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Designing a nuclear battery based on the Mo-99 radioactive source soluble in water and aqua regia in order to use in early tests

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Abstract Today, millions of electrocommunication, electric, medical, and industrial devices use battery. Batteries with long life and high energy density seem to be essential in medical, military, oil and mining, aerospace areas as well as conditions in which access is difficult and in situations where replacement or recharging of battery is costly. In this regard, the use of radiation energy resulting from radioactive materials and its conversion to electric energy can be effective in making batteries. In the present study, various Mo-99 radioisotope values with a half-life of 65.98 h were used as a soluble radioactive source in two materials of water and aqua regia. Then, by comparing the results of the Monte Carlo simulations program MCNPX for these two solutions, it was found that when the water is used instead of aqua regia (for idealization), the values of the superficial current of electrons, the volumetric flux of electrons, and the deposited energy in the volume containing the radioactive solution increased by 10.80, 4.10, and 13.80%, respectively. Also, the short-circuit current and energy conversion efficiency of this battery with a concentration of 0.01 molar, Mo-99 dissolved in the aqua regia are 0.79 μA and 16.47%, respectively.

Keywords Nuclear battery · Radioactive solution · MCNPX code · Mo-99

1 Introduction

Batteries are considered as one of the most important human inventions. Among the advantages of nuclear exchangers are their high energy density and durability [1]. The attempts for prolonging the lifetime and power density of a nuclear exchanger began in the early 1900s after the discovery of radiation, which has been continuing into the present [2]. First, batteries were only considered as a portable source. However, today millions of electrocommunication, medical, and industrial devices have to use batteries. Furthermore, lightweight and not very costly batteries especially in electronic pieces are felt needed with difficult accessibility conditions and nondischargeability [3].

Generally, in cases when we seek to enhance the lifetime of a battery or when the charging and replacement of batteries are difficult, nuclear exchangers are a very good option.

The electric energy of a nuclear battery is supplied by the decay energy of radioactive materials under a suitable conversion process. In designing a nuclear battery, selection of a proper and practical radioisotope plays a significant role. The output power of a battery, considering the area for which it is used, is an important factor in selecting a radioactive source. Other factors including accessibility and conservation are also important.

Radioisotopes should not generate large amounts of gamma ray, neutron radiation, or penetrating radiation. On the other hand, their half-life should be long enough to supply a relatively uniform energy for a logical period of time.

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Theoretically, any type of radiation (alpha, beta, gamma, and X-ray) can cause energy generation. However, beta-emitting radioisotopes are more widely used [4, 5].

There are several suitable beta emitters which are listed in Table 1.

The purpose of the present work is to design a nuclear battery with high energy density. The radioactive source used in this battery is soluble in water and aqua regia. In this regard, the Mo-99 radioisotope was used with a half-life of 65.98 h. For various values of the Mo-99 radioisotope, the superficial current of electrons, the volumetric flux of electrons, and the deposited energy in the active volume of the battery are obtained in two solutions of water and aqua regia by the Monte Carlo method.

2 Materials and methods

2.1 Mo-99

Mo-99 is produced by neutron capture during the ^{235}U(n, f)^{99}Mo reaction in reactors. Its half-life is 65.98 h [11]. Table 2 shows the most important beta radiations of Mo-99.

2.2 Calculating the final continuous beta spectrum of Mo-99

The probability density is calculated for each of the nine lines of beta energy with using the golden rule of Fermi (Eq. 1) and the computer code in Fortran language [13]

$$\lambda(W)\mathrm{d}W = \mathrm{const}(W_0 - W)^2 \rho W F(Z,W) C(Z,W)\mathrm{d}W, \quad (1)$$

where $W = \frac{E_\beta}{m_0c^2} = 1 + \frac{Te}{m_0c^2}$ and $W_0 = \frac{E_\beta^{\max}}{m_0c^2} = 1 + \frac{Te^{\max}}{m_0c^2}$.

The parameter F (Z, W) is the Fermi function [14].

$$F(Z,W) = 4L(Z,W)\left(2R\sqrt{W^2-1}\right)^{2(\gamma-1)} \mathrm{e}^{\pi\eta} \frac{|\Gamma(\gamma+i\eta)|^2}{|\Gamma(2\gamma+1)|^2}, \quad (2)$$

where $\eta = \pm Z\alpha W/\sqrt{W^2-1}$ for β^-/β^+ decays, α is the fine structure constant, and $\gamma = \sqrt{1-(\alpha Z)^2}$.

The function L (Z, W) is defined by Eq. (3) [15].

$$L(Z,W) = 1 + \frac{13}{60}(\alpha Z)^2 - \frac{\alpha ZRW(41-26\gamma)}{15(2\gamma-1)} - \frac{\alpha ZR\gamma(17-2\gamma)}{30W(2\gamma-1)}. \quad (3)$$

The parameter C (Z, W) is the weak interaction finite-size effect [16].

$$C(Z,W) = 1 + C_0 + C_1 W + C_2 W^2, \quad (4)$$

where

$$C_0 = -\frac{233}{630}(\alpha Z)^2 - \frac{(W_0R)^2}{5} + \frac{2}{35}W_0R\alpha Z,$$
$$C_1 = -\frac{21}{35}R\alpha Z + \frac{4}{9}W_0R^2,$$
$$C_2 = -\frac{4}{9}R^2.$$

Then, the graphs of each of the nine lines were plotted. Figure 1, respectively, from top to bottom, shows the energy lines 0.3537, 0.2287, 0.2159, 0.1856, and 0.1589 MeV. Figure 2 represents the energy lines 1.2151, 0.8487, 0.6863, and 0.4372 MeV.

Equation (5) is used to obtain the final continuous beta spectrum of Mo-99 [17].

$$\lambda_T = \sum_{i=1} I_i \lambda_i. \quad (5)$$

The parameter I_i is the energy intensity for each.

Table 2 The most important beta radiations for Mo-99 [12]

Maximum energy (MeV)	Probability × 100
158.9	0.0021
185.6	0.0019
215.9	0.111
228.7	0.021
353.7	0.146
437.2	16.4
686.3	0.057
848.7	1.16
1215.1	82.2

Table 1 Characteristics of suitable beta emitters

Nuclide	Half-life	Activity (Ci/g)	Maximum energy (keV)	Mean energy (keV)	Refs.
^{3}H	12.3 years	9664	18.6	5.7	[6]
^{147}Pm	2.6 years	800	225	73	[7, 21]
^{90}Sr	28.8 years	116	546	196	[8, 21]
^{63}Ni	100 years	57	65.9	17	[9, 21]
^{99}Mo	65.9 h	486,486	1214.3	389	[10]

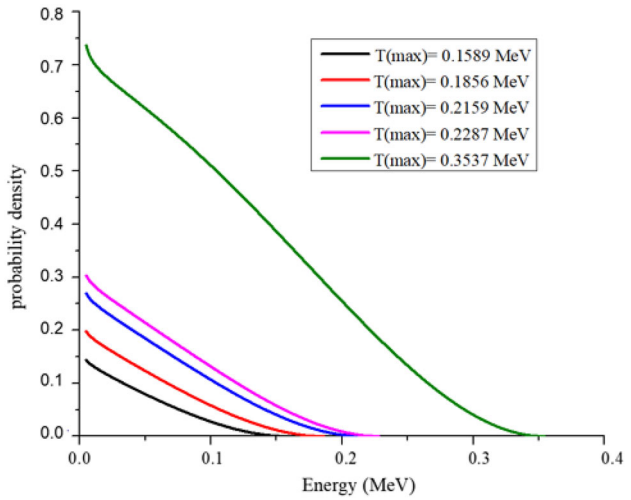

Fig. 1 Probability density plotted versus energy for beta lines 0.3537, 0.2287, 0.2159, 0.1856, and 0.1589 MeV using a Fortran code (Color online)

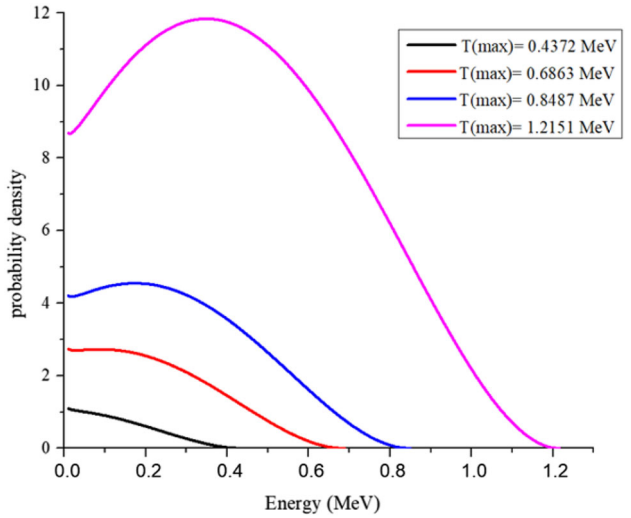

Fig. 2 Probability density plotted versus energy for beta lines 1.2151, 0.8487, 0.6863, and 0.4372 MeV using a Fortran code (Color online)

The final continuous beta spectrum of Mo-99 is shown in Fig. 3.

2.3 Monte Carlo simulation

The MCNP Monte Carlo code has been used to determine the amount of radioisotope radiation flux in different regions of material. In Sect. 2.2, the final continuous beta spectrum of Mo-99 was calculated which used this spectrum to define the source in the MCNPX code.

The studied geometry is demonstrated in Fig. 4. In this structure, the device includes a radioactive solution, located between two metal cylinders. The diameter of the internal cylinder is 0.05 cm and its height is 0.2 cm,

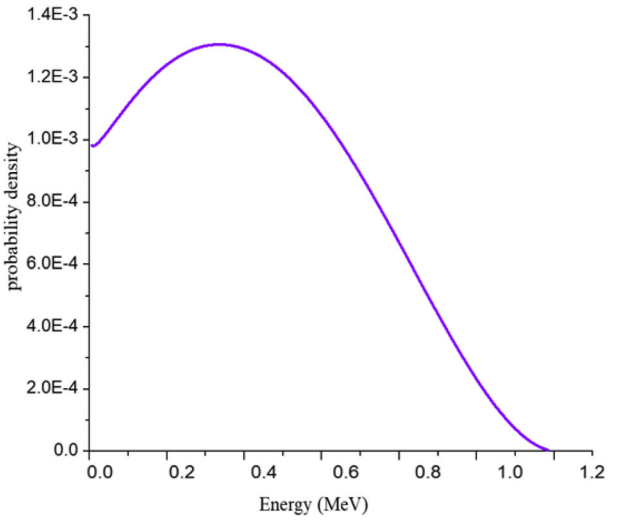

Fig. 3 Final continuous beta spectrum of Mo-99, which consists of nine energy lines of beta

starting from 0.011 cm in the point of origin along z axis direction. The cylinder diameter is 0.25 cm, the body thickness is 0.01 cm, and it is made of titanium. Figure 4a is a two-dimensional view, and Fig. 4b is a three-dimensional view of the nuclear battery studied.

According to Fig. 4b, the particle from the radioactive solution collides with atoms and other molecules to create negative and positive ions, which move to opposite electrodes and generate current.

In the nuclear battery studied by simulation, the material environment is designed in such a way that in addition to the electrons emitted from the radioactive solution, the secondary electrons produced by the interaction of beta particles with this environment also reach the positive electrode, and this will increase the current.

So in this battery, the current generated is due to two sources:

1. Electrons emitted from the radioactive solution.
2. Secondary electrons produced by the interaction of beta particles (electrons) with the material environment.

First, different values of the Mo-99 radioisotope were considered as soluble in water and aqua regia. Aqua regia is an acid which dissolves all metals except for titanium, iridium, tantalum, and osmium. Then, the superficial current of electrons, the volumetric flux of electrons, and the deposited energy in the volume containing the radioactive solution are obtained. Since the results obtained from the MCNPX code are for a particle, in each step the activity of the source is multiplied in the results. When the Mo-99 radioisotope is considered to be soluble in water and aqua regia at concentrations of 0.1, 0.01, 0.001, and 0.0001 M,

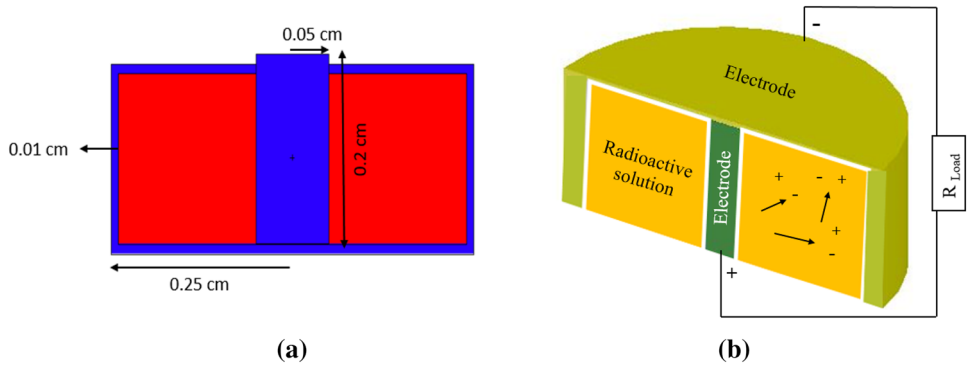

Fig. 4 **a** Two-dimensional view of the nuclear battery, **b** three-dimensional view of the nuclear battery (Color online)

the activity of radioactive solution in this battery is 183, 17.70, 1.85, and 0.185 Ci, respectively.

Based on Fig. 5, due to an increased number of interactions, the area under the curve related to the Mo-99 aqua regia solution is lower than that of Mo-99 that is assumed as water-soluble. To determine the magnitude of this difference, we calculated the relative difference in each stage. Further, for more accurate investigation, we plotted all of the four diagrams on a single diagram, as shown in Fig. 6.

According to Fig. 6, the use of water instead of aqua regia increases the superficial current of electrons by at least 9.4% and a maximum of 13.6%.

In each part of Fig. 5, the area under the curve represents the total current in each state. By obtaining this area and Eq. (6), the relative difference of the superficial current

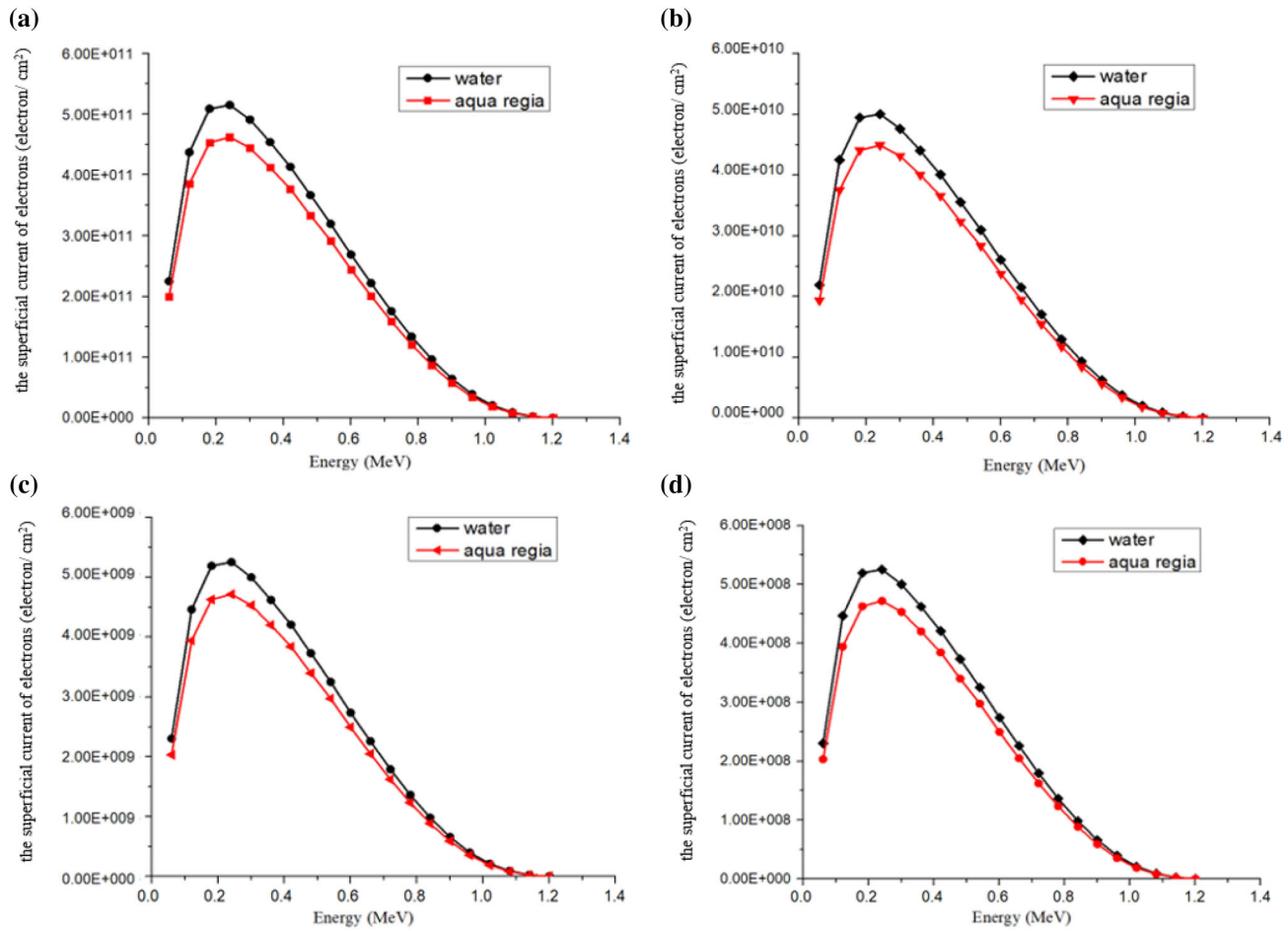

Fig. 5 Comparison of the superficial current of electrons in two solutions of water and aqua regia. The concentration of Mo-99 in the forms **a**, **b**, **c** and **d** is 0.1, 0.01, 0.001, and 0.0001 M, respectively (Color online)

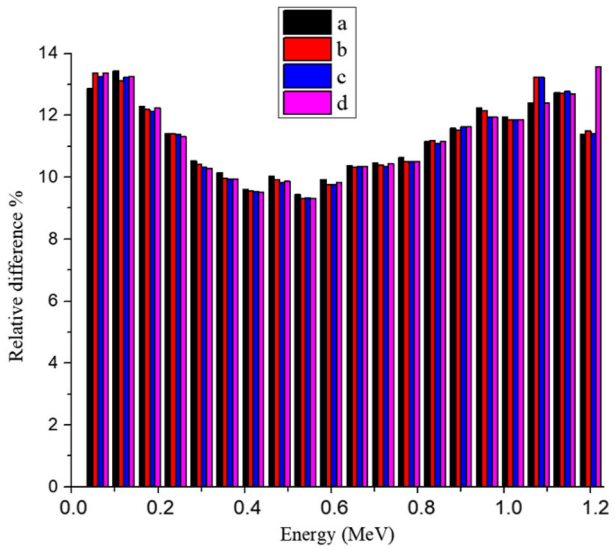

Fig. 6 Comparison of relative difference of the superficial current of electrons in two solutions of water and aqua regia. The concentration of Mo-99 in the a, b, c and d is 0.1, 0.01, 0.001, and 0.0001 M, respectively (Color online)

of electrons is calculated for various concentrations of Mo-99

$$D = \frac{\phi_{\mathrm{w}} - \phi_{\mathrm{a}}}{\phi_{\mathrm{a}}}, \tag{6}$$

where ϕ_{w} and ϕ_{a} are the superficial current of electrons in two solutions of water and aqua regia with different concentrations of Mo-99, respectively. D is the relative difference between these two states.

Based on the values in Table 3, the relative difference in the superficial current of electrons in each state is about 10.8% relative to the other state.

Also, the same stages were taken for the volumetric flux of electrons and the deposited energy in the volume containing the radioactive solution. The results are shown in Figs. 7 and 8.

Then, the relative difference was calculated to determine the exact amount of the differences in each of the cases in a–d in Figs. 7 and 8. Furthermore, for more accurate investigation, data plotted from all of the four diagrams were placed on a single diagram. The calculation results are shown in Figs. 9 and 10.

According to Figs. 9 and 10, whenever water is used instead of aqua regia, the values of the volumetric flux of electrons and the deposited energy in the volume containing the radioactive solution increased at least 2.50%, 12.40% with a maximum of 29.50%, 42.7%, respectively.

In each part of Figs. 7 and 8, the area under the curve represents the total of the volumetric flux of electrons and the total of the deposited energy in the volume containing the radioactive solution in each state, respectively. By obtaining this area and Eqs. (7) and (8), the relative difference of the volumetric flux of electrons and the deposited energy inside the active battery volume is calculated for various concentrations of Mo-99 in two solutions of water and aqua regia

$$G = \frac{\phi_i - \phi_j}{\phi_j} \tag{7}$$

$$J = \frac{D_m - D_n}{D_n}, \tag{8}$$

where ϕ_i is the volumetric flux of electrons in the solution of water with different concentrations of Mo-99; ϕ_j is the volumetric flux of electrons in the solution of aqua regia with different concentrations of Mo-99; D_m is the deposited energy in the solution of water with different concentrations of Mo-99; D_n is the deposited energy in the solution of aqua regia with different concentrations of Mo-99.

Also, G is the relative difference of the volumetric flux of electrons and J is the relative difference of the deposited energy inside the active battery volume in two solutions of water and aqua regia for various concentrations of Mo-99.

According to the results in Table 4, the relative differences in the volumetric flux of electrons and the deposited energy inside the active battery volume in each state are about 4.10% and 13.80% relative to the other state, respectively.

Table 3 Calculations of the relative difference of the superficial current of electrons in two solutions of water and aqua regia for different concentrations of Mo-99

Concentration of Mo-99 to mol	Solvent	Total of the superficial current of electrons	D (%)
0.1	Water	4.763E+12	10.95
0.1	Aqua regia	4.293E+12	
0.01	Water	4.627E+11	10.88
0.01	Aqua regia	4.173E+11	
0.001	Water	4.856E+10	10.84
0.001	Aqua regia	4.381E+10	
0.0001	Water	4.857E+9	10.87
0.0001	Aqua regia	4.381E+9	

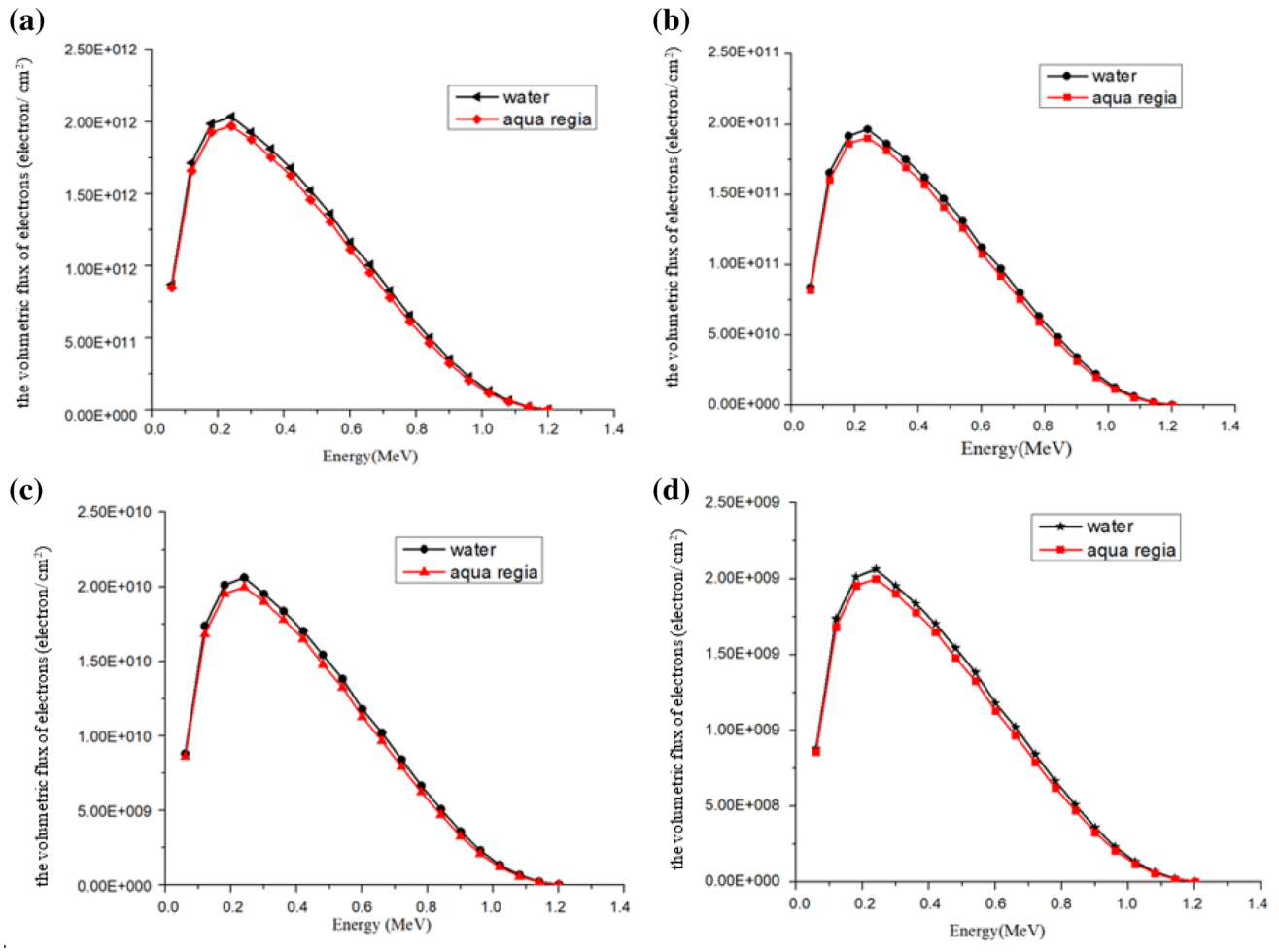

Fig. 7 Comparison of the volumetric flux of electrons in two solutions of water and aqua regia. The concentration of Mo-99 in the forms **a**, **b**, **c** and **d** is 0.1, 0.01, 0.001, and 0.0001 M, respectively

The short-circuit current of this battery for various concentrations of Mo-99 is given in Table 5.

By observing the results in Table 5, the relative difference of the short-circuit current is about 10.80% between the state when aqua regia was used and the case when the solvent was assumed as water.

Also, with the use of Eq. (9), energy conversion efficiency of this nuclear battery is calculated for various concentrations of ^{99}Mo, as shown in Table 6 [18].

$$\eta = \frac{P_{\mathrm{m}}}{P_{\mathrm{RD}}} \times 100\% = \frac{0.25 U_{\mathrm{oc}} I_{\mathrm{sc}}}{A \varepsilon_{\mathrm{avg}}} \times 100\% \tag{9}$$

In this equation, P_{m} is the maximum output power, P_{RD} is the power of radioactive decay, U_{OC} is the open-circuit voltage, Isc is the short-circuit current, A is the radioactive material activity in Becquerel, and $\varepsilon_{\mathrm{avg}}$ is the average energy of emitted radioactive particles in Joules [19].

$$U_{\mathrm{oc}} = I_{\mathrm{sc}} \times R_{\mathrm{leak}}. \tag{10}$$

This dependence R_{leak} in Ohm versus inter-electrode distance in millimeters can be approximated with suitable accuracy by the equation below [20].

$$R_{\mathrm{leak}} = 1.025 \times 10^{12} \times d^{0.83}. \tag{11}$$

Here, d is the distance between the two electrodes.

3 Conclusion

Based on Figs. 5, 7 and 8, which relate to the superficial current of electrons, the volumetric flux of electrons, and the deposited energy in the volume containing the radioactive solution, the use of water instead of aqua regia increased 10.80%, 4.10%, and 13.80% in each case, respectively.

Also, the short-circuit current increased by 10.80% when the solvent is water. Therefore, in nuclear batteries with liquid radioactive material, it is possible to use water instead of aqua regia for initialization and initial

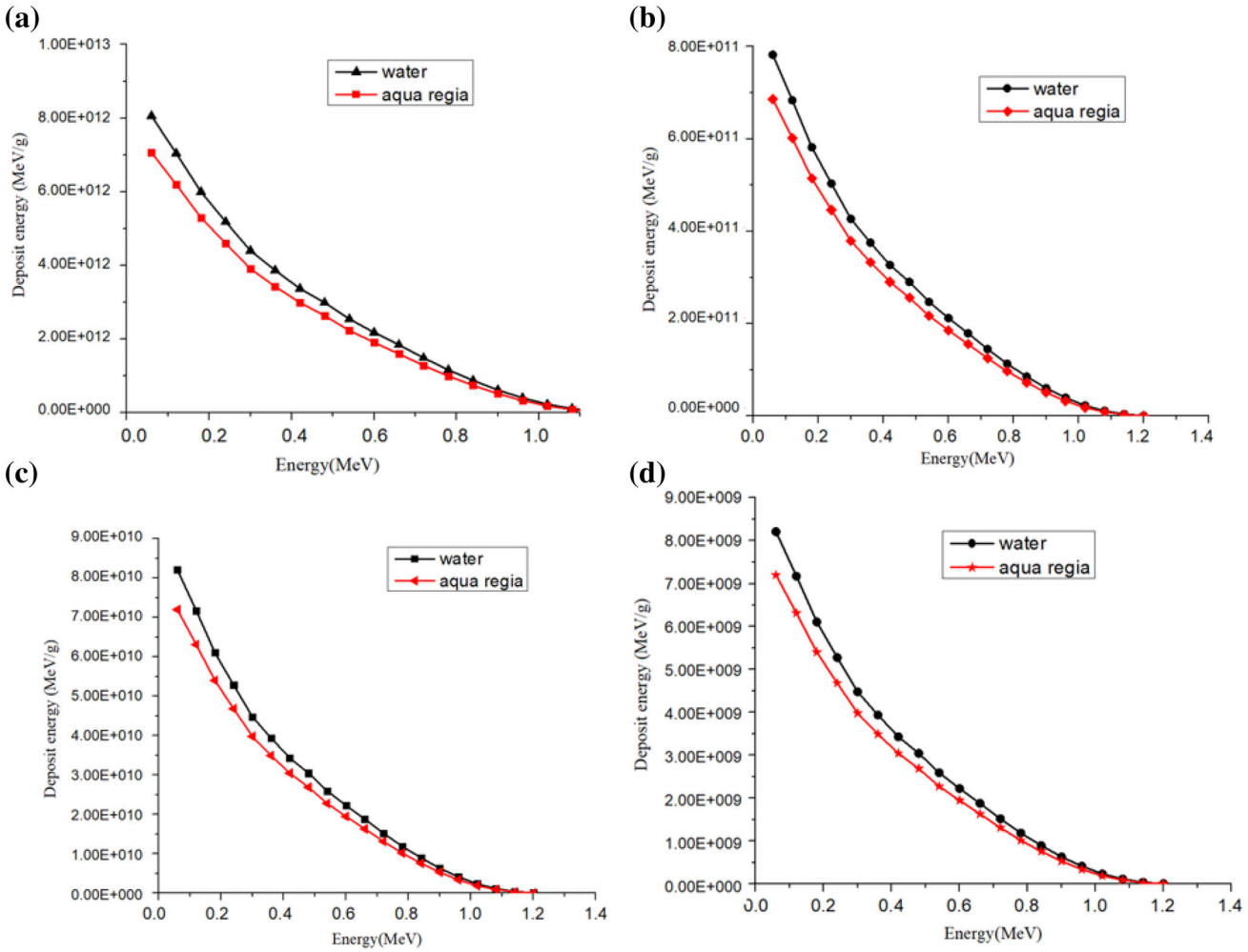

Fig. 8 Comparison of the deposited energy inside the active battery volume in two solutions of water and aqua regia. The concentration of Mo-99 in the forms **a**, **b**, **c** and **d** is 0.1, 0.01, 0.001, and 0.0001 M, respectively

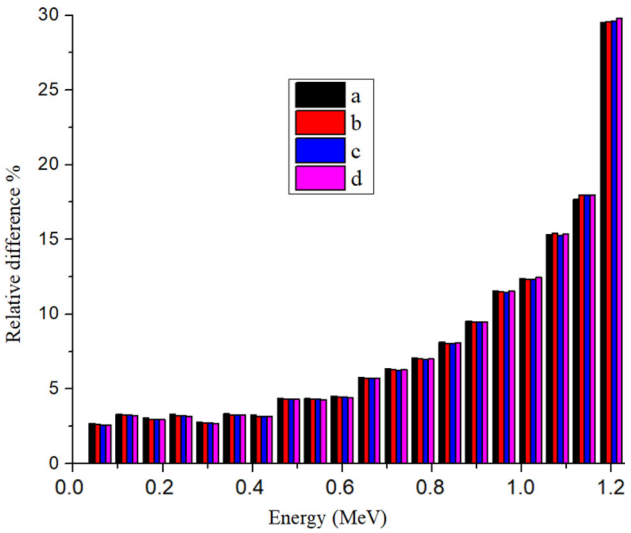

Fig. 9 Comparison of the relative difference of the volumetric flux of electrons in two solutions of water and aqua regia. The concentration of Mo-99 in the a, b, c and d is 0.1, 0.01, 0.001, and 0.0001 M, respectively (Color online)

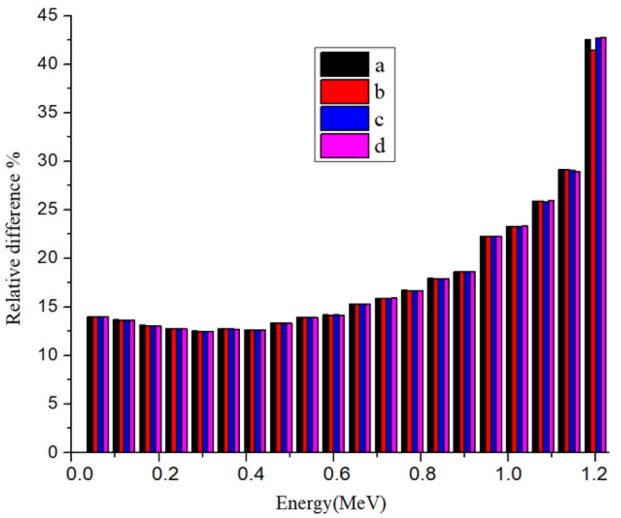

Fig. 10 Comparison of the relative difference of the deposited energy inside the active battery volume in two solutions of water and aqua regia. The concentration of Mo-99 in the a, b, c and d is 0.1, 0.01, 0.001, and 0.0001 M, respectively (Color online)

Table 4 Calculation of the relative difference of the volumetric flux of electrons and the deposited energy inside the active battery volume in two solutions of water and aqua regia for different concentrations of Mo-99

Concentration of Mo-99 to mol	Solvent	Total of the volumetric flux of electrons	Total of the deposited energy	G (%)	J (%)
0.1	Water	1.988E+11	5.245E+11	4.074	13.824
0.1	Aqua regia	1.907E+11	4.608E+11		
0.01	Water	1.922E+10	5.091E+10	4.173	13.791
0.01	Aqua regia	1.845E+10	4.474E+10		
0.001	Water	2.016E+09	5.342E+09	4.132	13.781
0.001	Aqua regia	1.936E+09	4.695E+09		
0.0001	Water	2.016E+08	5.342E+08	4.132	13.781
0.0001	Aqua regia	1.936E+08	4.695E+08		

Table 5 Calculation of the short-circuit current of the nuclear battery in two solutions of water and aqua regia for different concentrations of Mo-99

Concentration of Mo-99 to mol	0.1	0.01	0.01	0.001
Short-circuit current (water solvent) (μA)	7.621E−01	7.403E−02	7.770E−03	7.771E−04
Short-circuit current (aqua regia solvent) (μA)	6.869E−01	6.677E−02	7.010E−03	7.010E−04
Relative difference of the short-circuit current (%)	10.95	10.87	10.84	10.86

Table 6 Calculation of the energy conversion efficiency of the nuclear battery in two solutions of water and aqua regia for different concentrations of ^{99}Mo

Concentration of ^{99}Mo to mol	0.1	0.01	0.01	0.001
The efficiency of the nuclear battery (water solvent) (%)	2.028E+01	1.978	2.085E−01	2.086E−02
The efficiency of the nuclear battery (aqua regia solvent) (%)	1.647E+01	1.609	1.697E−01	1.697E−02

experiments. But, the correction factor must be taken into account in the obtained results. In the present work, the radioisotope of Mo-99 was used. Considering that half-life of ^{99}Mo is short, but due to its high activity it can generate a lot of current. As well as, it is not expensive and it is suitable for making such batteries. Further, radioisotopes that have a longer half-life and their beta energy spectrum similar to the beta energy spectrum of Mo-99 can be used.

References

1. T. Alam, M. Pierson, M. Prelas, Beta particle transport and its impact on betavoltaic battery modeling. Appl. Radiat. Isot. **130**, 80–89 (2017). https://doi.org/10.1016/j.apradiso.2017.09.009
2. M. Prelas, M. Boraas, F. De La Torre Aguilar et al., *Nuclear Batteries and Radioisotopes* (Springer, Columbia, 2016), p. 1
3. D. Dunn-Rankin, E.M. Leal, D.C. Walther, Personal power systems. Prog. Energy Combust. **31**, 422–465 (2005). https://doi.org/10.1016/j.pecs.2005.04.001
4. C. Honsberg, A. Doolittle, M. Allen, et al., GaN betavoltaic energy converters, *Paper Presented at the Thirty-first IEEE Photovoltaic Specialists Conference* (Lake Buena Vista, USA 3–7 Jan. 2005)
5. M. Schaldach, S. Furman (eds.), *Advances in Pacemaker Technology* (Springer, Berlin, 1975)
6. T.E. Adams, S.T. Revankar, Research, development and evaluation capabilities for betavoltaic power sources, *Paper Presented at 122nd ASEE Annual Conference and Exposition* (Seattle, 14–17 June 2015)
7. M. Prelas, M. Boraas, F. De La Torre Aguilar et al., *Nuclear Batteries and Radioisotopes* (Springer, Columbia, 2016), p. 5
8. K. Oh, M. Prelas, J.B. Rothenberger et al., Theoretical maximum efficiencies of optimized slab and spherical betavoltaic systems utilizing sulfur-35, strontium-90, and yttrium-90. Nucl. Technol. **179**, 234–242 (2012). https://doi.org/10.13182/NT12-A14095
9. S. Theirrattanakul, M. Prelas, A methodology for efficiency optimization of betavoltaic cell design using an isotropic planar source having an energy dependent beta particle distribution. Appl. Radiat. Isot. **127**, 41–46 (2017). https://doi.org/10.1016/j.apradiso.2017.05.005
10. R. Kowalsky, *Technetium Radiopharmaceutical Chemistry* (The University of New Mexico. Health Sciences Center, 2006), p. 7

11. S. Baba, N. Baba, H. Natsume, Half-lives of some fission product nuclides. J. Inorg. Nucl. **33**, 589–595 (1971). https://doi.org/10.1016/0022-1902(71)80409-8
12. I. Angeli, K.P. Marinova, Table of experimental nuclear ground state charge radii: an update. At. Data Nucl. Data Tables **1**, 69–95 (2003). https://doi.org/10.1016/j.adt.2011.12.006
13. S.M. Wong, *Introductory Nuclear Physics* (Wiley, London, 1999), p. 368
14. P. Huber, Determination of antineutrino spectra from nuclear reactors. Phys. Rev. C **84**, 024617 (2011). https://doi.org/10.1103/PhysRevC.84.024617
15. D.H. Wilkinson, Evaluation of beta-decay part II finite mass and size effects. Nucl. Instrum. Methods A **290**, 509–515 (1990). https://doi.org/10.1016/0168-9002(90)90570-V
16. D.H. Wilkinson, Evaluation of beta-decay part V the Z-independent outer radiative corrections for allowed decay. Nucl. Instrum. Methods A **365**, 497–507 (1995). https://doi.org/10.1016/0168-9002(95)00495-5
17. M. Shahrabia, H. Tavakoli-Anbaranb, calculating dosimetry parameters in brachiotherapy using the continuous beta spectrum of Sm-153 in the Monte Carlo simulation approach. Eur. Phys. J. Plus **130**, 1–8 (2015). https://doi.org/10.1140/epjp/i2015-15035-3
18. A. Kavetsky, G. Yakubova, M. Sychov, Tritium-charged capacitor. Nucl. Sci. Eng. **159**, 321–329 (2008). https://doi.org/10.13182/NSE159-321
19. A. Kavetskiy, G. Yakubova, Q. Lin et al., Promethium-147 capacitor. Appl. Radiat. Isot. **67**, 1057–1062 (2009). https://doi.org/10.1016/j.apradiso.2009.02.084
20. G.N. Yakubova, Dissertation (Department of Nuclear, Plasma and Radiological Engineering University of Illinois at Urbana-Champaign, 2010)
21. S. Kumar, Atomic batteries: energy from radioactivity. e-print arXiv:1511.07427 (2015)