

Italian National Agency for New Technologies, Energy and Sustainable Economic Development

157Gd and 155Gd (n,g) xs Project

From new measurements to new evaluations

JEFF Meeting 2017, OECD-NEA, April 26th 2017

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Structure of presentation

- Introduction
- Scientific motivations
- Scientific background
- ENEA S/U analysis for LWRs
- Experimental campaign at NTOF
- Further experiments
- Future developments of the project



Introduction

- At the Italian national nTOF meeting in March 201: idea of dedicating scientific efforts towards a better (n,g) xs for reactor applications.
- While present evaluations of these xs perform acce for improvements.
- At the international nTOF meeting in May 2015 in S Proposal to the ISOLDE and nTOF Committee was and positively accepted, opening the path to dedica experiments in Summer 2016.
- The Proposal received endorsements by, among or
- Later on, statements of interest arrived also from IF





Introduction

- Several persons and institutions are involved in the whole project.
- People involved at ENEA Bologna:
 - F. Rocchi (reactor physics aspects)
 - D. M. Castelluccio (experiments at nTOF and data analysis)
 - A. Guglielmelli (reactor physics aspects)
 - G. Clai (data analysis)
 - S. Lo Meo (spokesperson for Proposal at nTOF)



Scientific motivations

The main motivation is related to the use of "burnable neutron poisons" in nuclear reactors

- To increase the efficiency and economic performances of reactor fuel, it is necessary to increase the initial enrichment of ²³⁵U in the fuel itself.
- However high enrichments pose severe safety problems due to the high initial excess reactivity.
- This can be **inherently compensated** by loading the fuel with **"burnable neutron poisons"**, i.e. isotopes with very high capture cross section, that are depleted together with the fissile isotopes.

It is very important to assess the capture behavior of burnable poisons in order to evaluate:

- the economic gain due to the extension of fuel life;
- the **residual reactivity penalty** at EOL, in terms of reactor days lost (16 pins Gd-doped FAs for PWR = 5 full power days lost/year = 8 M€ for the electricity market in France);
- the **reactivity peak** for partially spent fuel for the criticality safety evaluations of Spent Fuel Pools.

Use in Gen. II & Gen. III Reactors

Current Gen. II and Gen. III nuclear reactors make extensive use of Gadolinium as:

- burnable neutron poison (Gadolinia: Gd₂O₃) for PWR, BWR, VVER fuels
- emergency shutdown poison (Gadolinium nitrate, GdNO₃), for CANDU.

The reason of this choice is the **extremely high neutron capture cross sections** of the odd Gd isotopes (155 Gd and 157 Gd) for low energy neutrons (thermal to $\approx 10 \text{ eV}$).





Use in CANDU Reactors

Emergency Shutdown Poison

- In CANDU reactors, in case of severe accidents due to or leading to criticality excursions, Gadolinium nitrate is injected into the moderator heavy water, to reduce (eliminate) criticality risks or excursions.
- However, uncertainties in the (n,γ) cross section of Gd odd isotopes may **impose special care** in the **safety** calculations for the licensing of CANDU reactors.





157Gd(n,g) thermal

Despite their importance, the capture cross sections of the odd Gd isotopes have not been so extensively studied and are **not known with the accuracy required** by present-day nuclear industry.

| Reference | Year | Thermal xs (b) | Deviation from ENDF/B-VII |
|--|------|--------------------|---------------------------|
| Pattenden 2 nd At. En. Conf. Geneva, 16 | 1958 | 264000 | +3.9% |
| Tattersall Jour. Nucl. Ener. A 12, 32 | 1960 | 213000 | -20% |
| Moller Nucl. Sci. Eng. 8, 183 | 1960 | 254000 | = |
| Groshev Izv. Akad. Nauk, SSSR, 26, 1118 | 1962 | 240000 | -6% |
| Sun J. Radioanal. Nucl. Chem. 256, 541 | 2003 | 232000 | -9% |
| Leinweber Nucl. Sci. Eng. 154, 261 | 2006 | 226000 | -12% |
| Mughabghab Evaluation (adopted in ENDF/B-VII) | 2006 | $254000 \pm 0.3\%$ | = |
| Choi Nucl. Sci. Eng. 177, 219 | 2014 | 239000 | -6% |

ENEL

CEA Melusine/GEDEON-II results



CEA Qualification Program for French LWR using the Melusine reactor in Grenoble in 1985 (2015 re-analysis based on JEFF 3.1.1 evaluations for EPR).







CEA Melusine/GEDEON-II results

| | lso | tope Conce | ntratior | ns (C/E | -1) | | | |
|--|--|---|--|--|--|--|--|-------|
| Nature | Position | Consumption [%] [¹⁵⁵ Gd] | ¹⁵² Gd/ ²³⁸ U [%] | ¹⁵⁴ Gd/ ²³⁸ U [%] | ¹⁵⁵ Gd/ ²³⁸ U [%] | ¹⁵⁶ Gd/ ²³⁸ U [%] | ¹⁵⁷ Gd/ ²³⁸ l [%] | |
| $UO_2 + Gd_2O_3$ | D07-270 [front | 26 | -3±5 | - 2 ± 1 | 1±3 | -3±1 | 7±5 | |
| 5% | UO ₂ + Gd ₂ O ₃ 5%] | | | | | | | |
| | D07-100 [front | 58 | -4±5 | -1±1 | -2 ± 2 | -1 ±1 | 2 ± 3 | |
| | $UO_2 + Gd_2O_3 5\%$ | | | | | | | 1. |
| | G04-270 [front H ₂ O] | 23 | -11 ± 5 | -1±1 | 0±6 | -2±1 | 9±10 | Larg |
| | G04-100 [front H ₂ O] | 32 | -12 ± 5 | -1±1 | 0±3 | -2±1 | 8±5 | |
| UO ₂ + Gd ₂ O ₃ 8% | D04-270 [angle H ₂ O] | 40 | -7±5 | -1±1 | 4±2 | -3±1 | 10±2 | diffe |
| | D04-100 [angle H ₂ O] | 33 | -8±5 | -1±1 | 6±1 | -4±1 | 14 ± 2 | hoty |
| | G04-270 [front H ₂ O] | 40 | -8±5 | 0±1 | 1±1 | -1±1 | 6±2 | Derv |
| | G04-080 [front H ₂ O] | 34 | -6±5 | -1±1 | 0±1 | -1±1 | 3 ± 2 | |
| | D07-270 [front | 53 | -11 ± 5 | -1±1 | 3 ± 2 | -3±1 | 13 ± 3 | |
| | $UO_2 + Gd_2O_3 8\%$ | | | | | | | · · |
| | D07-100 [front | 46 | -8±5 | -1±1 | 2 ± 2 | -2±1 | 10±2 | and |
| | UO ₂ + Gd ₂ O ₃ 8%] | | | | | | | Giria |
| | D10-270 [angle H ₂ O] | 66 | -12 ± 5 | -1±1 | 3±3 | -2±1 | 13 ± 4 | avna |
| | D10-100 [angle H ₂ O] | 58 | -9±5 | 0±1 | 1±3 | -2±1 | 8±3 | CAP |
| | K10-270 [angle H ₂ O] | 87 | -15 ± 6 | -1±1 | 17±9 | -3±1 | 43 ± 15 | |
| | K10-100 [angle H ₂ O] | 78 | -15 ± 5 | 0±1 | 1±4 | -1±1 | 10±6 | |
| | G10-270 [front H ₂ O] | 88 | -17 ± 6 | 0±1 | 9 ± 10 | -2±1 | 24 ± 17 | |
| | G10-100 [front H ₂ O] | 78 | -18 ± 5 | 0±1 | 4±5 | -1±1 | 16 ± 7 | |
| | K04–270 [angle H ₂ O] | 85 | -15 ± 5 | 0±1 | 7±9 | -1±1 | 25 ± 16 | |
| | K04-100[angle H ₂ O] | 75 | -14 ± 5 | -1±1 | -4±6 | -1±1 | 3±8 | |
| | K07-270 [front | 97 | -17 ± 5 | 0±1 | 56 ± 30 | -1±1 | 9 ± 40 | |
| | UO ₂ + Gd ₂ O ₃ 8%] | | | | | | | |
| | K07-100 [front | 89 | -15 ± 5 | 0±1 | 9±11 | -1±1 | 24 ± 20 | |
| | UO ₂ + Gd ₂ O ₃ 8%] | | | | | | | |

Large differences between calculations and experiment



CEA Melusine/GEDEON-II results

| ble 6 EL2005-11 rings/experi | iment compa | | cai cai | C.I. | | |
|---|--|---|--|--|--|-----------------|
| Nature | Positic | | Contents lists av | vailable at ScienceDirect Nuclear Energy | Marine of RELEDENT EXERCITY | |
| UO ₂ + Gd ₂ O ₃ 8% | D04-2 D04-1 C04-2 | ELSEVIER jo | urnal homepage: wwv | v.elsevier.com/locate/anucene | | Discharge 4GWd/ |
| | G04-0 D07-2 D07-1 D10-2 D10-1 K10-2 | Qualification of gadolin of MELUSINE/GEDEON- David Bernard *, Alain Santam CEA, DEN, DER, SPRC, Cadarache, F-13108 Saint-P | ium burnable po II spent fuel ana aarina ^{Jul-Lez-Durance, France} | oison: Interpretation lysis | 2015 !! | Discharge 7GWd/ |
| | K10-100 G10-270 G10-100 | [angle H ₂ O] [front H ₂ O] [front H ₂ O] | 78 88 78 | -0.2 ± 1.5 -1.0 ± 1.0 -0.8 ± 1.0 | -0.9 ± 1.5 -0.7 ± 1.0 -1.3 ± 1.4 | Discharge 10GWd |
| | K04-270 K04-100 K07-270 K07-100 | [angle H_2O] [angle H_2O] [front $UO_2 + Gd_2O_3 8\%$] [front $UO_2 + Gd_2O_3 8\%$] | 85 75 97 89 | $\begin{array}{c} -0.9 \pm 1.0 \\ 1.1 \pm 1.0 \\ -1.1 \pm 0.5 \\ -0.9 \pm 1.0 \end{array}$ | $\begin{array}{c} -1.0 \pm 1.0 \\ -0.4 \pm 1.0 \\ 0.0 \pm 0.3 \\ -0.6 \pm 1.0 \end{array}$ | Discharge 12GWd |

Some non-negligible biases are found for 157Gd, suggesting an underprediction of the capture xs.



AECL - Chalk River results

NUCLEAR DATA AND THE EFFECT OF GADOLINIUM IN THE MODERATOR

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AECL - Chalk River results





PSI 2008-2009 Validations for BWR Fuels

- PSI C-E comparisons using the <u>Leinweber data of 2006</u> within the PROTEUS reactor research programs (SVEA96+).
- Results for the total fission rate and 238U capture rate are generally much improved wrt using previous Gd evaluations.

<u>BUT</u>...

• ICSBEP LCT-035, LCT-005, HST-014 not well reproduced (JEFFDOC-1210, 2007)





PSI 2008-2009 Validations for BWR Fuels

| ICSBEP | Config. | K_ref | ENDF/B-VII | JEFF-3.1 | Leinweber | Improvement? |
|---------|---------|--------|------------|----------|-----------|--------------|
| HST-014 | C2 | 1.0000 | 1.00996 | 1.01304 | 1.01903 | N |
| | C3 | 1.0000 | 1.01827 | 1.01852 | 1.02636 | N |
| LCT-035 | C3 | 1.0000 | 0.99591 | 0.99556 | 0.99935 | Y |
| LCT-005 | C2 | 1.0000 | 1.00029 | 1.00006 | 1.00466 | N |
| | C3 | 1.0000 | 0.99907 | 1.00002 | 1.01651 | N |
| | C4 | 1.0000 | 0.99721 | 0.99846 | 1.01602 | N |
| | C6 | 1.0000 | 1.00684 | 1.00697 | 1.00962 | N |
| | C7 | 1.0000 | 1.00191 | 1.00258 | 1.00846 | N |
| | C8 | 1.0000 | 1.00163 | 1.00295 | 1.01213 | N |
| | С9 | 1.0000 | 1.00257 | 1.00379 | 1.01459 | N |
| | C10 | 1.0000 | 1.00135 | 1.00290 | 1.01474 | N |
| | C11 | 1.0000 | 1.00165 | 1.00342 | 1.01544 | N |
| | C13 | 1.0000 | 1.01309 | 1.01129 | 1.01303 | N |
| | C15 | 1.0000 | 1.01751 | 1.01750 | 1.02436 | N |



van der Marck 2012 Analysis

In 2012 S. C. van der Marcl ENDF/B-VII.1, JENDL-4.0, benchmarks (mainly ICSBE

The conclusion about Gd is to represent the experiment

| LSEVIER | Available online at www.sciencedirect.com SciVerse ScienceDirect Nuclear Data Steets 113 (2012) 2935-3005 www.sleevier.com/locate/uls | l cor sing |
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comprehensive analysis of ing MCNP6 over available

s above aren't good enough ainties included.



TABLE XXXVII: Average values for C/E - 1 (in pcm) for benchmarks containing Gd. N is the number of benchmarks ^{1.03}

| | | | | | 1.02 | L . | | | | 1 | | 1 | ENDF/E | 3-VII.1 | <u>⊢×</u> | 4 |
|---|--------------------------------|--|--|---|-----------------------------------|------|------|------|------------------------------------|--------------------------------|----------|-------|--------|---------|-------------|------|
| Category | N | ENDF/B-VII.1 | JENDL-4.0 | JEFF-3.1.1 | | | | | Ŧ | I | | | | | | |
| leu-comp-therm | 74 | -556 | -499 | -578 | 1.01 | ŀ | | | - | | k | | М. | _ | . 1 | |
| ieu-comp-therm | 2 | 285 | 224 | -24 | | | | | | | | | | | âL. | |
| heu-met-therm | 2 | 585 | 482 | 614 | 1 | | | | | | | N | I. | V | | |
| heu-sol-therm | 52 | 196 | 421 | 278 | 0.99 | M | | | | | | ++ | | | | |
| mix-sol-therm | 13 | -233 | 75 | -185 | | | | | ••• | it10 t15 | ti 7 | | | | | |
| mix-misc-therm | 6 | -1009 | -690 | -982 | 0.98 | - | | | 011001 | 부석 | а К | 0 | 5 | 4 (| | 1 |
| pu-sol-therm | 15 | -111 | 345 | 82 | 0.97 | lct3 | lct4 | lct5 | let3 | hst14 hst14 | hst1 | hst1 | hst2 | pst3 | msu mst/ | mit4 |
| leu-comp-therm ieu-comp-therm heu-met-therm heu-sol-therm mix-sol-therm mix-misc-therm pu-sol-therm | 74 2 52 13 6 15 | -556 285 585 196 -233 -1009 -111 | $ \begin{array}{r} -499\\ 224\\ 482\\ 421\\ 75\\ -690\\ 345\end{array} $ | -578 -24 614 278 -185 -982 82 | 1.01 1 0.99 0.98 0.97 | lct3 | lct4 | Ict5 | lett2 1ct35 1ct552 1ct552 | hst14 hmt10 hst15 hst15 htt | hsti7 | hst19 | hst25 | pst34 | meth | mst7 |

Benchmark uncertainty JEFF-3.1.1 H

ENEA S/U Analysis

- To understand and assess the importance and role of 157Gd and 155Gd in nuclear fuels, a Sensitivity and Uncertainty (SU) analysis on k for several different FAs has been performed at BOL, hot-full power (HFP) conditions using the US-NRC reference SCALE
 6.1 code system developed at ORNL.
- **Tsunami-2D** sequence with ENDF/B-VII.0 evaluations.



Covariance Data: 44-group library (based on ENDF/B-VII.0)



ENEA S/U Analysis

- BWR GE 10x10-8 results.
- Two different moderator densities tested.
- The region of highest sensitivity for k is between 0.1 and 1 eV.





ENEA S/U Analysis

| Nuclide-Reaction | Contrib. to Uncertainty in k (% Δk/k) | Rank |
|---|--|------|
| ²³⁵ U v | 2.70E-01 | 1.00 |
| ²³⁸ U(n,γ) | 1.97E-01 | 0.81 |
| ²³⁵ U(n,γ) | 1.43E-01 | 0.64 |
| ²³⁵ U(n,f) | 1.43E-01 | 0.56 |
| ²³⁵ U(n,f) / ²³⁵ U(n,γ) | 1.21E-01 | 0.54 |
| ²³⁸ U(n,n') | 1.20E-01 | 0.51 |
| ²³⁵ U χ | 1.13E-01 | 0.45 |
| ²³⁸ U $\overline{\nu}$ | 7.11E-02 | 0.32 |
| ¹⁵⁷ Gd(n,γ) | 6.03E-02 | 0.26 |
| ¹⁵⁵ Gd(n,γ) | 4.48E-02 | 0.20 |
| ⁹² Zr(n,γ) | 4.29E-02 | 0.16 |
| ¹ H(n,γ) | 3.67E-02 | 0.14 |
| ⁹¹ Zr(n,γ) | 3.48E-02 | 0.13 |
| ¹ H(n,n) | 3.13E-02 | 0.12 |
| ⁹⁰ Zr(n,γ) | 2.82E-02 | 0.10 |

The **uncertainty** on **Gd** cross sections gives the **largest contribution** to the uncertainty on k **after** ^{235,238}U.

Several cross sections in this list have already been measured at nTOF.



Measurement of the Gd Cross Sections

- Neutron capture measurements were performed by the time-of-flight technique using metallic Gd samples;
- ✓ The facility (CERN-nTOF) makes use of the spallation mechanism as a strong source of neutrons by using a proton beam impinging a lead target (1GeV/c -> ≈ 300 n);
- ✓ The source can be concentrated in short time pulses (\approx 7 ns rms) with a low duty cycle (0,5 Hz);
- ✓ Neutrons produced are canalized to an experimental area located ≈185 m downstream through a vacuum pipe to irradiate samples. Long flight path allows to gain high resolution in energy (10⁻³-10⁻⁴);
- ✓ Gamma capture measurements were performed by hydrogen-free deuteraded benzene (C6D6) detectors in combination with the Pulse Height Weighting Technique (PHWT).



155Gd – 157Gd Samples





- Samples were acquired by the nTOF collaboration from ORNL;
- ✓ 4 samples were isotopically enriched in either ¹⁵⁵Gd or ¹⁵⁷Gd;
- The quantity of Gd in the samples results from a compromise between the need of the reducing the requested beam time and the optimization of the expected count rate in the resonance region;
- Since cross section changes by orders of magnitude as a function of the neutron energy, two highly enriched samples for each isotope were measured: a very thin one up to 100 meV, and a thicker one for cross section determination above 100 meV.



| Isotope | Form | Geometry | Radius | Isotopic Purity | Weight | Areal |
|-------------------|----------|----------|--------|-----------------|-----------|-------------------------|
| | | | | [%] | [mg] | Density |
| ¹⁵⁷ Gd | metallic | disc | 1 cm | 88.32 | 191.6±0.1 | 61.0 mg/cm ² |
| ¹⁵⁷ Gd | metallic | disc | 1 cm | 88.32 | 4.7±0.1 | 1.5 mg/cm ² |
| ¹⁵⁵ Gd | metallic | disc | 1 cm | 91.74 | 100.6±0.1 | 32 mg/cm ² |
| ¹⁵⁵ Gd | metallic | disc | 1 cm | 91.74 | 10.0±0.1 | 3.2 mg/cm ² |



Samples preparation



- Samples were sandwiched between two Mylar foils (to prevent oxidation) and centered avoiding any damage (they are extremely fragile and have to be handled with maximum care) in an annular frame to be correctly positioned along the line during irradiation;
- All samples had the same dimension in order to cover the same fraction of the neutron beam



Experimental Campaign

- ✓ Measurements were performed from 17th June to 8th July 2016;
- In addition to the four Gd samples, a Gold sample, a Graphite sample and a Lead sample were used to study the background;
- ✓ Black resonance filters (Co, Ag, W, Cd) positioned along the flight path are used to determine the energy dependence of the background; they are chosen thick enough that the neutron beam is completely absorbed at the energies of the resonances;
- ✓ To validate the entire analysis procedure, 197Au(n, g) reaction cross section measurement was carried out with sample similar to Gd;
- ✓ Beam off measurements were carried out to characterize the room background;
- ✓ Energy calibration was performed using standard sources of ¹³⁷Cs, ⁸⁸Y, AmBe, CmC.



Experimental Setup



- ✓ For the detection of the prompt gamma rays resulting from the capture events, fast liquid scintillation detectors were used;
- ✓ The experimental setup for the measurements consisted of an array of four C6D6 scintillators opposite each other at 45 degree wrt the beam.



Details of time allocation for the experiments

| | ^ | lo Filters | With Filters | | | |
|-------------------------|----------|---------------------|--------------|---------------------|--|--|
| Sample | #Protons | Running Time [days] | #Protons | Running Time [days] | | |
| ¹⁵⁵ Gd thin | 3.54E+17 | 4.00 | - | - | | |
| ¹⁵⁵ Gd thick | 3.29E+17 | 4.00 | 4.17E+16 | 0.50 | | |
| ¹⁵⁷ Gd thin | 3.96E+17 | 5.00 | - | - | | |
| ¹⁵⁷ Gd thick | 4.12E+17 | 5.00 | - | - | | |
| Sample Out | 1.71E+17 | 1.00 | - | - | | |
| Beam Off | - | 0.25 | - | - | | |
| Calibrations | - | 0.87 | - | - | | |
| ^{nat} Pb | 5.75E+16 | 0.50 | - | - | | |
| ¹⁹⁷ Au | 3.63E+16 | 0.25 | - | - | | |
| ^{nat} C | 4.71E+16 | 0.20 | - | - | | |
| Total | 1.81E+18 | 21.07 | 4.17E+16 | 0.50 | | |



Further Experiments



EUROPEAN COMMISSION DG JOINT RESEARCH CENTRE Directorate G - Nuclear Safety & Security Unit G.2-Standards for Nuclear S EUFRAT Europeen facilities for reaction and decay data measure

A request for beamtime at the Gelina fa has been submitted in December 2016 JRC-Geel.

Moreover, if it proves possible, also the Research Reactor will be performed, ho

| | Unit G.2-Stand Security & Sat | dards for Nuclear Safety, feguards | | | |
|--|---|--|--|----------|-----------|
| A | PPLICATION F | ORM | Reserved for EUFRAT use (do not fill this area) PAC meeting: Experiment number: | | |
| Title of the propose | ed experiment | | | | e samples |
| Transmission exp | periment for 155Gd | and ¹⁵⁷ Gd at GELINA | | | |
| Spokesperson (nam Cristian Massimi Istituto Nazionale di Dipartimento di Fisie Via Irnerio 46, Bolog +39 0512091079 massimi@bo.infn.it | ee, address, phone, e- Fisica Nucleare (INF) ca e Astronomia - Uni gna – Italy | | sement from | | |
| Facility to be used | | Type of experiment | | | |
| GELINA | x | Collaborative X | Associated | | Budapest |
| Van de Graaff | | Equipment used (pleas | e specify in detailed prop | osal) | Judapoor |
| HADES | | | | , | |
| Radionuclide labora | atory 🗌 | standard JRC-Geel-G. | 2 research equipment | x | |
| Contact person at J Peter Schillebeeckx | RC-Geel-G.2 | more complex JRC-Ge | el-G.2 research equipmer | nt 🗆 | |
| Requested measure | ment time (hours) | own equipment alone | | | |
| 400 hr/year for 1 yea | r | own equipment combin | ed with JRC-Geel-G.2 ec | quipment | |
| Preferred measuren March-April 2017 | nent period | - | | | |
| In case of collaborat conditions in reimbu 2 weeks for 2 persons | tive project, travel co ursement rules) s (2 x 10 x 100 €) + Tr | est estimation for typicall ravel for 2 persons (2 x 40 | y two supported users (se 0 €) = 2800 € | e | |
| Potential safety pro | blems (radioactive ta | rgets and sources, gases, | high activation) | | |



Future developments of the project

After the nTOF data analysis is completed, and after publication of the results on EXFOR, INFN (C. Massimi et al.) and ENEA will try to produce new evaluations for the xs and will initiate the validation process making recourse to the relevant ICSBEP benchmarks, in particular:

- ✓ LEU-COMP-THERM-005 (Pacific Northwest-BNFL, 16 cases)
- ✓ LEU-COMP-THERM-035 (JAERI TCA, 3 cases)
- ✓ HEU-SOL-THERM-014 (Institute of Physic and Power Engineering, 3 cases)

A possible completion timeframe could be envisaged at around Spring 2018.



Thank you for your attention

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