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UK Chemical Nuclear Data Committee Progress Report: Data Studies During 1992

Edited by A L Nichols



February 1993

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# UK CHEMICAL NUCLEAR DATA COMMITTEE : PROGRESS REPORT, DATA STUDIES DURING 1992

Edited By

A L NICHOLS\*

#### SUMMARY

The basic nuclear data for commercial applications in the UK are monitored and peerreviewed by the UK Chemical Nuclear Data Committee (UKCNDC). Funding difficulties continue to undermine the work of the UKCNDC, resulting in the consolidation of these data reviewing and endorsing exercises within the main body of the committee. Work undertaken within the auspices of the UKCNDC includes the measurement and evaluation of radionuclide half-lives, decay parameters (eg emission energies and transition probabilities) and fission yields; these efforts are described in this document for information.

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# 1. **INTRODUCTION**

The UK Chemical Nuclear Data Committee (UKCNDC) has provided a forum within the UK for over 20 years to monitor the adequacy, quality and need for radionuclide decay data and fission product yields as applied to a wide range of fuel-cycle and environmental applications. The requirements and priorities for nuclear data measurements and evaluations are regularly reviewed within the UKCNDC. At the beginning of 1992, committee members debated the future requirements for the control and technical monitoring of nuclear data measurements and evaluations within the UK. These discussions took place within an environment of reduced Government funding in which the work load of the Data Library Sub-Committee was reconstituted under the direct auspices of the parent committee (ie UKCNDC). Two meetings of the UKCNDC were held during 1992 in April (combined with DLSC) and November (Chairman, A L Nichols (WTC) and Secretary, R W Mills (BNF plc on attachment to WTC)).

The UKCNDC is responsible for advising on measurement programmes and the maintenance of the following computerised libraries:

- (a) fission product yields,
- (b) fission product decay data,
- (c) activation product decay data,
- (d) heavy element and actinide decay data.

Significant progress during the year has involved the evaluation of activation product decay data, based on funding from AEA Reactor Services (to match BNF plc funding in this area over previous years). All of this evaluation effort is linked to the international Joint Evaluation File (JEF).

# 2. <u>MEASUREMENTS</u>

2.1 <u>Measurement and Evaluation of Decay Data</u> (P Christmas, D Smith, M J Woods, J L Makepeace, J P Sephton, S A Woods, S M Jerome, D H Woods, G R Worthington, J C J Dean, S E M Lucas, H C Paton Walsh, F E Clark and M T Curry (National Physical Laboratory, Teddington)).

Specific measurements have been completed or are under way:

- (a) The measurement of the half-life of Sr-90 has been completed and the results are being analysed.
- (b) Measurements of half-lives are in progress for radionuclides for which more accurate data are required, ie H-3, Mn-54, Co-57, Zn-65 and Se-75.
- (c) The half-life of the intermediate state of Se-75 has been determined by correlation counting.
- (d) NPL staff are reviewing the nuclear decay scheme data in evaluated data files
  (particularly JEF-2) with a view to being able to recommend the use of selected data to UK users (eg half-lives and principal gamma-ray emissions).
  Approximately 50 nuclides will be initially examined, primarily those for which NPL has supplied standards.

Various papers have been published or accepted for publication during 1992 associated with radioactivity measurements undertaken at NPL (1-9).

2.2 <u>Delayed Neutron Yield Measurements</u> (M Kellett and D R Weaver (University of Birmingham)).

An experimental project has just been started with a view to the measurement of the yield of delayed neutrons from the fission of U-238 at various neutron energies. The Birmingham Dynamitron will be used for these studies.

# 3. **DATA LIBRARIES: EVALUATIONS**

As noted in previous progress reports, funding for chemical nuclear data has declined significantly. Thus, it was agreed that the responsibility for defining and maintaining the various data libraries should return to the UKCNDC, and the Data Library Sub-Committee (DLSC) would be re-absorbed into the parent committee. This arrangement was agreed at the April 1992 meeting (combined UKCNDC and DLSC discussions).

A L Nichols organised and attended a three-day workshop at Idaho Falls, USA (5-7 October 1992) under the auspices of the International Committee for Radionuclide Metrology (ICRM). Specialists were invited to provide decay-data evaluation problems for assessment by their peers at the meeting. A series of recommendations evolved from the discussions, coupled with presentations of evaluation procedures for half-lives and decay data. The results of these in-depth discussions will be presented at the biennial meeting of the Non-neutron Nuclear Data Working Group of the ICRM to be held at NPL, Teddington in June 1993.

# 3.1 Data Library Development

The current status of the UKCNDC Data Libraries is summarised in Table 1. Progress continues to be made in the evaluation of fission yield and decay data. Efforts have also been made during the year to incorporate these data into the UK and JEF files.

## (a) <u>Fission Product Decay Data (A L Nichols (WTC))</u>

The decay data for a limited number of fission products have been evaluated (Sb-125, Sb-129, Cs-134, Sm-151, Eu-152 and Eu-155). However, because of the modest size of this effort, these data have been provisionally incorporated into the UK Activation Product Decay Data Library (UKPADD-2).

# (b) Activation Product Decay Data (A L Nichols (WTC))

Decay data for the following 63 radionuclides have been evaluated during 1992:

Li-8, Li-9, Be-8, Ca-45, Ca-47, Sc-50, Sc-50m, Ti-45, Ti-51, V-52, V-53, V-54, Cr-55, Mn-56, Fe-53, Fe-53m, Fe-55, Co-55, Co-56, Co-57, Ni-57, Cu-62, Cu-64, Cu-66, Sr-85, Sr-85m, Sr-90, Y-90, Y-90m, Ag-108m, In-114, In-114m, In-114n, In-116, In-116m, In-116n, Sn-119m, Sn-121, Sn-121m, Sb-125, Sb-129, Sb-129m, Te-129, Te-129m, I-125, Cs-134, Cs-134m, Ba-133, Ba-133m, Ce-139, Ce-139m, Sm-151, Eu-152, Eu-152m, Eu-152n, Eu-154, Eu-154m, Hf-175, Hf-181, W-181, Au-198m, Hg-197 and Hg-197m.

These data have been combined with the files assembled in previous years to produce the UK Activation Product Decay Data Library, UKPADD-2. Decay data have been recommended for 236 radionuclides, and these data sets have

# Table 1: UKCNDC DATA LIBRARIES: STATUS TABLE, DECEMBER 1992.

[		
Data	Present Status	File Development
Fission	Part of JEF-1 Decay Data Files, which	Pn values for delayed neutron
Product	include UK evaluations (ENDF/B-V	precursors from Swedish
Decay	format).	evaluation.
Data	UKFPDD-2 library superseded, but	Delayed neutron precursor spectra
	largely included in preliminary version of JEF-2.	from Kratz et al and Rudstam et al.
Activation	UKPADD-1 forms part of the	236 nuclides have been evaluated
Product	preliminary JEF-2 Decay Data Files,	for UKPADD-2 in ENDF/B-VI
Decay	which also include other UK evaluations	format. This work will be
Data	(ENDF/B-V format).	completed by 31 March 1993, and
		all evaluations will be submitted to
		JEF.
Heavy	Preliminary JEF-2 Decay Data Files	The heavy element and actinide
Element	include UKHEDD-2 evaluations	decay data library has been re-
and	(ENDF/B-VI format).	issued as UKHEDD-2, following
Actinide		the re-evaluation of the major decay
Decay		parameters within the files.
Data		
Fission	JEF-1 fission yields based on UKFY-1	Major reports on evaluations
Yields	evaluation of Banai and James. The	leading to UKFY-2 have been
	UKFY-2 evaluation of Mills and James	prepared and issued by James, Mills
	(completed January 1990 and revised	and Weaver. Weaver and James
ł	January 1991) has been incorporated into	have recommended integral yields,
	a preliminary version of JEF-2: 39 sets of	group data and spectra for delayed
	evaluated independent and cumulative	neutrons.
	yields for 21 fissile nuclides adjusted to	
	satisfy conservation of nucleons and	
l	charge.	

Spectral data from the decay data files may be accessed via the retrieval system described by Tobias (RD/B/5170N81, 1981).

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## Table 2 : Data Consistency : Comparison of the Percentage Deviation Between Effective Q-value and Calculated Q-value for UKPADD-1 and 2

Nuclide	UKPADD-1	UKPADD-2	Nuclide	UKPADD-1	UKPADD-2
н-3	0.2199	0.0000	C1-38	0.2634	-0.0079
He-6	not evaluated	0.0000	C1-38m	0.0000	-0.0640
He-8	not evaluated	incomplete	Ar-37	not evaluated	-0.0007
Li-8	not evaluated	0.0419	Ar-39	0.0000	0.0000
Li-9	not evaluated	incomplete	Ar-41	0.0522	-0.0384
Be-7	not evaluated	0.0111	Ar-42	not evaluated	0.0000
Be-8	not evaluated	-0.0092	K-38	not evaluated	0.0000
Be-10	0.0000	0.0000	K-38m	not evaluated	0.0000
Be-11	not evaluated	0.0915	K-40	-0.1619	-0.0011
B-12	not evaluated	0.0002	K-42	-0.0476	0.0066
в-13	not evaluated	-0.0238	К-43	not evaluated	-0.0474
C-14	0.0001	0.0000	K-44	not evaluated	0.1283
C-15	-0.0776	-0.0733	Ca-41	0.0038	0.0035
N-13	not evaluated	0.0000	Ca-45	-0.0016	0.0000
N-16	0.6385	0.0589	Ca-47	not evaluated	0.1213
0-19	-0.1895	-0.1655	Ca-49	not evaluated	-0.0229
F-18	not evaluated	0.0000	Sc-44	not evaluated	0.0171
F-20	not evaluated	0.0051	Sc-44m	not evaluated	0.0391
Ne-23	not evaluated	0.0183	Sc-46	0.0007	-0.0028
Na-22	0.0012	0.0018	Sc-46m	-0.5007	0.4710
Na-24	0.2474	0.0068	Sc-47	-0.0666	0.0017
Na-24m	0.0000	0.0003	Sc-48	-0.1006	0.0226
Na-25	not evaluated	0.1602	Sc-49	not evaluated	0.0000
Na-26	not evaluated	-0.0077	Sc-50	not evaluated	-0.0075
Mg-27	0.3764	0.0001	Sc-50m	not evaluated	-0.0276
Mg-28	not evaluated	0.1597	Ti-45	not evaluated	0.0022
Al-26	not evaluated	0.0250	Ti-51	not evaluated	0.0073
A1-26m	not evaluated	0.0000	V-48	not evaluated	-0.2125
Al-28	not evaluated	0.0022	V-49	not evaluated	0.0029
Al-29	not evaluated	-0.0207	V-52	not evaluated	-0.000é
A1-30	not evaluated	-0.0638	V-53	not evaluated	-0.2563
Si-31	not evaluated	-0.0014	V-54	not evaluated	incomplete
Si-32	not evaluated	0.0000	Cr-49	not evaluated	-0.0171
P-32	not evaluated	0.0000	Cr-51	0.0274	0.0053
P-33	not evaluated	0.0000	Cr-55	not evaluated	0.0000
P-34	not evaluated	-0.0065	Mn-54	0.8868	-0.0001
s-35	-0.1792	0.0000	Mn-56	0.3581	-0.0179
S-37	not evaluated	-0.0801	Fe-53	not evaluated	-0.2021
C1-34	not evaluated	0.0000	Fe-53m	not evaluated	0.1882
C1-34m	not evaluated	-0.0441	Fe-55	-0.0011	-0.0020
C1-36	-0.2000	0.0006	Fe-59	0.0433	-0.0172

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Table 2 : (continued)

Nuclide	UKPADD-1	UKPADD-2	Nuclide	UKPADD-1	UKPADD-2
Co-55	not evaluated	0.0305	Nb-95m	not evaluated	-0.0661
Co-56	not evaluated	0.5072	Mo-93	0.1008	-0.0074
Co-57	-0.0027	0.0020	Mo-93m	1.6087	0.0162
Co-58	0.2051	-0.0023	Mo-99	not evaluated	-0.0098
Co-58m	-0.3947	-0.3171	Tc-99	not evaluated	0.0000
Co-60	0.0065	0.0053	Tc-99m	not evaluated	0.0470
Co-60m	0.2040	-0.2943	Ru-103	not evaluated	-0.0914
Ni-57	not evaluated	-0.0314	Rh-102	not evaluated	-0.4531
Ni-59	0.3022	-0.0056	Rh-102m	not evaluated	0.0748
N1-63	0.0456	0.0000	Rh-103m	not evaluated	-0.3865
Ni-65	0.0249	0.0352	Rh-104	not evaluated	0.0233
Cu-62	not evaluated	0.0001	Rh-104m	not evaluated	-0.1843
Cu-64	-0.0019	-0.0007	Ag-107m	not evaluated	-0.0525
Cu-66	-0.1126	0.0166	Ag-108	not evaluated	0.0204
Zn-63	not evaluated	0.0286	Ag-108m	not evaluated	-0.0190
Zn-65	0.0061	-0.0075	Ag-109m	not evaluated	-0.1873
As-74	not evaluated	-0.2919	Ag-110	-0.2735	-0.0009
Se-75	not evaluated	-0.1033	Ag-110m	-1.0785	-0.3356
Br-79m	not evaluated	-0.0283	Cd-109	not evaluated	0.0330
Br-80	not evaluated	-0.0134	Cd-111m	not evaluated	-0.0834
Br-80m	not evaluated	-0.1960	Cd-113	not evaluated	0.0000
Br-82	0.0357	0.0729	Cd-113m	not evaluated	0.0012
Br-82m	-0.9768	0.5001	In-111	not evaluated	0.0372
Kr-79	not evaluated	-0.0466	In-111m	not evaluated	-0.0507
Kr-79m	not evaluated	-0.0728	In-113m	not evaluated	0.0014
Sr-85	not evaluated	-0.0051	In-114	not evaluated	-0.0031
Sr-85m	not evaluated	0.0031	In-114m	not evaluated	-0.1894
Sr-89	not evaluated	0.0000	In-114n	not evaluated	-0.0062
Sr-90	not evaluated	0.0000	In-115	0.2058	0.0000
Y-88	0.0001	0.0531	In-115m	1.1239	-0.0605
Y-89m	not evaluated	0.0024	In-116	-0.5528	0.6261
Y-90	not evaluated	0.0000	In-116m	0.5556	0.1921
Y-90m	not evaluated	-0.0151	'In-116n	0.0957	0.0632
Zr-89	not evaluated	0.0513	Sn-113	not evaluated	0.0204
Zr-89m	not evaluated	-0.0317	Sn-113m	not evaluated	-0.2109
Zr-93	not evaluated	0.0000	Sn-117m	not evaluated	-0.0006
Zr-95	not evaluated	-0.0121	Sn-119m	not evaluated	-0.1028
Nb-93m	0.9029	-0.3676	Sn-121	0.0000	0.0000
Nb-94	-0.1075	-0.0014	Sn-121m	0.0214	-0.0800
Nb-94m	-0.2252	-0.0290	Sn-123	not evaluated	-0.0042
Nb-95	not evaluated	0.0025	Sn-123m	not evaluated	-0.0694

#### Table 2 : (continued)

Nuclide	UKPADD-1	UKPADD-2	Nuclide	UKPADD-1	UKPADD-2
Sn-125	not evaluated	-0.0725	Eu-155	not evaluated	0.1705
Sn-125m	not evaluated	0.1417	Tb-157	1.1573	1.2277
Sn-126	not evaluated	0.0784	Dy-157	not evaluated	-0.0226
Sb-122	-0.1631	0.0041	Dy-159	not evaluated	0.0906
Sb-122m	1.3024	-0.0135	Hf-174	not evaluated	0.0155
Sb-124	-0.3591	-0.0495	Hf-175	0.4561	0.0337
Sb-124m	-0.0442	-0.3407	Hf-181	0.1206	0.0975
Sb-124n	-0.2882	-0.4979	Ta-179	not evaluated	0.8418
Sb-125	not evaluated	0.0434	Ta-180	not evaluated	0.0321
Sb-129	not evaluated	-0.6673	Ta-180m	not evaluated	0.1639
Sb-129m	not evaluated	incomplete	Ta-182	0.5610	-0.0057
Te-125m	not evaluated	-0.0051	Ta-182m	-1.4951	0.1654
Te-129	not evaluated	-0.0619	Ta-182n	0.2796	0.8531
Te-129m	not evaluated	-0.2257	W-181	0.0463	0.0684
I-125	not evaluated	0.0709	W-185	-0.1033	0.0001
I-126	-0.4034	0.0678	W-185m	-0.0138	-0.1514
Xe-125	not evaluated	-0.0292	W-187	0.0174	0.0631
Xe-125m	not evaluated	0.0606	Au-198	-0.0800	0.0018
Xe-127	not evaluated	-0.1451	Au-198m	incomplete	incomplete
Xe-127m	not evaluated	-0.0383	Hg-197	0.1156	0.1285
Cs-134	0.0004	0.0823	Hg-197m	0.1415	0.6335
Cs-134m	incomplete	-0.0812	Hg-203	0.0067	0.0271
Cs-135	not evaluated	0.0000	T1-204	not evaluated	0.0033
Cs-135m	not evaluated	-0.0280	Pb-204	not evaluated	-0.0097
Cs-136	0.1880	-0.2422	Pb-204m	not evaluated	0.0780
Cs-136m	incomplete	incomplete	Bi-207	not evaluated	0.1833
Cs-137	not evaluated	0.0000	Th-228	not evaluated	0.0122
Ba-133	-1.6779	0.0810	Th-231	not evaluated	-0.7156
Ba-133m	-1.2167	-0.0816	Np-239	not evaluated	-0.1364
Ba-137m	not evaluated	0.0176	Am-241	not evaluated	-0.0220
Ce-139	not evaluated	0.1622	Am-243	not evaluated	-0.0217
Ce-139m	not evaluated	0.0029			
Pm-145	0.0313	0.1990			
Sm-145	not evaluated	0.0659			
Sm-146	not evaluated	-0.0199			
Sm-151	not evaluated	-0.0006			
Eu-152	-0.7888	-0.1502			
Eu-152m	0.3645	-0.6753			
Eu-152n	incomplete	0.0207			
Eu-154	0.3257	0.0416		ļ	
Eu-154m	1.5263	0.0927			

been generated in ENDF/B-VI format (Table 2). The contents are based on a subset of the listing in reference 10, and are compared with an earlier library of evaluated data (11).

#### (c) <u>Heavy Element and Actinide Decay Data (A L Nichols (WTC))</u>

A detailed evaluation has been made of the decay data for Rn-223 (60 betaparticle and 275 gamma-ray emissions). These evaluated data include the halflife, average decay energies, beta-particle and gamma-ray energies and emission probabilities, internal conversion coefficients, and all associated uncertainties.

Doubts have developed in recent years concerning the values recommended for the absolute gamma-ray emission probabilities of Pa-234m. Thus, an initial assessment has been carried out of the emission probability of the primary 1001keV gamma ray of this radionuclide. A value of 0.00590(10) was adopted in previous evaluations on the basis of a single datum measurement in the 1960s; more recent studies are in good agreement to give a preliminary recommended value of 0.00837(12). An extensive evaluation effort is now required to refine this value and also determine a new set of relative gamma-ray emission probabilities and beta-particle decay data from these measurements (undertaken from 1986 to 1992).

(d) <u>Fission Product Yields</u> (M F James (Consultant to BNF plc), R W Mills (BNF plc) and D R Weaver (University of Birmingham)).

The work within the UK fission product yield evaluation programme has continued towards a new evaluated library (UKFY3) to update UKFY2 (reported in AEA-TRS-1015, 1018 and 1019). These studies have involved the following: expanding the experimental database (both with new data and with new types of data to aid in a greater understanding of the fission-product generation process), adapting the analysis procedure to use more of the data by iteration of the relative and ratio of ratio data, improving the technique of generating complete yield sets using the mass yields rather than the chain yields, and examining different models to explain the fission-product yield production process.

The UK group has continued involvement with the IAEA Collaborative Research Programme on Fission Yield Evaluation, and the NEA JEF evaluation programme.

#### 3.2 Interrogation and Extraction of Data

(a) <u>Decay Data</u> (A Hill, J Blackband and D R Weaver (University of Birmingham)).

Previous development work was described in terms of a computer program designed to provide a convenient way of displaying decay data from the JEF-2 library. This program has been subjected to testing and improvement during the year. Significant improvements have been made in the ease of use of the program, which is based around an interactive display of the Chart of the Nuclides. A number of enhancements have been made to the program and, at the same time, advantage has been taken of updates to the preliminary JEF-2 library. The program development was essentially completed in September 1992, with the aim by the NEA Data Bank (who sponsored the work) of making the code available for issue. At the time of writing, however, it is understood that the JEF community has decided that some further revision will be made to JEF-2 before this library is frozen and released, and it is expected that a further conversion of the original files to the binary format will be undertaken in the early months of 1993. This should allow the code to be issued through the NEA Data Bank. The program was demonstrated during 1992 at the IAEA Nuclear Data Section in Vienna, and at Brookhaven National Laboratory during the Conference on Nuclear Data Evaluation Methodology (October 1992).

(b) <u>Fission Yield Data</u> (A Hill and D R Weaver (University of Birmingham)).

Following the enthusiastic reception of the JEF-2 Decay Data display program, BNF plc sponsored a vacation studentship in 1992 to develop a similar package to provide an interactive means of displaying the fission yield data from the JEF-2 library. With the permission of the NEA Data Bank, the program was developed from the earlier code and maintains the same format. However, the new program displays independent and cumulative yields (plus uncertainties) for all the fissioning systems included within the JEF-2 library. Different options permit the display of these yields either individually or for (a) mass

chains, or (b) all radionuclides of a particular element. As in the original program the display can be graphical or numeric, and spooling to external devices is allowed. This version of the code is now being subjected to testing.

3.3 <u>Testing of Data Evaluation Methods</u> (S I Kafala and T D MacMahon (Centre for Analytical Research in the Environment, Imperial College at Silwood Park, Ascot), with P W Gray (INSCITECH Consulting Ltd, Woking)).

The problem of determining a recommended value and uncertainty from a discrepant set of data has been addressed by several authors with particular reference to radionuclide half-life data. Many sets of data have been generated by random selection of values from a normal distribution of known mean (unity) and standard deviation in order to carry out an objective testing of the various proposed data evaluation methods. Altogether 36 types of data set have been computer-generated. These data vary in:

- (i) number of values in each set, from 4 to 40;
- (ii) average standard deviation of the values in each set from 0.5% to 30%;
- (iii) spread of uncertainties for the values in each set by up to a factor of 10;
- (iv) uniform spread of uncertainties or uncertainties concentrated at the upper and lower limits of the spread;
- (v) assumed underestimation of the uncertainty by a hypothetical experimentalist from -50% to +60%, either fixed or randomly distributed;
- (vi) skewness and kurtosis, using a log-normal distribution.

For each of the 36 types of data set, 1000 sets of values were generated. Each data set was evaluated using ten evaluation procedures. The number of data sets which resulted in a mean value  $\bar{x}$  and an uncertainty  $\sigma_{\bar{x}}$  satisfying  $1 - \sigma_{\bar{x}} < \bar{x} < 1 + \sigma_{\bar{x}}$  was determined, remembering that the 'true' mean of each data set was unity. It is assumed that the ideal evaluation procedure would be that for which the above criterion would be satisfied in 68.3% of the data sets. Percentages below this value would indicate that the procedure was underestimating the recommended uncertainty and, conversely,

higher percentages would indicate that the procedure was overestimating the recommended uncertainty.

On the above basis, the evaluation procedure which provides the 'best' results is a proposed modified Bayesian procedure, where the weighted mean is taken as the recommended value and the recommended uncertainty is the uncertainty of the weighted mean multiplied by

$$\sqrt{\frac{\chi^2}{N-2}}$$

irrespective of the value of  $\chi^2$ .

## 4. <u>CONCLUSIONS</u>

As in previous years, UK measurements and evaluations of nuclear data depend on a low level of effort and dedicated expertise. Judicious investments by BNF plc and AEA Reactor Services have maintained skeletal evaluation effort in 1991/92 and 1992/93. However, suitable personnel are retiring or being redirected towards other studies, which undermines the country's abilities to undertake such work; this loss of expertise and application needs to be viewed with considerable concern at the highest level as the situation continues to deteriorate.

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