Study on using the Monte Carlo simulation code for characterization of nuclear materials

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Abstract: Uranium is one of the most important nuclear materials (NMs) for nuclear safeguards purposes, thus the characterization of the nuclear materials (measurements of the mass content and the enrichment of 235U isotope) is very useful in fuel-fabrication plants, nuclear safeguards inspections and waste characterization at nuclear facilities. Gamma radiation detectors can be used as non-destructive assay (NDA) techniques for NMs characterization and measurements. Those detectors such as germanium detectors, NaI(Tl) detectors and Cadmium Zinc Telluride (CZT) detectors. The dependency on NMs standards which are necessary for other relative or semi-absolute methods could be eliminate during NMs measurements using absolute methods,. In the present work, Monte Carlo (MCNP) method could be used (a) as an alternative approach which was investigated, where the calibration is performed through Monte Carlo simulation (MCNP5) instead of experiment in advance, as the measurement bias was reduced to be around 5 % (b) as an absolute method to verify for the nuclear materials which used in the nuclear fuel cycle and (c) to fully simulate the experiments and consequently calibrate them mathematically in which detailed information about measuring system are available. Aim of this research is to evaluate and explore an approach to measure the enrichment of the samples using the efficiency factor method by the help of the Monte Carlo simulation code for nuclear safeguards purposes instead of depending on the standards nuclear materials which are not available for all geometries of the measured samples.

Keywords: Monte Carlo simulation; detection efficiency; nuclear and radioactive materials; nondestructive assay; gamma-ray spectrometry; enrichment

1 Introduction

One of the most widely detectors used in the field of nuclear safeguards is the high-purity germanium detectors because of their high efficiency beside high energy resolution. They are used in non-destructive assay (NDA) techniques to determine the 235U enrichment in combination with some softwares such as the multi-group analysis for uranium (MGAU). To use MGAU, reference sources are required for prior detectors calibration. Because most of the measured samples in the field can be different and since the enrichment calibration constants depends on more parameters such as collimators, container well and sample chemical composition, MCNP modeling may represent an alternative approach to experimental determination of the calibration curve and constants when there is lack of uranium reference standards.

More research studies had been done in that direction such as; efficiency transfer method using Monte Carlo code has been applied to determine the full energy peak efficiency of a gamma spectrometry system based on coaxial n-type HPGe detector for three Certificate Reference Materials. Thereafter, the calculated efficiency was used to calculate the activity concentration of the detected radionuclides in those materials. A good agreement was found between the reported and simulated activity concentration within mean deviations of 5% [1]. Also, a proposed MCNP model was applied to determine the response curve for a 152Eu source filled in matrix soil using two reference point sources (241Am, 137Cs) at five locations in front and around the detector, using the same geometry (Marinelli beaker). Thereafter, the obtained value for the efficiency was used to estimate accurately the

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activity of 137Cs on a soil samples [2]. MCNP application for modeling a semiconductor HPGe well detector was applied to assure its validation by comparison of two volumetric sources geometries, which were both simulated and measured experimentally. Both results were in agreement and the proposed application was validated [3]. The Monte Carlo method was presented and verified for the ability of to be used as a good approach for the determination of accurate full-energy peak efficiencies; as MCNP method was used for optimization the thickness of detector dead layer to obtain matching between the measured and simulated efficiency [4]. The Monte Carlo method has been concluded an efficient and powerful tool to obtain the efficiency calibration curves for two scintillation detectors; CsI(Tl) and NaI(Tl) detectors using gamma-ray energies from the terrestrial samples, in order to determine precise and accurate activity concentrations for those samples [5]. The Monte Carlo code was evaluated for calibrations in situ gamma-ray spectrometry using HPGe detectors, at five different places in Sweden to determine activity levels in soil for 137Cs from the 1986 Chernobyl accident. Moreover, MCNP-calculated efficiency calibration factors were in good agreement compared with corresponding values calculated using a more traditional semi-empirical method [6]. To develop accurate simulation model for the detector, the Monte Carlo was used to optimize the dead-layer thickness of HPGe detectors using a low-energy source (*e.g.*, Am-241) at different locations. Thereafter, the model was validated by calibrating the detection efficiency [7]. Monte Carlo simulation is very powerful and useful tool for complementing experimental calibration in low-level gamma-ray spectrometry. It can be successfully applied to estimate the correction factors for the efficiency in order to overcome for discrepancies between the measurement and simulation results [8]. One of the advantages of the Monte Carlo method is, the obtained efficiency factor by direct Monte Carlo computation depends much stronger on the parameters of detector and the cross section data of the Monte Carlo model, than the correction factors of detection efficiency. In order to obtain precise and accurate results of the Monte Carlo simulated efficiencies it is important to compare those results against experimental data for several test measurements. If some discrepancies are found, it usually can be eliminated by optimization of some detector parameters. In a recent study, strong deviations between experimental results and MCNP calculations were obtained based on the physical dimensions of the detector as provided by the manufacturer. Where smaller value for the active detector volume was detected than that stated by the manufacturer. At this value, experimental and calculated values agree within 4% over the entire energy range from 30 to 1500 **keV**. MCNP model could be more convenient and applicable for applications such as 235U enrichment determination [9]. Similar Monte Carlo analysis has been recently carried out with a germanium detector and environmental radioactive samples has indicated that Monte Carlo methods can be used for the uncertainty analysis of gamma-ray spectrometers and represent a valuable tool for the detector response curve. This consequently minimizes/eliminates the need for standard radioactive sources [10]. The results of Monte Carlo Performance drawn the ability for building models with considerable geometric and material complexity [11]. A study has been carried out to be extended for sample geometry optimization for bottle-like geometries so that gain with three detectors could reach the required value three, enhancing the sensitivity [12]. However the accurate results of MC modeling, discrepancies in the observed detection efficiency were found. This is commonly due to the non-accurate dead layer thickness of the detector which stated by the manufacturer. Hence, this thickness is adjusted in the model to obtain matching for Monte Carlo calculated efficiencies with experimental efficiencies. Thus, Monte Carlo simulation has been showed applicable model alternative method can be used to determine HPGe detector efficiency.

For DL adjustment, a valid method using experimental approach and Monte Carlo simulation has been developed to estimate thickness of the inner dead-layer of HPGe detector because of the discrepancies between simulated and experimental efficiencies. Varying the dead layer, leads to reducing the discrepancy to ≤ 3 %, also a good linear correlation was observed in the energy range of 122–1408 **keV** [13]. [14] Showed an increase of up to 7.5 **mm** in the thickness of the outer crystal dead-layer for an n-type detector leads to a good agreement between MCNP results and experimental efficiencies. In contrast with the nominal DL (0.4 **mm**) value which results in the strongest deviation from measurements. Many authors studied the DL influence on the efficiency performance of co-axial Ge detectors for medium to high gamma-ray energy range [15–18]. MCNP5-code consider the photons and electrons transport radiation in its calculations, thus F8 tally (pulse height distribution) has been used to determine the response function of NDA instruments such as HPGe detectors [7,19,20].

2 Monte Carlo calculations

2.1 Monte Carlo description

MCNP is a general Monte Carlo N-particle transport code developed by Las Alamos national laboratory [21], is commonly applied to NDA systems as modeling tool for NDA equipment [22], to optimize its performance, to predict its response in different configurations, and as a computational calibration technique [3,7,23]. Computational codes that based on MC method allow modeling of complex geometries and determination the response of an NDA instrument without the need for reference standards foe calibration. MC method obtains answers by simulating individual particles and recording some tallies of their average behavior such as tally F8 for efficiency calculations which will be described in the next paragraph.

MCNP code treats an arbitrary three-dimensional configuration of materials in geometric cells bounded by surfaces. The main features of the code include:

Nuclear data and reactions,
Source specifications,
Tallies and output,
Estimation of errors,
Variance reduction.

The MCNP code can be used for modeling the detector response, since it contains a tally, F8 (absolute full energy peak efficiency of HPGe detector), which is specific for detector pulse height determination being output of the MCNP input file. The fraction of absorbed photons in the detector active volume with certain energy represents its absolute full energy peak efficiency at that energy. The data provided by the detector's manufacturer were used to construct the MCNP input file.

An input file contains information about the problem will be created, such as:

- The geometry specification,
- The description of materials,
- The location and characteristics of the radiation source,
- The type of answers (tallies) desired, and
- Any variance reduction techniques used to improve efficiency.

That input file is subsequently read by MCNP.

MCNP uses continuous-energy nuclear and atomic data libraries. The primary sources of nuclear data can be achieved from the Evaluated Nuclear Data File (ENDF) system, the Evaluated Nuclear Data Library (ENDL) and the Activation Library (ACTL). Each data table available to MCNP is listed on a directory file, XSDIR. A specific data table could be selected through unique identifier, called ZAID. The data in the photon interaction tables allow MCNP to account for coherent and incoherent scattering, photoelectric absorption with the possibility of fluorescent emission, and pair production.

Source Specification; it is the generalized user-input source capability of MCNP allows specifying wide variety of source conditions without making a code modification. Independent probability distributions may be specified for the source variables of energy, time, position and direction, and for other parameters such as starting cell(s) or surface(s). Information about the geometrical extent of the source can also be given. Also, source variables may depend on other source variables (for example, energy as a function of angle). All input distributions could be biased.

Tallies and Output file; MCNP provides seven standard neutron tallies, six standard photon tallies, and four standard electron tallies. These basic tallies can be modified in many ways. The code has to be instructed to make various tallies related to particle current, particle flux, and energy deposition. Tallies are normalized to be per starting particle except for a few special cases with critical sources. Currents can be tallied as a function of direction across any set of surfaces, surface segments, or sum of surfaces in the problem. A pulse height tally provides the energy distribution of pulses created in a detector by radiation. Particles may be flagged when they cross specified surfaces or enter designated cells. In addition to the tally information, the output file contains tables of standard summary information that can give insight into the physics of the problem and the adequacy of the simulation.

Statistical relative error corresponding to one standard deviation is given with each tally. Estimation of Monte Carlo Errors, MCNP tallies are normalized to be per starting particle and are printed in the output file followed with the estimated relative error (R) defined to be one estimated standard deviation of the mean (S_x) divided by the estimated mean (x). For a well-behaved tally, R will be proportional to ($1/\sqrt{N}$) where N is the number of histories. Thus, to obtain R, the total number of histories must increase fourfold. For a poorly behaved tally, R may increase as the number of histories increases. It is extremely important to note that these confidence statements refer only to the precision of the Monte Carlo calculation itself and not to the accuracy of the result compared to the true physical value.

Errors and Uncertainty Estimation a measurement is an attempt to determine the value of a certain parameter or quantity. Anyone attempting a measurement should keep in mind the following two axioms regarding the result of measurement:

Axiom1 No measurement yields a result without an error.

Axiom 2 The result of a measurement is almost worthless unless the error associated with that result is also reported.

2.2 Monte Carlo detector model

The HPGe detector of high resolution allows for several energies with very proximate values to be discriminated. Also, the use of an HPGe detector for gamma spectrometry required a more precise determination of the detection efficiency and the response curve. Based on the energy of photons The MCNP5 version of the code has been used for modeling the detector response, since it contains a tally, F8 (absolute full energy peak efficiency of HPGe detector), which is specific for detector pulse height determination. The fraction of gamma-rays with certain energy which absorbed in the detector active volume represents its absolute full energy peak efficiency at that energy.

2.3 Dead layer (DL) optimization

Because of some discrepancies/dissimilarities between the simulation and the measurement results as in Fig.1a, therefore, the dead-layer thickness should be modified in the initial simulation model. By increasing the dead-layer thickness step by step in the simulation process, it was possible to obtain acceptable agreement with the measured data, as shown in Fig.1b. Also, Fig.2 shows that the thickness of the inner dead-layer was estimated to be approximately 2.14 **mm**, twice the nominal value stated by the manufacture.

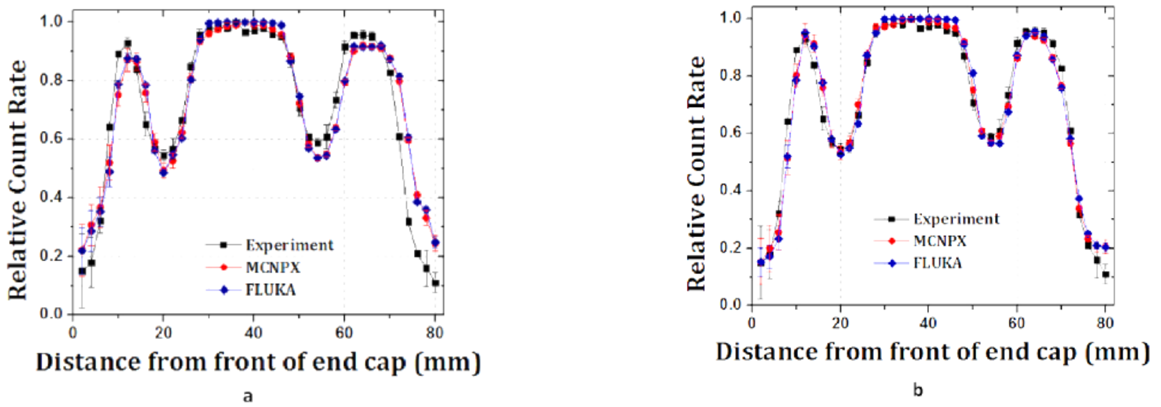

Fig.1 Comparison of simulated results against experimental data for; (a) primary model and (b) corrected model.

Experimental case study for DL optimization

Varying the DL step by step in the simulation model has shown that increasing the inactive (dead) layer thickness decreases the calculated efficiency. Finally, results indicated that a good agreement between simulated and measured efficiencies is obtained of ratio ≈1.07 when the deviation from the experimental efficiency was less than 3 % as shown in Fig.3 using a modified value for DL thickness approximately six (2.45 **mm**) in comparison with (0.389 **mm**) by the detector manufacturer. This reveals that the MCNP procedure is more effective as a useful approach in building simulation model for re-characterization the HPGe aged detector.

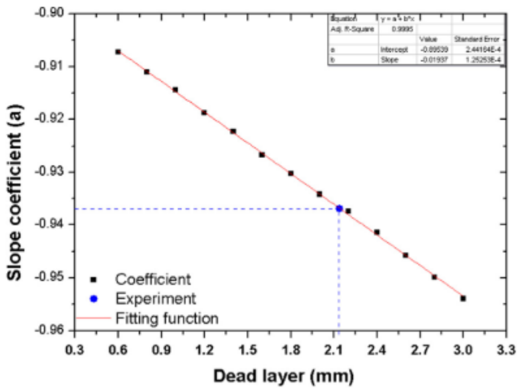

Fig.2 Fitting of slope coefficient of efficiency curve versus thickness of the inner dead-layer [13].

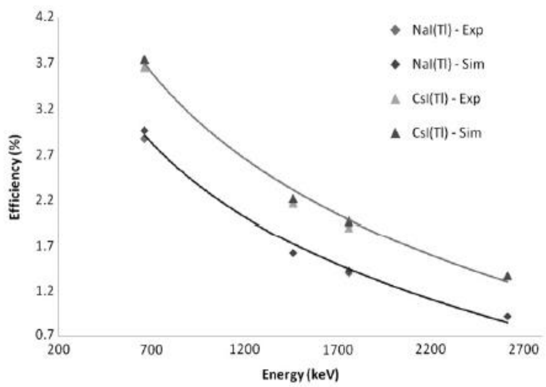

Fig.4 Full-energy peak efficiencies curves of 3″×3″ CsI(Tl) and NaI(Tl) detectors using a cylindrical soil sample (IAEA-375) [5].

3 Validations of Monte Carlo results

3.1 Comparison the simulated efficiency with the experimental efficiency

High-purity germanium (HPGe) detectors are widely used in gamma spectrometers, mainly because of their high resolution, and the possibility of application in radioisotopes analysis/ characterization of the radioactive and nuclear material samples. To obtain accurate results, detection efficiency of the HPGe detector should be precisely known. Experimental method to determine efficiency requires standard samples, which must have the same geometry as the measured samples. This may be difficult to provide all the necessary standard sources for all samples.

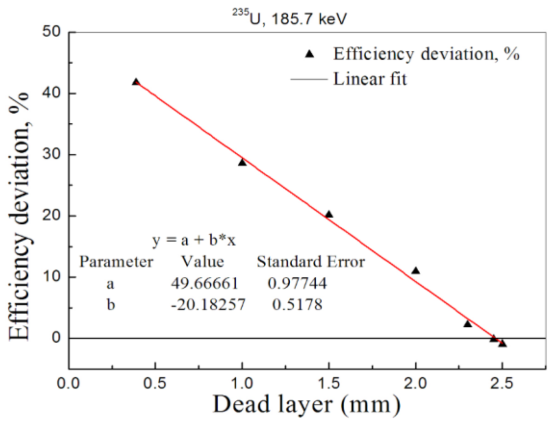

Fig.3 Deviation for both, simulated and experimental efficiencies for natural uranium sample.

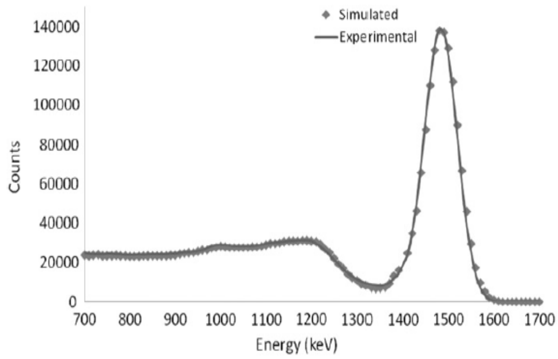

Fig.5 Simulated spectrum of cascade decay processes of 208Tl [5].

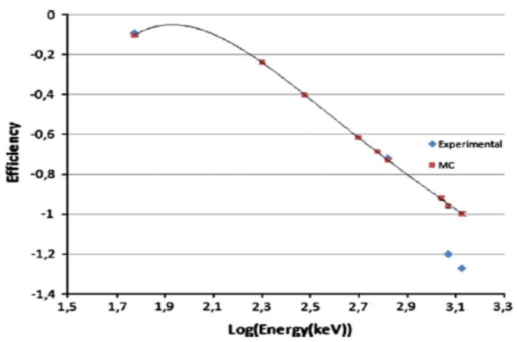

Fig.6 Efficiency curves for small plastic vessel inside the detector's well. Both uncertainties, for Monte Carlo and experimental values, were kept under 0.5% [3].

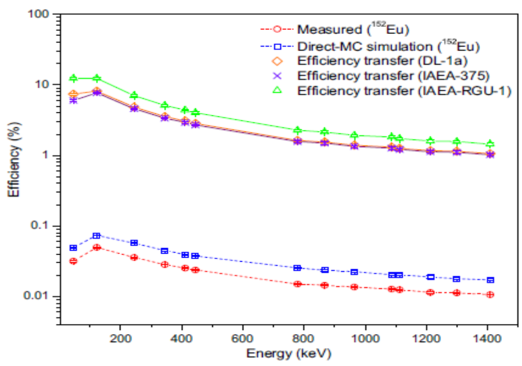

Fig.7 Measured and computed photo-peak efficiency. The dotted lines correspond to the experimental and the MC calculated efficiency using the nominal parameters of the detector. The solid lines represent the efficiency after application the efficiency transfer [24].

Figure 4 shows the matching/ agreement between simulated and experimental full-energy-peak efficiencies calibration curves of the 300″ ×300″ CsI(Tl) and NaI(Tl) detectors using a cylindrical reference material Soil-375 as described above. The spectrum is shown in Fig.5 proved that the overall agreement is good. The Monte Carlo method concluded a useful and simple tool for the full-energy efficiency calibration curve in routine measurement laboratories, by avoiding the difficulties and time consuming of preparing standard sources and eliminating the need for the usual experimental calibrations for many different sample configurations.

Figure 6 shows the efficiency curves for both simulated and calibrated/experimental data for the small plastic vessel. The counting uncertainties for this case were kept under 0.5% – too small to be seen on the graphic.

Figure 7 shows the comparisons between the measured peak and MC calculated efficiencies using manufacturer's data for 152Eu reference source; as well as the efficiency curves for the three CRM after transfer calculation. The discrepancies obtained between the experimental and calculated efficiencies for the reference source even exceed the 50%. This can be overcome by application the efficiency transfer method to correct the geometric differences.

4 Application of Monte Carlo for nuclear materials characterization

In this section the Monte Carlo simulation code was tested and verified for nuclear materials characterization through the calculations of the enrichment of the investigated samples as shown in table 1. First, with the help of Monte Carlo code (MCNP) the efficiency factor was acquired by simulation [25]. The efficiency factor method depends strongly on the modeling accuracy of the measurement setup[12].

For the gamma ray line 185.71 **keV** of 235U

$$A_5 = \frac{I_5(E_i)}{B_5 f_\varepsilon} \qquad (1)$$

Where A5 is the 235U activity, the efficiency factor $f_\varepsilon = \varepsilon_5(E_i)\tau_5(E_i)\Omega_5$, for the experiment and the gamma ray energy *Ei*=185.71 **keV**. In MCNP5 model, the photons and electrons transport are considered, so F8 tally (pulse height distribution) has been used for photons and electrons. Since the experiment setup is modeled with the well-known samples and gamma spectrometer, the efficiency factor of the full energy peak of 185.71 **keV** can be given by the Monte Carlo simulation. Hence the absolute 235U activity is obtained with the energy peak intensity $I_5(E_i)$, which had been extracted from gamma spectrum using MGAU code then got M5. Consequently the enrichment of 235U of the samples can be calculated according to the equation [26]

$$\eta_5 = \frac{M_5}{M_T} \qquad (2)$$

Where M5 is the mass content of 235U isotope and MT is mass content of uranium element for sample. For the samples of uranium isotopes not in equilibrium with its daughters, the enrichment meter method can be employed to measure the 235U enrichment. The bias of the measurement results is too large to lead a reliable judgment. With the help of the efficiency factor given by Monte Carlo simulation the result is more accurate and well improved with low uncertainty ~7 % in comparison with the enrichment meter ~17 % as shown in table 1. The conclusion acquired from this work that the SEF proposed acceptable alternative method with precise results than MGAU method. Since it can eliminates the standard reference sources required for calibration, it can be used in the routine inspection of the nuclear material. In the future work, MC simulation can be used to study how the precise characteristics of the sample and detector parameters provided by the manufacture can affect the measured result.

Results from Monte Carlo-based calibration methods were found better than measurement uncertainties to estimate 235U enrichment as shown in table 2. The drawn conclusion that, it is safe to apply Monte Carlo to estimate more complex geometries that is difficult to be handled by the semi-empirical calibration method.

Table 1 Comparison the measured ^{235}U enrichment using MGAU with those based on MCNP efficiency factor.

Sample ID.	η_5 with simulated f_ε (wt. %)	Rel. uncertainty (%)	η_5 enrichment mode (wt. %)	Rel. uncertainty (%)
SN	0.729 ± 0.049	6.7	0.812 ± 0.135	16.6
SL	2.989 ± 0.198	6.6	2.856 ± 0.458	16.0

Table 2 Show some previous results of acceptable uncertainty using efficiency factor based on MCNP5.

Sample	**Aim**	**Spectrometer**	**Method**		**Ref**
			MGAU (%)	**MCNP (%)**	
Uranium samples	234U/235U Activity ratio (%)	coaxial HPGe detector (Ortec)	31.1 ± 50	25.43±7.9	[27]
UO2	235U-Enrichment	HPGE planar detector(Canberra)	1.49±34.99	1.39±2.46 1.38	[28]
certified reference NM(N,D,EU)	235U-Enrichment	HPGE detector(Canberra)	0.942 ± 11.4 2.779 ± 12.9 4.473 ± 15.8	0.7119 ±0.0006 2.9492 ± 0.0021 4.4623 ± 0.0032	[29]
radioactive waste	235U-Enrichment	HPGE detector(Canberra)	0.715 ± 3.916	0.692±0.9	[30]

5 Conclusions

The use of the MCNP Monte Carlo computer code for the response function calculations for different sample geometries can be eliminate preparing several standard sources, especially in the lack of proper isotopes, saving both time, financial resources and effort, supporting a precise and powerful tool in the field of nuclear and environmental materials verifications. A good agreement between experimental and simulation detector's response function was obtained; the maximum deviation is less than 5% for different isotopes. The methodology for determining the dead layers of HPGe well detector by Monte Carlo simulation proved to be effective and can be applied, which plays an important role in the determination of detector's response curves.

Nomenclature

MCNP: Monte Carlo
MGAU: Multi-group analysis for uranium
NDA: Non-destructive assay
DL: Dead later
NMs: Nuclear materials

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