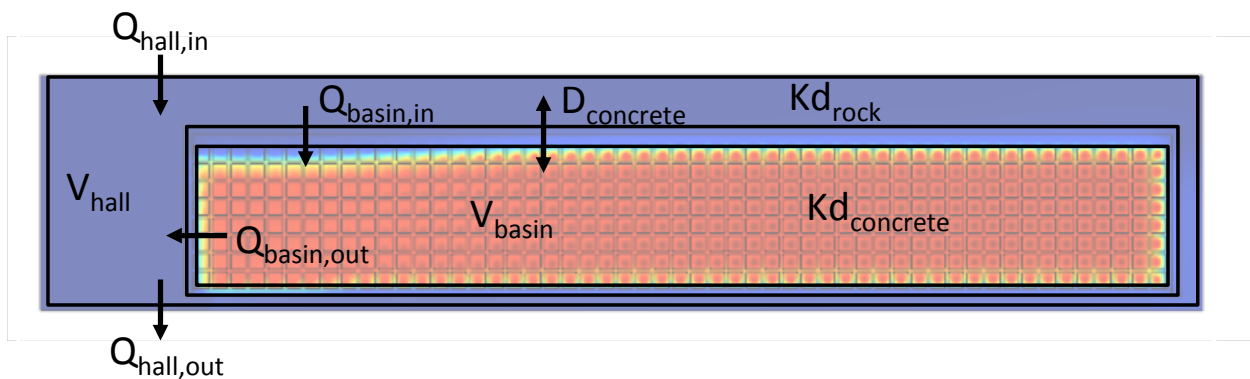
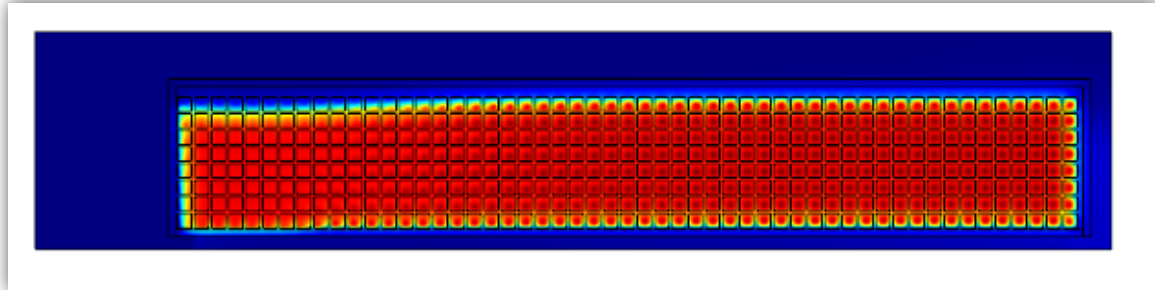
## Groundwater flow (2)



## Groundwater flow (3)



500 years after the closure





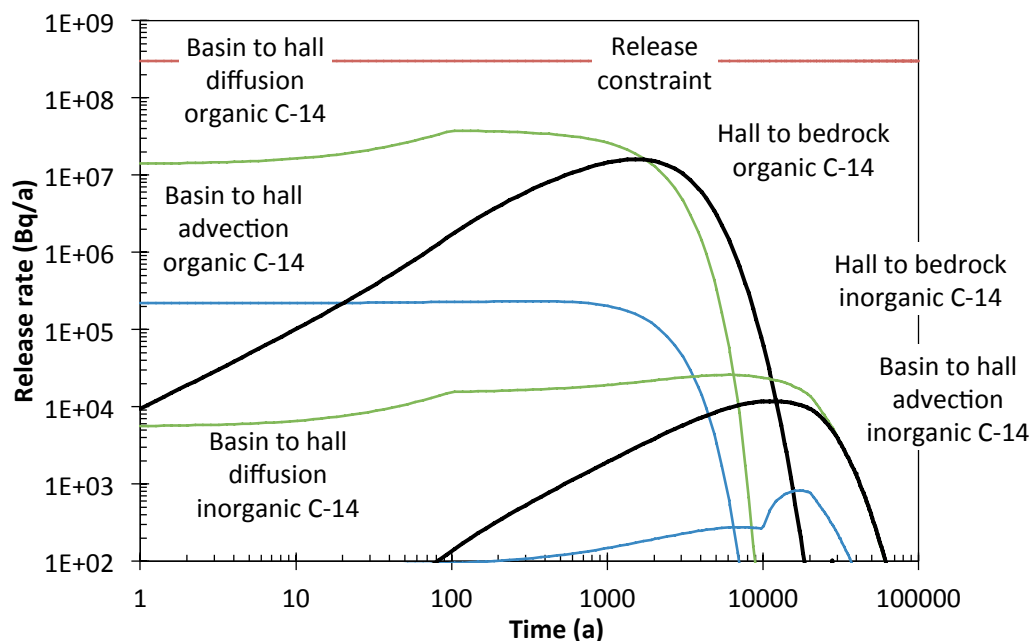
# Parameter selections



	C-14 organic	C-14 inorganic
Activity (GBq)	75	425
Kd <sub>concrete</sub> (m³/kg)	1E-5...1E-3	0.5...5
Kd <sub>rock</sub> (m³/kg)	1E-5...1E-3	1E-4...1E-2
Diffusion coefficient in concrete (m²/s)		
- 0...100 a	3.50E-12	
- 100...10 000 a	1.00E-11	
- 10 000...20 000 a	5.00E-11	
- 20 000...100 000 a	1.00E-10	
Groundwater flow (m³/a) through		
	Hall	Basin
- 0...10 000 a	2.50	0.01
- 10 000...20 000 a	2.50	0.11
- 20 000...100 000 a	2.50	1.10

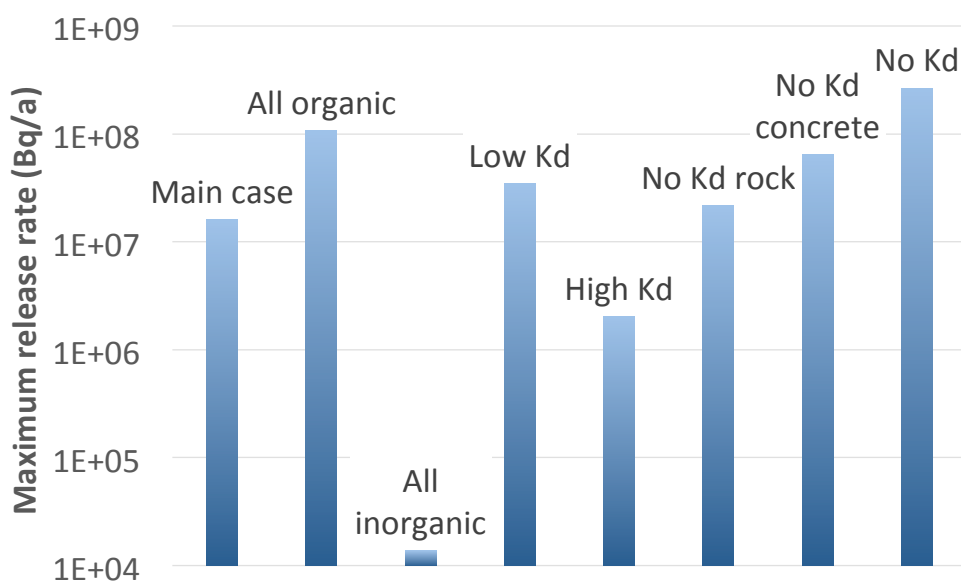


## Release rates – main case





## Release rates – sensitivity cases



## Expected output from CAST



- Support to C-14 speciation
- Sorption data
- Enhance understanding



## Concluding remarks



- Ready to start disposal
- Long-term safety demonstrated
  - Reduction of uncertainties



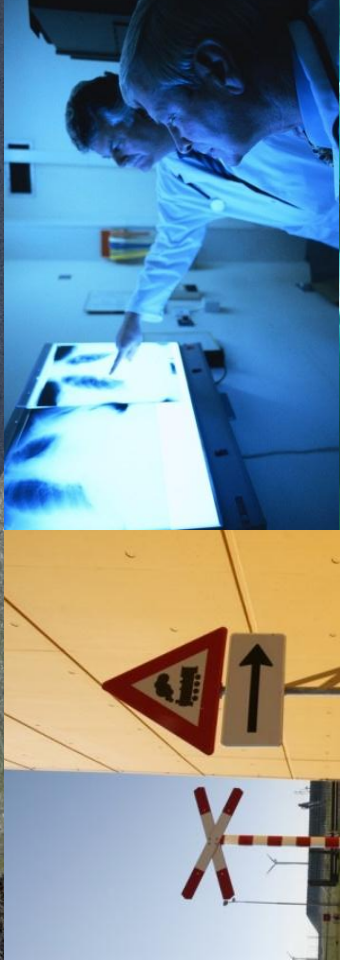
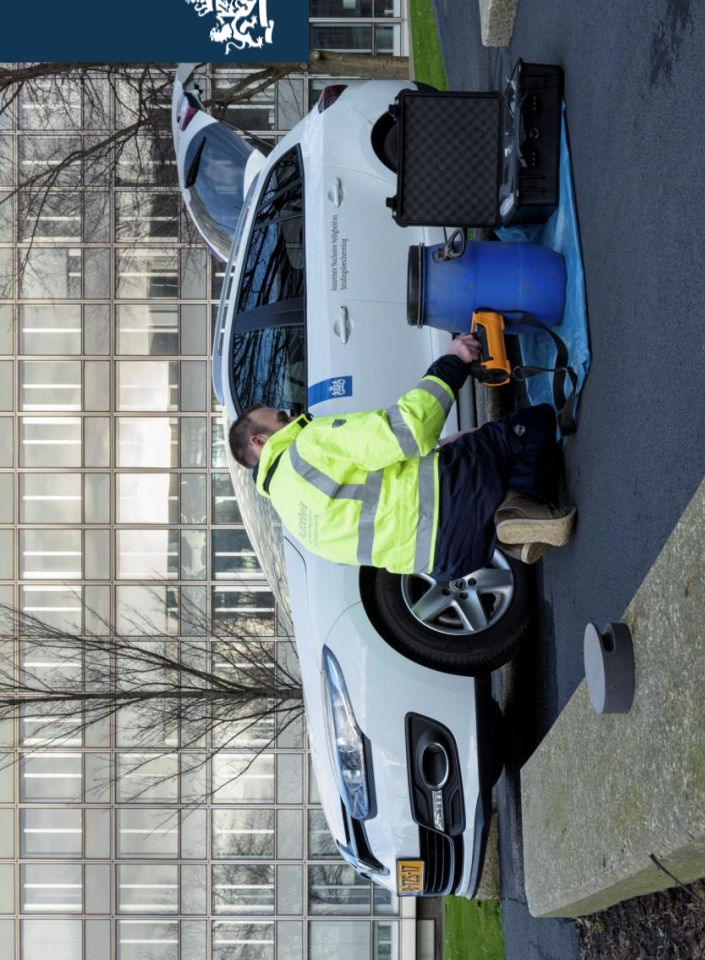




## Presentation No. 16



Authority for Nuclear Safety and  
Radiation Protection



## ANVS Introduction to CAST (Carbon-14 Source Term) October 5, 2016

Thierry Louis

19 September 2016



# Establishment of the Authority for Nuclear Safety and Radiation Protection (ANVS)



1. Strengthen nuclear safety and radiation protection by combining the existing knowledge and expertise



Combination of existing regulatory staff in one organisation:  
ANVS (1-1-2015, part of Ministry of the Environment)



IAEA

2. Follow recommendations of the IAEA and Euratom for an independent regulator



Formalisation of ANVS as an independent regulator in the Nuclear Energy Law in the Netherlands (foreseen 1-1-2017)

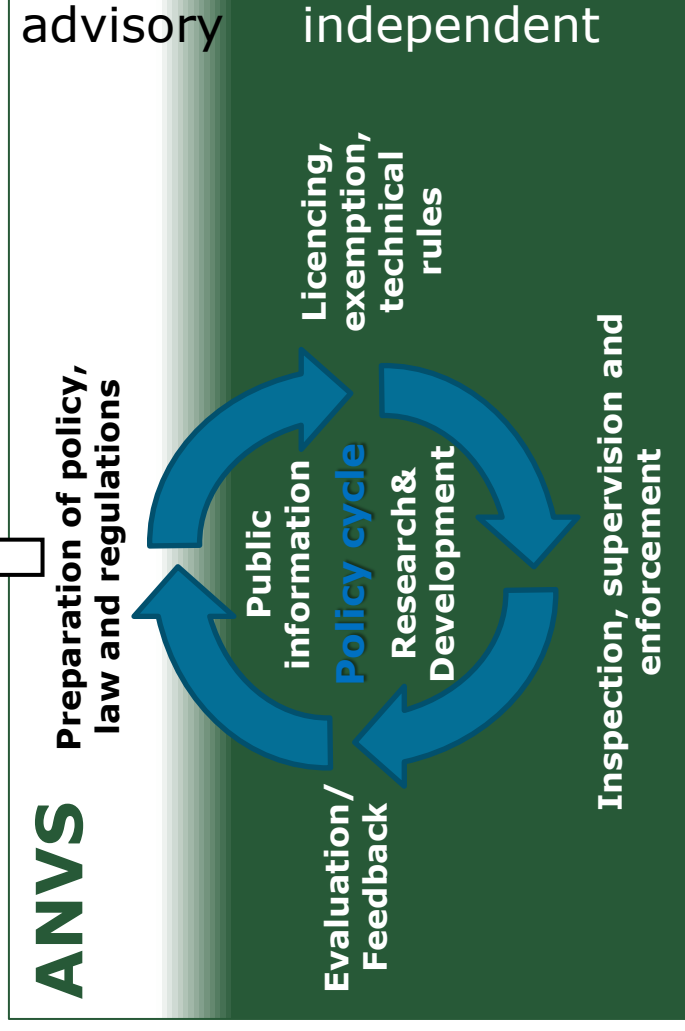
# Regulatory tasks

## Responsibilities

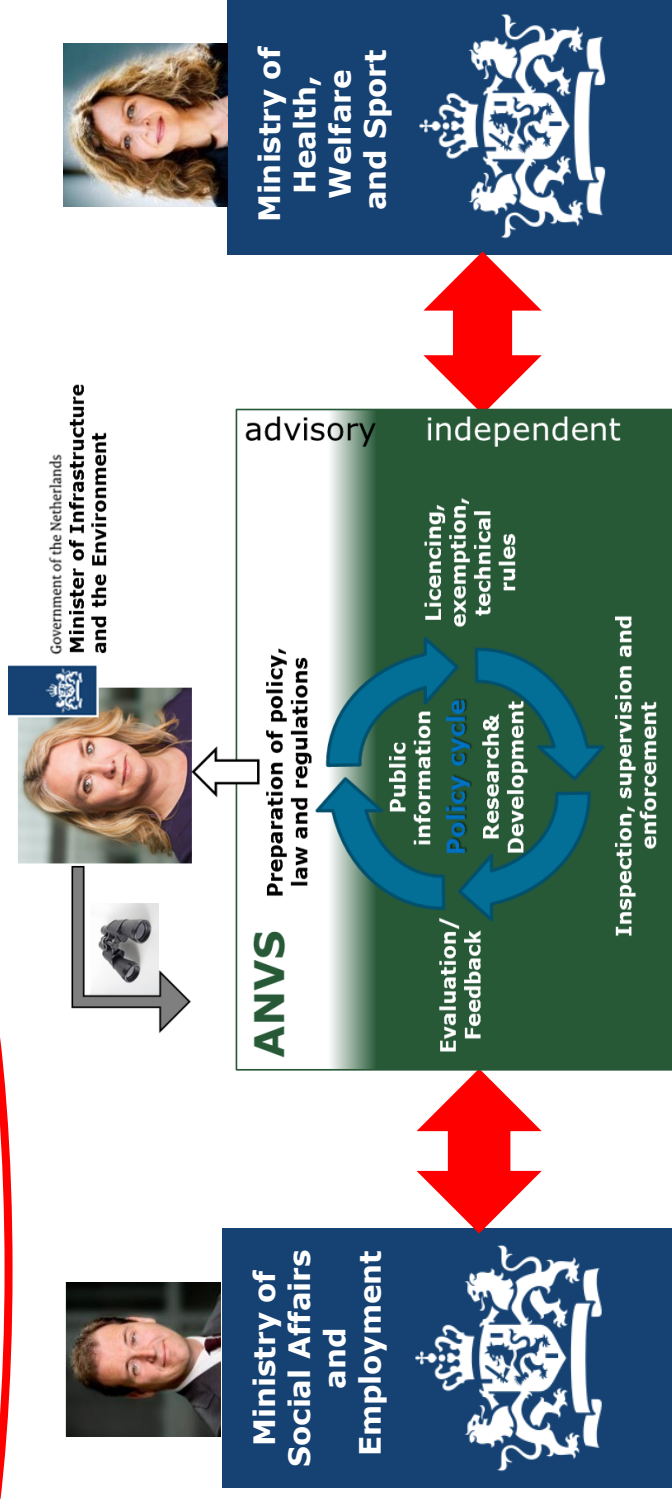
- Nuclear safety
- Radiation protection (public & environment)
- Emergency preparedness and response
- Security
- Safeguards
- Spent fuel and radioactive waste



Government of the Netherlands  
**Minister of Infrastructure  
and the Environment**



# Radiation Protection



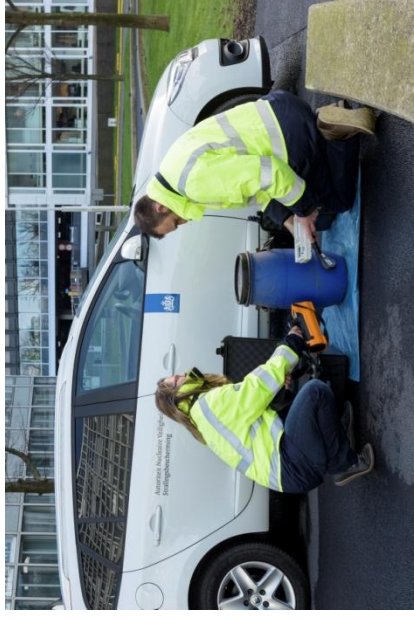




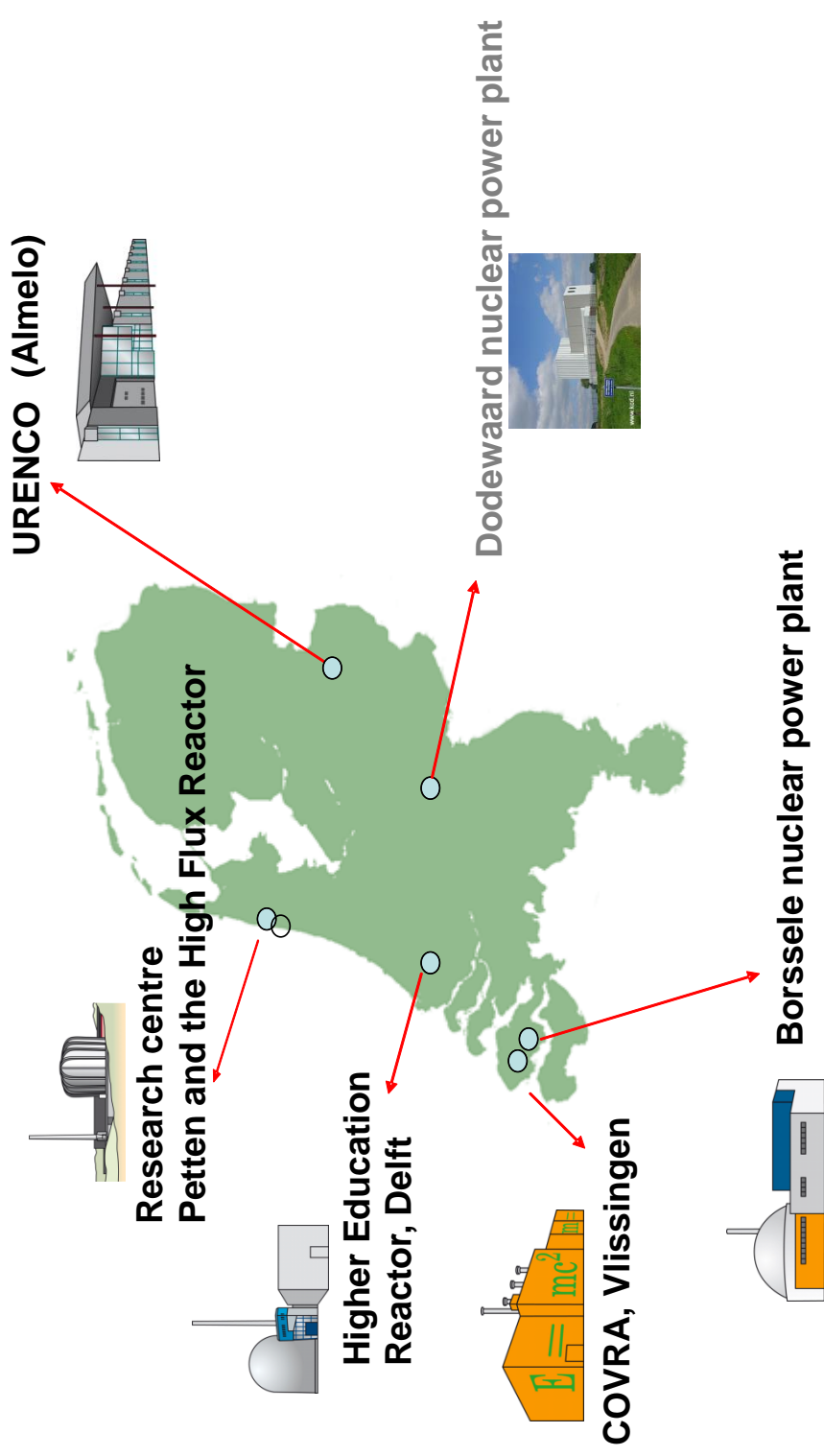
# Application of ionising radiation

## *Regulating ionising radiation*

- 1,000 Licensees (+100 security)
- 10,000 Individuals subject to a notification requirement (dentists, veterinarians, ...)
- 30 Compound licences (>100 sources)
- 10,000 Transports per year



# Nuclear Sites







# CLASSICAL TRIANGLE

authority

- policy

- laws

- licenses

- inspection

- polluter pays, *cost efficient*

responsibilities

waste

- prevent, minimize
- notify, deliver
- payment

liability

WMO

- infrastructure

- acceptance criteria

- financing

- execution



# Dutch policy on spent fuel and radioactive waste

- Minimisation of waste production: prevention, reuse and use of radioactive decay.
- Prime responsibility rests with the waste producers (i.e. license holders); the license holders shall also bear the associated costs.
- COVRA is the central organisation for the management of spent fuel and radioactive waste.
- Above-ground storage for at least 100 years; final disposal is envisaged around 2130.
- A dual-track policy: a national geological disposal facility as well as the option of cooperating with one or more countries.
- The disposal facility shall have passive safety features and during the operational period of the disposal facility the waste shall be retrievable.



# Overview of the national framework of legislation

## The Nuclear Energy Act:

- is the basis of Dutch regulations

## Governmental decrees:

- the Radiation Protection Decree (Bs): this lays down the most important rules for handling radioactive waste substances;
- the Nuclear Installations, Fissionable Materials and Ores Decree (Bkse): this lists the most important rules for handling spent fuel;
- the Transport of Fissionable Materials, Ores and Radioactive Substances Decree (Bvser): this Decree regulates the transport of spent fuel and radioactive waste.



# Collection and management of radioactive waste

## – the COVRA

- The sole organisation in the Netherlands for the collection and storage of spent fuel and radioactive waste.
- Very low level radioactive waste (ZELA) may be disposed of at specified landfill sites, or reused.
- 100% of the shares in COVRA are held by the State.
- Reserves and manages financial resources for long-term aboveground storage and for the implementation of (geological) disposal of the waste.
- Coordinates a research programme.



# Financial resources

- Low level and intermediate level radioactive waste:
  - lists of charges
  - standard waste packaging
  - paid for each waste package received
- Spent fuel and high level radioactive waste:
  - construction costs and operating costs of HABOG are borne by the waste generators



# Research Programme Disposal of Radioactive Waste (OPERA)

- Results are expected in 2016
- Coordinated by COVRA
- Financed 50-50 by government and the nuclear sector
- COVRA is obliged to finance the costs by charging on these costs via its standard charges
- Before OPERA there have been other multiyear research programmes

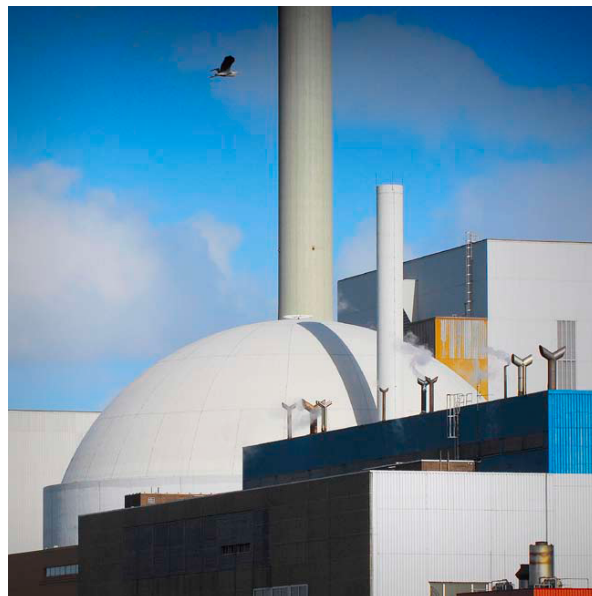


Type of Liability	Long term strategy	Funding of Liabilities	Current Practice/ Facilities	Planned facilities
Spent Fuel	Geological disposal	Costs for management and final disposal covered by all waste producers. Money for disposal stored in separate COVRA fund	Interim long-term storage (COVRA)	Extension of HABOG
Nuclear Fuel Cycle Waste				
Other Radioactive Waste				
Decommissioning Liabilities	Green field	Legislation requiring financial securities	1 NPP in safe enclosure & 1 RR under de-commissioning	None
Disused Sealed Sources	Same as 'other radioactive waste'			

## Presentation No. 17

1


1



**Jan Wieman**  
**Fuel Cycle Manager EPZ**

2



- 
1. EPZ responsibilities towards radwaste
  2. processes resulting in radioactive waste
    1. plant operations
    2. fuel cycle
    3. decommissioning
  3. summary

- EPZ is licensed to possess nuclear materials for production of energy, under specific conditions such as:
  - transfer operational waste within 5 years to COVRA
  - minimize the on-site inventory of spent fuel
  - maintain adequate decommissioning resources
- EPZ adheres to the principles of the UN Global Compact, including care for environmental impact in its supply lines
- management of spent fuel (reprocessing) is subject of French-Netherlands intergovernmental agreements
- decommissioning fund managed by independent foundation

- Low level waste such as gloves, scrapped tools, non washable clothes ... pressed in 100 l drums
- Evaporator concentrates, used resins, ... cemented in 200 l drums



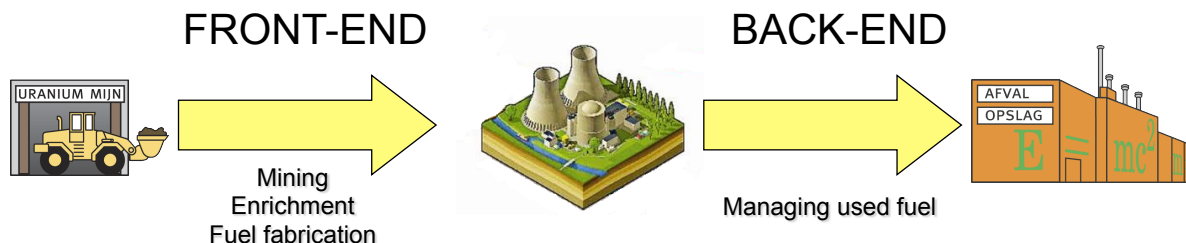
5



Waste Storage Facility licensed to store waste up to 5 years.  
Then it is shipped to COVRA for take-over.

6

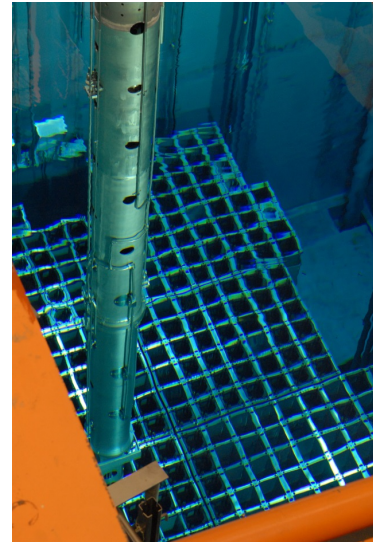
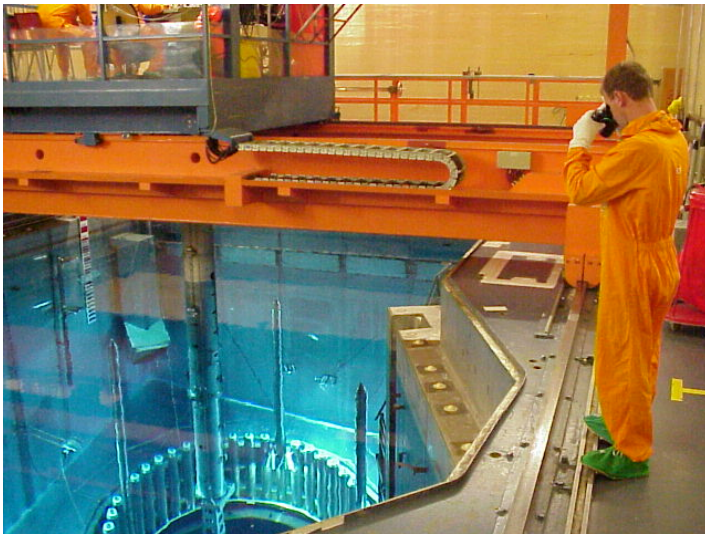
EPZ manages its front- and backend by itself



EPZ takes its responsibility for waste arising in all phases of the fuel cycle (UN Global Compact commitment)

- Dominated by environmental impact of uranium mining
- Industrial activities all outside The Netherlands except URENCO in Almelo
- EPZ expects from its suppliers good environmental and safety performance, according to international standards (ISO 14001, OHSAS 18001 etc.)





- Used fuel must be handled under water
- On-site storage as short as practical

9

- In 1979, 2009 and 2012 exchange of letters between France and The Netherlands on reprocessing Borssele fuel
- Used fuel is to be 100% reprocessed in France including last core
- Uranium and plutonium are being re-used for reactor fuel
- High Level radioactive waste returned to The Netherlands within 8 years of reprocessing

10



Reprocessing used fuel results in 2 types of radioactive waste:

- Vitrified Fission Product  
(heat production ~ 1000 W / canister)
- Compacted metallic parts  
(hulls and end-pieces)



EPZ contracted COVRA to:

- design, build and operate HABOG facility
- receive the high level residues from reprocessing
- take title of residues and store these for 100 years
- manage the funds for ultimate geological disposal



- Legal framework for planning of plant decommissioning
- Project Schedule and Cost Estimate re-issued every 5 years
- Minister to concur with Project Schedule and Cost Estimate
- EPZ dotates annually into fund managed by dedicated legal entity, fund should grow to 0,5 B€ by 2034
- Cost of decommissioning included in Borssele production cost.

- EPZ takes its responsibility for the radioactive waste arisings from its industrial activities
- EPZ cares for the impact of its fuel supply lines in foreign places
- High level waste solutions under contract with COVRA
- Decommissioning funds managed safely
- Good co-operation with COVRA





Presentation No. 18

1

The COVRA logo, with "COVRA" in a bold, dark blue sans-serif font and a small "NV" in a lighter blue font to its right. The logo is positioned over a background of several overlapping light blue ovals.The background image shows a large, rust-colored metal structure, likely a containment vessel, with a large green 'H' logo painted on its side. A person in a blue hard hat and dark clothing is walking in the foreground at the bottom left, providing a sense of scale. The sky is clear and blue.

# ROLE & RESPONSIBILITIES COVRA

Ewoud Verhoef

The COVRA logo, with "COVRA" in a bold, white sans-serif font and a small "NV" in a lighter blue font to its right.

CAST Workshop 5-6 October 2016, Nieuwdorp



# ROLE

**provide continuous  
care for radioactive  
waste in the  
Netherlands to  
protect man and the  
environment**

**1982 | state-owned enterprise |  
20 million turn-over | 60 people  
Waste-ownership**



**COVRA<sub>NV</sub>**

3

# RESPONSIBILITIES

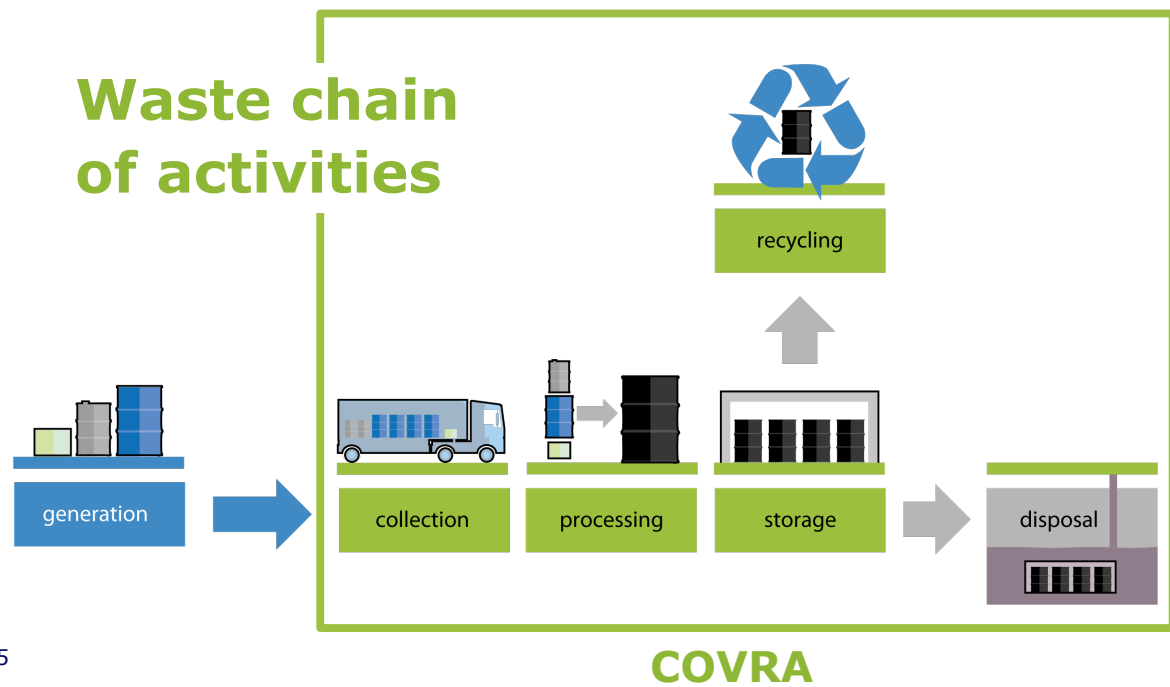
- **OPERATIONAL:**

- collecting, processing and storing all types of radioactive waste;
- coordinating research on geological disposal;

**COVRA<sub>NV</sub>**

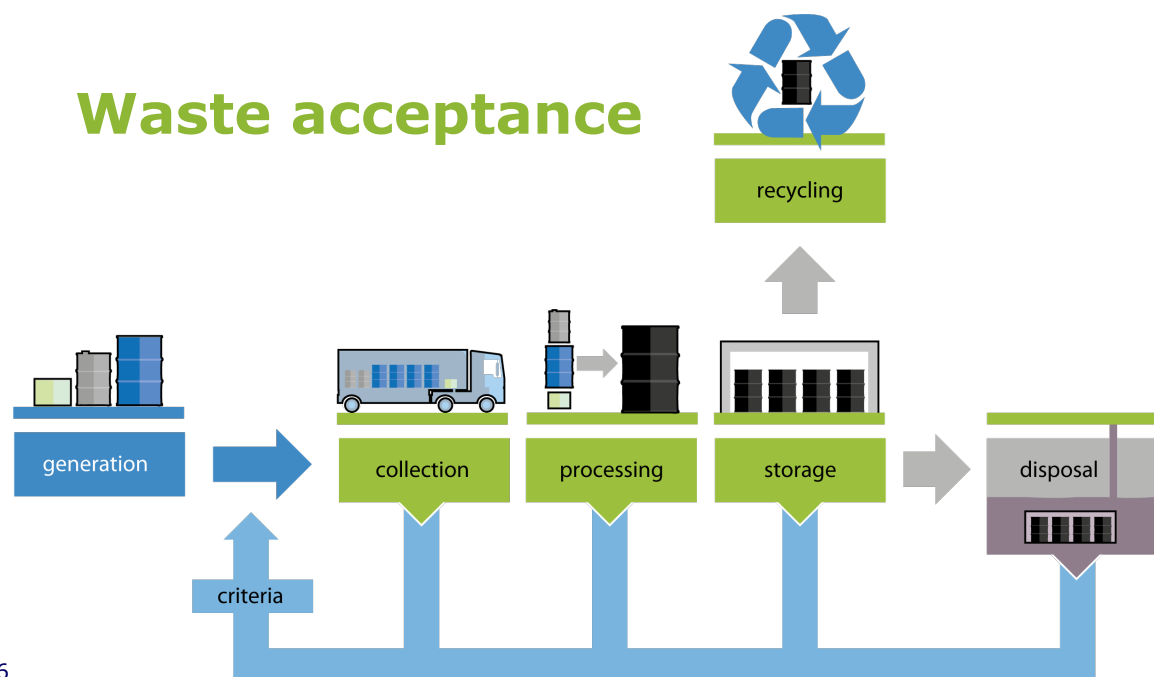
4

# OPERATIONAL



5

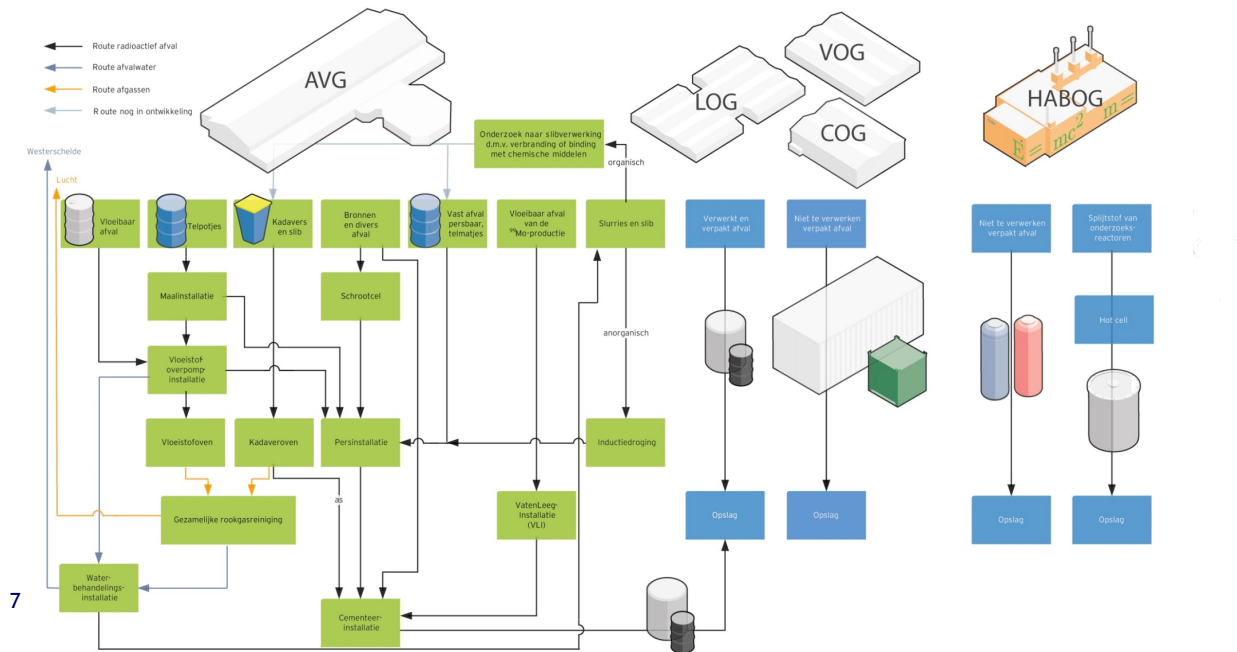
# OPERATIONAL



6

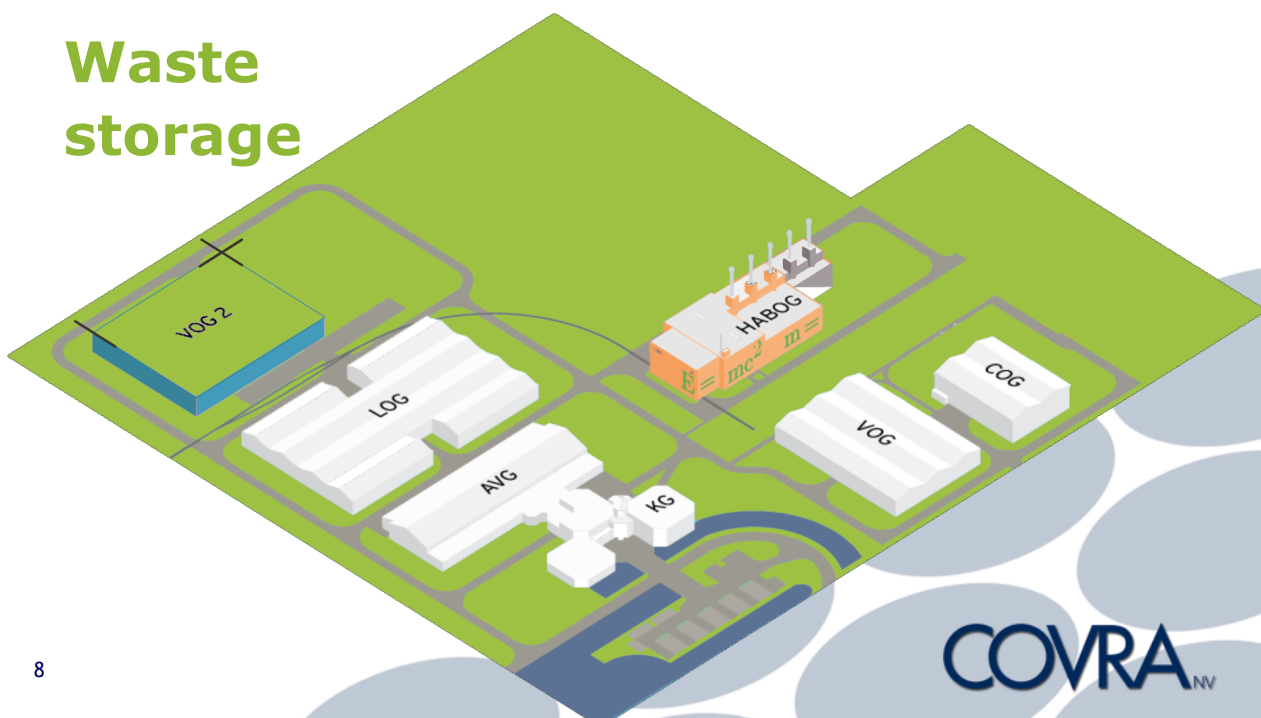
# OPERATIONAL

## Waste processing

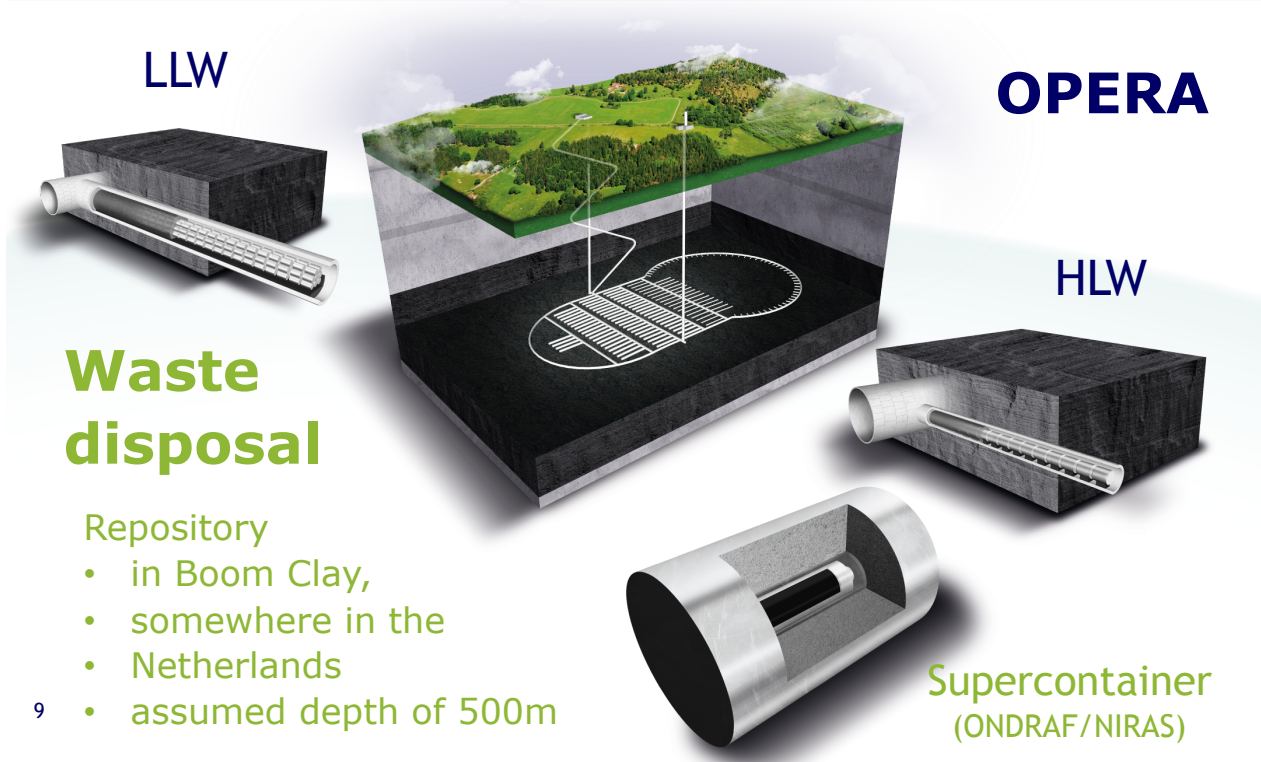


# OPERATIONAL

## Waste storage



# OPERATIONAL



# RESPONSIBILITIES

- **OPERATIONAL:**

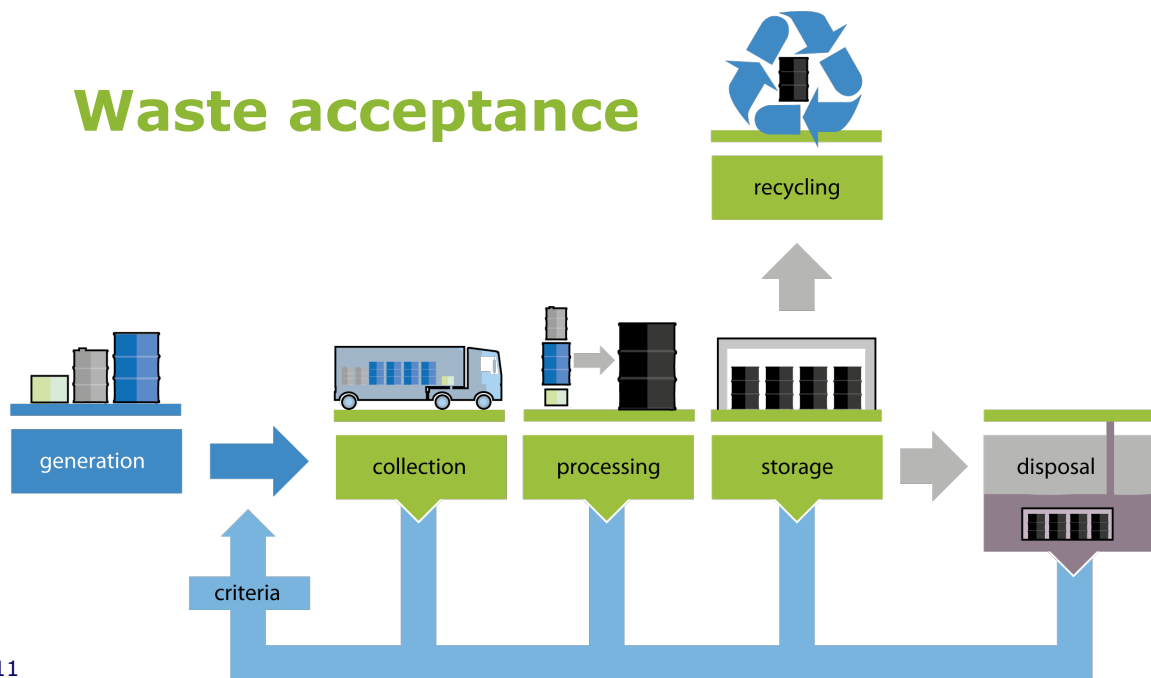
- collecting, processing and storing all types of radioactive waste;
- coordinating research on geological disposal;

- **FINANCIAL:**

- reserving and managing the financial means for long-term above ground storage;
- preparing geological disposal, including reserving and managing the financial means for execution;

# MANAGEMENT

## Waste acceptance



11

# RESPONSIBILITIES

## • OPERATIONAL:

- collecting, processing and storing all types of radioactive waste;
- coordinating research on geological disposal;

## • FINANCIAL:

- reserving and managing the financial means for long-term above ground storage;
- preparing geological disposal, including reserving and managing the financial means for execution;

## • KNOWLEDGE:

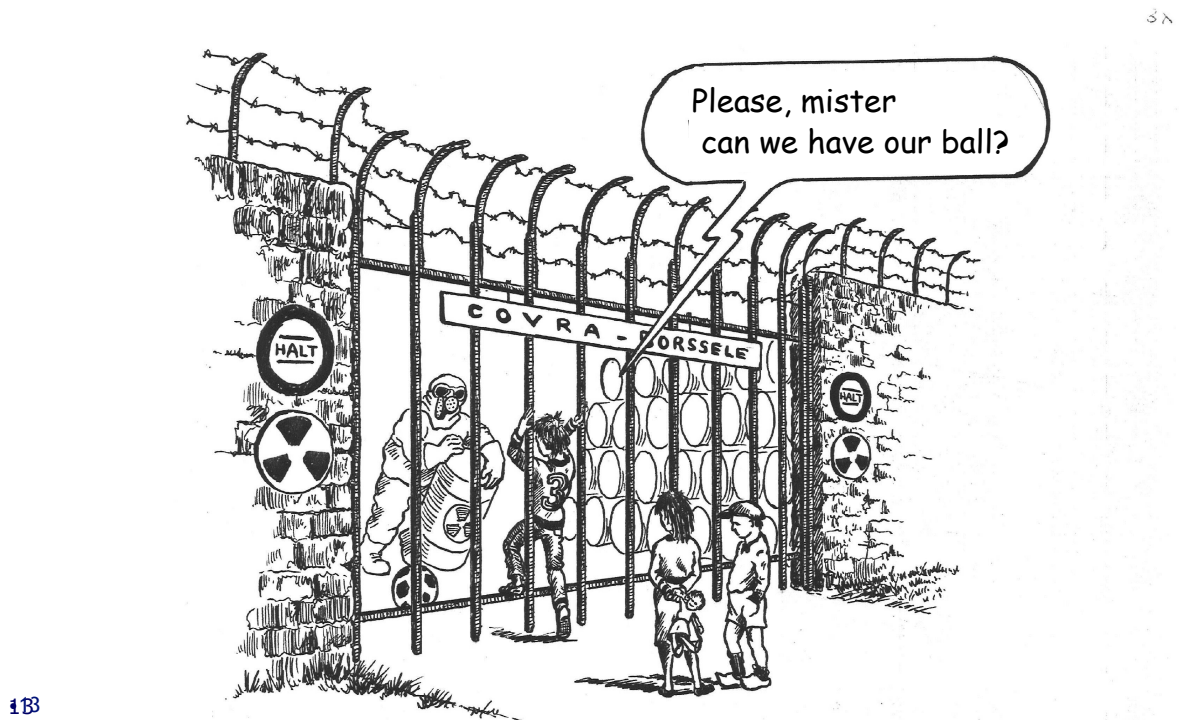
- serving as a knowledge centre for government, industry and society, including educational aspects; and
- actively participating in various international settings in the field of radioactive waste management.

12

**Communicate!**

**COVRA<sub>NV</sub>**

# COMMUNICATION

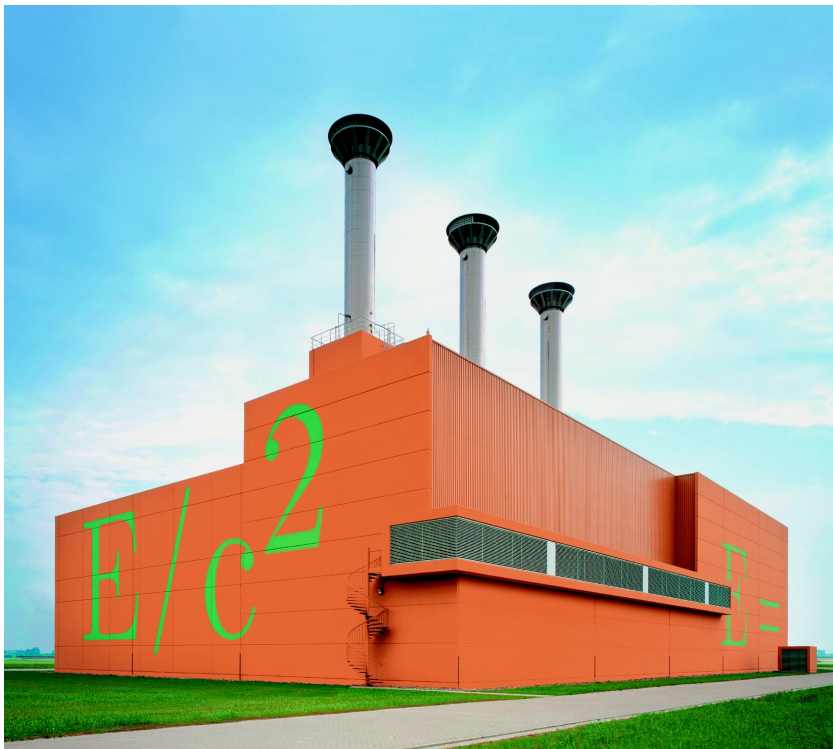


# COMMUNICATION

- **HABOG is a work of art:**  
**METAMORPHOSIS**  
*(by William Verstraeten)*
- **show at the outside what is happening inside**
- **when inside, look outside through the walls of 1.70 m concrete**

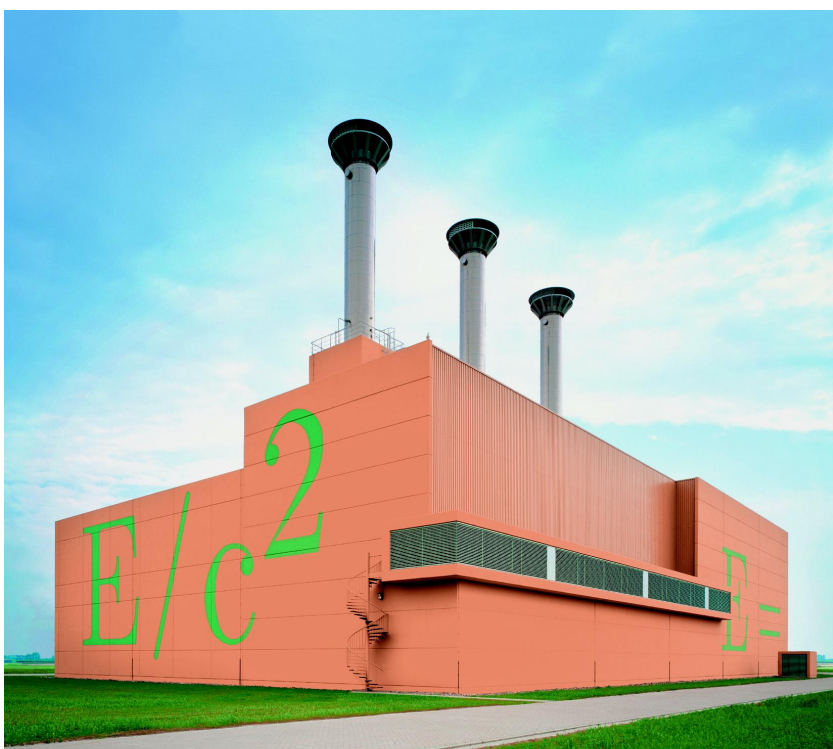


2003



COVRA<sub>NV</sub>

2023



COVRA<sub>NV</sub>

2043



COVRA<sub>NV</sub>

2063



COVRA<sub>NV</sub>



2083



COVRA<sub>NV</sub>

2103

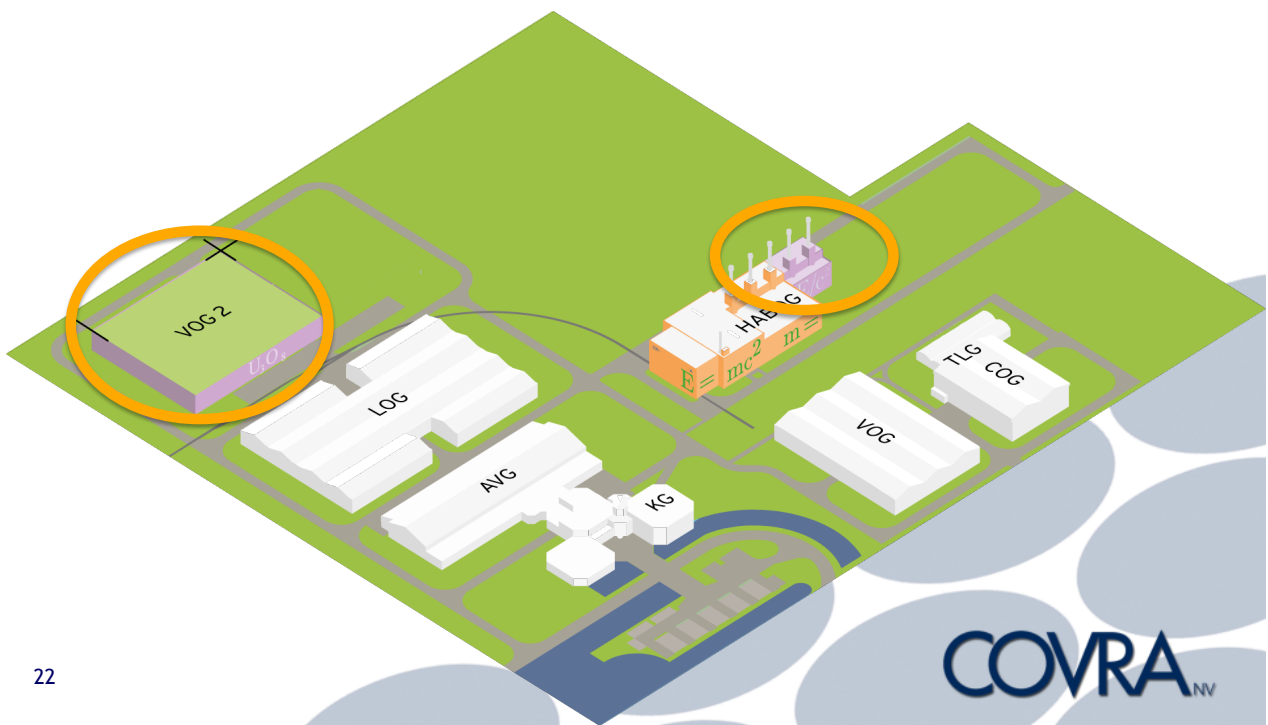


COVRA<sub>NV</sub>

SAFE = BEATIFUL



NEW BUILD



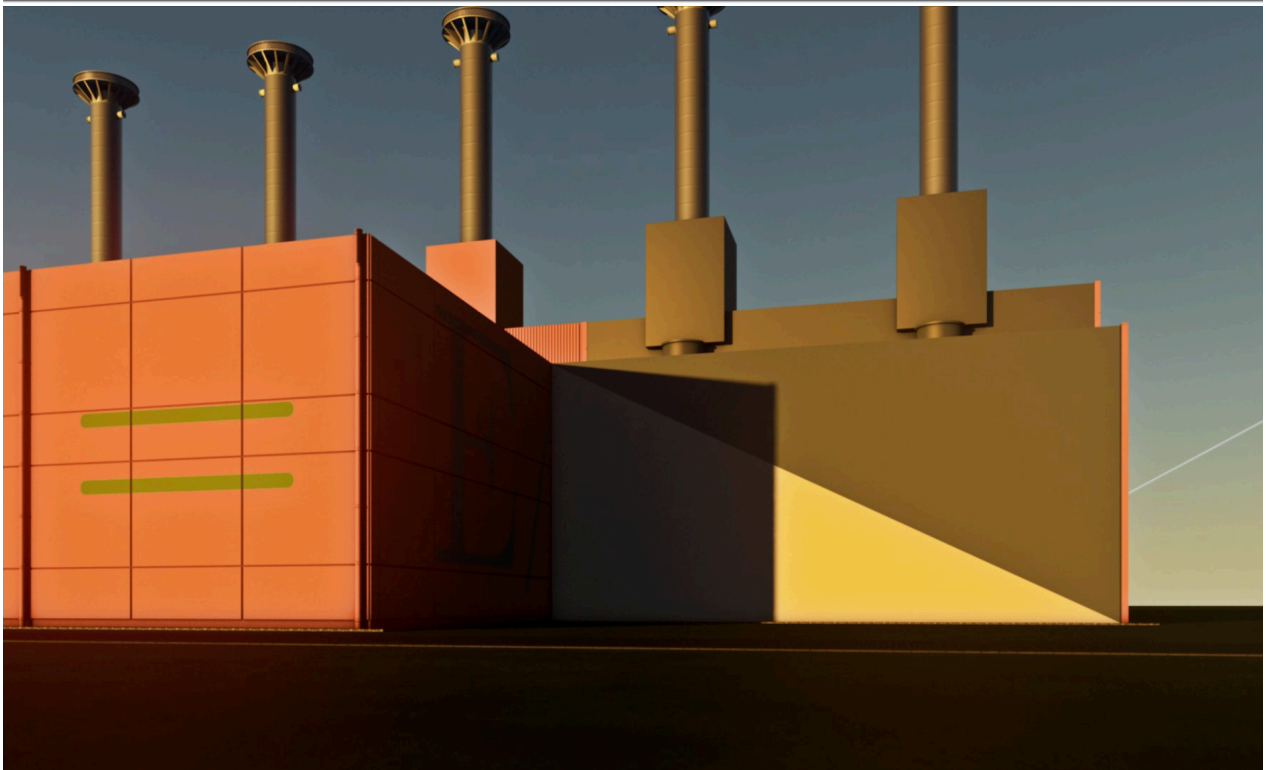
# EXTENSION HABOG



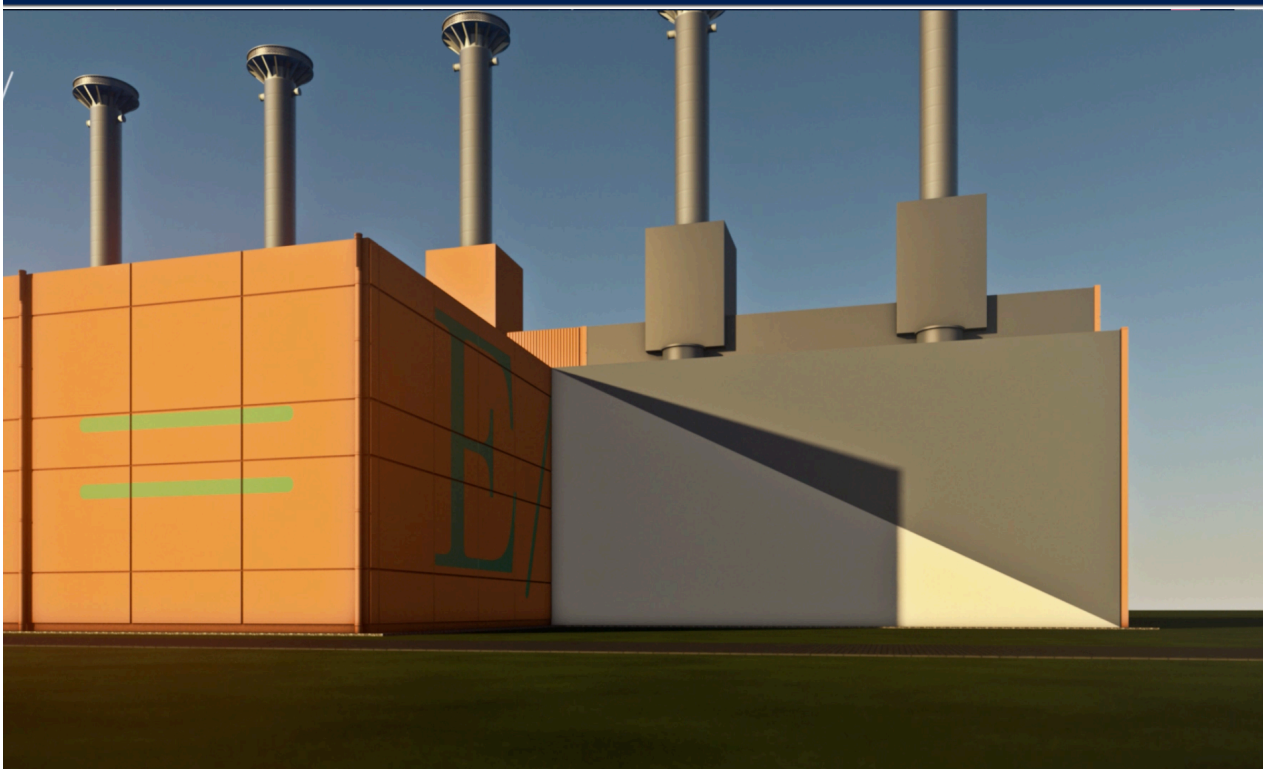
# EXTENSION HABOG



# EXTENSION HABOG

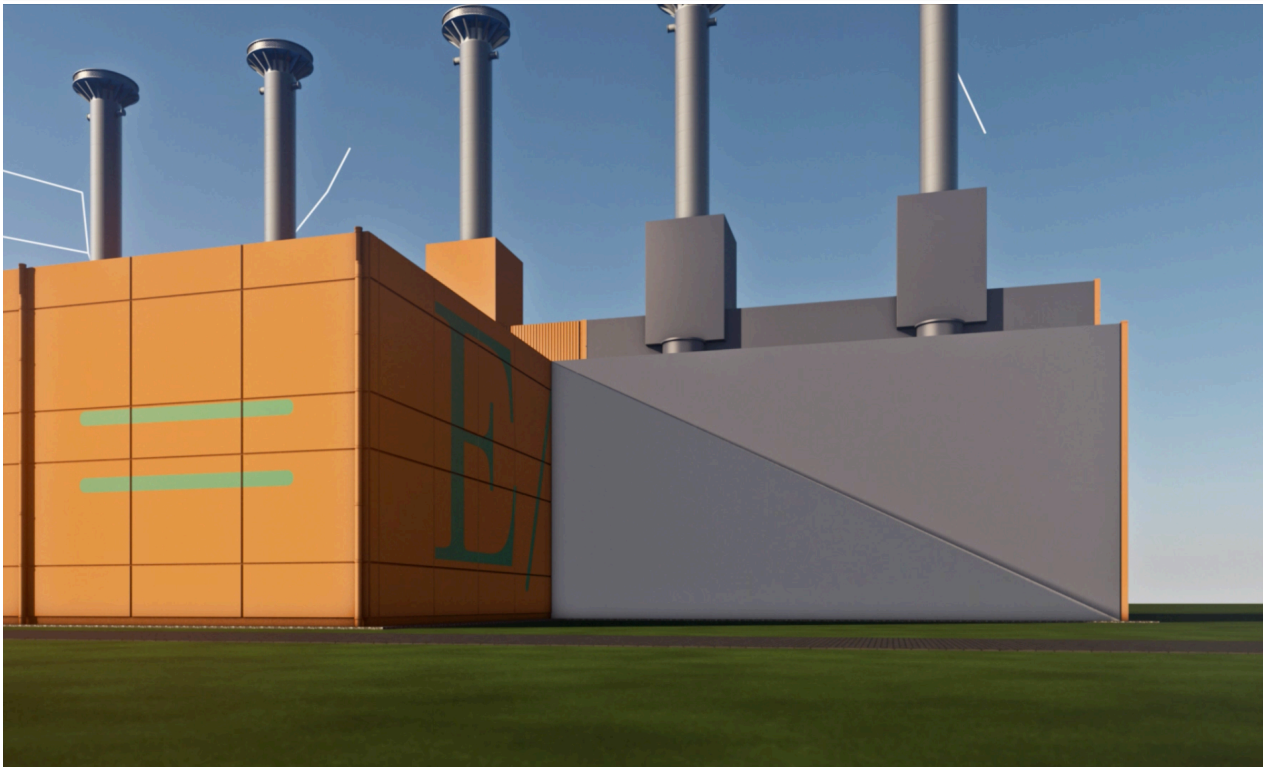


# EXTENSION HABOG

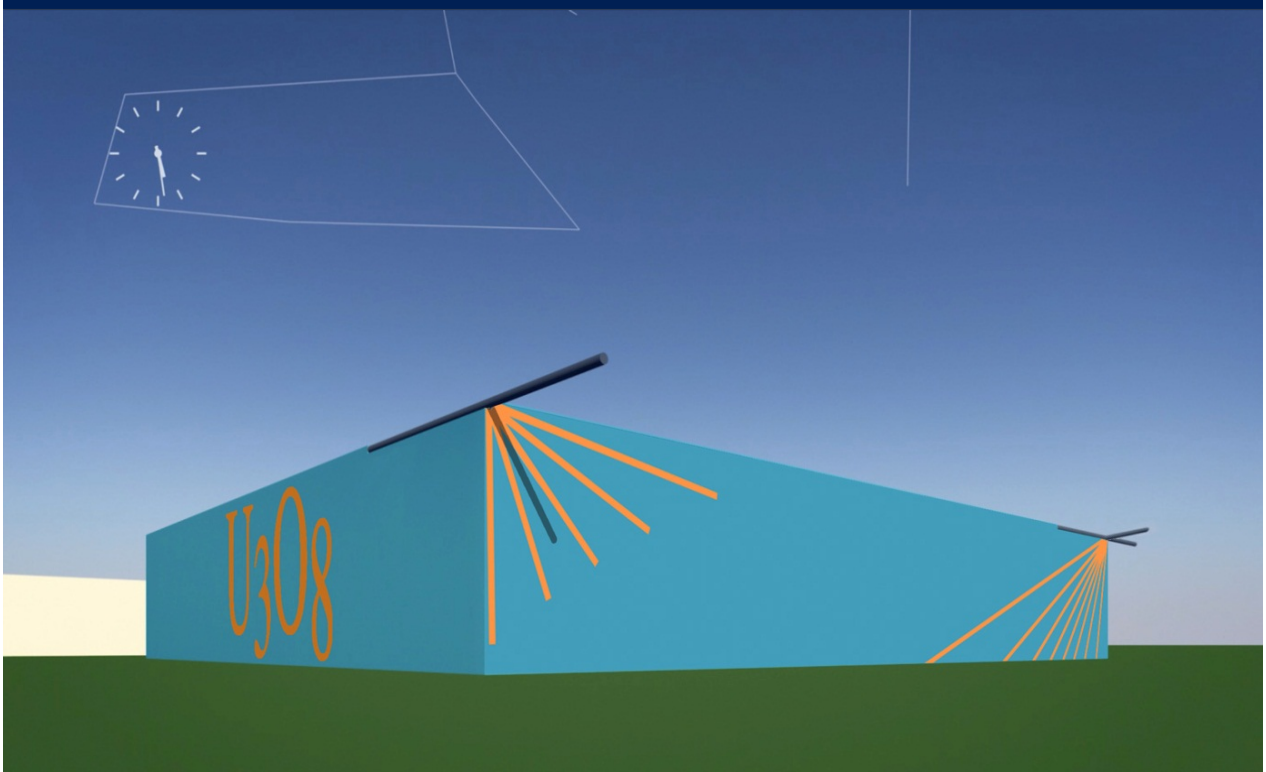




# EXTENSION HABOG



# VOG-2







Presentation No. 19

1

# **CARbon-14 Source Term CAST**

## **An example of the PA of graphite**

Dalia Grigaliuniene, Asta Narkuniene, Povilas Poskas  
Lithuanian Energy Institute

6<sup>th</sup> October, 2016



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.





# An example of the PA of graphite



## Lithuanian case

- Description of waste (package) intended to be disposed.
- Conceptual model:
  - Degradation of waste matrix / package, release mechanism carbon-14;
  - Assumptions made to implement conceptual model;
  - Substantiation of assumptions;
- Outcomes of calculations.
- (Expected) contribution from CAST.


3



An example of the PA of graphite – Lithuanian case

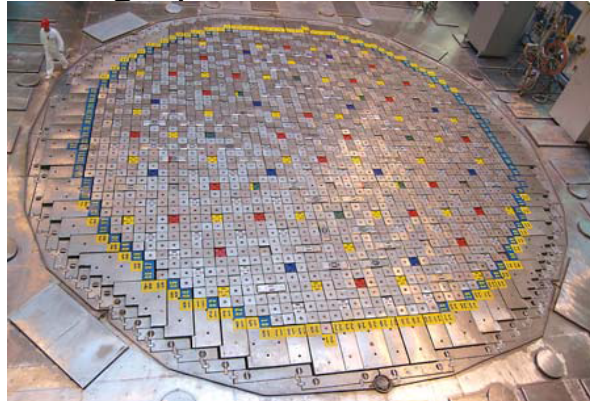
## Description of waste (package) intended to be disposed



- The main C-14 source is irradiated graphite from Lithuanian nuclear power plant at Ignalina site.
- 
- Two units of Ignalina NPP were equipped with RBMK type reactors containing graphite as moderator and reflector leading to app. **3800 tones** of graphite blocks and sleeves.

4

- The major contamination sources for graphite:
  - neutron activation of raw graphite material – carbon;
  - impurities activation in the graphite matrix;
  - impurities activation within the pore space;
  - activation of reactor coolant gases;
  - technological incidents during the reactor operation.

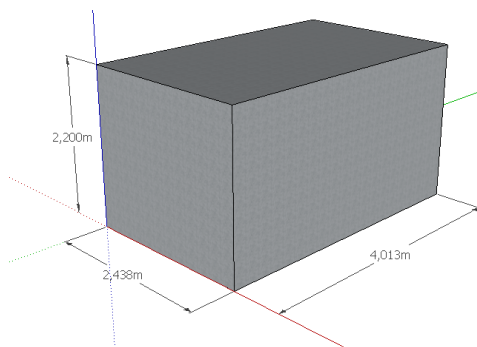


5

- Type of graphite:
  - Blocks – GR-280 (approx. 95 % of graphite waste);
  - Sleeves – GR-2-125.
- Assumptions on activity and substantiation:
  - Based on modelling;
  - Maximal initial impurity content (N – 70 ppm) in all – open and – closed pores;
  - Activity of blocks is higher than of graphite sleeves – all graphite is of grade GR-280.
- Specific activity –  $9.9 \times 10^5$  Bq/g.

6

- No final decision on long-lived LILW (and graphite) disposal.
- After dismantling graphite will be stored in steel containers at interim storage facility.
- No decision on graphite waste treatment.

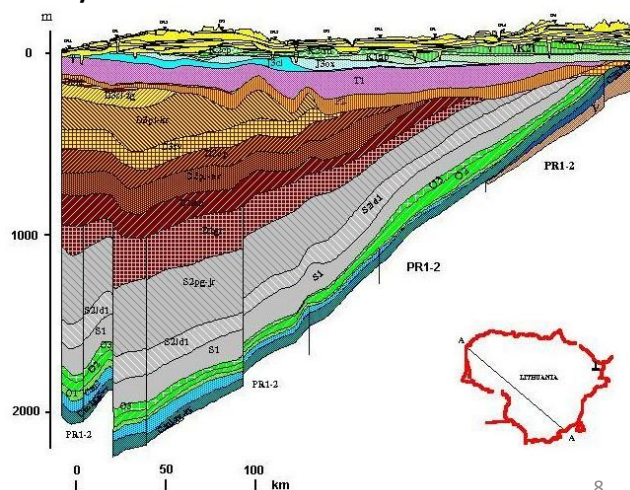


- Assumptions on waste packages:
  - Alternative 1 – 4 m long box stainless steel container (based on Nirex concept);
  - Alternative 2 – the same container, but with cement based material as encapsulant (thickness of encapsulant– 0.15 m).

7

## Geological Repository Concept

- Assumptions:
  - Graphite is disposed of at the same deep geological repository as SNF but at a certain distance;
  - Hypothetical repository in crystalline rocks.
  - Disposal at 500–600 m depth with overlying sedimentary rock cover;
  - Sedimentary rocks: sandstone, limestone, clay, Quaternary deposits (forming aquifers and aquitards).



„PR-1“ represents the crystalline basement

8

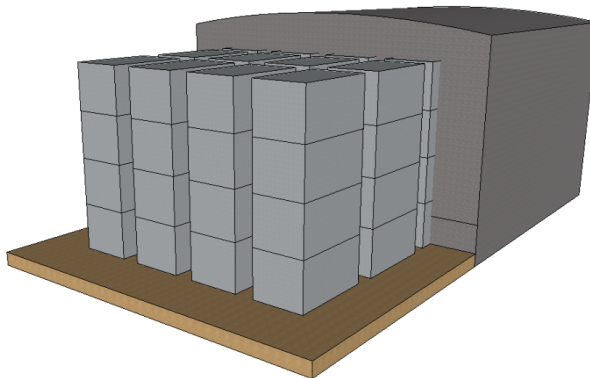


## An example of the PA of graphite – Lithuanian case

Description of waste (package) intended to be disposed (cont.)



- Four containers in horizontal and vertical directions could be stacked;
- Disposal tunnel of 16 m × 16 m (width × height);



- Cementitious backfill (NRVB-Nirex Reference Vault Backfill).

9

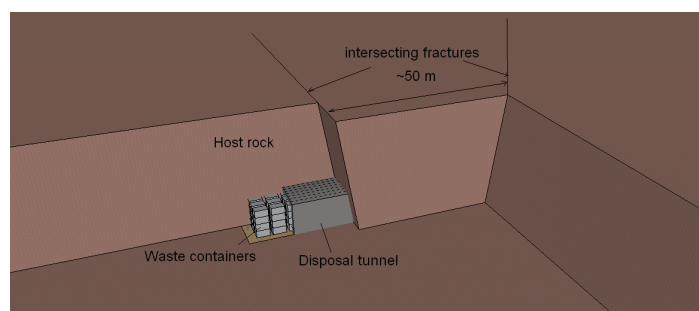


## An example of the PA of graphite – Lithuanian case



# Conceptual model

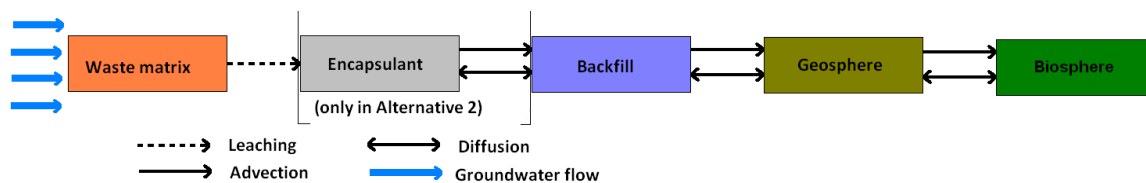
- One groundwater pathway scenario:
  - $^{14}\text{C}$  release from the graphite matrix upon contact with water;
  - Transport through engineered barriers up to the fracture intersecting the disposal tunnel;
  - Transport through the sedimentary cover upwards to the groundwater discharge area.



10

### Conceptual model (cont.)

- Two alternatives analysed:
  - Alternative 1 – stainless steel container without encapsulant;
  - Alternative 2 – stainless steel container, with encapsulant.
- Processes considered:
  - Leaching;
  - Advective/diffusive transport;
  - Solubility;
  - Sorption;
  - Radioactive decay.



11

### Conceptual model (cont.)

- Degradation of waste matrix/package:
  - No credit for the metallic containers (steel containers are corroded soon after repository closure);
  - No changes in encapsulant properties.

12



- C-14 release mechanism:
  - C-14 release from i-graphite at low temperatures in aqueous solutions is not well known at present;
  - Experimental results indicate C-14 release at an initially higher release rate (C-14 onto the graphite surfaces and pores) followed by a decreased release rate in the long-term (C-14 incorporated in the crystal lattice);
  - This trend was observed also with RMBK-1500 irradiated graphite;
  - Different for treated and non-treated graphite;
  - Increase in C-14 release to solution with increase in geometric surface area of the graphite (powder/block).

13



- Assumptions on C-14 release rate:
  - empirical approach – release rate is proportional to the activity of C-14 in the i-graphite;
  - 7 different release rates are assumed in modelling.

Variant	Release rate, 1/yr	Comment
Variant A	$1.83 \cdot 10^{-5}$	Experimental value
Variant B1	<10 yr - 0.1 >10 yr - 0.01	Non-treated waste
Variant B2	<10 yr - 0.1 >10 yr - 0.001	Non-treated waste
Variant C1	0.1	Treated waste, powder
Variant C2	0.01	Treated waste, granular
Variant C3	0.001	Treated waste, block
Variant D	instant release	

14



- **C-14 speciation, sorption, solubility:**
  - C-14 can be released as organic and inorganic compounds (mainly  $\text{CO}_2$  and  $\text{CH}_4$ ) with different retention;
  - Depends on the nature, history and irradiation conditions, geological environment;
  - No ratio organic/inorganic compounds is available;
  - Two bounding cases regarding sorption in the near field considered (based on literature survey):
    - ✓  $K_d = 0.2 \text{ m}^3/\text{kg}$  and
    - ✓  $K_d = 0 \text{ m}^3/\text{kg}$ .
  - Two cases regarding solubility limit considered (based on literature survey):
    - ✓ unlimited solubility and
    - ✓ solubility limit of  $0.01 \text{ moles}/\text{m}^3$ .

15

- **Calculation cases for Alternative 1 (no encapsulant):**

Parameter	Case 1	Case 2	Case 3
K <sub>d</sub> , m <sup>3</sup> /kg	0	0	0.2
Solubility limit, moles/m <sup>3</sup>	0	0.01	0.01
Release rate, 1/yr	7 variants:		
	Variant	Release rate, 1/yr	
	Variant A	1.83·10 <sup>-5</sup>	
	Variant B1	<10 yr - 0.1 >10 yr - 0.01	
	Variant B2	<10 yr - 0.1 >10 yr - 0.001	
	Variant C1	0.1	
	Variant C2	0.01	
	Variant C3	0.001	
	Variant D	instant release	

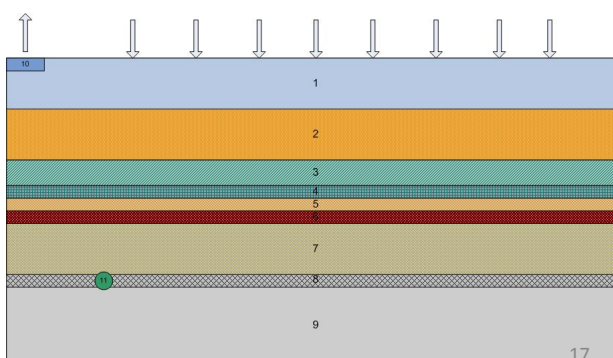
- **Alternative 2 (with encapsulant): the same calculation cases as for Alternative 1**

16

## Conceptual model (cont.)

- Other assumptions:
  - Repository saturation immediately after its closure;
  - No changes in material properties;
  - Geological and hydro-geological characteristics remain constant during the analysed period of time;
  - No sorption in geosphere;
  - Hydrogeological data of sedimentary cover are based on data for southern Lithuania.

Layer no.	Depth (m)	k (m/s)	n (m <sup>3</sup> /m <sup>3</sup> )	Description
1	0–100	$1.2 \cdot 10^{-5}$	0.25	glacial loam
2	100–200	$2.3 \cdot 10^{-5}$	0.35	sand
3	200–250	$5.8 \cdot 10^{-5}$	0.5	anhydrite
4	250–270	$1 \cdot 10^{-6}$	0.5	limestone
5	270–290	$5.8 \cdot 10^{-5}$	0.05	sandstone
6	290–310	$1.16 \cdot 10^{-10}$	0.13	clay
7	310–410	$6 \cdot 10^{-6}$	0.05	sandstone
8	410–420	$1.16 \cdot 10^{-7}$	0.01	weathered crystalline rocks
9	420–520	$1.16 \cdot 10^{-13}$	0.0038	monolithic crystalline rocks



## Conceptual model (cont.)

# Model implementation

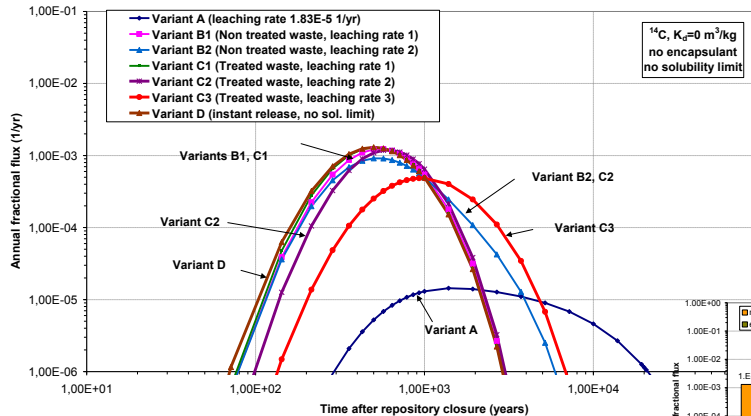
- Near field:
  - Computer tool AMBER;
  - Compartmental approach;
  - Transfer between compartments are described by transfer coefficients.
- Far field:
  - Computer tool Petrasim (TOUGH2).
- Output:
  - Fractional flux to geosphere and to biosphere (1/yr).



# Outcomes of calculations

## Case 1: $K_d=0 \text{ m}^3/\text{kg}$ , no solubility limitation

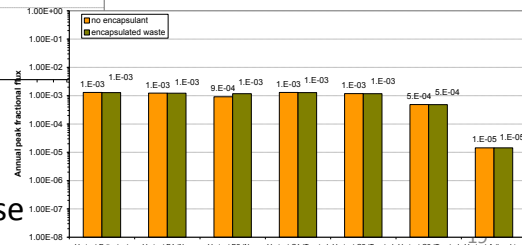
- Alternative 1 (no encapsulant)



Differences between leaching rates of the order of  $10^{-1} - 10^{-2} \text{ 1/yr}$  and instantly released inventory do not result in significant differences of peak fractional flux to the geosphere.

$^{14}\text{C}$ ,  $K_d=0 \text{ m}^3/\text{kg}$ , no solubility limitation

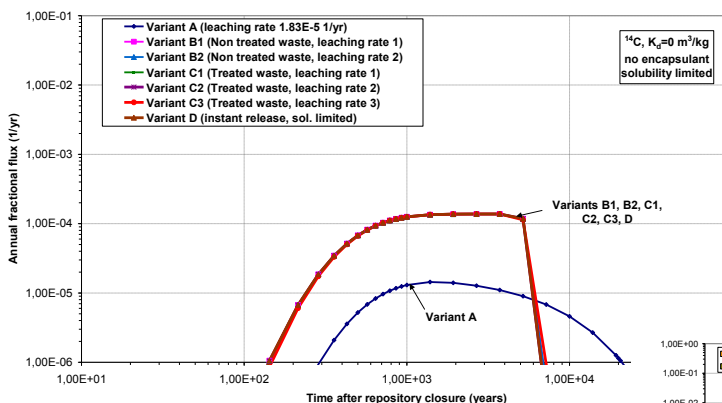
- Alternative 2 (with encapsulant):
  - encapsulation does not result in decrease or delay of C-14 peak flux to geosphere.



## Outcomes of calculations (cont.)

## Case 2: $K_d=0 \text{ m}^3/\text{kg}$ , solubility limit=0.01 moles/ $\text{m}^3$

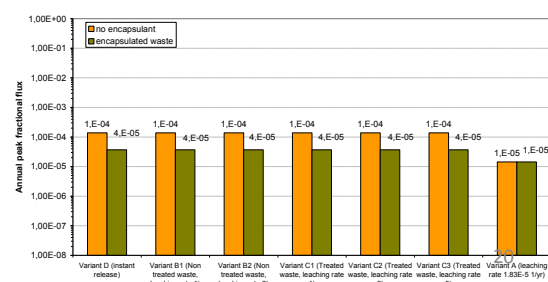
- Alternative 1 (no encapsulant)



If the assumed  $^{14}\text{C}$  release rate from waste is  $10^{-1} - 10^{-3}$  or instant release is the case, the release is controlled by solubility limitation.

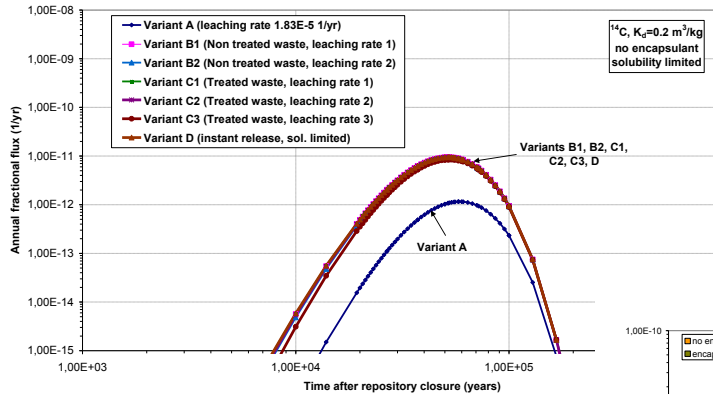
$^{14}\text{C}$ ,  $K_d=0 \text{ m}^3/\text{kg}$ , solubility limited (0.01 moles/ $\text{m}^3$ )

- Alternative 2 (with encapsulant):
  - the peak flux is decreases by a factor of 2.5 due to contribution of encapsulant as additional barrier.



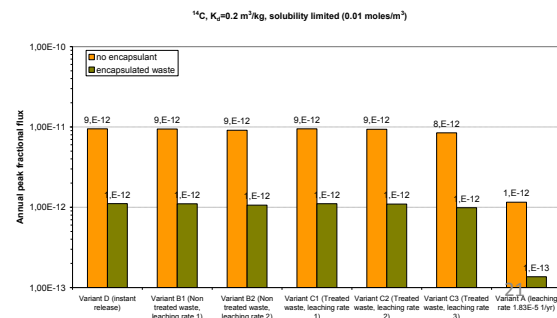
### Case 3: $K_d=0.2 \text{ m}^3/\text{kg}$ , solubility limit= $0.01 \text{ moles}/\text{m}^3$

- Alternative 1 (no encapsulant)



Flux is not limited by the solubility limit.

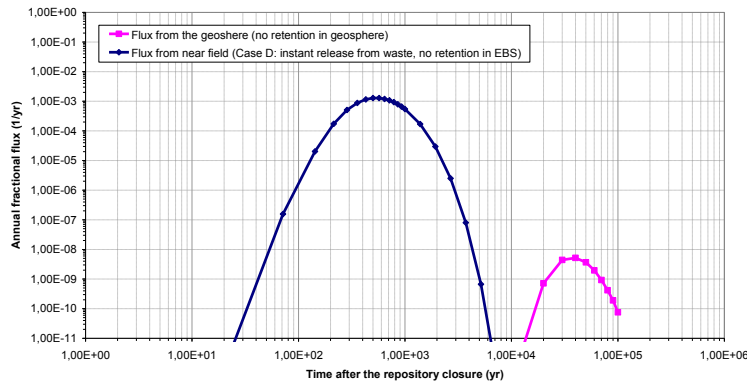
- Alternative 2 (with encapsulant):
  - the peak flux is decreased app. by one order of magnitude due to contribution of encapsulant as additional chemical barrier.



### Comparison of maximal fractional fluxes to geosphere

Parameter	Case 1	Case 2	Case 3
$K_d$ , $\text{m}^3/\text{kg}$	0	0	0.2
Solubility limit, moles/ $\text{m}^3$	0	0.01	0.01
Maximal fractional flux to geosphere, 1/yr	Alternative 1 (no encapsulant)		
	1E-03	1E-04	1E-11
	Alternative 2 (with encapsulant)		
	1E-03	5E-05	1E-12

## Transport through natural barriers



Alternative 1 (no encapsulant);  
Case 1 (no sorption, no solubility limit);  
Variant D (instant release)

- Significant impact of overlying system of aquitards and aquifers (sedimentary rocks)

23

## Summary of outcomes

- The peak fractional flux from the near field would vary within app. 1-2 orders of magnitude for the assumed different leaching rates (difference in the leaching rate was within app. 5 orders of magnitude).
- Encapsulation in a cement based material could give benefit to the C-14 flux reduction to app. one order of magnitude
- The natural barriers contribute to the delay and decrease of dissolved C-14 flux from the hypothetical repository with RBMK-1500 irradiated graphite by 5 orders of magnitude.

24



## (Expected) contribution from CAST

- **WP5** – to increase understanding of the factors determining release of  $^{14}\text{C}$  from irradiated graphite (i-graphite) under geological disposal conditions.
- **Defined tasks:**
  - Review of CARBOWASTE and other relevant R&D activities to establish the current understanding of inventory and release of  $^{14}\text{C}$  from i-graphites;
  - Characterisation of the  $^{14}\text{C}$  inventory;
  - Measurement of release of  $^{14}\text{C}$ ;
  - Consideration of new waste forms and  $^{14}\text{C}$  decontamination techniques.

25



## (Expected) contribution from CAST (cont.)

### **Conclusions from the review of public available results on $^{14}\text{C}$ compounds released from i-graphite in alkaline environment** (performed by INR, Romania)

- Only a very small fraction of the C-14 is released (less than 1%), which can be correlated with the C-14 activated from the nitrogen adsorbed on the surface of the graphite;
- Both organic and inorganic C-14 species are released in alkaline solutions, their amounts depending on the irradiation history and graphite properties;
- C-14 amount released in solution seems to be larger than C-14 amount released in gas phase;
- $\text{CO}_2$ /carbonate dominates the C-14 release in solution, while CO and  $\text{CH}_4$  are considered the major compounds released in gas phase;
- C-14 leaching rate decreases in time with 1-2 orders of magnitude, trending to establish a very slow release rate.

26



### **Conclusions from the review of C-14 leaching from French i-graphite** (performed by EDF/ANDRA, France):

- C-14 leaching rate is very slow for the stack graphite and faster leaching rate was observed for the sleeve graphite;
- In most of cases, a quasi-steady state leaching rate appears to be achieved after the elapse of around 100 to 200 days;
- release rate depends on the shape of the samples (block/powder);
- For the experiments reviewed, the nature of the leaching liquid (deionised or lime or soda water) does not evidence any clear impact on the C-14 leaching behaviour.

27



### **Conclusions from the review of C-14 release from Oldbury Graphite** (performed by RWM, UK)

- Under baseline conditions (anoxic, under pH 13 solution, ambient temperature) about 0.07% of the C-14 inventory was released into solution in one year.
- For powdered sample the release was higher by ~65% than from single piece.
- About 1% of the released C-14 was released to the gas phase.
- An initial phase of rapid C-14 release was observed (in ~28 days), which was followed by a longer term phase of slower release.
- Gaseous C-14 was predominantly in the form of hydrocarbons and other volatile organic compounds and CO.
- The ratio of C-14 in hydrocarbons / organic compounds to CO was approximately 2:1.
- Less than 2% of the gas-phase release under alkaline conditions was in the form of  $^{14}\text{CO}_2$ .

28



### **Conclusions from the review of C-14 inventory in RBMK-1500 i-graphite** (performed by LEI, Lithuania):

- Modelling of Ignalina NPP graphite revealed that there could be four major fractions of C-14:
  - In graphite matrix;
  - Associated with activation of N impurities incorporated in the virgin graphite matrix;
  - Associated with N adsorption from cooling gases;
  - Associated with N penetration from cooling gases into the graphite pores.
- The best agreement between modelled and measured activity was for initial N impurity of 13-15 ppm.

29



### **Main findings from the review of C-14 inventory and release from i-graphite to be used for the updated PA**

- Updating inventory:
  - Reducing conservatism based on the most recent investigation results.
- Release rate:
  - Potential for reducing conservatism assuming lower fraction of rapidly releasable C-14 (for non-treated graphite);
  - Very slow release of C-14 was measured (e.g. 0.07% of the C-14 inventory was in one year from Oldbury graphite) – potential for assumptions of more realistic release rate values.
- Gaseous release:
  - Indication of low importance of gaseous release (only 1% of the released C-14 is released to the gas phase).
- Speciation:
  - Inorganic CO<sub>2</sub>/carbonate dominates the C-14 release in solution, while inorganic CO and organic CH<sub>4</sub> dominate in gas phase; therefore, only small fraction of organic C-14 can be expected.

30



An example of the PA of graphite – Lithuanian case  
(Expected) contribution from CAST (cont.)



## Harmonized leaching procedure

- A harmonized leaching procedure was developed by FZJ (Germany) and is based on:
  - Review of the leaching approach;
  - Critical discussions (with input from CIEMAT and NDA/RWM);
  - Previous experience of leaching tests at FZJ.
- The developed leaching approach can be used by other project participants, to enable better inter-laboratory comparison of results.

31



An example of the PA of graphite – Lithuanian case  
(Expected) contribution from CAST (cont.)



## Experimental work ongoing/foreseen in CAST WP5

- Behaviour of  $^{14}\text{C}$  during reactor operation (IPNL, France).
  - C-13 is used to simulate C-14 release from the matrix carbon.  
Conclusion: Whatever the irradiation regime and even for temperatures as high as  $1000^{\circ}\text{C}$ , the implanted C-14 is not released from the graphite matrix.
- Inventory of C-14 in the i-graphite arising from TRIGA 14MW reactor thermal column, inorganic/organic ratio (INR, Romania).
- Gaseous and liquid phase originated during leaching of i-graphite from Vandellós I NPP; total C-14 and organic/inorganic compounds (CIEMAT, Spain).
- Inventory of C-14 in the i-graphite arising from thermal column of VVR-S Reactor and RAW containing organic and inorganic C-14 compounds (IFIN-HH, Romania).
- A possible new decontamination method based on exfoliation-like approach has been studied (ENEA, Italy).

32



An example of the PA of graphite – Lithuanian case  
(Expected) contribution from CAST (cont.)



### Modelling work ongoing/foreseen in CAST WP5

- New models for numerical estimation of RBMK-1500 graphite activation are under development (LEI, Lithuania):
  - Updating model;
  - Gathering of new information relevant to the RBMK graphite activation issues and application for modelling;
  - Combining of new experimental results and new models for better estimation of C-14 inventory in i-graphite of the whole core of Ignalina NPP Unit 1 reactor.
- Comparison of results from activation calculations of C-14 in i-graphite with experimental measurements for EDF CO<sub>2</sub>-cooled reactors (UNGG) (EDF, France).

33



An example of the PA of graphite – Lithuanian case  
(Expected) contribution from CAST (cont.)



Current PA	From review	From CAST results
<b>Inventory</b>		
Based on modelling with conservative assumptions	Reduced conservatism based on the most recent investigation results.	Reduced conservatism based on updated modelling and measurement results.
<b>Release rate</b>		
7 different release rate	Potential for reducing conservatism assuming lower fraction of rapidly releasable C-14 and lower release rate.	Substantiation for making assumptions about: <ul style="list-style-type: none"><li>· rapidly and slowly releasable fractions;</li><li>· release rate.</li></ul>
<b>Speciation</b>		
Not addressed explicitly	Indication of small fraction of organic compounds in solution.	Substantiation for making assumptions about: <ul style="list-style-type: none"><li>· released compounds;</li><li>· inorganic/organic compounds ratio.</li></ul>
<b>Gaseous release</b>		
Not considered	Indication of low importance of gaseous release.	Substantiation for assumptions about gaseous release (ratio liquid/gaseous releases).





An example of the PA of graphite – Lithuanian case  
(Expected) contribution from CAST (cont.)



- **Released publications:**

- D5.2 Annual Progress Report on WP5 - Year 1;
- D5.3 Report on graphite categories in the RBMK reactor;
- D5.4 Harmonised Leaching methodology;
- D5.5 Current understanding of inventory and release of C-14 from irradiated graphite;
- D5.6 Annual Progress Report on WP5 - Year 2;
- D5.7 C-14 in irradiated graphite from research reactor VVR-S;
- D5.8 C-14 Speciation in solution and gas from French graphite waste.

<http://www.projectcast.eu/publications>

35



An example of the PA of graphite – Lithuanian case



**Thank you for your attention**

36



## Presentation No. 20

# Carbon-14 Source Term

## CAST

Irradiated Zircaloy – The Netherlands

Name: **Jaap Hart**

Organisation: **NRG, Petten**

Date: **6 October 2016**



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.

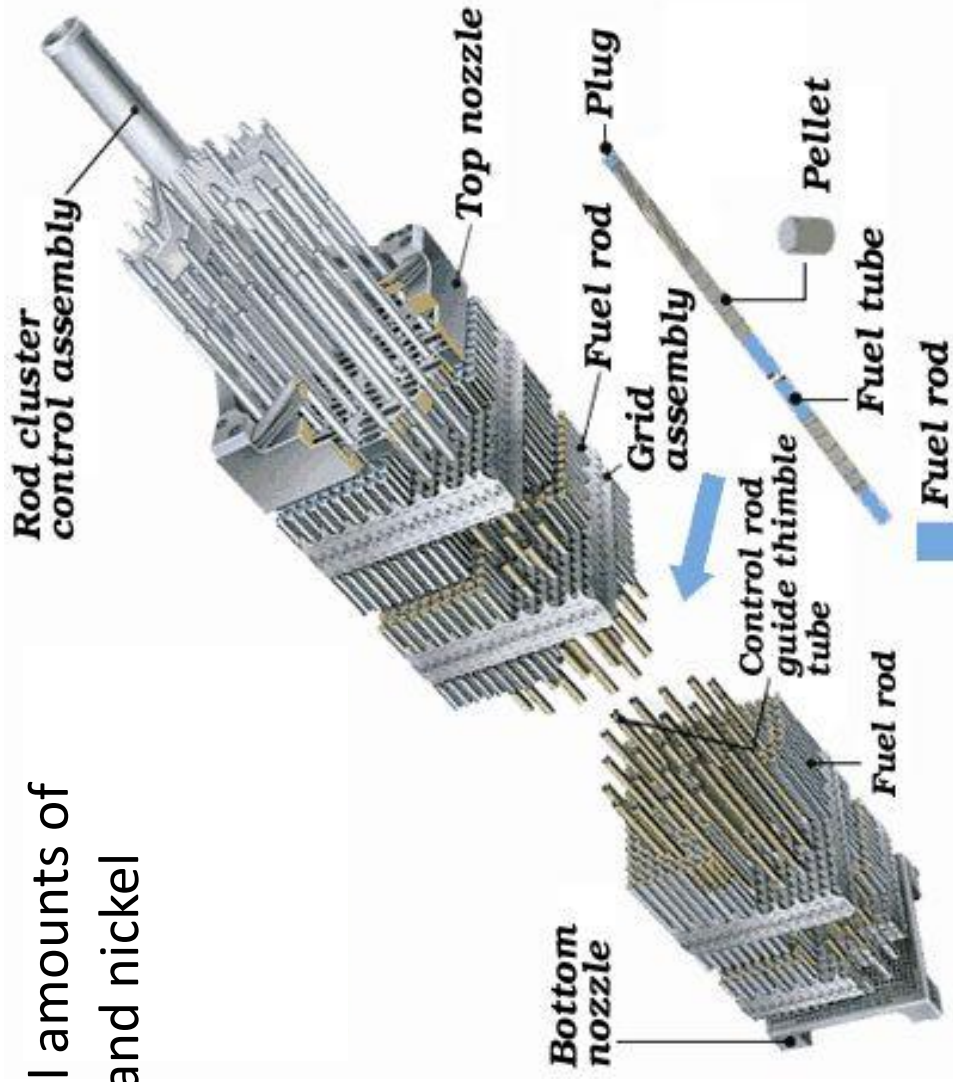
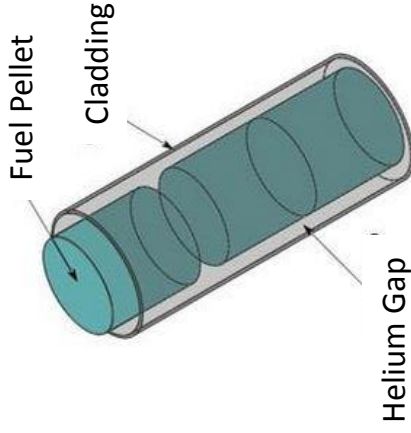
# Contents

- Zircaloy and <sup>14</sup>C
- Dutch disposal concept
- OPERA performance assessment
- CAST performance assessment
- Conclusions and outlook



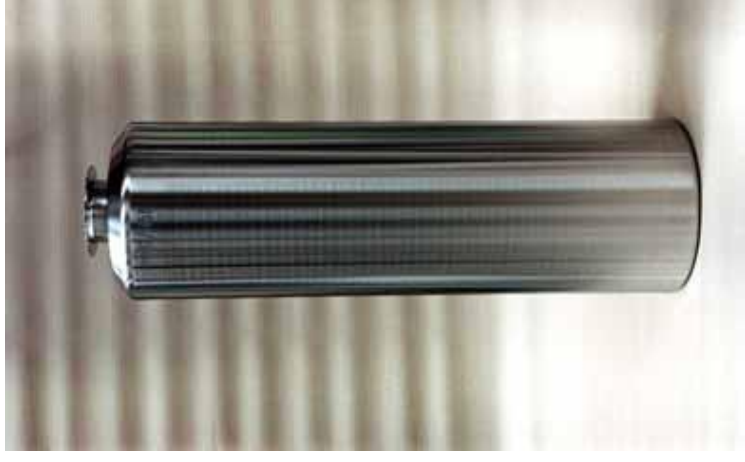
# Zircaloy

- Zircaloy widely used in nuclear industry - **cladding** materials for BWRs and PWRs
- Zirconium alloy, includes small amounts of tin, niobium, iron, chromium and nickel
  - Mechanical strength
  - Corrosion resistance
  - Transparent to neutrons
- Dimensions 0.6-0.7 mm



# Zircaloy

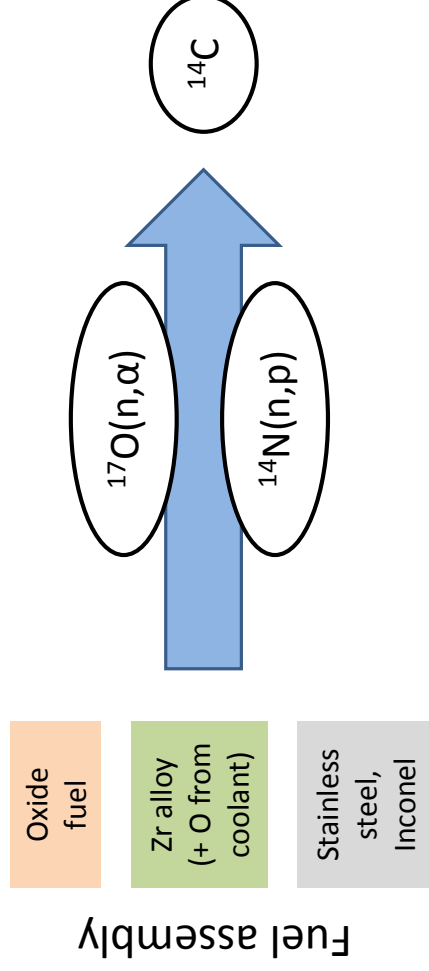
- Zircaloy hulls and end-pieces separated from spent fuel
  - Collected in standard compacted waste containers
    - CSD-C containers
- Colis Standard de Déchets - Compactés



# $^{14}\text{C}$ Production in Zircaloy



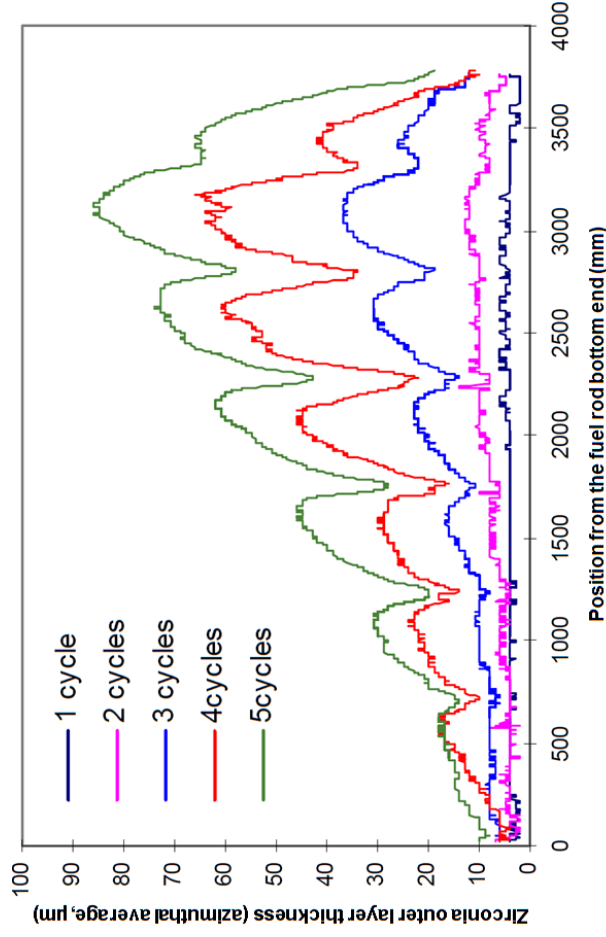
- $^{14}\text{C}$  is generated in light-water reactors by:
  - $^{17}\text{O}(n,\alpha)^{14}\text{C}$  reactions in oxide fuels and reactor coolants
  - $^{14}\text{N}(n,p)^{14}\text{C}$  reactions with nitrogen present as an impurity in fuel assembly components, reactor coolants, and structural hardware



- In **Zirconium** alloy claddings, neutron activation of  $^{14}\text{N}$  impurity is the main source of  $^{14}\text{C}$

# $^{14}\text{C}$ Production in Zircaloy

- For an irradiated fuel of burn-up 45 GWd/ $t_U$  the production of  $^{14}\text{C}$  in zircaloy cladding is about  $30 \pm 10 \text{ kBq.g}^{-1}$
- For PWR fuels of burn-up  $\geq 45 \text{ GWd}/t_U$ , the oxide layer  $\text{ZrO}_2$ , typically  $20\mu\text{m}$  thick for a cladding thickness of  $0.75 \text{ mm}$ , contains  $\leq 20\%$  of the  $^{14}\text{C}$  inventory
- The  $\text{ZrO}_2$  layer thickness depends on many factors such as burn-ups, operating conditions, cladding types

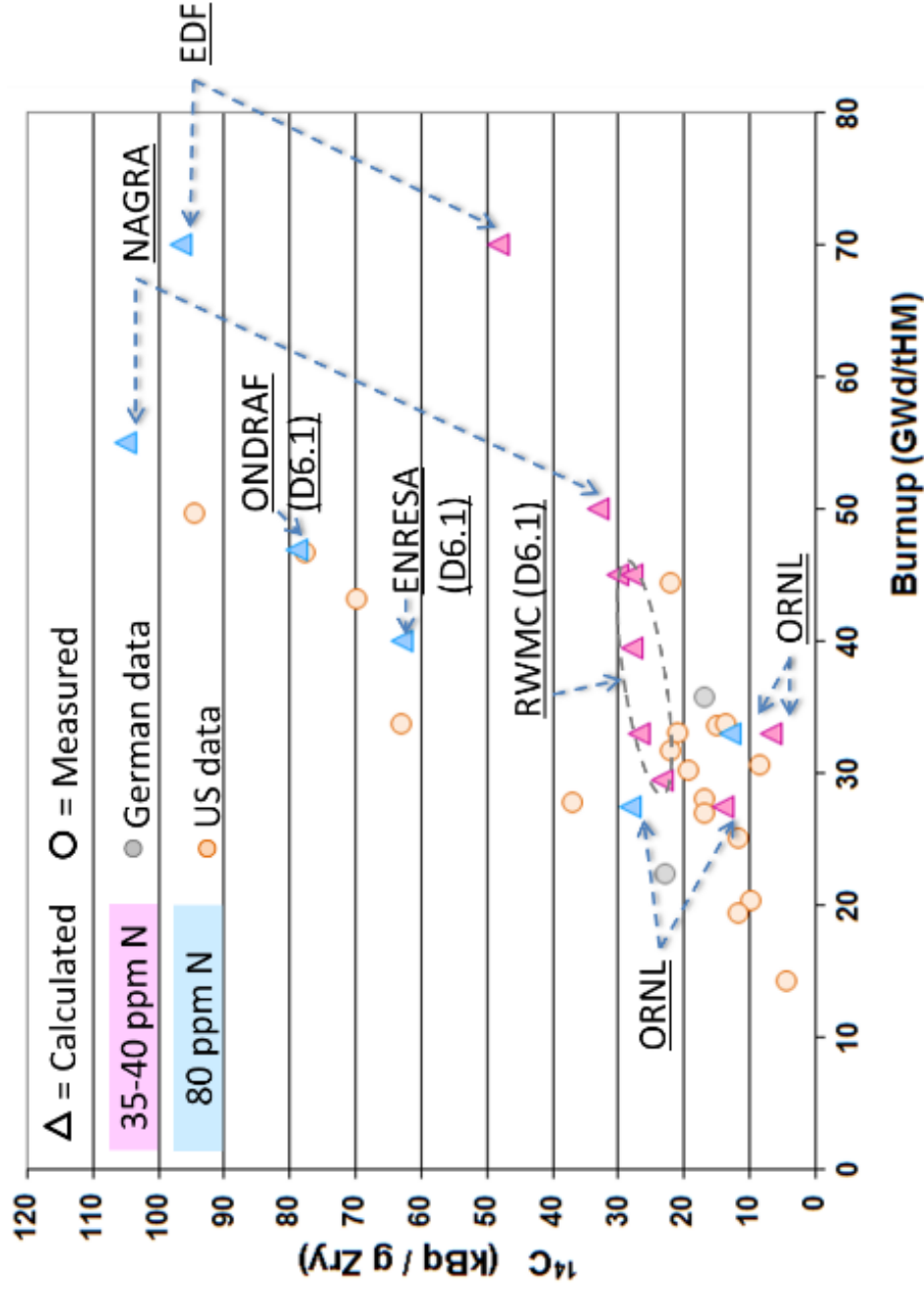




# $^{14}\text{C}$ Inventory in Zircaloy



Dependency of C-14 inventory in Zircaloy on burnup and N impurity (CAST D3.2)



Adapted from (Gras J.M., Report D3.2)

# $^{14}\text{C}$ Release from Zircaloy

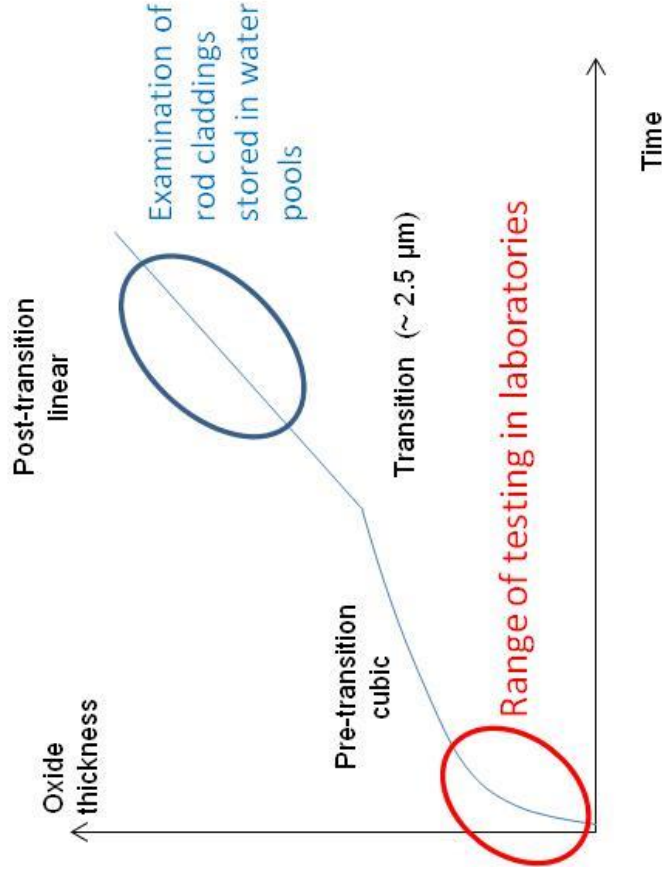


- Investigated in CAST Task 3.3
- Release rates of  $^{14}\text{C}$  inventory from cladding hulls are very low in alkaline or anaerobic waters (repository conditions)
- $^{14}\text{C}$  Release rates seem to be controlled by the **uniform corrosion** of zirconium
- Localized/stress corrosion appears **unlikely** under such conditions
- Previously assumed upper bound corrosion rate:  $20 \text{ nm.yr}^{-1}$  (**excessive**)
- **Recent** experimental findings indicate corrosion rate of 1 to  $2 \text{ nm.yr}^{-1}$
- This corresponds to:
  - A lifetime of  **$2.5 - 4 \cdot 10^5$  years** dissolution time of Zry cladding hulls or
  - Approx.  $10^{-6}$  fractional release  $\text{yr}^{-1}$
- Compare:  $^{14}\text{C}$  half life 5730 yr

# $^{14}\text{C}$ Release from Zircaloy



- At a critical  $\text{ZrO}_2$  thickness of  $2.5\mu\text{m}$ , a change in corrosion kinetics is observed from parabolic to pseudo-linear >> greater release rates
- Additional testing on thicker ( $>10\mu\text{m}$ ) oxide layer to assess this effect



Typical corrosion-time curve of Zircaloy in neutral and alkaline waters  
Range explored with corrosion tests (CAST D3.1)

# $^{14}\text{C}$ Release from Zircaloy



- During Zircaloy hull corrosion:
  - $^{14}\text{C}$  incorporation in the oxide film, followed by
  - release by diffusion to the surface, or
  - release during the  $\text{ZrO}_2$  layer dissolution
- $\text{ZrO}_2$  formed on the cladding surface is very stable and protective for the underlying Zircaloy metal
- The chemical forms of released  $^{14}\text{C}$  calculated for the C-H-O system assuming thermodynamic equilibrium: dominated by  $\text{CO}_3^-$  and  $\text{CH}_4$
- Organics and inorganics identified in leaching tests of irradiated or activated hulls, including short-chain fatty acids, alcohols and aldehydes
- In absence of data it is usually assumed that the corrosion rate of zirconium/zircaloy determines the radionuclides release rate

# $^{14}\text{C}$ Release from $\text{ZrO}_2$

**CAST WP6:** safety assessment studies consider an IRF Instant Release Fraction for  $^{14}\text{C}$  release from  $\text{ZrO}_2$

- IRF
  - 20% (Nagra, Ondraf)
  - 10% (SURAO)
  - 10% from Zircaloy hulls [Smith et.al. 1993] (GRS)
  - IRF based on cladding types (Andra)
- Speciation of the release:
  - Uncertain, assumed conservatively as methane(g) or organics(aq.)
  - GRS/ENEA : release as  $\text{CO}_2$ -gas after container failure (salt repository)
- Hypothesis regarding distribution:
  - $^{14}\text{C}$  often considered as homogeneously distributed in the cladding => release rate depends linearly on corrosion rate
  - $^{14}\text{C}$  inventory in oxide layer assumed roughly twice its concentration in the base metal to account for the  $^{17}\text{O}(n,\alpha)^{14}\text{C}$  reaction (Andra, RWMC)

# $^{14}\text{C}$ Speciation



- The speciation of  $^{14}\text{C}$  released from the claddings is uncertain
- The aqueous C-H-O system in complete thermodynamic redox equilibrium would be dominated by carbonate and methane
- **Assuming C inorganic** (relatively well known)
  - $^{14}\text{C}$  originating from the oxide layer considered to be released in inorganic form
  - Transport via solute diffusion
  - In clay isotope exchange may occur between calcite and carbonate > efficient retardation, potentially resulting in virtually infinite transport time for inorganics
- **Assuming C organic** (not so well known)
  - Usually conservatively assumed as methane gas
  - Can dissolve into pore water / might migrate as free gas (mixed with carrier gas)
  - Retardation processes conservatively neglected or marginally considered in SA

# $^{14}\text{C}$ Source Term in CSD-C

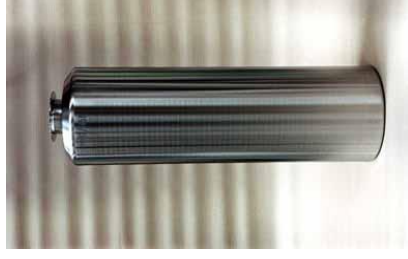


- CAST D6.2: *Accounting for an instant release fraction from compacted waste is still a matter of debate*
  - An IRF used for the cladding of directly disposed spent fuel should also be applicable to CSD-C
  - The compaction process is not expected to reduce the specific surface area
  - The acid treatment of the material during reprocessing *might* remove accessible  $^{14}\text{C}$  from the surfaces
    - IRF for CSD-C may range from 0% to 20%
- GRS: No IRF assumed due to the acid treatment of the material which removes the “accessible inventory”
- ANDRA:  $^{14}\text{C}$  inventory assumed to be located in the thinner pieces in order to assume also a congruent release
- NAGRA:
  - 20 % Inorganic (Instant Release Fraction)
  - 80% Organic (Congruent Release)

# $^{14}\text{C}$ Inventory in CSD-C



	$^{14}\text{C}$ Activity (GBq/package)	[N] cladding (ppm)
ONDRAF (47 GWd/tHM)	45	30-80
Andra (60 GWd/tHM)	20	30-40
GRS (33 GWd/tHM)	13.8	45
Nagra (33 GWd/tHM)	15	70-80
COVRA (39 GWd/tHM)	13.8	



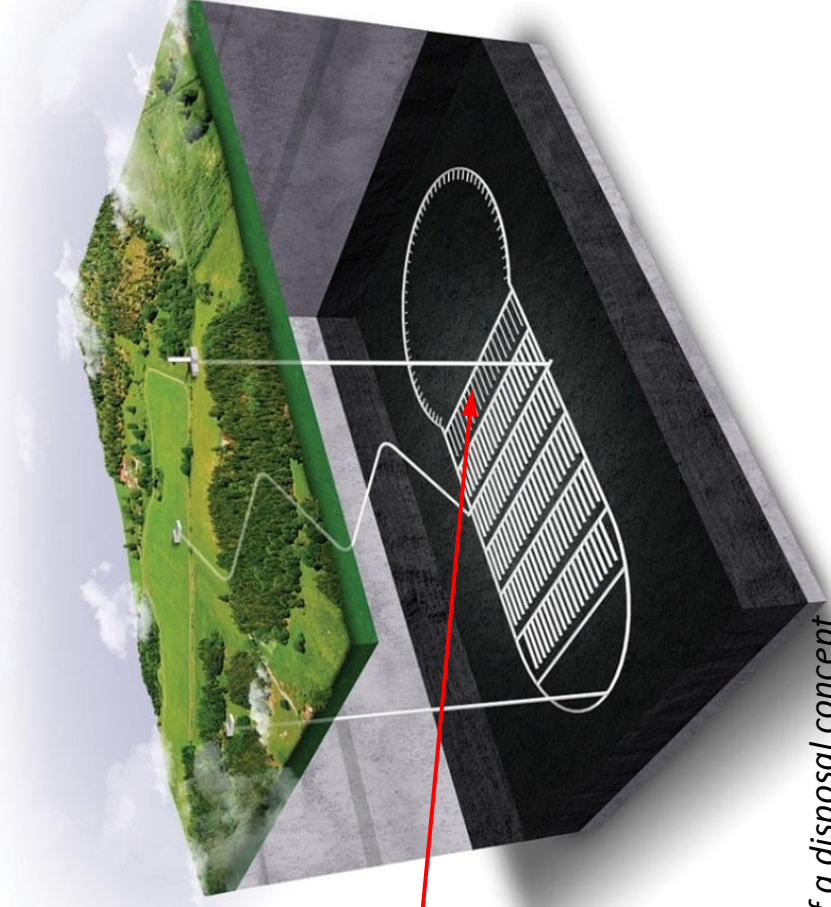
- Andra: Measurement (tritium tracer); Modelling: CESAR
- GRS: OREST simulation; N impurities: pessimistic assumptions
- Nagra: Specification from producer; Burn-up calculations (ORIGEN-S/ARP)
- COVRA: Specification from producer



# Dutch Disposal Concept

- **OPERA** Disposal concept
- Onderzoeks Programma Eindberging Radioactief Afval
- Research Programme into the Geological Disposal of Radioactive Waste (2011-2016)
- Boom Clay
- Generic depth: 500 m
- Separate sections for different waste types

CSD-C

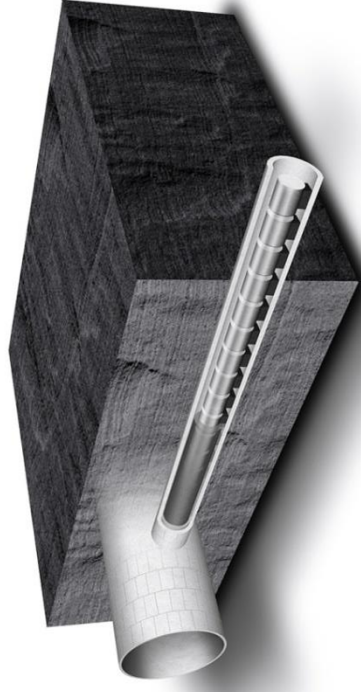


*Verhoeef E, Neeft E, Grupa, J, Poley A, Outline of a disposal concept in clay, First update, OPERA-PG-COV008, 13 november 2014*

# Dutch Disposal Concept



- CSD-C container overpack:
  - OPERA Supercontainer
  - Adopted from Belgian supercontainer concept
  - Carbon steel overpack, concrete buffer and stainless steel envelope
- Disposal galleries:
  - Concrete backfill
  - Concrete lining
- $^{14}\text{C}$  inventory: 13.8 GBq /container



# OPERA

## Performance Assessment



### **Objectives of the OPERA PA**

- Model the transport of radionuclides through the deep underground to the earth's surface, as well as the subsequent uptake by humans
- Provide a robust projection of the long-term safety of deep geological disposal of radioactive substances in Boom Clay
- Provide extended perceptions in the phenomena that are relevant for the transport of nuclides from the repository to the biosphere

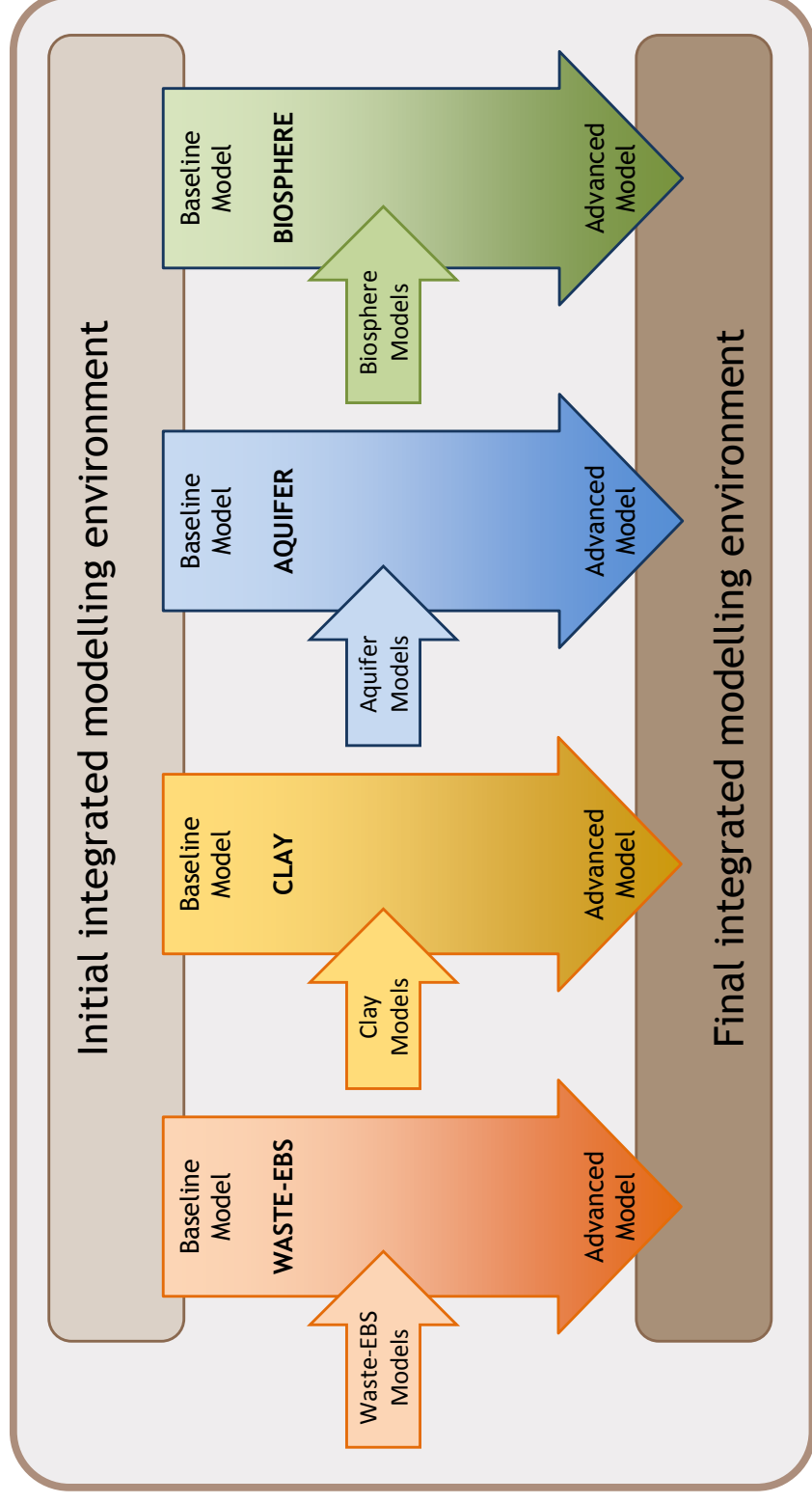
### **Restrictions of the OPERA PA**

- No formal safety assessment for obtaining a license to construct, operate, or close a disposal facility

# Formulation and Implementation of Assessment Models



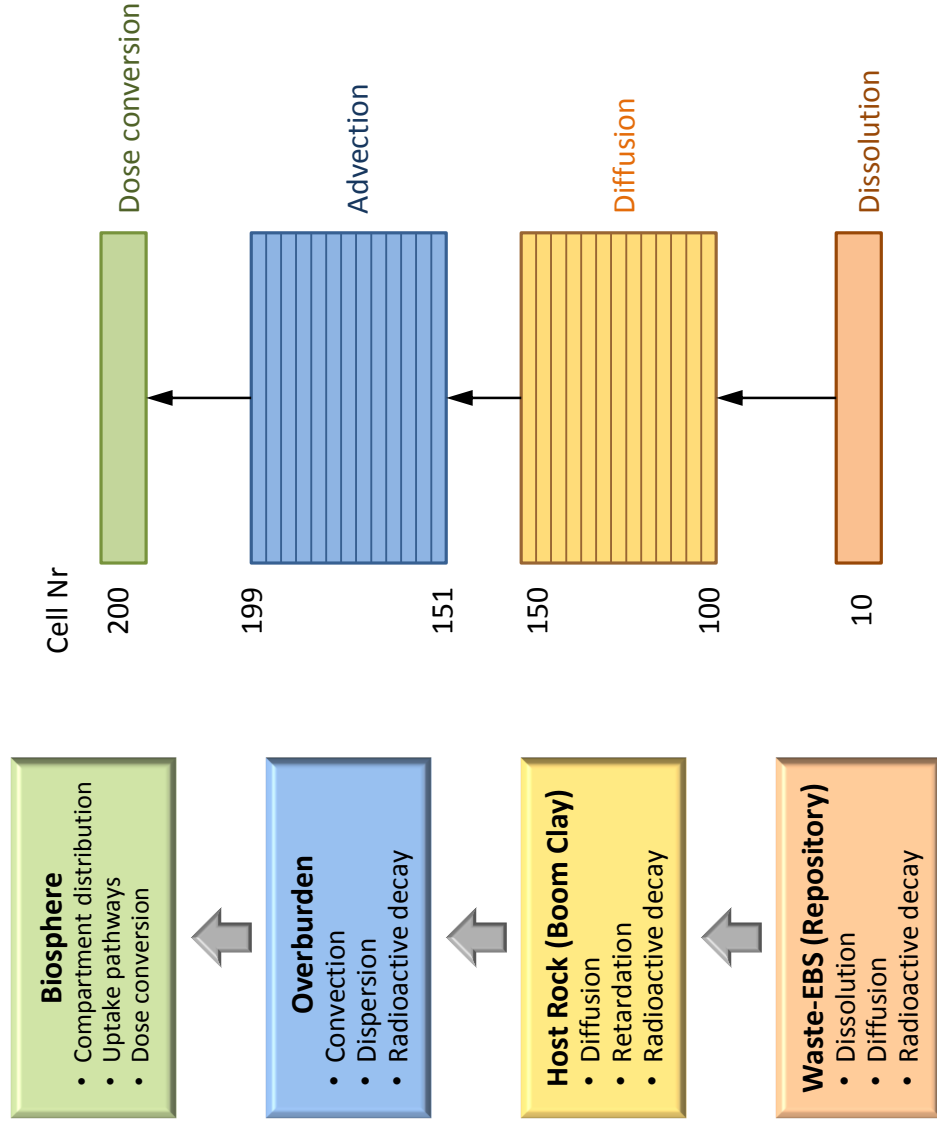
Translation of information from OPERA WPs/Tasks into models and data  
for the ORCHESTRA integrated conceptual model



# OPERA Performance Assessment



- Conceptual model
- Computer code: ORCHESTRA

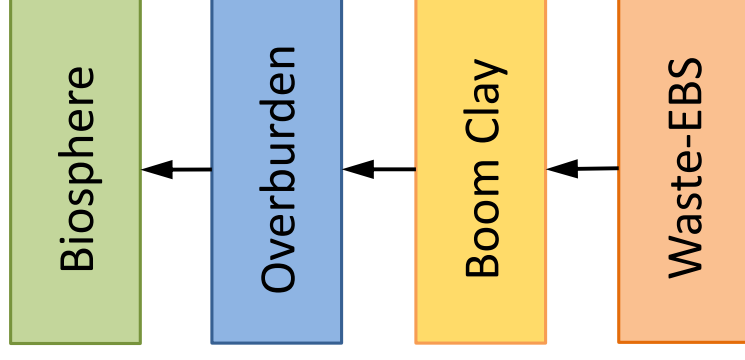


## Objects Representing CHEmical Speciation and TRAnsport

- Open source code
- Calculate chemical speciation and reactive transport processes
- Simulate **equilibrium** reactions, **kinetically** controlled reactions, radioactive **decay**, **transport**
- .....(many more)
- [www.orchestra.meeussen.nl](http://www.orchestra.meeussen.nl)

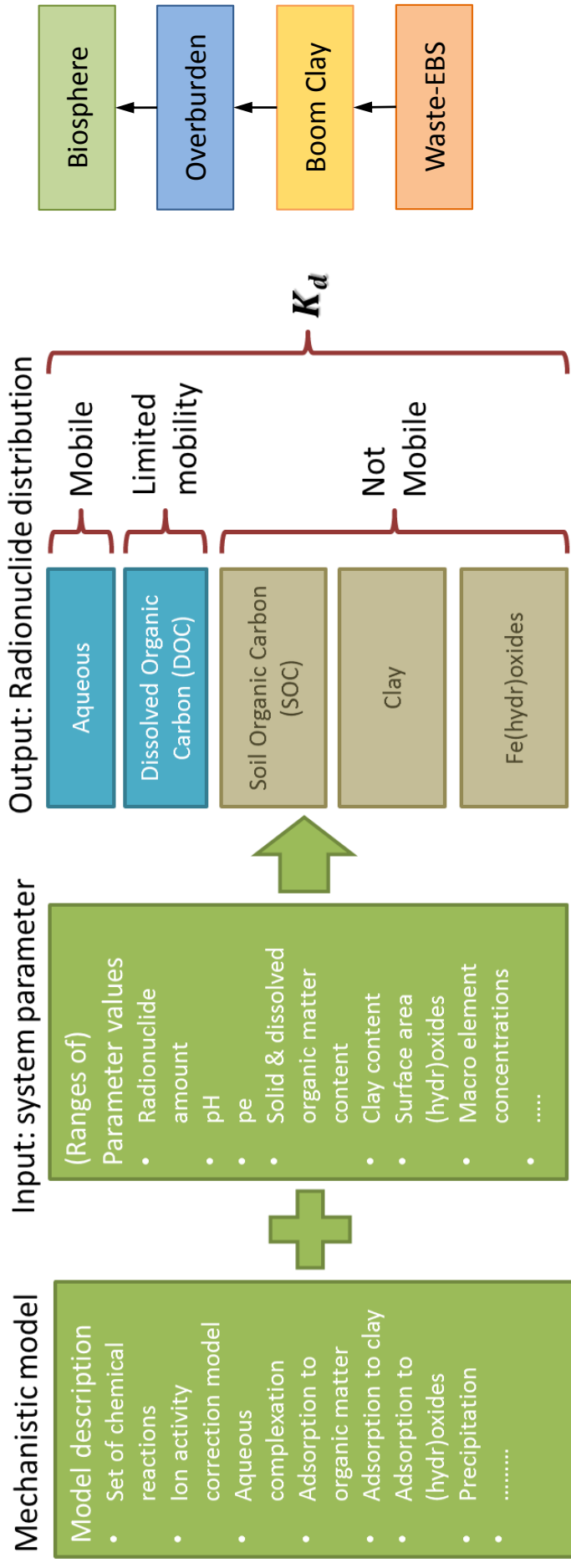
# Compartment Waste-EBS (OPERA)

- Separate compartments for different waste classes
- Inventories determined for each waste class
- Start and duration of RN release depend on conditioning
- OPERA Supercontainer CSD-C – time of failure
  - Lower bound: 1'500 years
  - Expert value: 15'000 years
  - Upper bound: 700'000 years



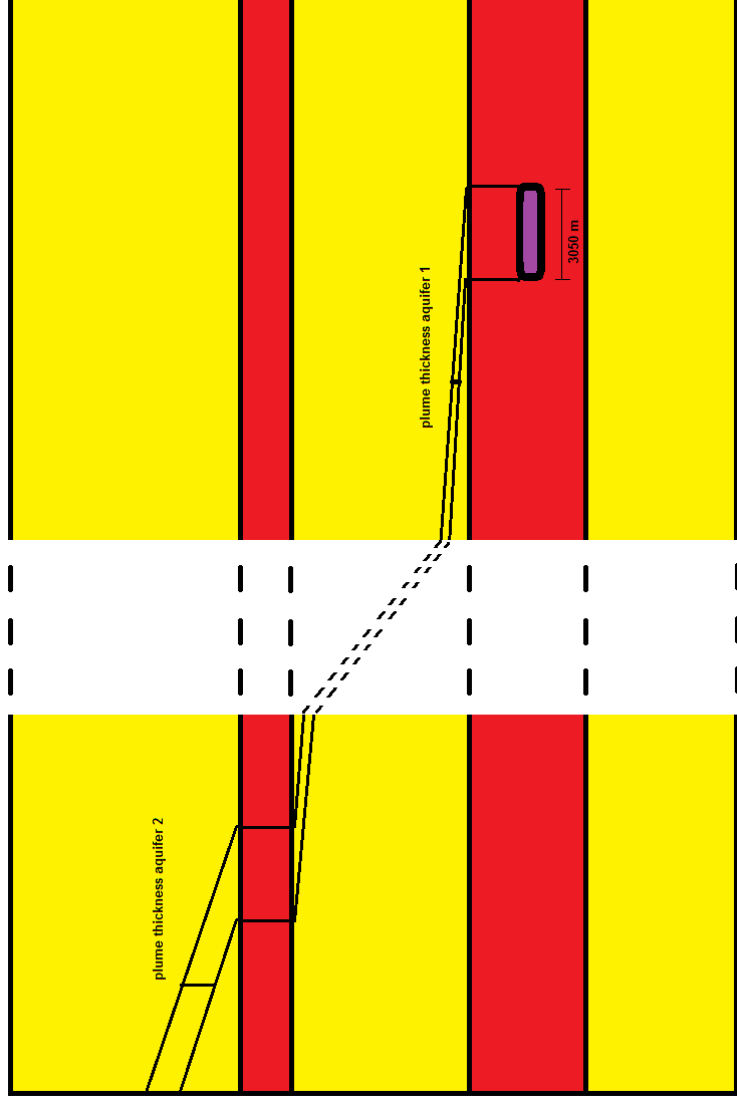
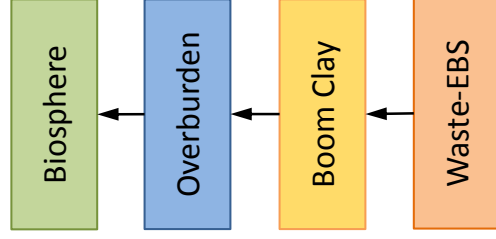
# Compartment Boom Clay (OPERA)

- Mechanistic model calculates distribution of nuclides over different system phases using set of thermodynamic and physical parameters as input
- This information is used to calculate mobility (retardation factor), using ORCHESTRA



# Compartment Overburden (OPERA)

- Aquifer: advective flow + dispersion
  - Main parameters:
    - path length
    - effective flow velocity
    - RN sorption coefficients
- (ORCHESTRA)

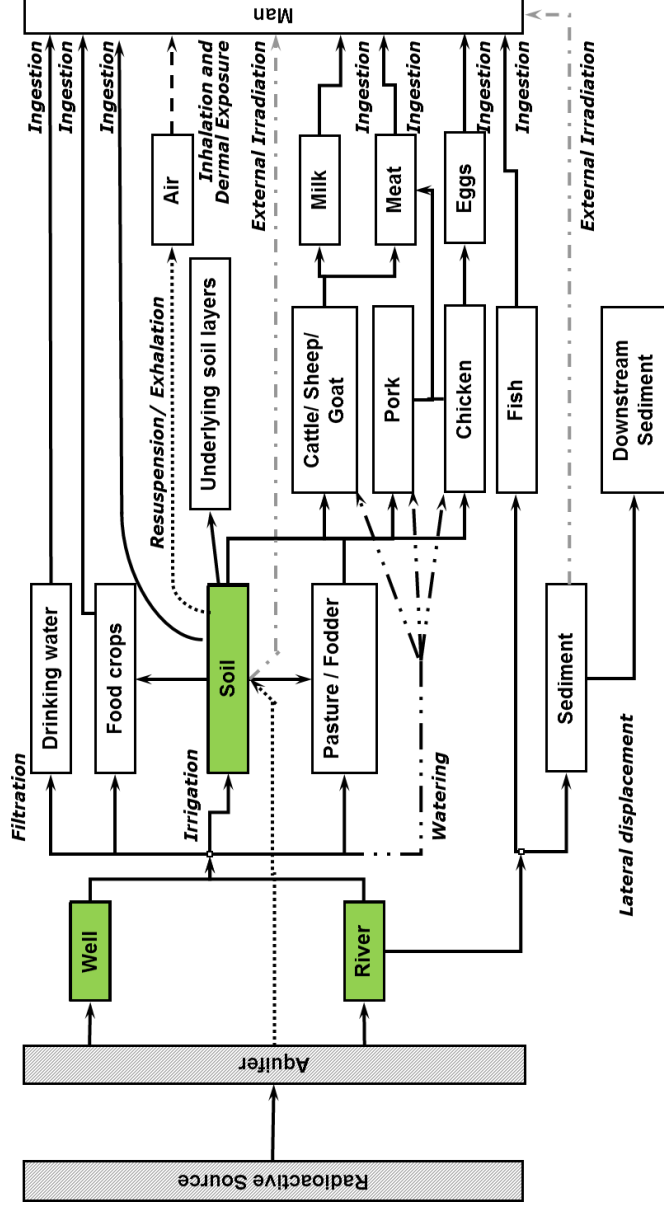




# Compartment Biosphere (OPERA)

Main parameters:

- Dilution
- Dose Conversion Coefficient (DCC)

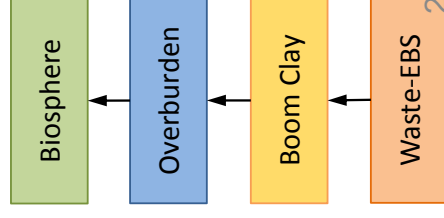


5 climate states:

- Present temperate maritime
- Warmer ("Mediterranean")
- Colder (boreal)
- Periglacial (polar tundra)
- Glacial (polar frost)

➤ Affect:

- Transfer to plants, animals
- Human habits, consumption rates
- DCC - **Dose Conversion Coefficient**



# CAST Performance Assessment

## Sensitivity/uncertainty analysis

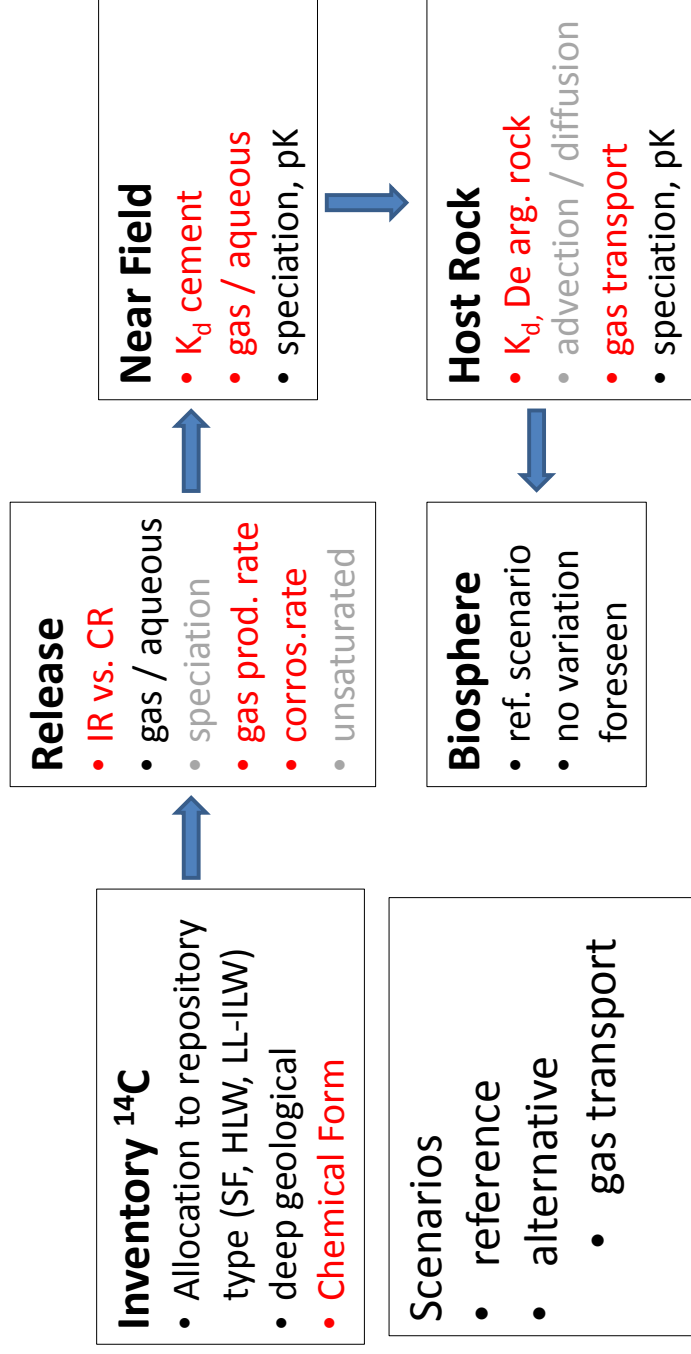


- At the CAST technical meeting of March 2016, the WP6 organisations decided to place their calculations in the perspective of **sensitivity/uncertainty analysis**
- Assess the influence of key factors on  $^{14}\text{C}$  release and transport:
  - Corrosion rates
  - IRF versus congruent release
  - Ratio of organic/inorganic release
  - Sorption coefficient ( $K_d$ -value)
  - Gas versus aqueous transport
- Ranges of parameter values defined for each of these factors

# Uncertainty Analyses Argillaceous Rock



CAST Document: Handling of C-14 in safety assessment - Guidance for uncertainty analysis, Manuel Capouet (ONDRAF/NIRAS)



The objective is not to compare concepts, but to illustrate possible impacts and identify future areas of investigation

# Uncertainty Analyses Argillaceous Rock

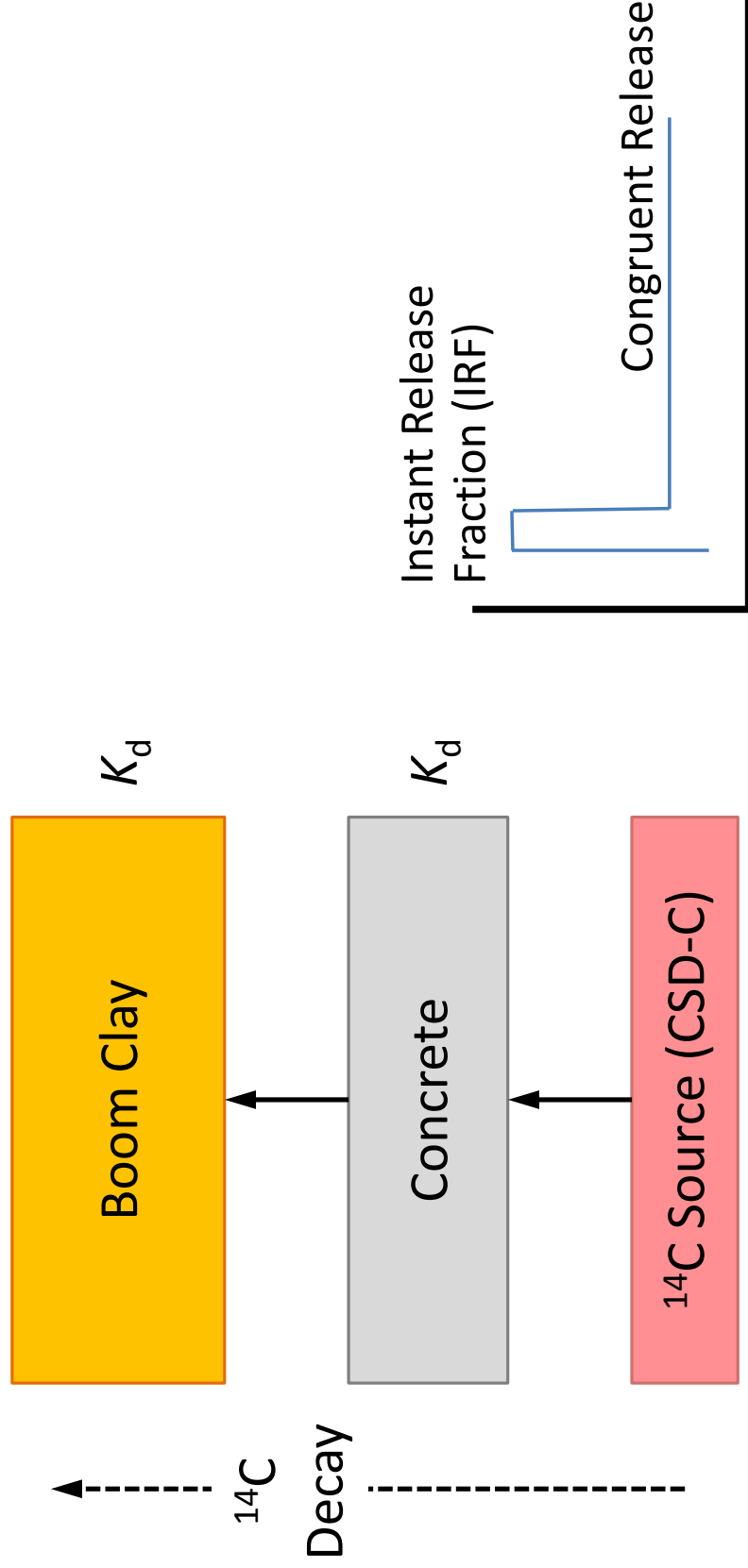


<b>CAST Key Factors</b>	<b>Implementation NRG</b>
<sup>14</sup> C Inventory	$1.38 \cdot 10^{10}$ Bq/CSD-C
<b>250</b> Simulations with variation of:	
IRF versus congruent release	IRF: 1-20% of inventory
Corrosion rates	Zircaloy cladding: 1-10 nm/y
Solid/liquid partition coefficients ( $K_d$ -value)	$10^{-5} - 10^{-3}$ m <sup>3</sup> /kg Concrete+Boom Clay
Ratio of organic/inorganic release	Presently assumed <b>inorganic</b>

# Conceptual model



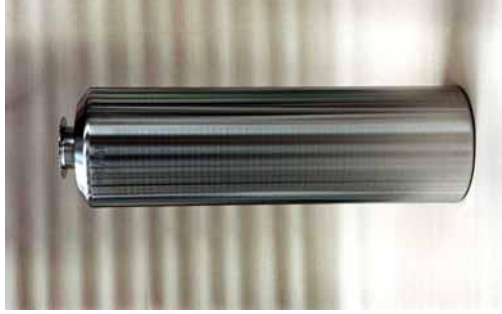
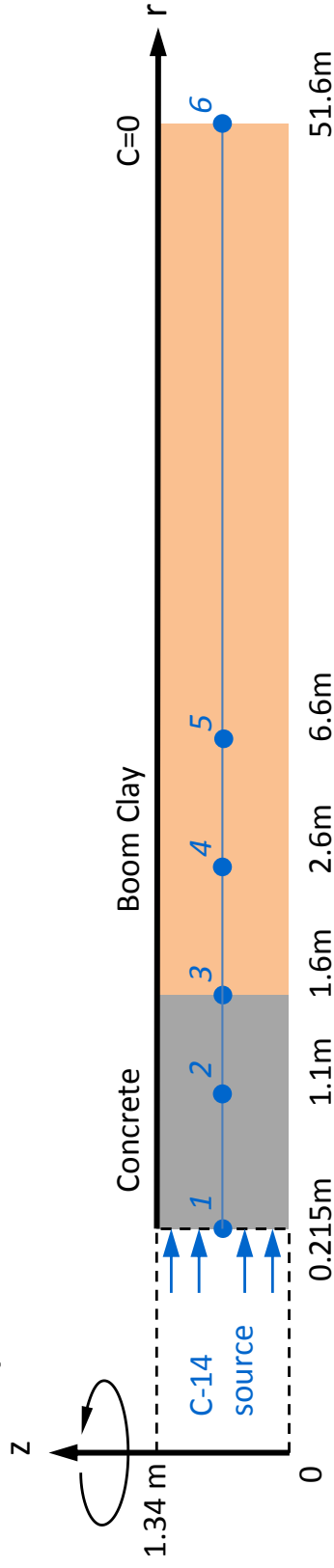
CAST: reduced version of OPERA conceptual model



# Conceptual model



## Conceptualization in ORCHESTRA



### CSD-C Canister

- OPERA Supercontainer (concrete)
- $z = 1.34$  m (CSD-C canister length)
- $r1 = 0.43/2 = 0.215$  m
- $r2 = 1.1$  m (Supercontainer radius)
- $r3 = 1.6$  m (Concrete liner radius)
- $r4 = 2.6$  m (1 m into Boom Clay)
- $r5 = 6.6$  m (5 m into Boom Clay)
- $r6 = 51.6$  m (50 m into Boom Clay)



# Test Simulation – Conservative Assumptions



OPERA Supercontainer / CSD-C – time of failure

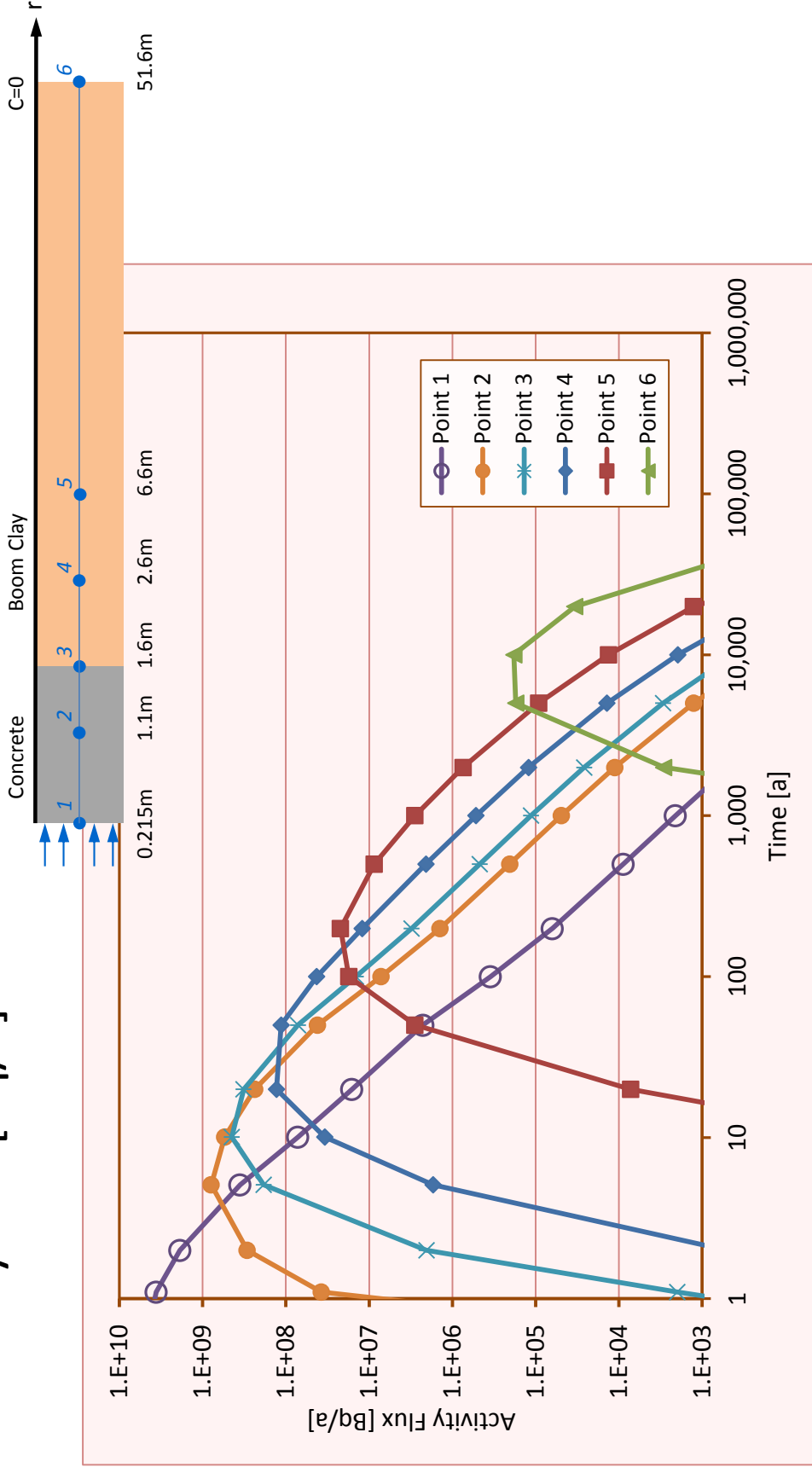
- Upper bound: 700'000 years
- Expert value: 15'000 years
- Lower bound: 1'500 years
- Assumed for CAST: 0 years

Sensitivity Parameter	Value	
<sup>14</sup> C Instant release fraction	20%	Upper bound
Zircaloy corrosion rate	10 nm/a	Upper bound
<sup>14</sup> C Congruent release duration Assumed flat Zry plates, 0.65 mm	65'000 year	Lower bound
K <sub>d</sub> Concrete	10 <sup>-5</sup> (m <sup>3</sup> /kg)	Lower bound
K <sub>d</sub> Argillaceous rock	10 <sup>-5</sup> (m <sup>3</sup> /kg)	Lower bound

# Calculation Results

Test Simulation – Conservative Assumptions

$^{14}\text{C}$  Activity flux [Bq/a]

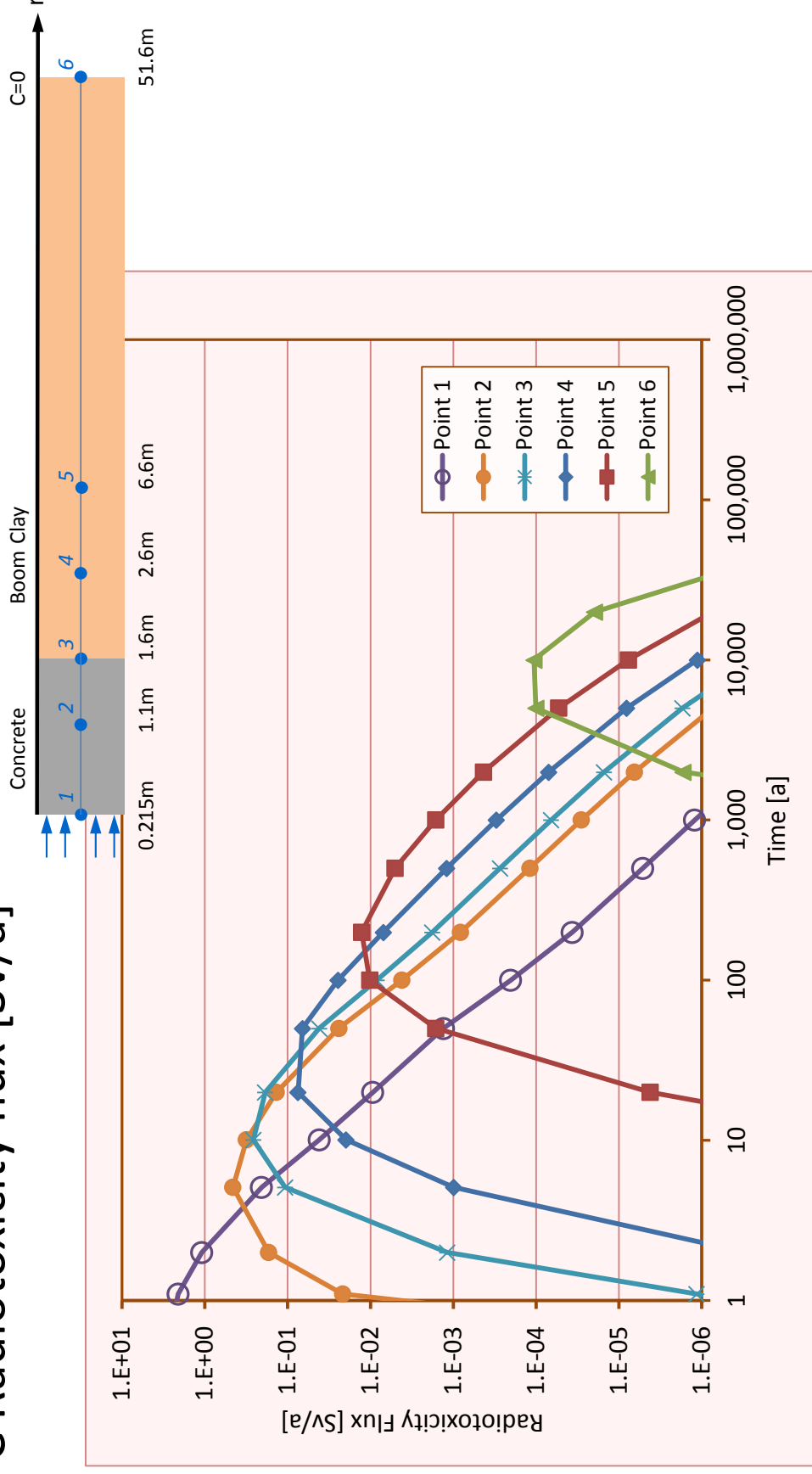




# Calculation Results

Test Simulation – Conservative Assumptions

$^{14}\text{C}$  Radiotoxicity flux [Sv/a]

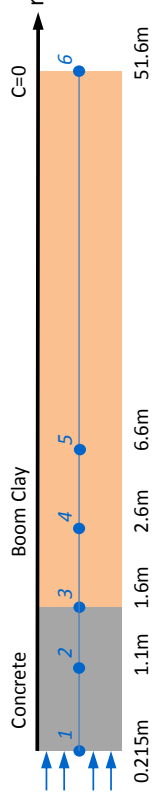
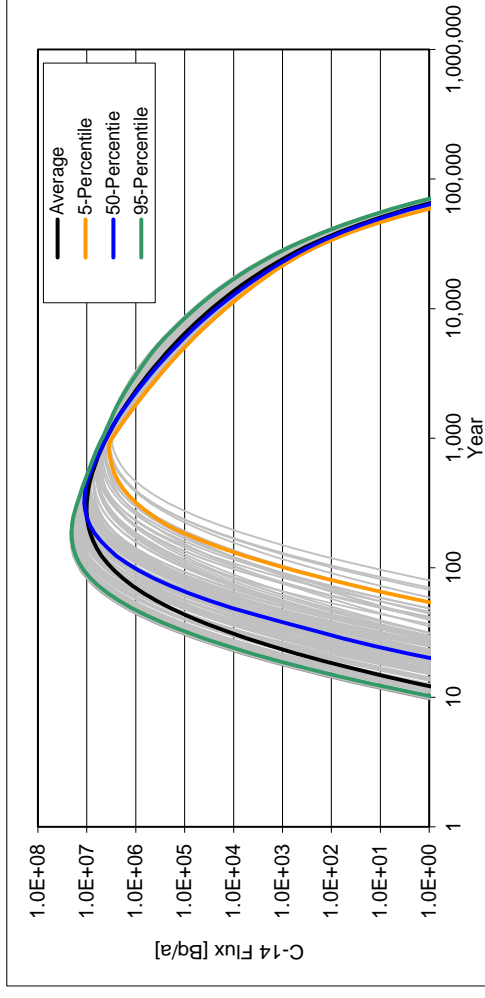


# Calculation Results

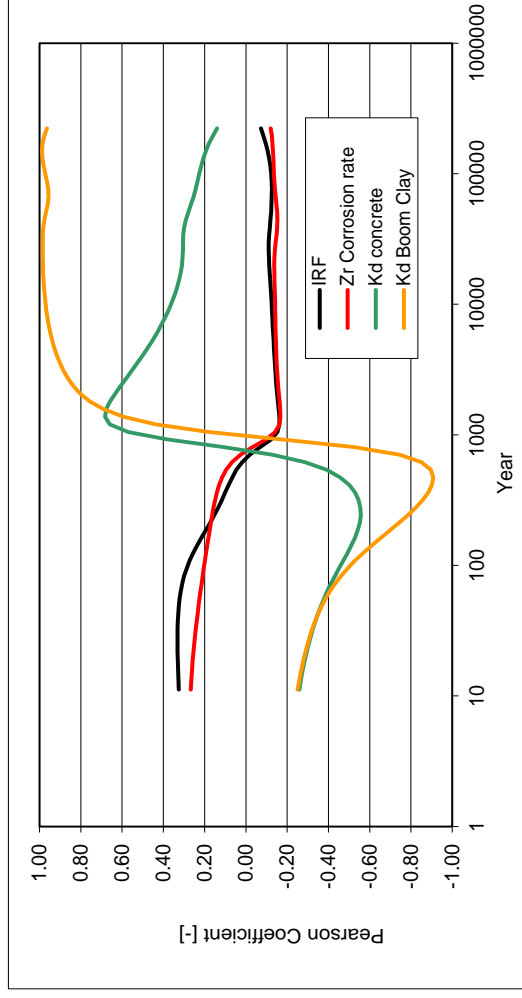
UA/SA Test Simulation – 100 runs



## $^{14}\text{C}$ Activity flux [Bq/a] – Point 5



- Preliminary results – need to be verified!
- 5 m into the Boom Clay
- Largest influence:  $K_d$  Boom Clay (Pearson coefficient)
- Further analysis required



# Calculation Results

Test Simulation – Conservative Assumptions



## Observations

- $^{14}\text{C}$  Release from waste compartment:
  - Increases  $^{14}\text{C}$  concentrations in concrete/Boom Clay
  - Settles  $^{14}\text{C}$  concentration differences between waste and concrete/Boom Clay
  - Decreases  $^{14}\text{C}$  release rate from waste compartment to concrete
- $^{14}\text{C}$  Release from Boom Clay (50 m of clay; **conservative assumptions**):
  - Peaks around 5'000-11'000 years
  - Amounts approx 65 mSv/a for the expected total of 600 CSD-C canisters (Dutch disposal concept)
  - Accumulates to approx 17% of the initial inventory (decay)

# Remaining Uncertainties

## $^{14}\text{C}$ and Zircaloy



- CAST contribution to further reduction of remaining uncertainties:
    - Speciation of  $^{14}\text{C}$  released from  $\text{Zry/ZrO}_2$  – organic/inorganic/gas
    - Instant Release Fraction – likely an important aspect for long-term safety (at least for argillaceous rock)
    - $^{14}\text{C}$  Carbonate sorption on concretes (backfill, lining, ...)
    - $K_d$  values in Boom Clay ( $^{14}\text{C}$  species interactions)
  - Other (outside CAST): assumed/expected lifetimes of CSD-C containers (and overpacks like the Supercontainer)
- 
- **Less conservatism in PA/SA?**
  - **Arguments to reduce the impact of  $^{14}\text{C}$  on long-term safety?**

# Conclusions and the Way Forward



- CAST WP3 made considerable progress on the formation, release, and speciation of  $^{14}\text{C}$  in Zircaloy
- Transport of  $^{14}\text{C}$  in concrete, Boom Clay (and other materials) is presently being addressed in CAST WP6
- Preliminary PA results show that, under conservative assumptions,  $^{14}\text{C}$  may reach the top of the Boom Clay
- The way forward (CAST WP6)
  - Perform uncertainty/sensitivity analysis
  - Determine most influential parameter(s) for  $^{14}\text{C}$  transport (IRF, Zircaloy corrosion rate,  $K_d$  concrete/Boom Clay)
  - Further analyze  $^{14}\text{C}$  transport in concrete and Boom Clay (speciation,  $K_d$ , gas, ...)

# Acknowledgement



- The research leading to these results has received funding from the Dutch research programme on geological disposal OPERA. OPERA is financed by the Dutch Ministry of Economic Affairs and the public limited liability company Elektriciteits-Productiemaatschappij Zuid-Nederland (EPZ) and coordinated by COVRA
- The project has received funding from the European Union's European Atomic Energy Community's (Euratom) Seventh Framework Programme FP7/2007-2013 under grant agreement no. 604779, the CAST project.





Presentation No. 21

1



# CArbon-14 Source Term CAST

Name: **N. Diomidis** (J.Mibus, V.Cloet, M.Pantelias, B. Volmert)

Organisation: **Nagra**

Date: **6/10/2016**



The project has received funding from the European Union's Seventh Framework Programme for research, technological development and demonstration under grant agreement no. 604779, the CAST project.





An example of

# **Safety assessment of $^{14}\text{C}$ release from irradiated steels**



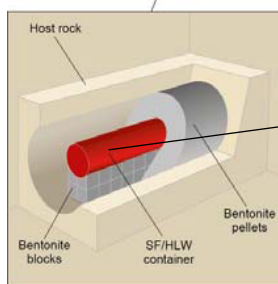
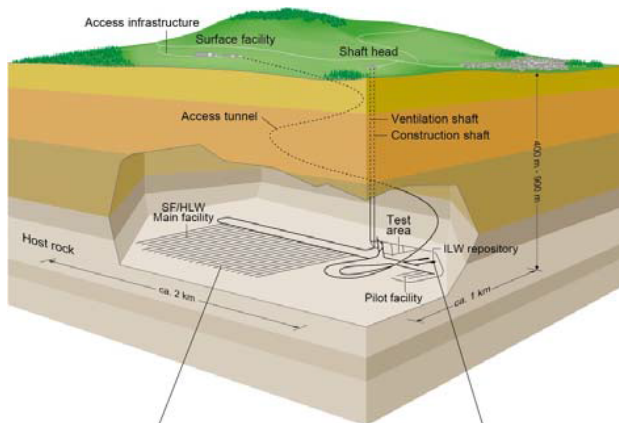
## Outline



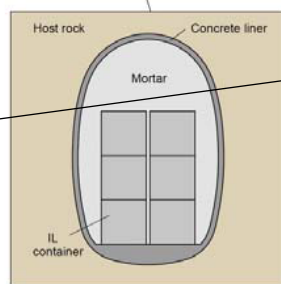
- Swiss disposal concept
- Waste inventory and description
- Safety assessment



# Geological disposal of SF/ HLW/ILW



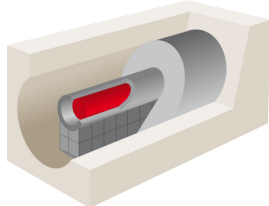
Emplacement drift for SF/HLW



Emplacement tunnel for ILW

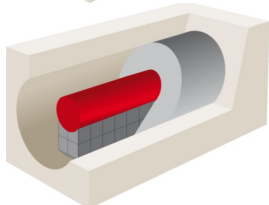


## Safety barriers for SF/HLW



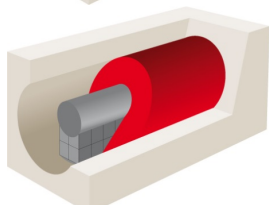
### Waste matrix

- Containment of RNs in glass and fuel pellets
- Low glass dissolution
- Low  $\text{UO}_2/\text{MOX}$  dissolution and Zy corrosion



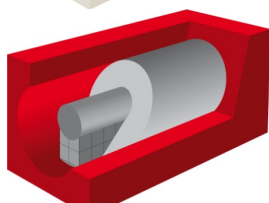
### Disposal canister

- 10'000 years absolute containment
- Corrosion induces favorable local chemistry
- Corrosion products absorb radionuclides



### Bentonite

- Low saturation times, self-sealing
- Low radionuclide solubility in porewater
- Diffusion-limited system
- Sorption of radionuclides

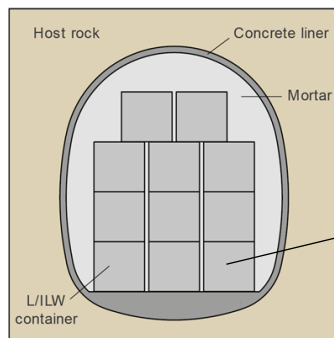
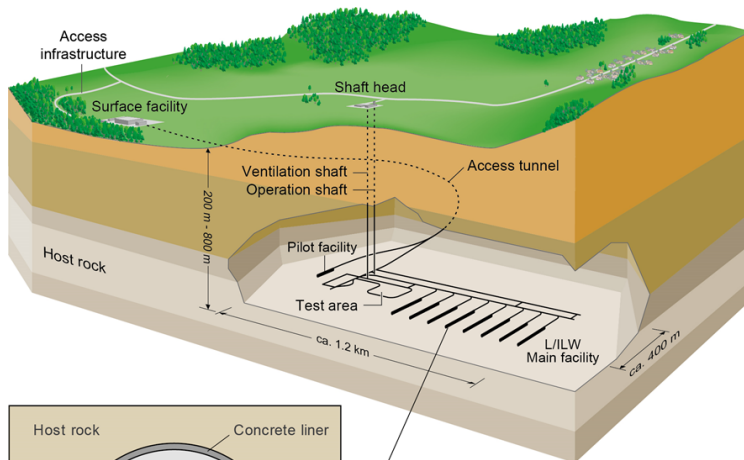


### Opalinus Clay

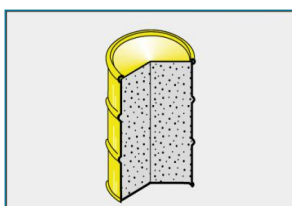
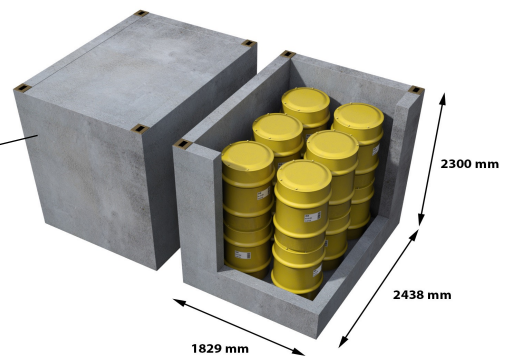
- Jurassic claystone 180M yrs. old
- Very low porosity and hydraulic conductivity
- Self-sealing
- Sorption and dilution

Isolation of waste.  
Decay of majority of  
nuclides within the  
disposal system.  
Attenuation of release  
of remaining nuclides.



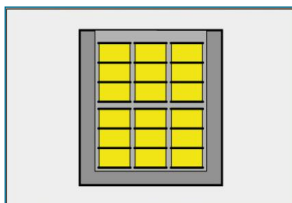


Emplacement tunnel for L/ILW



## Waste in cement matrix

- Slow corrosion of metals
- Slow degradation of organics



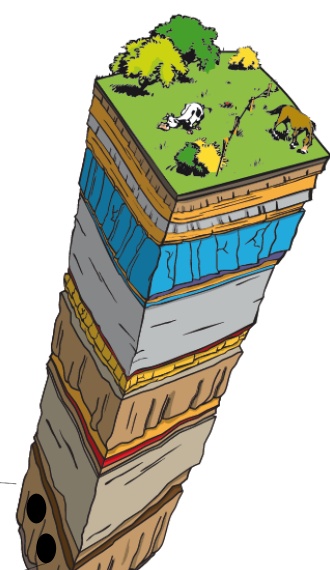
## Concrete containers filled with mortar

- Low solubility of RNs in cementitious near field
- Sorption of RNs



## Disposal cavern

- Low groundwater flux in cement and host rock



L/ILW repository



# Overview of $^{14}\text{C}$ inventory

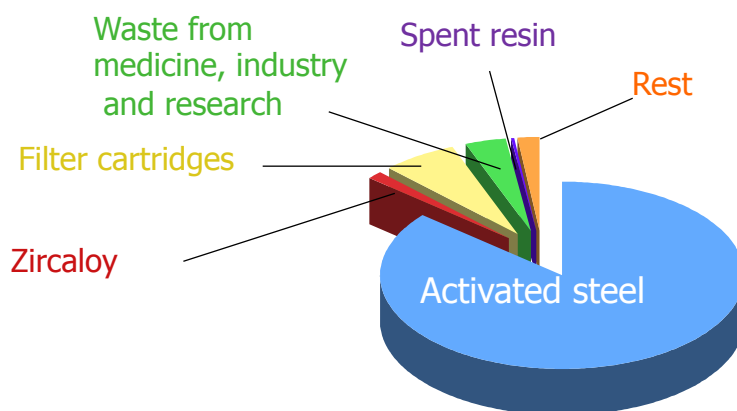


- HLW repository:  $2.7 \times 10^{14}$  Bq
- L/ILW repository:  $1.9 \times 10^{14}$  Bq

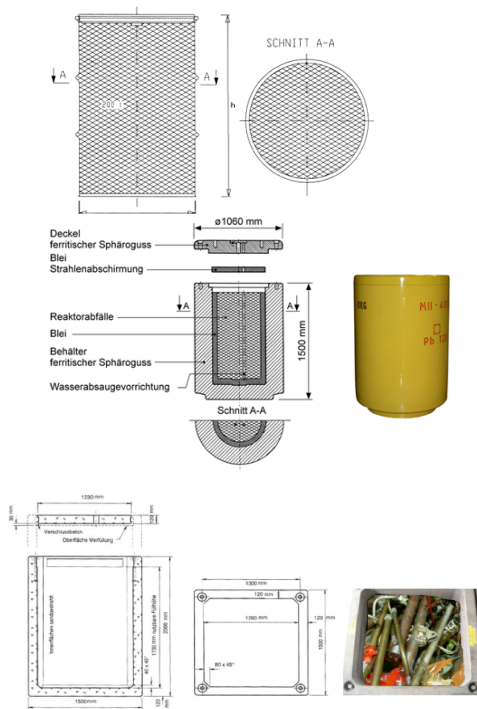
Waste type	$^{14}\text{C}$ inventory	N (ppm)	Determination method
Fuel assemblies: PWR/BWR 35 to 65 GWd/tHM	55 – 105 GBq/tHM	25	Info from waste producers and depletion/burnup calcs. (ORIGEN-S/ARP). International standards used for validations
Fuel assemblies: cladding (zircaloy)	0.05 – 0.08 GBq/kg	70 – 80	
Fuel assemblies: structural parts (steel)	0.2 – 0.4 GBq/kg	400 – 800	
Reactor core internals	< 0.7 GBq/kg	90	Neutron transport and activation calcs. (MCNP/ORIGEN-S)
Reactor pressure vessel	< 0.02 GBq/kg	50	
Ion Exchange Resins	0.002 – 0.01 GBq/kg		Radiological analyses of representative samples
Vitrified HLW (CSD-V)	1 – 3 GBq/package	75	Info from waste producers
Compacted ILW metallic waste (CSD-C)	15 GBq/package	Zircaloy: 70 – 80 Steel: 400-800	



## $^{14}\text{C}$ in L/ILW



# Typical waste package



## Activated Steel

- Reactor internals, decommissioning waste, shielding
- In 200L-drums, Mosaik II container, LC-84, KC-T12
- Backfilling with cement (200L drum, LC-84, KC-T12)
- No backfilling (Mosaik II)
- Material inventory of waste package available (divided into organic, inorganic and steel components)
- Calculated nuclide inventory for waste package



## Waste properties (MIRAM)



**Miram 14 - Basis Scenario**

**SA-G-MS-L3-SMA - Metall: Stahlabflle in LC84**

**SCHEMATISCHE DARSTELLUNG DES REPRSENT**

**Miram 14 - Basis Scenario**

**SA-G-MS-L3-SMA - Metall: Stahlabflle in LC84**

**RADIOLOGISCHE KENNDATEN FR DAS REPRSENT**

**Nuklidinventar**

Nuklid	Aktivitt [Bq]	Nuklid	Aktivitt [Bq]	Nuklid	Aktivitt [Bq]	Nuklid	Aktivitt [Bq]
Mittel	Maximal	Mittel	Maximal	Mittel	Maximal	Mittel	Maximal
H-3	8.4E+07	2.2E+08	Nb-94	2.5E+07	6.7E+07	Pm-145	4.5E+03
C-14	4.0E+07	1.0E+08	Mo-93	1.1E+06	3.0E+06	Pm-147	2.1E+01
Cl-36	3.6E+06	9.3E+06	Tc-99	1.5E+05	4.1E+05	Sm-151	3.0E+05
Ar-39	2.2E+06	5.9E+06	Ag-108	8.3E+05	2.2E+06	Eu-152	8.9E+07
K-40	1.0E+04	3.1E+04	Ag-108M	9.5E+06	2.5E+07	Eu-154	3.8E+06
Ca-41	1.2E+05	3.0E+05	Cd-113M	4.9E+00	1.2E+01	Eu-155	2.0E+03
Fe-55	2.0E+08	5.1E+08	Sn-121	1.3E+02	3.7E+02	Tb-157	2.0E+05
Co-60	2.8E+09	7.3E+09	Sn-121M	1.7E+02	4.7E+02	Ho-166M	3.0E+04
Ni-59	1.8E+07	4.7E+07	Sn-126	6.9E+01	5.3E+01	Ti-204	1.1E+06
Cu-63	1.5E+09	4.0E+09	Sb-125	1.6E+03	1.8E+04	Tb-231	1.2E+01
Se-79	1.5E+04	4.3E+04	Sb-126	2.7E+00	7.4E+00	Tb-234	5.7E+00
K-95	1.5E+01	3.9E+01	Ag-116M	1.9E+01	6.3E+01	Bn-242	5.7E+00

**Schematic representation of waste package**

**Calculated nuclide inventory**

**MATERIALKENDATEN FR DAS REPRSENTATIVE GEBULDE**

**Materialinventar und Herkunft**

Material	Masse [kg]	*	C	D	E	F	G
			Abfallprodukt	Extrakt	Fllmaterial	Endsubstanz	Lebensmittel
Zementstein --Micropoz	3.50E+02	A			3.50E+02		
Zementstein --Wasser	1.10E+03	A		3.95E+02	7.07E+02		
Zementstein --Zement HTS	2.14E+03	A		9.75E+02	1.17E+03		
Baustahl	6.69E+03	M	6.69E+03				
Clinoptilolith	1.75E+02	A			1.75E+02		
Diethanolamin	6.29E+00	O			6.29E+00		
Formaldehyd	1.93E+00	O			1.93E+00		
Glucosure	2.91E+00	O			2.91E+00		
Harnstoff	4.19E+00	O			4.19E+00		
Kies	4.89E+03	A		4.89E+03			
LaurylaminPEGlycoether	8.62E-01	O			8.62E-01		
Ligninsulfonate	1.78E-01	O		1.78E-01			
Polycarboxylate	2.93E+00	O		2.93E+00			
Quarzsand	1.82E+03	A			1.82E+03		
Siliciumdioxid SiO2	7.80E+01	A			7.80E+01		
Stahl	1.02E+03	M	1.02E+03				
Stahl III SIA 162	3.27E+02	M			3.27E+02		
Triethylphosphat C12H27O4P	6.99E-02	O			6.99E-02		
<b>Total</b>	<b>1.86E+04</b>		<b>7.71E+03</b>	<b>6.67E+03</b>	<b>4.24E+03</b>	<b>0.00E+00</b>	<b>0.00E+00</b>
* total anorganisch/nichtmetallisch	1.06E+04	A		6.34E+03	4.22E+03		
metallisch	8.04E+03	M	7.71E+03	3.27E+02			
organisch	1.94E+01	O		3.11E+00	1.63E+01		

**Oberflchen/Massen-Verhltnisse von Metallen**

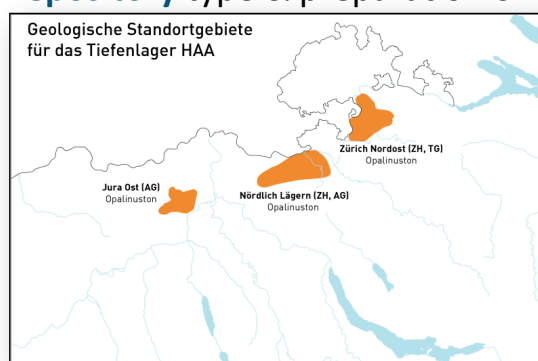
Material	m <sup>2</sup>	kg	m <sup>2</sup> /kg
Baustahl	2.01E+02	6.69E+03	3.01E-02
Stahl	3.89E+02	1.02E+03	3.82E-01
Stahl III SIA 162	1.67E+01	3.27E+02	5.10E-02



# Swiss Sectoral Plan for Deep Geological Repositories



- Demonstration of **disposal feasibility** (L/ILW: 1988, HLW: 2006)
- **Site selection** («sectoral plan»)
  - Stage 1: selection of **siting regions**
  - Stage 2: selection of **site for surface facility** within siting regions, **narrowing down of siting regions** to at least 2 for each repository type (provisional safety analyses & safety-based comparison)
  - ➔ Stage 3: selection of **a site for each repository** type & preparation of general license application
- **General license: ~2022**
- **Construction license: ~2045**
- **Operation license: (~2055)**
- **License for closure**



## Safety assessment



- Nagra's last safety analyses within Stage 2
- So-called provisional safety analyses to compare different siting regions
  - Not a full Safety Case (methodology is similar)
- Reference case describes expected and most plausible evolution
- Alternative cases prescribed by regulator





# Basic principles of $^{14}\text{C}$ speciation and release



- Activation leads to organic species
- Contamination leads to inorganic species
- Metallic materials release  $^{14}\text{C}$  via uniform corrosion -> congruent release (Al -> inst. rel.)
- All other materials conservatively assumed to be released instantaneously
  - Organics: organic
  - Concrete: inorganic



## $^{14}\text{C}$ speciation in the waste



- ILW & L/ILW:
  - Homogeneous metallic waste from NPPs and nuclear research facilities: CR in organic form (except Al: instantaneous release (IR))
  - Homogeneous concrete waste from NPP and CERN: organic IR
  - Homogeneous concrete waste from Zwiilag and surface facilities of a future repository: inorganic IR
  - Reinforced concrete from nuclear research facilities: 50 % organic IR, 50 % organic CR
  - Operational waste from PSI: organic IR
  - Operational and decommissioning waste from PWR: 70 % inorganic and 30 % organic (both IR)
  - Operational and decommissioning waste from BWR: 90 % inorganic and 10 % organic (both IR).



# Basic principles of $^{14}\text{C}$ migration



- Formation of aqueous and volatile species assumed
- Focus on transport in aqueous phase, consideration of gas transport as What-if scenario in a former safety case
- Sorption of  $^{14}\text{C}$  inorg on cement and calcite in the host rock by isotopic exchange with stable carbon (as carbonate)
- Weak sorption of  $^{14}\text{C}$  org on cement considered (measured experimentally)
- Diffusion of  $^{14}\text{C}$  inorg (as  $\text{H}^{14}\text{CO}_3^-$ ) and  $^{14}\text{C}$  org as small organic molecules in the host rock



## Reference case ILW & L/ILW



- Containment for 100 yrs
- Congruent release from steel for 10 Kyr
- Linear sorption of  $^{14}\text{C}(\text{inorg})$  and  $^{14}\text{C}(\text{org})$  in cementitious near-field
- Diffusion through host rock Opalinus Clay OPA and confining rocks
- Linear sorption of  $^{14}\text{C}(\text{inorg})$  in OPA, no sorption of  $^{14}\text{C}(\text{org})$





# Alternative cases



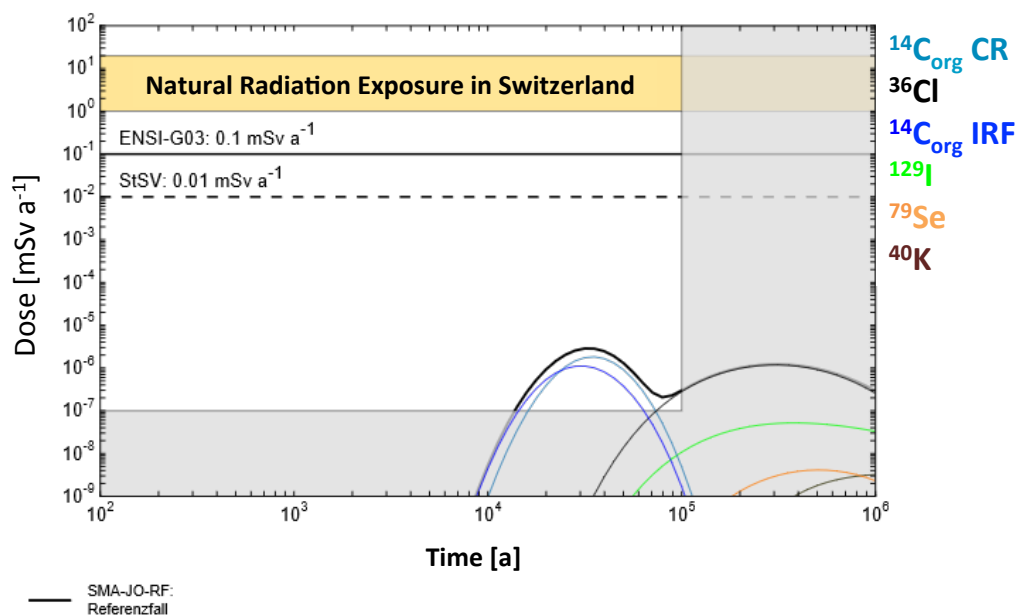
- Increased water flux through the repository
- Unfavourable diffusion coefficients
- Increased solubility limits for radionuclides
- Decreased sorption coefficients in the near field and in the host rock
- Consideration of alternative climate variants
- Increased dissolution rate of spent fuel elements
- SF/HLW canister lifetime reduced to 1,000 years.



## Example L/ILW Repository Siting Area Jura East



### Calculated Dose Curves: Reference case

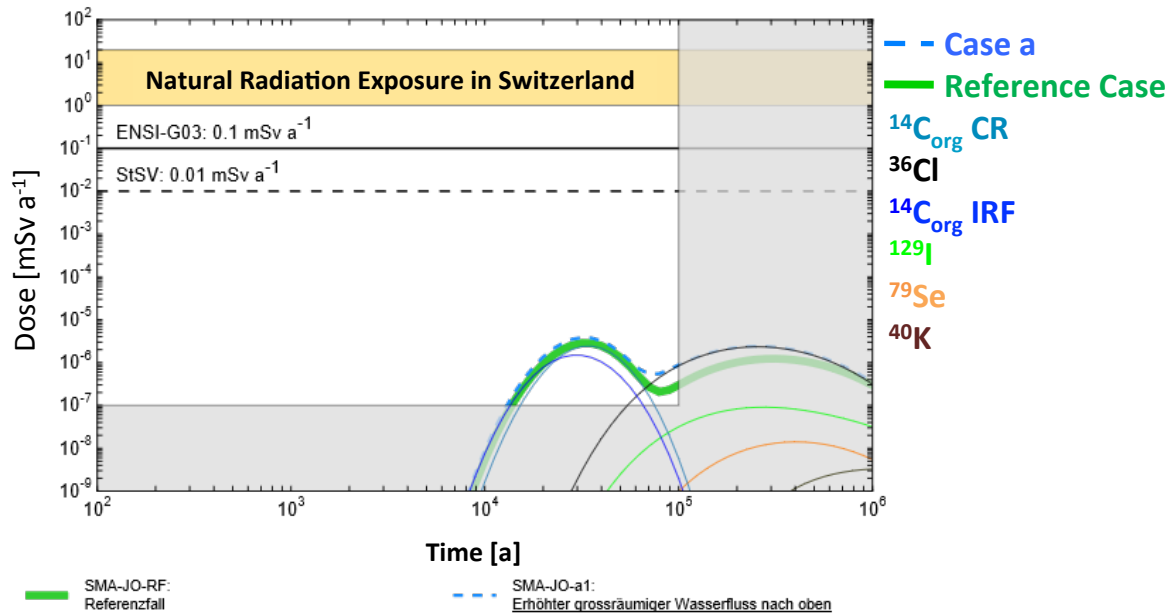




# Example L/ILW Repository Siting Area Jura East



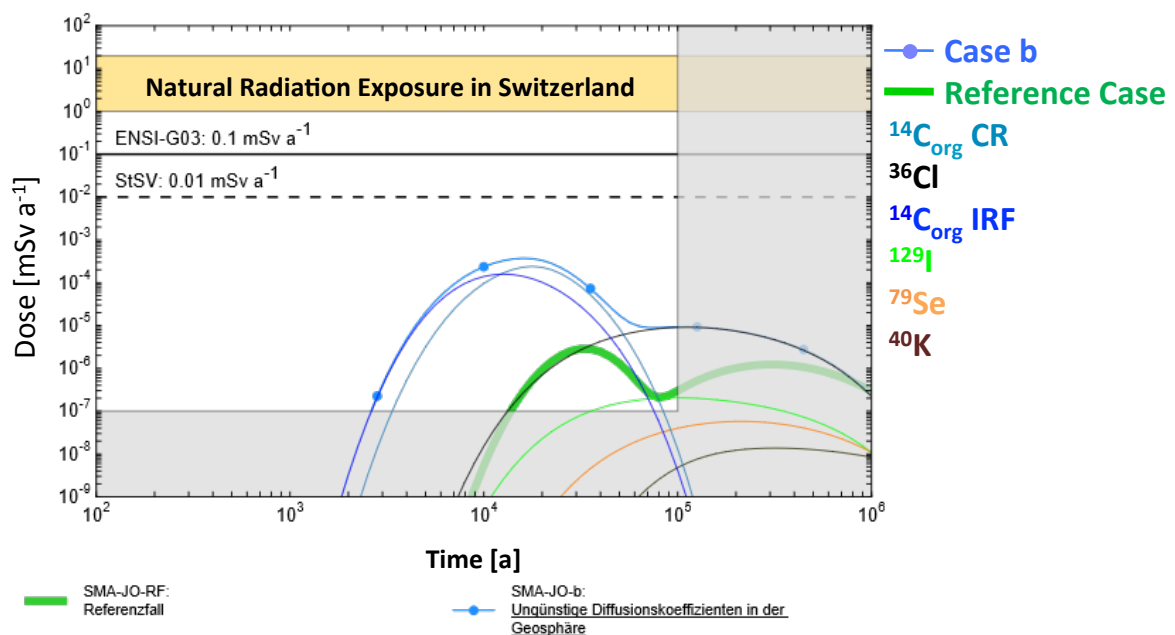
## Calculated Dose Curves: Increased Water Flux (Case a)



# Example L/ILW Repository Siting Area Jura East



## Calculated Dose Curves: Unfavourable Diffusion Coefficients (Case b)

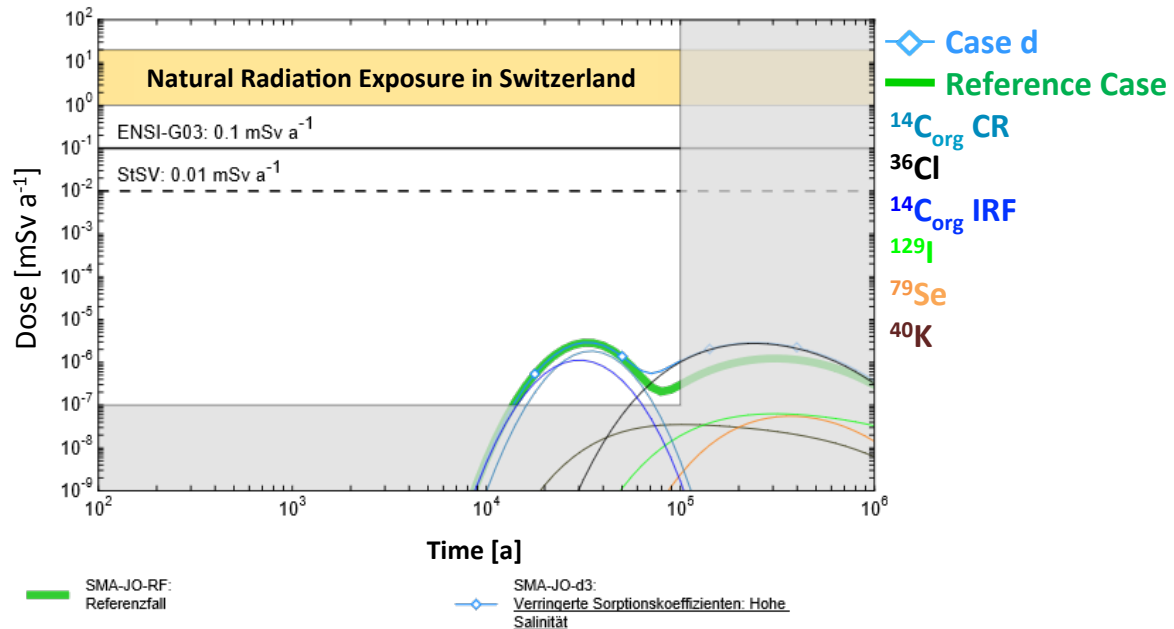




# Example L/ILW Repository Siting Area Jura East



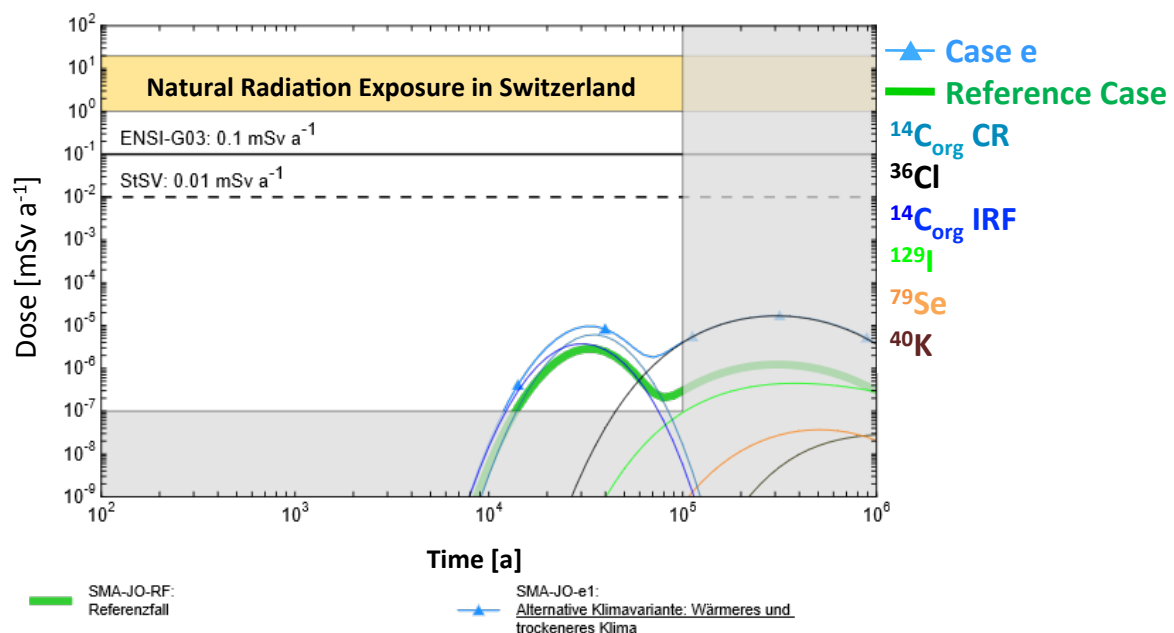
## Calculated Dose Curves: Decreased Sorption Coefficients (Case d)



# Example L/ILW Repository Siting Area Jura East



## Calculated Dose Curves: Warm and Dry Climate (Case e)





# Open questions



- Uncertainties in the inventory
- Speciation of  $^{14}\text{C}$  after release from the waste
- Stability and potential retention of the different  $^{14}\text{C}$  species in a cementitious or clayey environment
- Understanding and modelling of volatile  $^{14}\text{C}$  species through gas pathways will be reviewed and refined if necessary



## Expected contribution from CAST



- Speciation of  $^{14}\text{C}$  released from different materials
- Quantitative relation between release and corrosion rate (scale to real waste)
- Non-metallic waste forms: reduce conservatism in assumed release rate
- Validate our activation models and reduce uncertainty in inventory



Thank you for your attention!

For difficult questions:

[jens.mibus@nagra.ch](mailto:jens.mibus@nagra.ch)



# CArbon-14 Source Term CAST-workshop

Country: **SLOVENIA**

Organisation: **ARAO**

Name: **T. ŽAGAR, L. KEGEL**

This presentation **can** be used for the Proceedings of the workshop that will be published at CAST website





# Irradiated Steels



- What are the amounts you expect to have / will be generated?
  - Origin(s):

NPP Krško decommissioning waste - RPV and internals. Total expected mass of irradiated steels is 349 tons. NPP Krško is still operating and is expected to operate until 2043. Decommissioning of NPP Krško is not expected to begin before 2044.
  - Image(s) of waste (processed) package: /
  - How is the carbon-14 content determined / specified?

C-14 content in the irradiated steel of the NPP has not been measured. C-14 content in the irradiated steel was estimated using the scaling factors developed by SKB, Sweden.
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal:

C-14 activity in the irradiated steels is estimated for NPP Krško operation until 2043. The total C-14 activity is estimated to accumulate up to  $4.3\text{E}+13$  Bq ( $1,23\text{E}+8$  Bq per kg of irradiated steel)
- What is the designated end-point of this waste?

Proposed end-point is deep geological disposal facility



# Irradiated Zircaloy



- No such type of waste is existing in Slovenia
- What are the amounts you expect to have / will be generated?
  - Origin(s)
  - Image of waste (processed) package
  - How is the carbon-14 content determined / specified?
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal
- What is the designated end-point of this waste?



# Spent Ion-exchange resins



- What are the amounts you expect to have / will be generated?
  - Origin(s):  
Primary spent ion exchange resins (PR) and blow-down spent ion exchange resins (SR) from NPP Krško operation (PR resins are conditioned by in drum drying system (IDDS), SR resins are conditioned by vermiculite)
  - Image of waste (processed) package: /
  - How is the carbon-14 content determined / specified?  
For NPP operational waste C-14 content is not measured. C-14 content is estimated using the scaling factors developed by SKB, Sweden.
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal:  
Total calculated (estimated) activity of C-14 in ion-exchange resins is  $2.2\text{E}+12$  Bq. ( $2.2\text{E}+12$  Bq per  $153\text{ m}^3$  of spent ion-exchange resins)
- What is the designated end-point of this waste?  
LILW repository



# Irradiated Graphite



- What are the amounts you expect to have / will be generated?
  - Origin(s):  
Graphite reflector from Triga research reactor decommissioning. Triga reactor is still operating. Triga research reactor is expected to operate at least until 2026. No decommissioning is expected in next ten or more years.
  - Image of waste (processed) package: /
  - How is the carbon-14 content determined / specified?:  
Estimation is based on comparison with Triga reactor in Vienna
  - What is the activity of  $^{14}\text{C}$  per waste package / kilogram metal:  
Total C-14 activity in irradiated graphite is expected to total to  $3.5\text{E}+10$  Bq ( $5\text{E}+7$  Bq per kilogram of graphite).
- What is the designated end-point of this waste?  
Proposed end-point is LILW repository, final decision is still pending.





**Agenda item 22**  
**Wrap-up and closure**  
**Gunnar Buckau**

**CAST Workshop 1: Wrap-up and Closure by Gunnar Buckau (JRC) *with added comments by Erika Neeft***

The objectives of the workshop are to:

- a) Contribute to an integrated view on the management of carbon-14 containing waste between Regulators, Waste Generators and WMOs, and
- b) Further identify synergies for (future) co-operation.

1. Does the Project contribute to an integrated view / common understanding?
  - a. As formulated, could compromise integrity (of the regulator).
  - b. The project allows for mutual information of on-going work.
  - c. The project provides a possibility for regulators to express their expectations.
  - d. Link between work and safety issues not visible.
  - e. Integrated view on C-14 generation routes, speciation in the waste, speciation the disposal system, mobile species, volatile species, metabolism in the biosphere: Discussed in the workshop, but not visible from workshop information. It is outside the scope of work packages 2, 3, 4 and 5.
  - f. Specify the generation route from each waste type, in particular identify the most important one, and provide for Plausibility/Verification/Validation of the outcome.
  - g. Work on confidence in codes is included.
  - h. KIT and PSI has done speciation and compared with modelling.
  - i. Determination of nitrogen as a source for C-14 should be added to characterization of source material.
  - j. The need for code benchmarking is identified in CAST, but its implementation is beyond the CAST workplan.
  - k. Potential lack in analytical capability and competence to measure C-14 in relation to speciation: Need to further develop methods based both on extraction from material matrix and in-situ.

*In the preparation of the CAST project plan, waste management organisations and research organisations have expressed their interest. The definition of research activities in this plan is based on a mutual interest with the beneficiaries working in CAST. The workshops organised in CAST have been envisaged for European waste management organisations, European regulators and European waste generators / producers because they have an implementation responsibility. This audience for the workshop is different as a group than the beneficiaries working in CAST. Consequently, there can be a different mutual interest in research activities. In the above list, two subjects i.e. under e (integrated view on C-14 generation routes...) and j (code benchmarking) are a result of this difference in mutual interest. The following three items are also outside the scope of CAST but attempts will be made to include them in the next workshop:*

- l. There is lack of common understanding concerning representative sampling, including to which extent material investigated is representative for what.
- m. There is lack in understanding concerning far-field behaviour/migration the C-14 containing organic species (geochemical behaviour, geo-microbial fate, ).
- n. For some radionuclides and elements, there is a need to use scaling from other radionuclides: Here there is deviating views concerning confidence in the applicability.

**2. If not, how could this be achieved?**

- a. Verification/Validation of the chemical composition of C-14 containing species is difficult, specifically for resins and steel (in general, lack of material with composition and c-14....). The present project and its workprogramme cannot provide the necessary results and interpretation. Consequently, there is a need to formulate the needs for a follow-up project. Nevertheless, there is a great deal of work done and the outcome needs to be communicated better in preparation of well designed follow-up projects.
- b. There is a great deal of work done, needs to be communicated and disseminated better (high on the agenda for next WS)

**3. Next WS\*:**

- a. Representativity of samples and lab results.
- b. Up-scaling from lab-scale to real system.
- c. Status and conclusions concerning speciation of C-14 containing compounds in the source material and through the biosphere, in particular from comparison of the speciation and modelling with respect to C-14 compounds in all parts of the system. For that purpose, identify and invite analytical competence for characterisation of C-14 species also from outside the project.
- d. Transfer of Knowledge and its application to the Safety Case.
- e. It is presently felt that Knowledge is not sufficiently mature for providing training. Identify sufficiently mature areas. Where there is sufficient confidence, consider providing training after the second workshop.
- f. Identify and convey positive messages: Emphasize that we have good Knowledge that has increased through the CAST project, and present areas that would benefit from further investigations (preparation for follow-up project).

**4. Are there synergies between the actors from the Project?**

- a. The workshop is an important contributor to Knowledge sharing, but the Knowledge sharing to a broader audience and on a longer term is mainly through reports and published output.
- b. One should consider if the synergy of the different types of actors could contribute to identifying what is actually needed for a Safety Case.

**5. Is the Project progressing in accordance with (your) expectations?**

- a. Identify safety relevance: Revisit with all actors for final reporting. This includes the numerical analysis but also the confidence in the scientific basis (well founded, substantiated with robust arguments - process understanding)
- b. Elaborate on the management of uncertainty.

\*: the next/second/final CAST workshop will take place around Jan. to March 2018. All project partners, including associated groups, End-User groups,..., are encouraged to provide proposals for topics to be dealt with (e-mail to Erika Neeft).