DE LA RECHERCHE À L'INDUSTRIE

WORK PLAN FOR IMPROVING THE DARWIN2.3 DEPLETED MATERIAL BALANCE CALCULATION CONCERNING SOME IMPORTANT ISOTOPES FOR FUEL CYCLE

ATOMS FOR THE FUTURE – TECHNICAL SESSIONS

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JUNE 28TH, 2016

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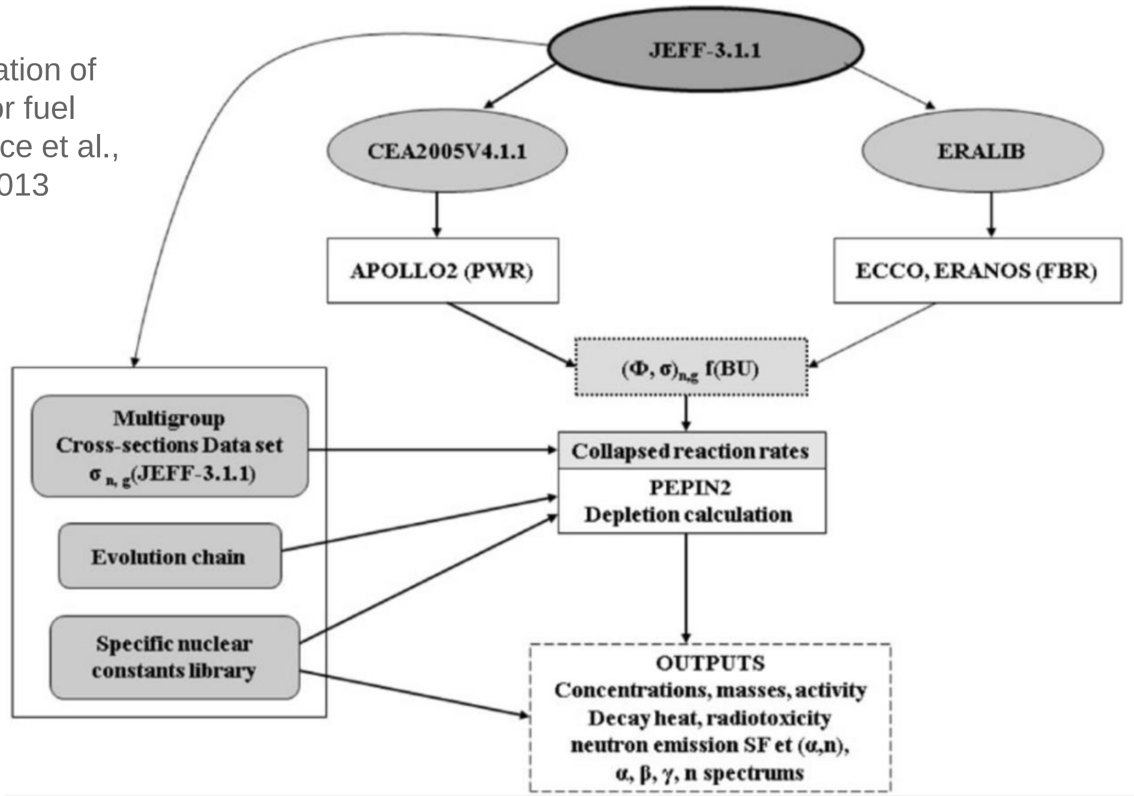
INTRODUCTION AND CONTEXT

CONTEXT – THE DARWIN PACKAGE

- DARWIN2: reference package for fuel depletion calculations – developed by CEA\DEN and industrial partners
  → Based on APOLLO2 / ERANOS2 for deterministic neutron transport calculation, and DARWIN/PEPIN2 for depletion calculation

Source: Experimental validation of the DARWIN2.3 package for fuel cycle applications, San-Felice et al., Nuclear Technology, 184, 2013



CONTEXT – DARWIN EXPERIMENTAL VALIDATION

Post-Irradiation Experiment (PIE) : fuel pellet dissolution + chemical analysis

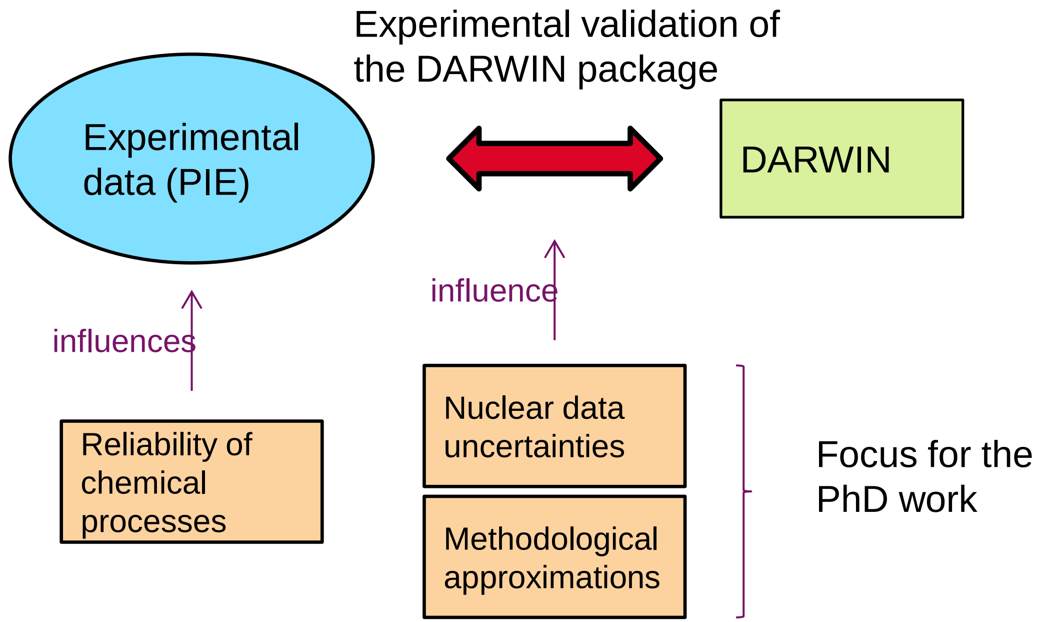
ATALANTE hot cell
Source: International conference "ATALANTE 2016" record on cea.fr

DARWIN2 package is experimentally validated
→ calculation vs experiment ("C/E" or "(C-E)/E") values
→ Large French fuel data base (UOX and MOX PWR)

PWR	Burn-up (GWj/t)	239Pu/238U		137Cs/238U	
		(C-E)/E (in %)	σ (in %)	(C-E)/E (in %)	σ (in %)
UOX Gravelines 4.5% 235U	25	**-3.2**	0.7	**-7.5**	2.2
	40	**-1.9**	0.9	**-6.4**	2.3
	50	**-0.2**	1.0	**-6.9**	2.1
	60	**0.0**	0.7	**-6.0**	1.5
MOX SLB1 5.6% 239Pu	10	**0.4**	0.6	**-2.4**	1.5
	28	**0.5**	0.8	**-0.7**	1.2
	40	**0.2**	0.9	**-1.2**	1.1

Source: Experimental validation of the DARWIN2.3 package for fuel cycle applications, San-Felice et al., Nuclear Technology, 184, 2013

CONTEXT – DARWIN EXPERIMENTAL VALIDATION



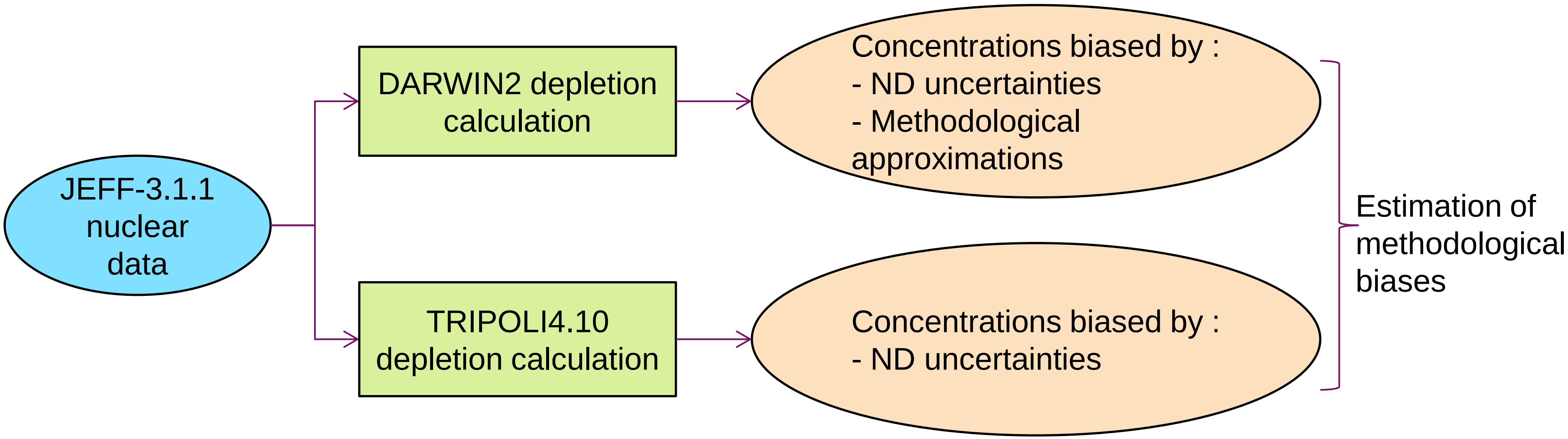
- Several ways to improve the calculation of isotopic concentrations in the DARWIN2 package:
  - Proposing new experiments to improve isotopic concentration measurements
  - Improving nuclear data: integral experiments, cross sections experimental validation, analytic experiments, production of covariance data…
  - Quantifying potential methodological biases induced by the deterministic approximations in the DARWIN package resolution scheme

QUANTIFICATION OF METHODOLOGICAL BIASES

TRIPOLI4.10 DEPLETION CALCULATIONS

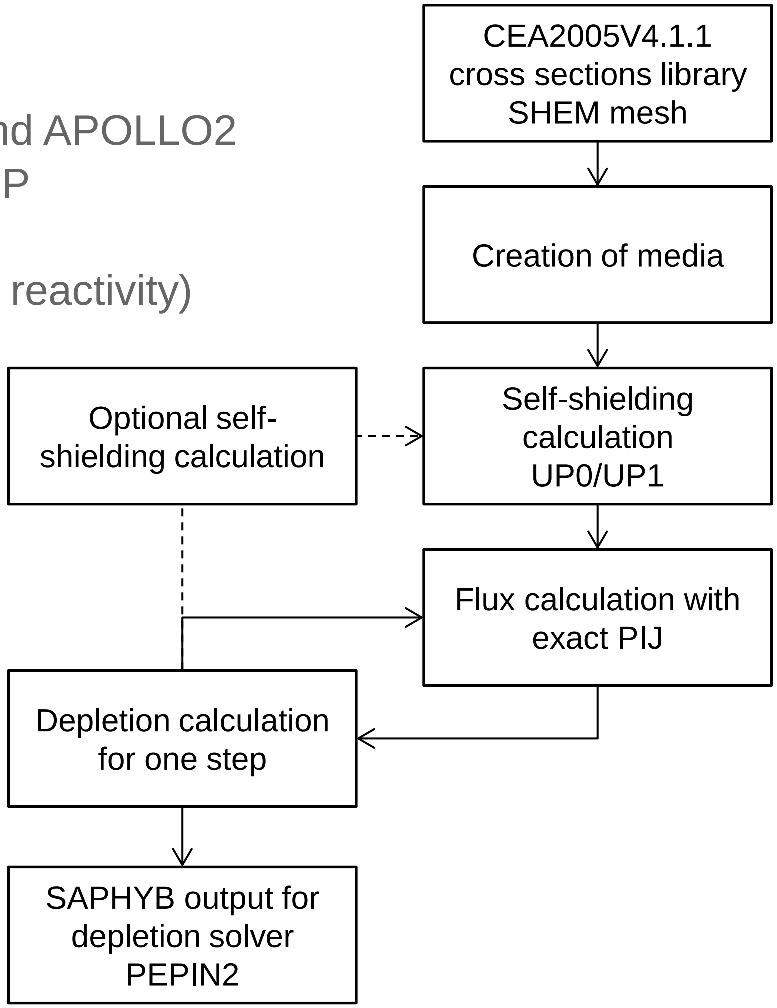
- Monte-Carlo method currently used for reference stationary calculations
  → Few modelling approximations compared to deterministic methods
  → Residual biases supposed only due to nuclear data

- Recent advances in coupling:
  - TRIPOLI4 stochastic transport code (developed by CEA and its industrial partners)
  - MENDEL new depletion solver developed by CEA (new software architecture)



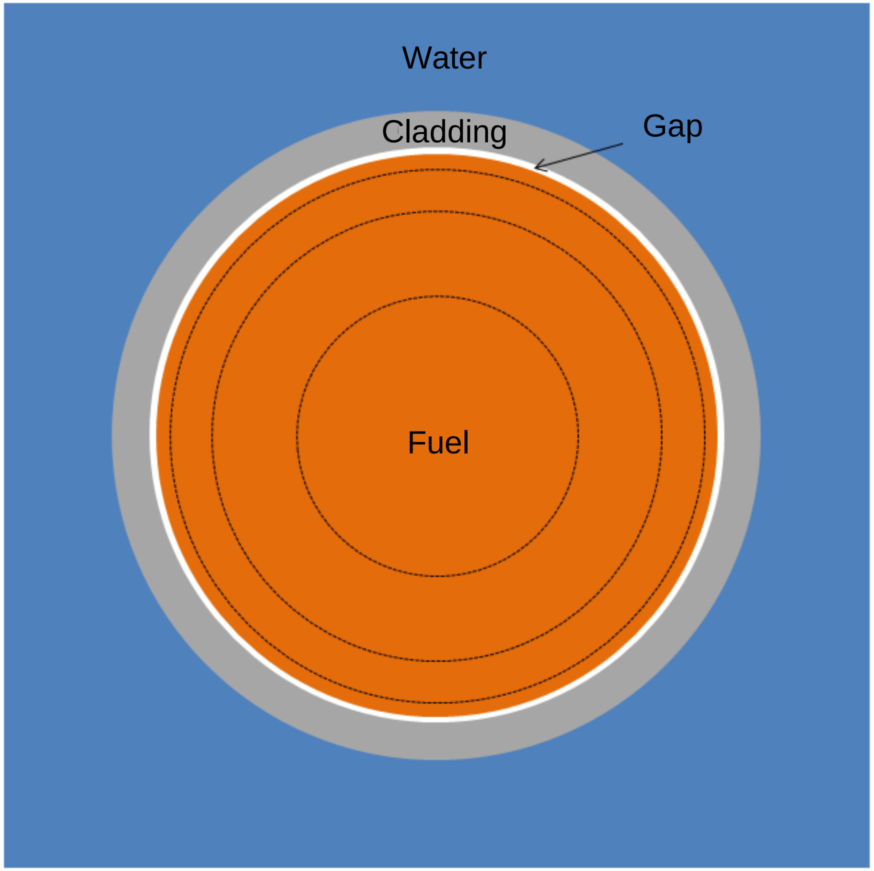
APOLLO2 CALCULATION SCHEME

- CYCLE2008-REP calculation scheme used for PWR depletion calculation in the DARWIN package
- Comparison made between TRIPOLI4.10 and APOLLO2 (instead of DARWIN) using CYCLE2008-REP
- 160 isotopes calculated (optimized chain for reactivity)



COMPARISON AGAINST APOLLO2

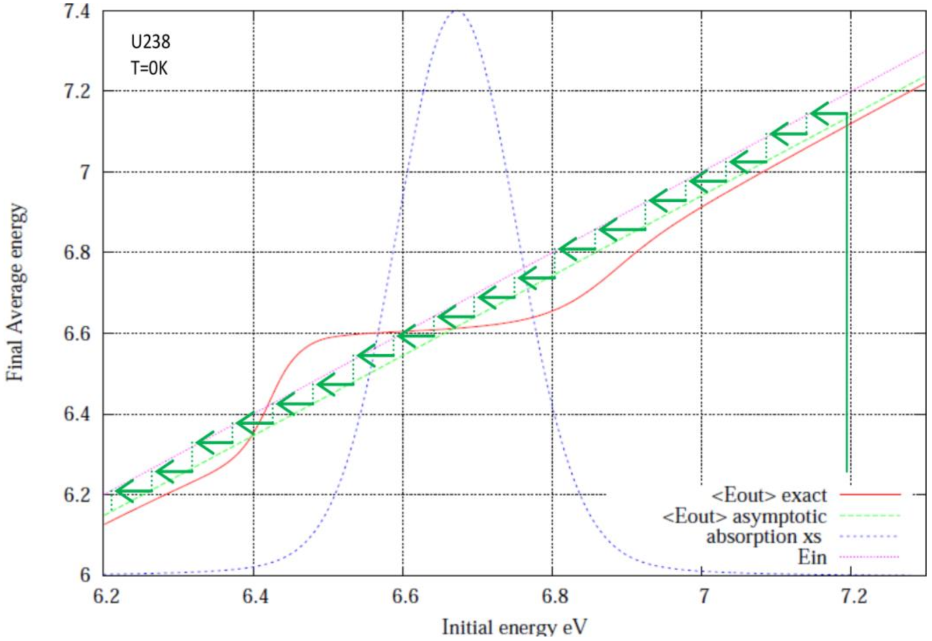
- 3.7% enriched PWR UOX cell irradiated until 60GWd/t, divided in four sections to take into account spatial flux heterogeneities
- Cross sections library based on JEFF-3.1.1
- Work in progress
  → Paper for ND2016

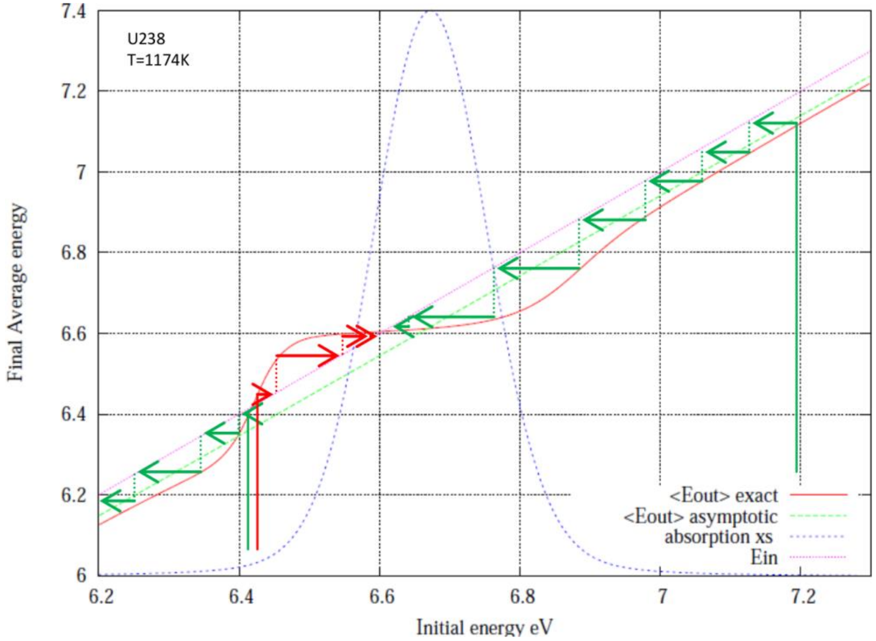


RESONANT UPSCATTERING PHENOMENON

- Possible loss of lethargy around nuclides resonances
  → Historically neglected for heavy nuclides
  → Assessment of its influence on isotopic fuel calculations

- Originally requested by AREVA for BWR reactor calculations, then developed and integrated in APOLLO2

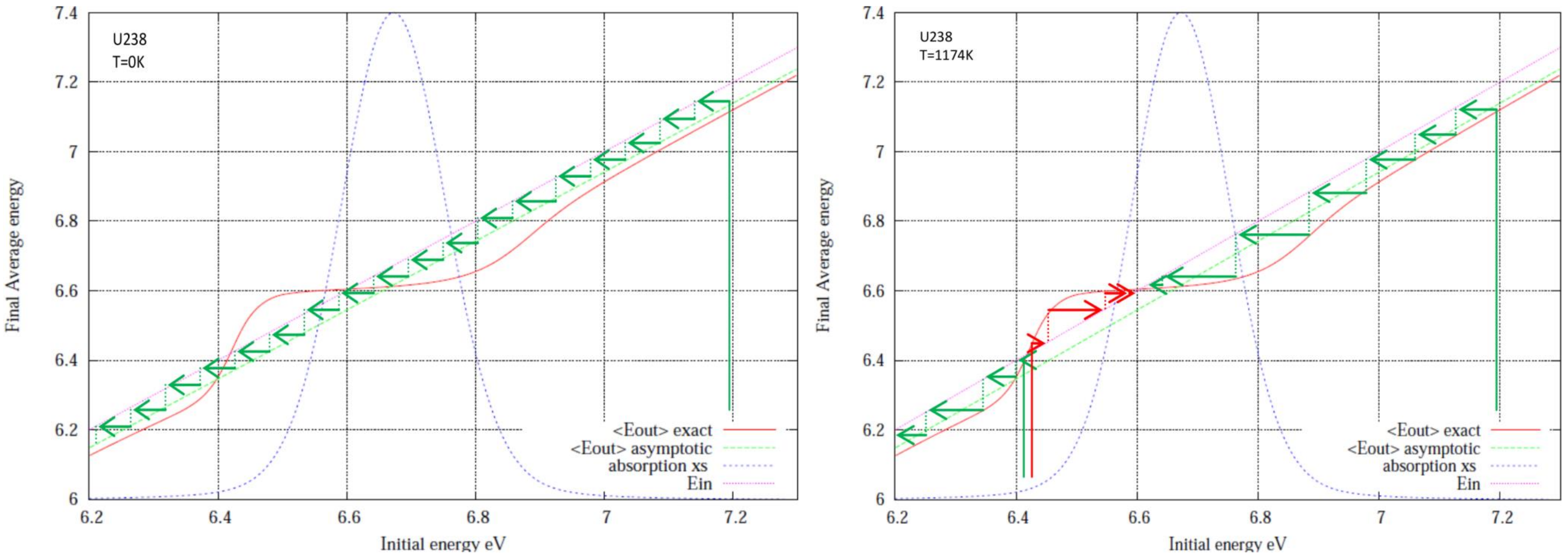




RESONANT UPSCATTERING PHENOMENON

- Physical consequences:
  - Higher capture rate → Loss of reactivity (low burnup)
  - Higher 239Pu production rate → Gain of reactivity (high burnup)
  - Higher production of minor actinides (Curium for inst.)
  - Fission products production rates will be different

- Work in progress → Paper for ND2016



EXPERIMENTAL VALIDATION OF SOME NUCLEAR DATA

PRINCIPLE AND OVERVIEW

- Feasibility study of an irradiation experiment in the MINERVE experimental reactor at Cadarache to validate important nuclear data for fuel cycle calculations
- Principle : dedicated experiment to assess nuclear data involved in the buildup of important isotopes
  → Concentration calculation of some important isotopes with DARWIN is not experimentally validated

14C example
ß- emitter (mean energy 49.5 keV)
half life: 5700 years

Major buildup contribution for PWR UOX and MOX fuels :
56% from 17O(n,α)
39% from 14N(n,p)
3% from ternary fissions

THE 14C CASE - OVERVIEW

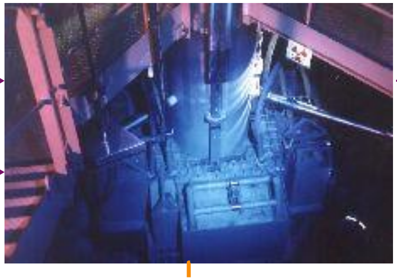
Initial composition

Neutron spectrum characteristics + fluence

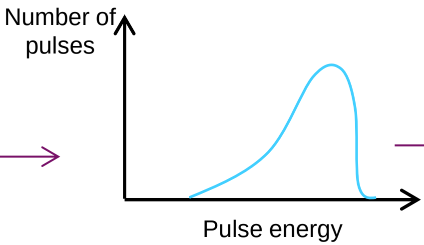
Estimation of 14N(n,p)

14N-enriched material (without 17O)
ex : C3H6N6

MINERVE experimental reactor



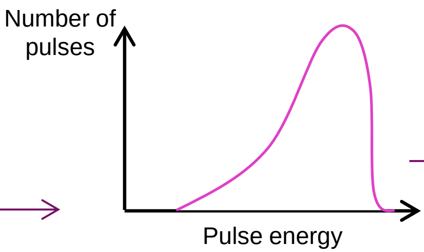
14C production only *via* 14N(n,p)



Measurement of 14N(n,p)-produced 14C

17O-enriched material (without 14N)
ex : enriched water

14C production only *via* 17O(n,α)



Measurement of 17O(n,α)-produced 14C

Initial composition

Neutron spectrum characteristics + fluence

Estimation of 17O(n,α)

CONCLUSION AND PERSPECTIVES

CONCLUSION AND PERSPECTIVES

- Ways of improving inventory calculation in DARWIN for this PhD :
  - Quantification of methodological biases → TRIPOLI4 depletion calculations
  - Improvement on nuclear data → feasibility study for an experimental validation of nuclear data

- Long-term objectives:

  - Quantification of methodological biases on assembly calculations + comparison with data provided in the DARWIN2.3 experimental validation report

  - Improvement on nuclear data with integral experiment assimilation, production of covariance data (to propagate nuclear data uncertainties on isotopic concentration calculation for non qualified isotopes)

  - Combination of these studies results to improve inventory calculation in DARWIN for important fuel cycle isotopes

Thank you for your attention ! ☺

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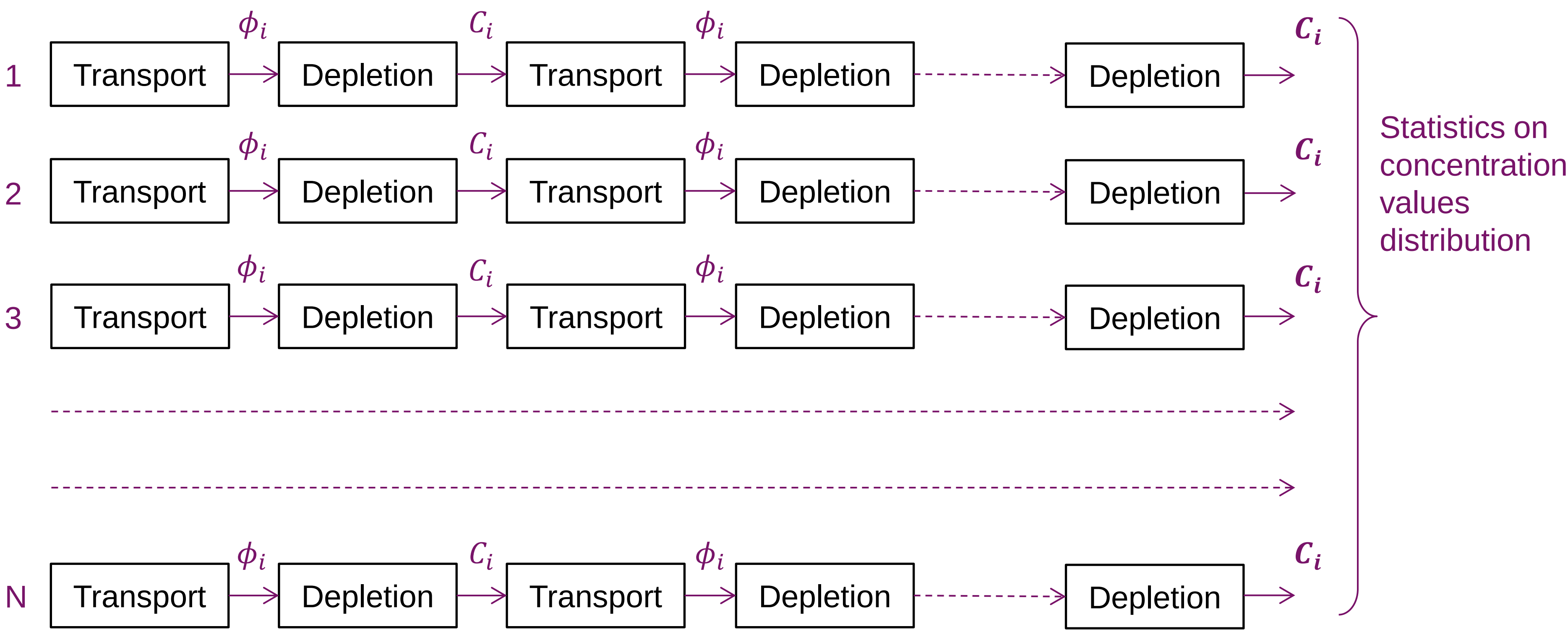
Laboratoire d'Etude du Cycle

TRIPOLI4.10 DEPLETION CALCULATIONS

Calculation based on N independent replicas with different random seeds

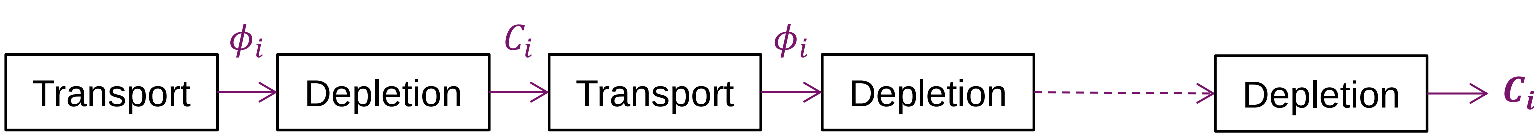


TRIPOLI4.10 DEPLETION CALCULATIONS

Some important sources of bias in Monte-Carlo depletion calculation
Transport calculation → statistic biases due to a finite number of simulated neutrons
Depletion calculation → systematic biases due to deterministic resolution of Bateman equation
Several burnup steps → systematic biases due to propagation of others sources of uncertainty along the depletion calculation

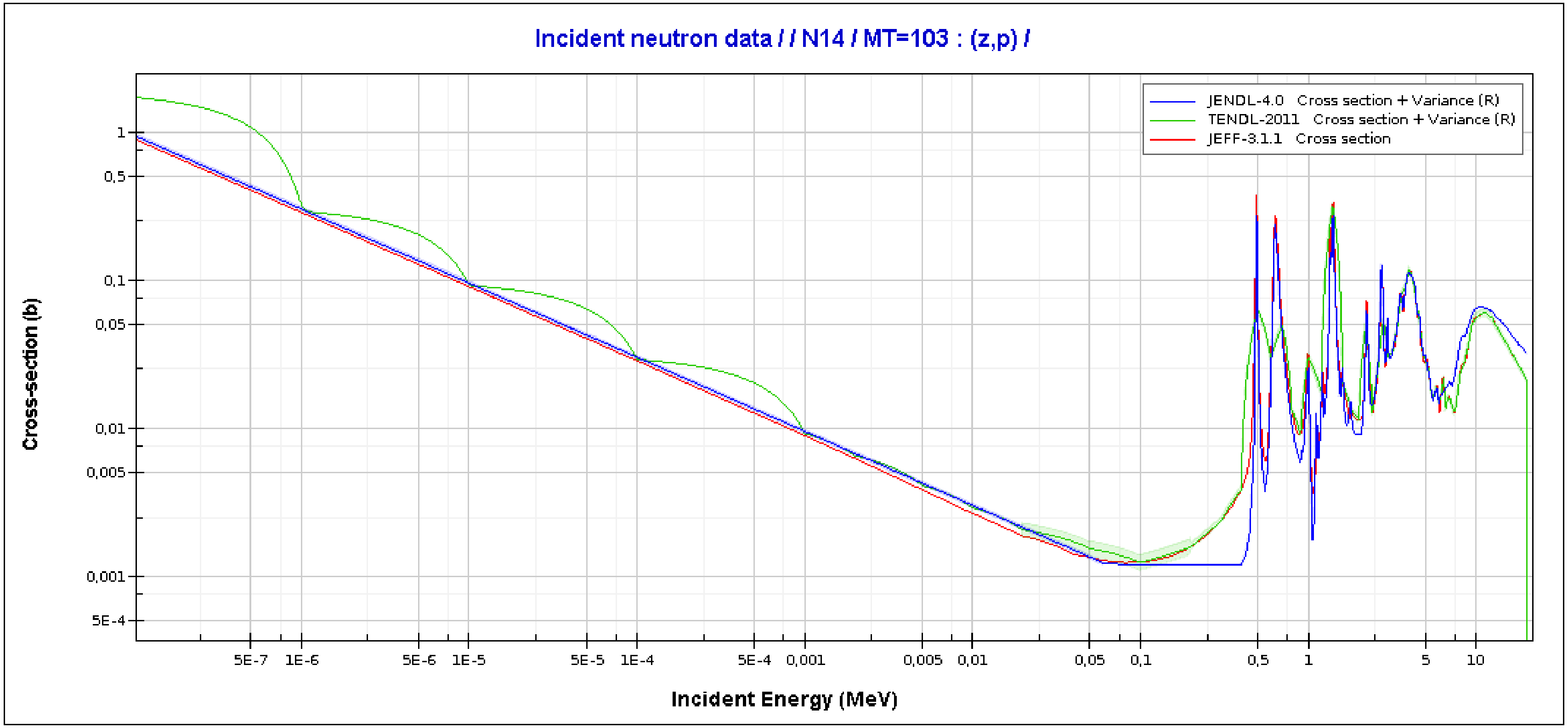
- Independent replicas method used to compute a statistical distribution of the TRIPOLI4 calculated concentrations

- E. Brun et al., *Systematic Uncertainty due to Statistics in Monte Carlo burnup codes: application to a simple benchmark with TRIPOLI4-D*, Nucl. Sci. And Tech., **2**, pp.879-885
→ becomes negligible for a sufficient number of simulated particles per simulation



THE 14C CASE

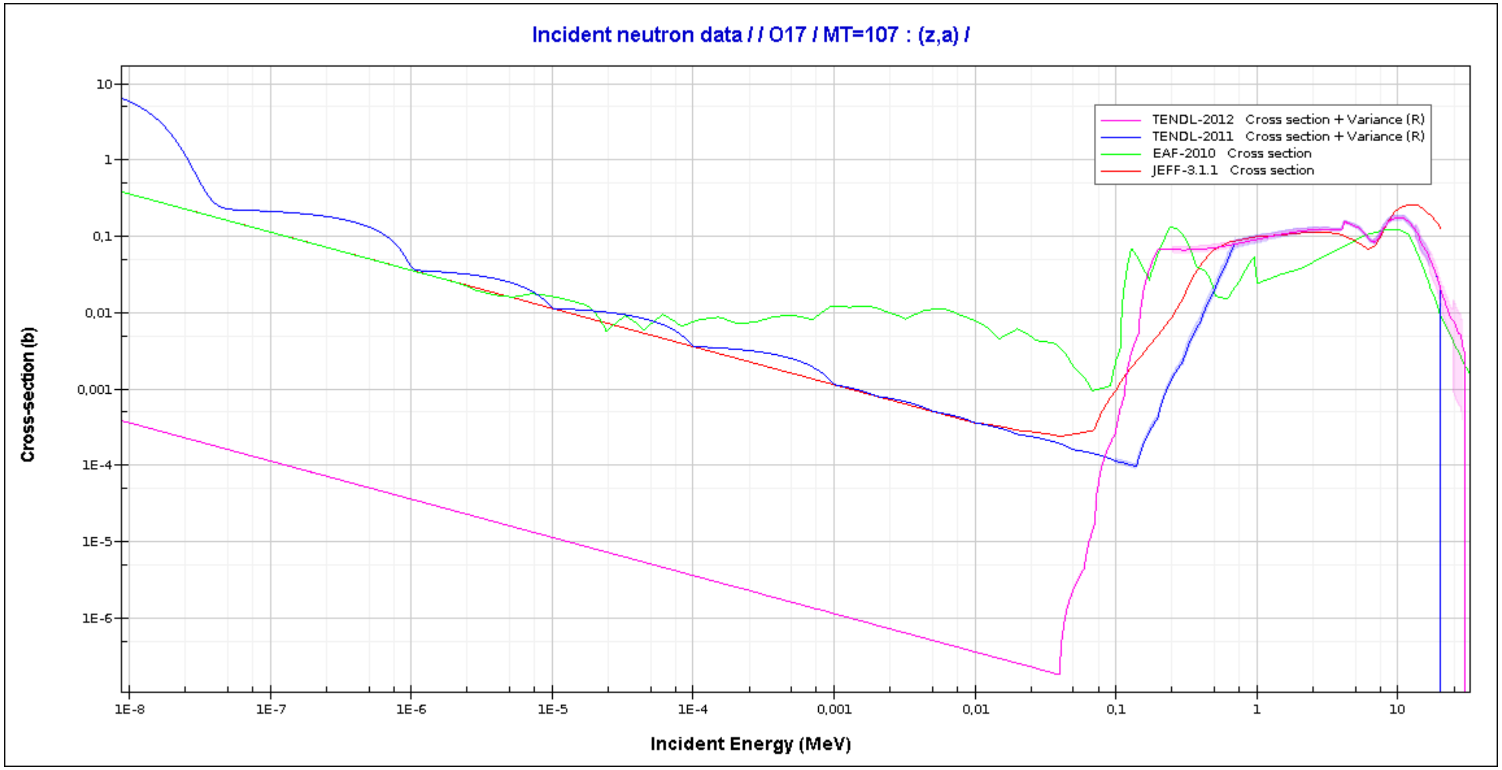
- Importance of the 14N(n,p) validation : discrepancies between different evaluations → to compare with 2,9% (1σ)



- For this cross section, JEFF-3.1.1 = EAF-2010 = ENDF-BVII.1 = TENDL-2012

THE 14C CASE

- Importance of the 17O(n,α) validation : discrepancies between different evaluations → to compare with 8,4% (1σ)

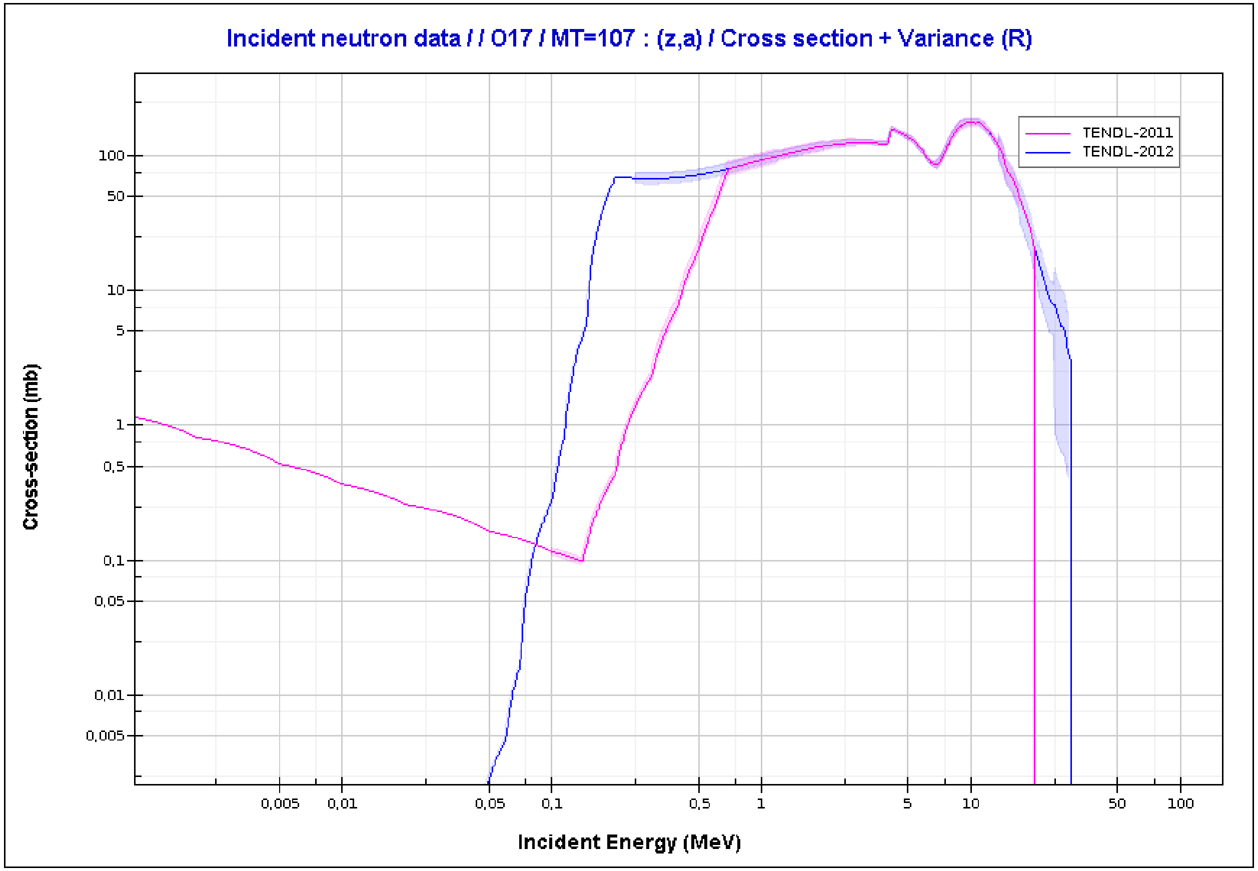


- For this cross section, JEFF-3.2 = TENDL-2012

THE 14C CASE

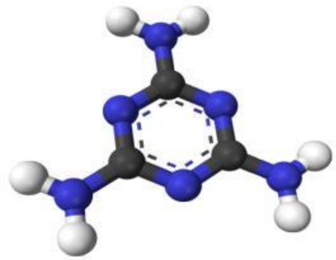
- Uncertainty data only available for fast neutrons…

- Importance of the 17O(n,α) validation : discrepancies between different evaluations
  → to compare with 8,4% (1σ)



THE 14C CASE

14N(n,p) cross section



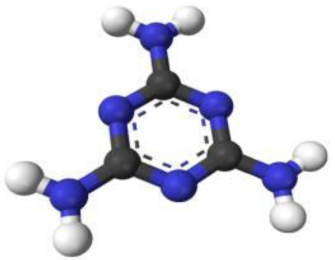
- Feasibility study of the irradiation of melamine (C3H6N6, 1,57g/cm3) in MINERVE with a PWR-like neutron spectrum, to produce enough 14C to detect and measure its concentration
  → No 17O in the melamine

- 14C is a β- emitter (mean energy : 49,5 keV – max energy : 156 keV) with a 5700 years half-life :
  → Chemical process selected t: liquid scintillation

- Experimental requirements (discussions with LARC)
  - Target activity = 1 Bq (precision ~5%)
  - Avoid 3H formation : measurement perturbation

THE 14C CASE

14N(n,p) cross section



- DARWIN calculation of 14C and 3H activity in MINERVE with R1-UO2 core configuration with CEAV5.1.2 library, for a 6 hours irradiated melamine sample : 0,65 Bq/cm3
- Several irradiation cycles
  → higher activity for better results
  → fluence monitoring with Au neutron activation dosimetry
- Non negligible tritium activity : 0,44 Bq/cm3
  → Separation of 14C and 3H

THE 14C CASE

17O(n,α) cross section

- Feasibility study of the irradiation of 17O-enriched water (80%) in MINERVE with a PWR-like neutron spectrum, to produce enough 14C to detect and measure its concentration
  → No 14N in the enriched water

- Experimental requirements (discussions with LARC)
  - Target activity = 1 Bq (precision ~5%)
  - Avoid 3H formation : measurement perturbation

17O(n,α) cross section

- DARWIN calculation of 14C and 3H activity in MINERVE with R1-UO2 core configuration with CEAV5.1.2 library, for of 6 hours irradiated enriched water sample : 0,102 Bq/cm3

- Several irradiation cycles
  → higher activity for better results
  → fluence monitoring with Au neutron activation dosimetry

- Negligible tritium activity : 0,001 Bq/cm3
  → No separation of 14C and 3H