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Title page

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4 MCNP model of L-54 M nuclear research reactor: development and verification

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31 decommissioning activities [1], such as CeSNEF (*Centro Studi Nucleari Enrico Fermi*)
32 L-54M nuclear research reactor (50 kW nominal thermal power) [2]. It was
33 commissioned by Politecnico di Milano to Atomics International [3], construction works
34 started in 1958 and the first criticality condition was reached in 1959. The reactor was
35 used for research and didactic purposes until 1979, when it was shut-down, since Safety
36 Competent Authority (ISPRA, Italian Institute for Environmental Protection and
37 Research) did not grant the license to restart normal operation due to large growth of
38 urban fabric around the site. Politecnico di Milano decided to manage the structure
39 according to deferred dismantling strategy [4], namely to keep the plant under safe
40 conditions until successful removal of regulatory radiological constraints. In the
41 following decades, to pursue facility safe storage and safety of workers and public,
42 Politecnico di Milano carried out several activities according to National and
43 International well-established guidelines [5] [6]. In 1979, the fuel solution was
44 transferred to a well-shielded tank and, in 1994, sent to the Italian spent nuclear fuel
45 reprocessing facility. In the same years, the primary circuit was decontaminated.
46 Afterwards, in 2001, the most activated and contaminated part of the plant, *i.e.* the
47 secondary case envelope containing the core and all its components, was moved to a
48 well-shielded container, to be safely stored *in-situ*. In 2014, the Ra-Be neutron start-up
49 source was transferred from the graphite monolith to a neutron shielding container and
50 moved away from the plant. Recently, preliminary radiological characterization of the
51 site has started to obtain a preliminary estimation of radionuclide inventory and volume
52 of radioactive waste that will likely arise from forthcoming decommissioning operations
53 [7]. In 2015, an environmental radiological characterization was carried out to assess
54 unrealistic contamination of the surrounding area and obtain the reference radiological
55 blank [8]. In 2016, Politecnico di Milano joined the IAEA collaborative research project
56 on irradiated GRAPhite Processing Approaches (GRAPA) [9].

57 This paper describes the 3D neutronic model of L-54M reactor developed in order
58 to accurately assess the neutron activation of the graphite stack occurred in 20 years of
59 operations [10] and better address the future radiological characterization efforts [11].
60 MCNP6 (Monte Carlo N-Particle 6, version 1) software was selected for the modeling

61 due to its high-level performance and reliability in reactor physics modeling [12]. To
62 build an accurate MCNP model, rigorous geometrical and material information of the
63 main reactor components were gathered by a thorough bibliographic research. Specific
64 experimental analyses have been performed on unirradiated graphite and heavyweight
65 concrete samples to accurately simulate neutron transport. The L-54M reactor pile has
66 been proficiently modeled in detail. The capability of the implemented model to
67 accurately describe L-54M neutron source was verified evidencing promising accordance
68 between the simulated data and the available experimental ones. Yet, deep neutron
69 transport from the source to structural materials has to be verified before assessing their
70 activation. Afterwards, the main production reactions of the most relevant radionuclides
71 have to be identified and simulated, in order to calculate activity concentration maps to
72 be point-to-point compared with available experimental values, thus demonstrating
73 validity and robustness of the developed approach.

74 **Theory**

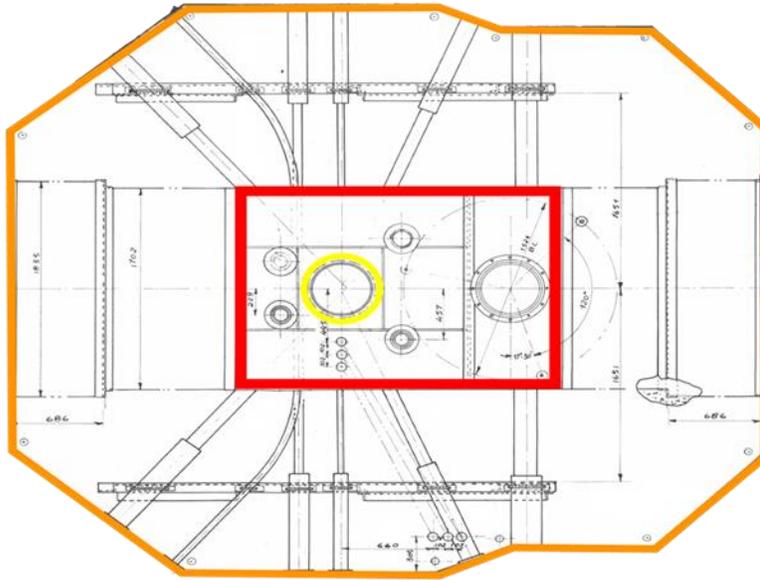
75 A gradual approach has to be followed. First of all, the MCNP neutronic model of
76 the nuclear reactor has to be developed by implementing geometrical and material
77 composition structural information in dedicated input file sections, named cell, surface
78 and material cards. Then, the model must be verified with experimental criticality and
79 fluences data. Afterwards, the main nuclear activation reactions of the radionuclides of
80 interest for the material to be decommissioned have to be identified and simulated in
81 order to obtain reaction rates. Finally, the calculated reaction rates have to be processed

82 to obtain activity concentrations distribution for each radionuclides to be compared with
83 available experimental data from radiological characterization to validate the model.

84 A thorough investigation of reactor geometry and materials has been carried out
85 consulting documents by Atomics International [2] [13] in order to find essential data
86 required for L-54M MCNP model [13]. Due to the restricted policy applied by the
87 vendor, this research turned out to be not sufficient to set up an accurate geometrical and
88 materials description. Consequently, reliable data from reactor safety reports [3] as well
89 as from academic works [14] [15] were properly integrated in the model.

90 In Fig. 1 a sketch of the reactor is depicted. The spherical stainless steel core (20 cm
91 radius) accommodating the uranyl sulfate liquid fuel was crossed by a longitudinal
92 stainless steel channel devoted to material irradiation and by 4 vertical channels
93 containing the stainless steel control rods enclosing boron carbide pellets. An overflow
94 chamber was soldered to the top of the core to host accidental liquid fuel losses caused by
95 extreme power excursions. The gases produced by fuel solution radiolysis were
96 recombined and reintroduced in the core, preventing the accumulation of a significant
97 amount of detonating mixture.

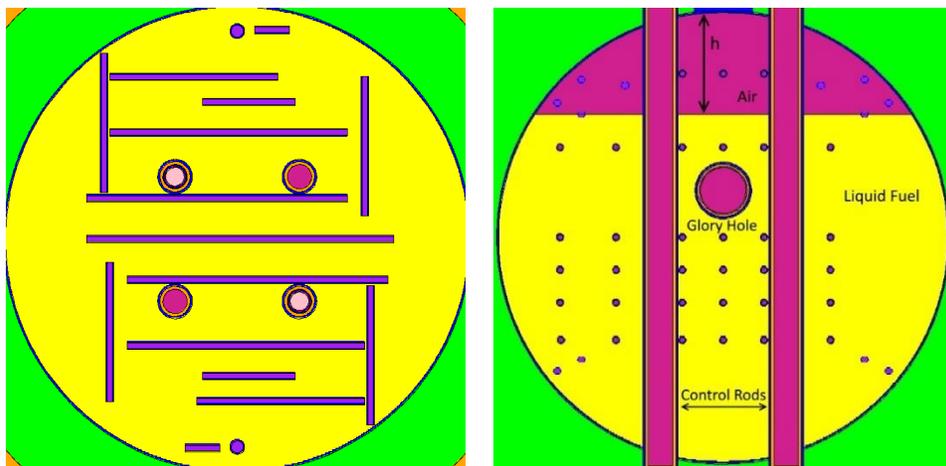
98 The heat generated by fission events was removed by a cooling coils system [3].
99 Although neutron moderation already took place in the fuel solution, a further moderating
100 effect was guaranteed by filling the secondary case with nuclear grade AGOT (Atcheson
101 Graphite Ordinary Temperature by US National Carbon Company) graphite [16].
102 Furthermore, the core was surrounded by a graphite stack to reflect escaping neutrons, for
103 overall 11 tons of graphite inventory. A heavyweight magnetite-colemanite concrete shell
104 (about 700 tons) was used as biological shield to reduce the radiation emission from the
105 reactor pile.



106

107 **Fig. 1** Reactor horizontal section showing the secondary case accommodating the spherical core (marked in yellow), the
108 graphite reflector case (in red), the heavyweight concrete shielding case (in orange) and several exposure tubes

109 The L-54M reactor pile has been proficiently modeled up to the outer boundary of the
110 concrete shielding. The core, the secondary envelope with the graphite moderator, the
111 aluminum case containing the graphite reflector, the exposure tubes and the heavyweight
112 concrete biological shield have been accurately implemented. Furthermore, an accurate
113 MCNP implementation of the cooling coils, one of the most complex structures of the
114 core, was achieved, even if quite demanding due to lack of precise data. The input script
115 limitations have been overlooked by substituting curved connections with simpler
116 components. In order to guarantee the best model accuracy, this simplification process
117 has been carried out paying attention to mass conservation and spatial distribution for
118 each array. Conversely, some other complex parts of the plant, essential for reactor safety
119 during operation, are irrelevant for this study as they do not influence neutrons
120 production neither their diffusion, have not been included. An example of model
121 implementation is shown in Fig. 2.



122 **Fig. 2** Details of central exposure tube (“Glory hole”), control rods and cooling coils. Left:horizontal section; right:
123 vertical section

124 An additional research has been carried out to find essential information about materials
125 used inside the reactor. As far as nuclear fuel is concerned, the original uranyl sulfate
126 solution contained 6623 g of U enriched in ^{235}U at 19.94 wt% dissolved in 26.5 L of
127 water. In order to ensure satisfactory stability of the solution and avoid salts precipitation,
128 chemical additives were used, leading to 1.346 g cm^{-3} final density [14]. Nuclear grade
129 stainless steel used for structural components, cooling system and all main steel parts of
130 the reactor is AISI 347/347H Niobium Stabilized. Nuclear grade graphite used for
131 neutrons moderation and reflection inside the L-54 M reactor is AGOT graphite [17]. It is
132 an extremely pure material with impurities at ppm or ppb level. Even if AGOT graphite
133 has been widely used, scarce impurities information could be gathered from the literature
134 to the best of author’s knowledge. In order to bridge this knowledge gap and accurately
135 investigate graphite activation inside the reactor, virgin graphite samples were analyzed
136 by ICP-MS. The biological shield was created with a monolithic pouring of heavyweight
137 concrete, with mean density of $3.66\text{ g}\cdot\text{cm}^{-3}$. A specifically designed heavyweight concrete
138 characterized by an inert phase of magnetite with Fe content above 64% and colemanite
139 rocks ($\text{Ca}_2\text{B}_6\text{O}_{11}\cdot 5\text{H}_2\text{O}$) with B_2O_3 content of about 41% was used. The homogeneous
140 distribution of all components in the pouring phase was experimentally verified in this
141 work by ICP-MS analyses on heavyweight concrete samples collected from different
142 positions along the outward biological shield.

143 Afterwards, several system properties described by the code, such as criticality data and
144 neutron fluxes in different core positions (e.g. in experimental exposure channels) have to
145 be compared with available experimental values in order to verify the model accuracy.
146 *KCODE* card is used to calculate neutron multiplication factor (k_{eff}) in a multiplying
147 medium at ambient temperature (20 °C). The flux of particles with determined energy
148 averaged over a surface or volume is calculated, in criticality condition, by *F2* and *F4*
149 tallies respectively. If MCNP fluence (expressed in cm^{-2}) is normalized on the ratio
150 between the real power produced by the reactor (obtained from official operational
151 records) and the energy deposited by fission events averaged on the core region cell
152 (calculated by *F7* tally), it can be converted to the more widespread $\text{cm}^{-2}\cdot\text{s}^{-1}$ unit.

153 Preliminary to simulation of activation distribution, the material has to be properly
154 covered by a dense mesh and the neutron flux has to be calculated for all the defined
155 volumes. Once the radionuclides of interest for decommissioning purposes have been
156 identified, their correspondent activation reaction rate has to be calculated in all the
157 volumes, taking into account the precursors concentration (*i.e.* the material composition),
158 the main reactions (*i.e.* the cross sections) and the neutron flux and spectrum.

159 Finally, the activity concentration of each radionuclide in all single element volumes has
160 to be calculated by solving the following differential equation:

$$161 \quad dN = q \cdot E - \lambda \cdot N$$

162 where N is the number of radioisotope nuclei, λ is the correspondent decay constant, q is
163 the reaction rate and E is the scaling factor (*i.e.* the energy produced by the reactor).

164 Since the simulation of materials activation is a time-dependent problem, the reactor
165 operational history is a required information. In fact, it influences both activation and

166 decay of radionuclides. These data were recorded on a monthly timescale [3]. The total
167 energy produced during L-54M operating history was about $1.2 \cdot 10^{12}$ J, with mean power
168 production of approximately 2 kW.

169 The final validation of the neutronic model is performed by comparing the simulated
170 activity concentrations with the available radiometric measurements deriving from
171 preliminary radiological characterization of the material under investigation.

172

173 **Experimental**

174 Microstructural and porosity data were already available in literature for nuclear grade
175 graphite [18] [19] [20] although only few data were available for AGOT graphite [16]
176 [17]. In this work, bulk and skeletal density and porosity of L-54M virgin graphite were
177 determined by the well-established mercury porosimetry technique and turned out to be
178 in accordance with literature data (see Table 1).

179 **Table 1** Comparison between density and porosity of virgin L-54M graphite (this work) and literature values [17]

	This work	Literature
Skeletal Density [g cm^{-3}]	2.29 ± 0.02	-
Bulk Density [g cm^{-3}]	1.69 ± 0.03	1.7
Porosity (%)	26.4 ± 0.7	25

180 To correctly simulate neutron transport inside graphite and biological shield, ICP-MS
181 analysis were performed to quantitatively characterize its macro-constituents and
182 impurities.

183 Few literature works describing nuclear grade graphite chemical composition and
184 impurities content are available [21] [22], but no data for AGOT graphite traces elements
185 were found. Graphite powder has been collected from a virgin bar using a clean drill bit.

186 A weighed amount (2.5 g) of graphite powder was placed in a ceramic crucible. The
187 sample was heated on a hot-plate at 250°C for few minutes and then in a muffle at 650°C
188 for about 24 hours, covering the crucible with a lid in order to prevent any sample loss
189 during oxidation. The duration of this step turned out to be dependent on the powder
190 grain size. This step allowed the complete oxidation of the sample. The resulting ashes
191 were treated with concentrated of HNO₃ (65 %), HCl (36 %) and HF (48 %) on a hot-
192 plate at 250°C until complete dissolution of the residue. In order to get rid of the
193 remaining fluorine, the solution was repeatedly heated to incipient dryness and the
194 precipitated dissolved by concentrated HNO₃. Finally, the remaining solution has been
195 diluted to suitable cations concentrations by 1 % HNO₃. All reagents used to dissolve and
196 digest ashes were certified ultrapure and analytically grade, so as to reduce any possible
197 elemental contamination deriving from reagents. In order determine the total yield of this
198 procedure, a known amount of Yb carrier was added to the powder. The choice of this
199 element was driven by its low concentration in nuclear grade graphite (below ppb).
200 Moreover, it is not a precursor of long lived neutron activated radionuclides of interest.
201 The chemical composition of AGOT graphite obtained by ICP-MS analysis is
202 summarized in the following Table 2. The results are comparable with those of nuclear
203 graphite in the literature. Nonetheless, the quantification of one of the main activation
204 precursor, boron, was not possible, because below the limit of detection (20 ppb). In
205 order to overcome this issue, the equivalent boron content was estimated from impurities
206 determined by ICP-MS and resulted to be 0.088 ± 5.0 % ppm [23]. Further analyses will
207 be carried out in order to quantify B, N, and Cl, important activation precursors of ³H, ¹⁴C
208 and ³⁶Cl.

209 **Table 2** L-54M AGOT graphite by ICP-MS analysis

Element	ppm	Element	ppm
Li	0.019 ± 2.7 %	Nb	0.005 ± 8 %
Mg	3.140 ± 0.2 %	Mo	0.145 ± 1.3 %
Al	1.179 ± 2.9 %	Pd	0.007 ± 28.5 %
K	1.219 ± 11 %	Cs	5·10 ⁻⁴ ± 4.8 %
Ca	7.971 ± 1 %	Ba	0.059 ± 1.9 %

Ti	0.622 ± 4.8 %	La	0.003 ± 2.9 %
Cr	0.032 ± 3.6 %	Ce	0.003 ± 4.6 %
Mn	0.031 ± 1.7 %	Pr	8·10 ⁻⁴ ± 2.7 %
Fe	2.809 ± 0.3 %	Nd	4·10 ⁻⁴ ± 29.4 %
Co	0.009 ± 1.5 %	Sm	5·10 ⁻⁴ ± 9.7 %
Ni	1.000 ± 1.3 %	Eu	6·10 ⁻⁴ ± 4 %
Cu	0.208 ± 1.6 %	Pb	0.069 ± 1.7 %
Zn	0.188 ± 2 %	Th	9·10 ⁻⁴ ± 5 %
Sr	0.023 ± 1.3 %	U	7·10 ⁻⁵ ± 33.3 %

210

211 Several subsamples of about 0.25 g of unirradiated concrete powder were treated at
 212 600 W (55 min) by microwave-assisted acid digestion using a mixture of concentrated
 213 ultrapure acids: HCl, H₃PO₄, HClO₄ and HF. After treatment and cooling, HCl and
 214 HClO₄ were added to the digested solutions. A blank was prepared following the same
 215 sample procedure. The macro-constituent and impurities content of L-54M heavyweight
 216 concrete are reported in Table 3 and Table 4.

217 **Table 3** L-54M heavyweight concrete macro-constituents by ICP-MS and CHNS (H only).

Element	Wt%
H	0.5 %
B	0.219 %
Mg	1.567 %
Al	0.356 %
K	0.234 %
Ca	3.376 %
V	0.075 %
Mn	0.093 %

Fe	42.82 %
Sr	0.045 %

218
219

Table 4 L-54M heavyweight concrete impurities by ICP-MS analysis

Element	ppm	Element	ppm
Li	32.4 ± 14 %	Ce	55.5 ± 1 %
Cr	17.7 ± 12 %	Pr	6.48 ± 1 %
Co	99.4 ± 2 %	Nd	28.3 ± 4 %
Ni	190 ± 2 %	Sm	4.54 ± 11 %
Cu	40.2 ± 6 %	Eu	0.86 ± 1 %
Zn	39.5 ± 7 %	Yb	4.97 ± 13 %
Pd	10.8 ± 60 %	Pb	8 ± 4 %
Cs	8.64 ± 3 %	Th	15.3 ± 4 %
Ba	43.2 ± 1 %	U	1.08 ± 1 %
La	26.6 ± 2 %		

220

221 **Results and discussion**

222 Several criticality simulations were run, and the simulated outcomes were compared with
 223 the available literature data to verify the MCNP model and its accuracy. All simulated
 224 data are provided with statistical MCNP error. Rod worth, reactivity excess, control rod
 225 inventory and calibration are the four criteria selected. A *KSRC* fission point source was
 226 positioned in the origin (0, 0, 0), *i.e.* in the center of the core. The *KCODE* card was used
 227 for all criticality calculations of this work, with about 4000 cycles and 6000 neutrons
 228 each. At least the first 30-50 cycles were discarded to remove the spatial dependence of
 229 fission reactions due to the initial guess [24]. A variance reduction technique was used in
 230 order to increase the number of neutrons coming to the furthest regions (especially to the
 231 heavyweight concrete biological shield) and improve statistics, without recurring to
 232 excessive computational time and effort. In particular, the geometry splitting and Russian

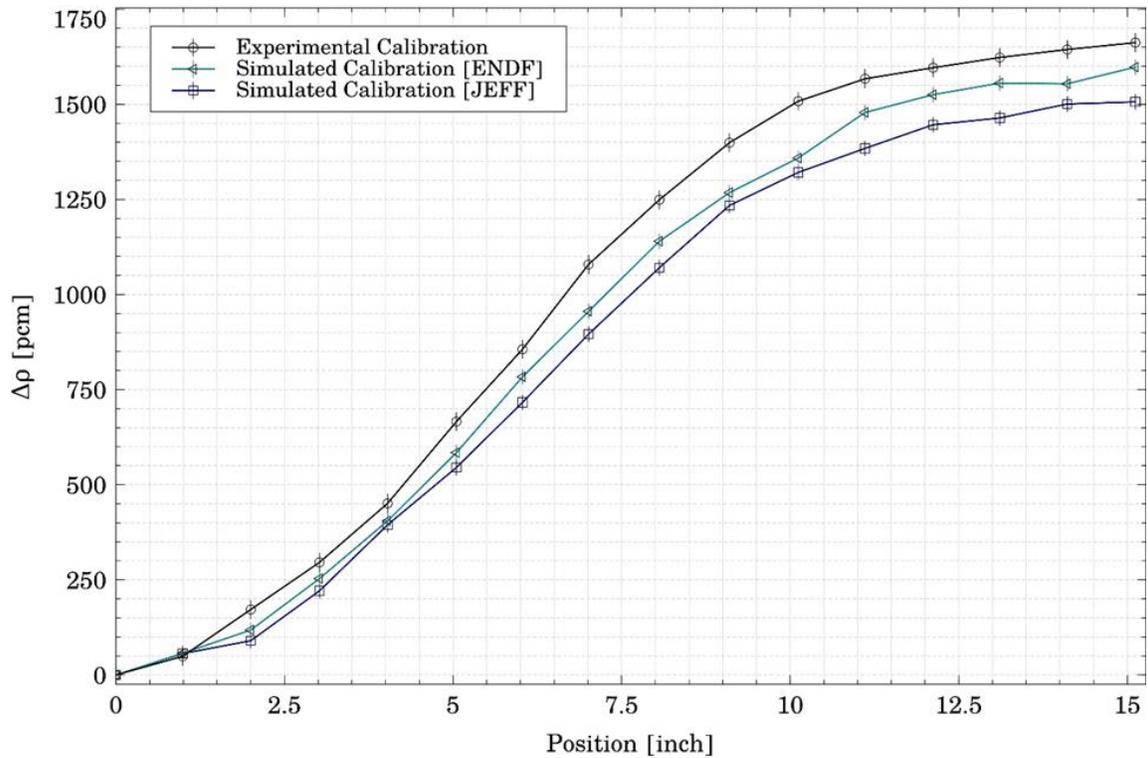
233 Roulette built-in variance reduction techniques were adopted for artificial population
 234 control [25]. In order to increase statistics in the outer cells far from the core and
 235 characterized by the presence of highly absorbing materials, importance was set greater
 236 than 1, locally enhancing events statistics without altering it. The operational critical state
 237 was successfully reproduced obtaining a unitary k_{eff} with one control rod fully inserted
 238 and another halfway inserted in the core, accordingly with actual reactor operating
 239 condition [3]. Maximum subcritical and supercritical conditions were evaluated at 20 °C
 240 by completely inserting and withdrawing the control rods. The sum of these values is the
 241 total rod worth, *i.e.* the overall reactivity associated to the control rods set. These
 242 calculations were run using *ENDF/B-VII.0* [26] and *JEFF-3.2* [27] cross-sections
 243 libraries. Satisfactory agreement between simulated and literature data is shown in Table
 244 3, even if slight differences between the nuclear datasets are evidenced: the core excess
 245 reactivity is better predicted by *JEFF-3.2* libraries, while the total rod worth by *ENDF/B-*
 246 *VII.0* libraries. This discrepancy could be explained by lack of resolution of *JEFF* cross
 247 section libraries in epithermal region and by different contribution and behavior of
 248 molecular scattering for *JEFF* and *ENDF* libraries [28]. Furthermore, some literature data
 249 are provided without associated error and they were just calculated and not
 250 experimentally measured, being reactivity values too large.

251 **Table 3** Comparison between simulated (this work) and literature reactivity values in pcm (10^{-5})

	MCNP [<i>ENDF/B-VII.0</i>]	MCNP [<i>JEFF-3.2</i>]	Literature
Subcritical Reactivity (20 °C)	-4587 ± 16	-3493 ± 16	-
Supercritical Reactivity (20 °C)	2385 ± 14	3115 ± 14	3500 [3]
Total Rod Worth	6972 ± 22	6608 ± 21	6950 ± 150 [14]

252 In order to further verify the model, the typical sinusoidal pattern of reactivity variation
 253 associated to the insertion or removal of the 4th control rod was simulated, moving it from
 254 the fully inserted position to the fully extracted one with all other rods withdrawn. The
 255 simulated reactivity variation was compared with those of the original calibration

256 experiment [14]. As outlined in Fig. 3 the model can proficiently describe the typical
 257 sinusoidal experimental calibration curve, with discrepancies below 100 pcm.



258

259 **Fig. 3** ENDF/B-VII.0 and JEFF-3.2 simulated calibration curves (this work) versus experimental one [14]

260 The reactivity worth of each control rod was simulated with *ENDF/B-VII.0* libraries in
 261 supercritical conditions as difference between reactivity values with all rods withdrawn
 262 and with each single rod inserted. Satisfactory agreement between simulated and
 263 literature data is evidenced in Table 4. The slight difference could be attributed to some
 264 lack of knowledge in the available documentation on absorbers, steels and alloys
 265 composition and structural dimension.

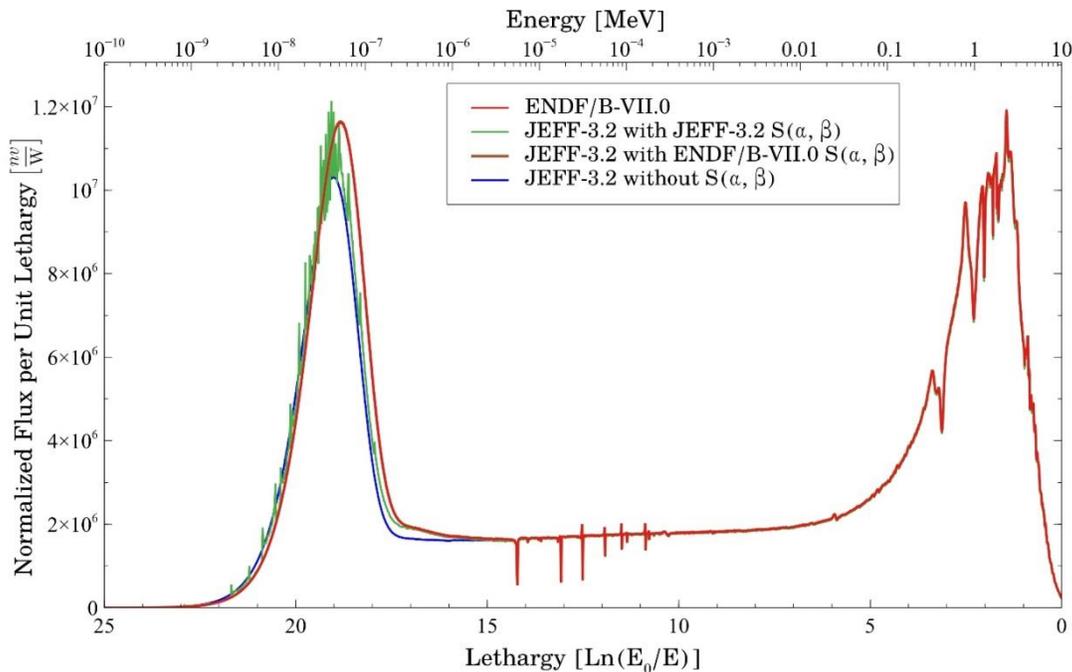
266 **Table 4** Comparison between simulated (this work) and literature [14] control rods worth in pcm (10^{-5})

Rod Worth	MCNP	Literature
1 st Rod	1624 ± 21	1750 ± 50
2 nd Rod	1625 ± 21	1750 ± 50

3 rd Rod	1637 ± 21	1750 ± 20
4 th Rod	1618 ± 21	1700 ± 30

267

268 All subsequent flux and activation simulations were performed in criticality condition.
 269 The emission density could be considered nearly proportional to fission density and,
 270 consequently, to neutron flux density, since the core is homogeneous. The calculated
 271 thermal and fast neutron profiles are reported in Fig. 4, as the most relevant lethargy flux
 272 spectra averaged over the fuel region.

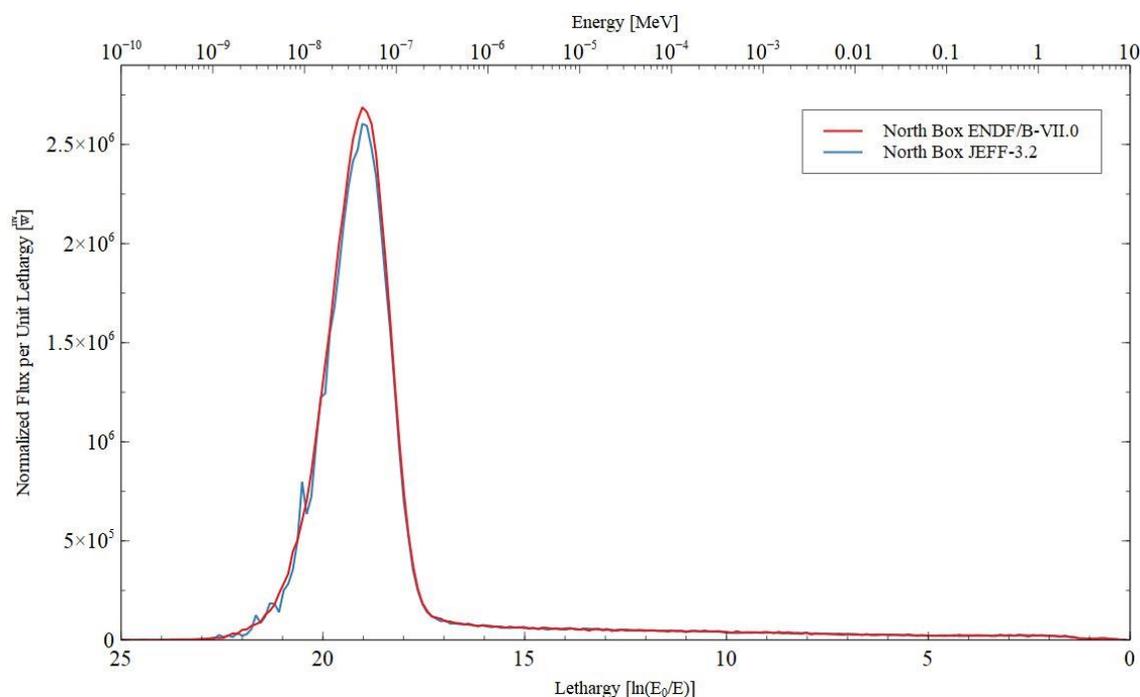


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Fig. 4 Comparison between neutron lethargy (and energy) spectra inside the fuel region obtained with ENDF/B-VII.0 and JEFF-3.2 cross-sections data.

276 Although ENDF/B-VII.0 and JEFF-3.2 give essentially the same spectra, adding the
 277 corresponding S (α , β) inelastic scattering cross-sections causes an alteration of the
 278 spectrum in the thermal region, shifting and broadening the peak. However, this effect is
 279 different in magnitude between the two series and in addition, the JEFF-3.2 S (α , β)
 280 introduce also jags and spikes in the thermal peak. This discrepancy could be attributed to
 281 the differences between S (α , β) energy dependent cross-sections of these two series
 282 although they are quite similar, especially for graphite. Since no experimental data are

283 available from reactor operations, it is paramount to validate the calculations on suitable
284 benchmark. To the best of authors' knowledge, there are no publications dealing with
285 neutron transport in uranyl sulfate solutions. Since water is the main constituent, a
286 benchmark can be established with works focused on neutron transport in water [29]. In
287 addition, the neutron profiles were calculated in the graphite reflector regions directly
288 facing the core. In order to further deepen cross-section data effect, both ENDF/B-VII.0
289 and JEFF-3.2 libraries were tested in the flux simulation. For example, the comparison
290 between the neutron profiles obtained with the two cross-section libraries in the north
291 graphite region is reported in Fig. 5. Unlike abovementioned criticality issues, the choice
292 of different cross-section libraries entails small impact on activation calculations. In fact,
293 from Fig. 5 it could be inferred that satisfactory agreement between results was obtained,
294 with thermal flux discrepancies below 4%.

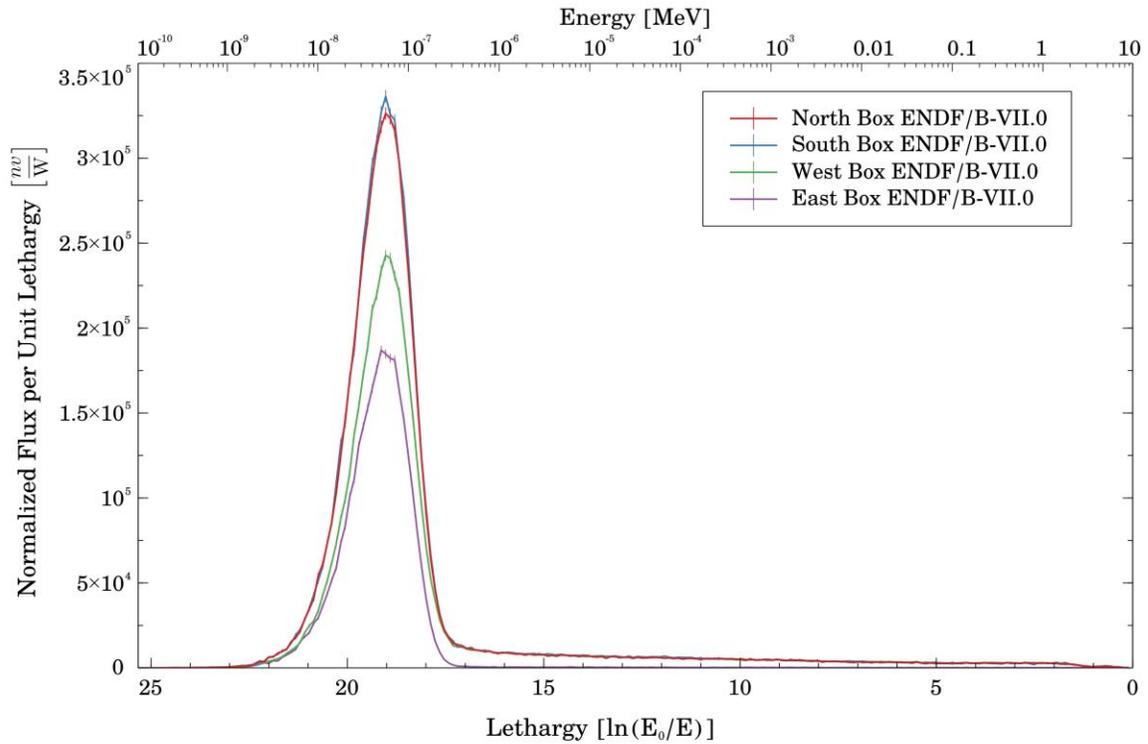


295

296 **Fig. 5** Neutron lethargy (and energy) spectra in the north graphite reflector region obtained with ENDF/B-VII.0 and
297 JEFF-3.2 cross-sections data

298 Consequently, the well-studied and already implemented in the code ENDF/B-VII.0
299 cross-section libraries were selected for subsequent simulations. The neutron profiles

300 calculated in the four graphite reflector regions with ENDF/B-VII.0 are reported in Fig.
301 6. These profiles are reliable due to validated neutron transport in the fuel solution.



302

303 **Fig. 6** Neutron lethargy (and energy) spectra in the graphite reflector region obtained with ENDF/B-VII.0 cross-
304 sections data.

305 In order to study activation of graphite, the verification of the correct source
306 characterization is not sufficient. In fact, the model still has to be verified from the point
307 of view of deep transport from source to structural components. This task is on-going.

308 Conclusions

309 In this work, a MCNP model of L-54M nuclear research reactor has been developed to
310 assess the activation of graphite reflector. The information gathered by the bibliographic
311 research could be considered fair enough to develop a satisfactorily accurate model.
312 Despite remaining uncertainties, a preliminary verification of the developed MCNP
313 model has been satisfactorily acquired by comparing reactor parameters simulated using
314 different cross-section libraries with the available experimental equivalents.

315 As natural prosecution of the herein described work, further simulations, including
316 sensitivity analysis, and experiments are being performed. In particular, an ultimate
317 model verification is being achieved by comparing simulated and experimental neutron
318 fluxes at different positions inside the monolith. The activity concentration of the main
319 radionuclides of concern are being simulated and compared with the few available
320 experimental measurements. Further radiometric analyses on recently collected irradiated
321 graphite samples will be performed to determine γ -emitting and pure β -emitting
322 radionuclides and complete the model validation.

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